Optimal control methodologies for the optimisation of maintenance scheduling and production in processes using decaying catalysts

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I would like to dedicate this thesis to my loving parents ...

Declaration

I hereby declare that except where specific reference is made to the work of others, the contents of this dissertation are original and have not been submitted in whole or in part for consideration for any other degree or qualification in this, or any other university. This dissertation is my own work and contains nothing which is the outcome of work done in collaboration with others, except as specified in the text and Acknowledgements. This dissertation contains fewer than 65,000 words including appendices, bibliography, footnotes, tables and equations and has fewer than 150 figures.

Saidarshan Adloor
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Abstract

Optimal control methodologies for the optimisation of maintenance scheduling and production in processes using decaying catalysts

In this thesis, optimal control methodologies are developed for solving problems involving the optimisation of maintenance scheduling and production in processes using decaying catalysts. Previously, such problems were solved using a category of methods which involve making decisions of discrete as well as continuous nature, called mixed-integer optimisation techniques. However, these techniques are combinatorial in nature and can solve differential equations only by approximations as collections of steady state equality constraints, and such features can cause these techniques difficulties in obtaining optimal and accurate solutions for these problems. The goal behind developing optimal control methodologies is to effectively solve these problems while overcoming the drawbacks that mixed-integer optimisation techniques face or would face in solving these problems.

First, an optimal control methodology is developed to optimise maintenance scheduling and production in a process containing a reactor using decaying catalysts. This methodology involves using a multistage mixed-integer optimal control problem (MSMIOCP) formulation and obtaining solutions as a standard nonlinear optimisation problem, without using mixed-integer optimisation techniques. Two different solution implementations are required, each which has its own relative advantages. The methodology using the second procedure is particularly successful in effectively obtaining solutions within the stipulated tolerances. Further, the methodology possesses features of robustness because it enables a relatively small problem size, reliability because it solves differential equations using state-of-the-art integrators, and efficiency because it is not combinatorial in nature. These features indicate the methodology's success in overcoming the drawbacks of using mixed-integer optimisation

techniques to solve this problem.

Next, the abovementioned methodology is extended to form an optimal control methodology to optimise maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts. This methodology, when applied to a case study of such a process, is also able to effectively obtain solutions within the stipulated tolerances. Further, the solutions obtained, once again, possess features of robustness, reliability and efficiency, which indicate that the methodology can overcome the drawbacks that mixed-integer optimisation techniques would face, if used to solve such problems.

And lastly, an optimal control methodology is developed for considering uncertainties in kinetic parameters in the optimisation of maintenance scheduling and production in a process containing a reactor using decaying catalysts. The methodology involves using a multiple scenario approach to consider parametric uncertainties and formulating a stochastic MSMIOCP, which is solved as a standard nonlinear optimisation problem as per the previously developed procedure. The results obtained provide insights into the effects of parametric uncertainties and the number of scenarios generated on the optimal operations, and indicate that the methodology is capable of solving this problem. Further, the robust, reliable and efficient nature of the results obtained suggest that the methodology can overcome the disadvantages that mixed-integer methods would introduce in the conventional methodologies, if such methodologies are used to solve such problems.

Saidarshan Adloor

List of peer-reviewed publications

This work has resulted in the following peer-reviewed publications:

Adloor, S.D., Pons, T. and Vassiliadis, V.S. (2020). An optimal control approach to scheduling and production in a process using decaying catalysts. *Computers & Chemical Engineering*, 135:106743.

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Further, in the work leading to the preparation of this thesis, the following short note was published, which, however, does not form a part of this thesis:

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Nomenclature

Acronyms / Abbreviations

AD	Automatic differentiation
COIN-OR	Computational Infrastructure for Operations Research
CPU	Central processing unit
CSTR	Continuous stirred tank reactor
DAE	Differential-algebraic equation
DICOPT	Discrete and Continuous Optimiser
DLTLBO	Diversity Learning Teaching Learning Based Optimisation
GAMS	General Algebraic Modelling System
GBD	Generalised Benders Decomposition
GB	Gigabyte
GHz	Gigahertz
IPOPT	Interior point optimiser
MIDO	Mixed-integer dynamic optimisation
MILP	Mixed-integer linear programming
MINLP	Mixed-integer nonlinear programming
MSMIOCP	Multistage mixed-integer optimal control problem
NLP	Nonlinear programming

OA	Outer Approximation
OCP	Optimal control problem
ODE	Ordinary differential equation
RAM	Random access memory
RSD	Relative standard deviation
SQP	Sequential Quadratic Programming
SUNDIALS	Suite of Nonlinear and Differential-Algebraic Equation Solvers

Roman Symbols

a	The activity of a catalyst in a generic chemical reaction
Ar	The pre-exponential factor in the Arrhenius expression of the rate of the product formation reaction in the industrial process, in all problems considered
$\overline{Ar}$	Average of the range of values considered for the pre-exponential factor in the Arrhenius expression for the product formation reaction of the industrial process in the problem involving uncertainty in kinetic parameters
$base\_cof$	The cost, before inflation, of raw material per unit volume in the industrial process, in all problems considered
$base\_crc$	The cost, before inflation, of replacement of a catalyst load in a reactor in the industrial process, in all problems considered
$base\_pen$	The penalty cost, before inflation, of unmet demand per unit of product in the industrial process, in all problems considered
$base\_psp$	The sales price per unit of product, before inflation, in the industrial process, in all problems considered
c	Set of constraints in a generic OCP formulation
c_1	Coefficient of linear controls in the set of constraints in a generic MSMIOCP formulation

The symbols may contain additional subscripts depending on the problem involved

c_2	Component of constraints independent of linear controls in a generic MSMIOCP formulation
cat_act	The activity of a catalyst in the reactor in problems considering a single reactor industrial process
cat_act1	The activity of a catalyst in reactor 1 in the problem considering an industrial process with parallel lines of reactors
cat_act2	The activity of a catalyst in reactor 2 in the problem considering an industrial process with parallel lines of reactors
cat_act3	The activity of a catalyst in reactor 3 in the problem considering an industrial process with parallel lines of reactors
cat_act4	The activity of a catalyst in reactor 4 in the problem considering an industrial process with parallel lines of reactors
cat_age	The age of a catalyst in the reactor in the problem considering a single reactor industrial process
cat_age1	The age of a catalyst in reactor 1 in the problem considering an industrial process with parallel lines of reactors
cat_age2	The age of a catalyst in reactor 2 in the problem considering an industrial process with parallel lines of reactors
cat_age3	The age of a catalyst in reactor 3 in the problem considering an industrial process with parallel lines of reactors
cat_age4	The age of a catalyst in reactor 4 in the problem considering an industrial process with parallel lines of reactors
$\overline{C}$	Average of a set of constraints over all scenarios in a generic stochastic MSMIOCP formulation
c_m	The set of inequality constraints in a generic mixed-integer programming problem
Cnc	The reactant concentration in a generic chemical reaction

The symbols may contain additional subscripts depending on the problem involved

cof	The cost of raw material per unit volume in the industrial process, in all problems considered
cR	The concentration of reactant at the exit of the reactor in problems considering a single reactor industrial process
$CR0$	The concentration of reactant entering a reactor in the industrial process, in all problems considered
$cR1$	The concentration of reactant at the exit of reactor 1 in the problem considering an industrial process with parallel lines of reactors
$cR2$	The concentration of reactant at the exit of reactor 2 in the problem considering an industrial process with parallel lines of reactors
$cR3$	The concentration of reactant at the exit of reactor 3 in the problem considering an industrial process with parallel lines of reactors
$cR4$	The concentration of reactant at the exit of reactor 4 in the problem considering an industrial process with parallel lines of reactors
crc	The cost of replacement of a catalyst load in a reactor in the industrial process, in all problems considered
cum_inc	The cumulative inventory cost of the industrial process, in all problems considered
ΔG	The change in total free energy in a generic chemical reaction
$demand$	The demand for the product in the industrial process, in all problems considered
E	The activation energy in a generic chemical reaction
Ea	The activation energy in the Arrhenius expression of the rate of the product formation reaction in the industrial process, in all problems considered
$\overline{Ea}$	Average of the range of values considered for the activation energy in the Arrhenius expression for the product formation reaction of the industrial process in the problem involving uncertainty in kinetic parameters

The symbols may contain additional subscripts depending on the problem involved

F	Label for a problem in the series of problems solved when applying the penalty term homotopy technique to a generic MSMIOCP formulation
f	Set of differential equations in a generic OCP formulation
f_1	Coefficient of linear controls in the set of differential equations in a generic MSMIOCP formulation
f_2	Component of differential equations independent of linear controls in a generic MSMIOCP formulation
ffr	Feed flow rate to the reactor in problems considering a single reactor industrial process
$ffr1$	Feed flow rate to reactor 1 in the problem considering an industrial process with parallel lines of reactors
$ffr2$	Feed flow rate to reactor 2 in the problem considering an industrial process with parallel lines of reactors
$ffr3$	Feed flow rate to reactor 3 in the problem considering an industrial process with parallel lines of reactors
$ffr4$	Feed flow rate to reactor 4 in the problem considering an industrial process with parallel lines of reactors
Fs	Label for a problem in the series of problems solved when applying the penalty term homotopy technique to a generic stochastic MSMIOCP formulation
FU	The upper bound for the feed flow rate in the industrial process, in all problems considered
g	Set of algebraic equations in a generic OCP formulation
G, Gp	Label for a problem in the series of problems solved when applying the penalty term homotopy technique to the MSMIOCP formulation of the industrial process
g_1	Coefficient of linear controls in the set of algebraic equations in a generic MSMIOCP formulation

The symbols may contain additional subscripts depending on the problem involved

g_2	Component of algebraic equations independent of linear controls in a generic MSMIOCP formulation
g_m	The set of equality constraints in a generic mixed-integer programming problem
GRS	The gross revenue from product sales in the industrial process, in all problems considered
Gs	Label for a problem in the series of problems solved when applying the penalty term homotopy technique to the stochastic MSMIOCP formulation of the industrial process
H	The Hamiltonian in the generic MSMIOCP formulation
h	Set of junction conditions in a generic OCP formulation
h_1	Coefficient of linear controls in the set of junction conditions in a generic MSMIOCP formulation
h_2	Component of junction conditions independent of linear controls in a generic MSMIOCP formulation
icf	The inventory cost factor in the industrial process, in all problems considered
$inflation$	The rate of annual inflation in the industrial process, in all problems considered
inl	The inventory level of the industrial process, in all problems considered
J	Generic junction condition
Kd	The rate constant of deactivation of a catalyst in the industrial process, in all problems considered
$\overline{Kd}$	Average of the range of values considered for the catalyst deactivation rate constant in the industrial process in the problem involving uncertainty in kinetic parameters
Kr	The rate constant of the rate of product formation reaction in the problem considering a single reactor industrial process

The symbols may contain additional subscripts depending on the problem involved

$Kr1$	The rate constant of the rate of product formation reaction in reactor 1 in the problem considering an industrial process with parallel lines of reactors
$Kr2$	The rate constant of the rate of product formation reaction in reactor 2 in the problem considering an industrial process with parallel lines of reactors
$Kr3$	The rate constant of the rate of product formation reaction in reactor 3 in the problem considering an industrial process with parallel lines of reactors
$Kr4$	The rate constant of the rate of product formation reaction in reactor 4 in the problem considering an industrial process with parallel lines of reactors
L	Continuous performance index in a generic OCP formulation
L_1	Coefficient of linear controls in the continuous performance index in a generic MSMIOCP formulation
L_2	Continuous performance index independent of linear controls in a generic MSMIOCP formulation
L_m	The objective function in a generic mixed-integer programming problem
M, Mp, Ms	Weight term in the objective function in the series of problems solved when using the penalty term homotopy technique
max_cat_age	The maximum allowable age of a catalyst in a reactor in the problems involving industrial processes with a single reactor and parallel lines of reactors
min_cat_act	The minimum allowable activity of a catalyst in a reactor in an industrial process in the problem involving uncertainties in kinetic parameters
n	The maximum number of catalyst replacements allowed for a reactor in the industrial process, in all problems considered
NC	The net costs of the industrial process in the problems involving industrial processes with a single reactor and parallel lines of reactors

The symbols may contain additional subscripts depending on the problem involved

NC_{st}	The net costs in the industrial process in the problem involving uncertainties in kinetic parameters
NM	The total number of months in the time horizon of the industrial process, in all problems considered
NP	Total number of stages in a generic MSMIOCP formulation
$NPUD$	The net penalty for unmet demand of product in the industrial process, in all problems considered
NS	Number of scenarios generated in the stochastic MSMIOCP formulation of the industrial process
pen	The penalty cost of unmet demand per unit of product in the industrial process, in all problems considered
Pr	The pressure in a generic chemical reaction
psp	The sales price per unit of product in the industrial process, in all problems considered
Q	Product in the chemical reaction of the industrial process, in all problems considered
R	Reactant in the chemical reaction of the industrial process, in all problems considered
r	The rate of a generic chemical reaction at any time
r_0	The initial rate of a generic chemical reaction
$r_1(a)$	A function dependent only on the catalyst activity
rD	Label for the expression of the rate deactivation of a catalyst
R_g	The universal gas constant
rR	Label for the expression of the rate of the product formation reaction
$sales$	Quantity of product sales in the industrial process, in all problems considered

The symbols may contain additional subscripts depending on the problem involved

SN	Number of scenarios generated in a generic stochastic MSMIOCP formulation
$start\_cat\_act$	The activity of a fresh catalyst in the industrial process, in all problems considered
T	Temperature of the reactor in problems considering a single reactor industrial process
t	Time
t_0	Initial time
$T1$	Temperature of reactor 1 in the problem considering an industrial process with parallel lines of reactors
$T2$	Temperature of reactor 2 in the problem considering an industrial process with parallel lines of reactors
$T3$	Temperature of reactor 3 in the problem considering an industrial process with parallel lines of reactors
$T4$	Temperature of reactor 4 in the problem considering an industrial process with parallel lines of reactors
$TCCC$	The total cost of catalyst changeovers in the industrial process, in all problems considered
Te	The temperature in a generic chemical reaction
t_F	Final time
TFC	The total cost of feed of raw material in the industrial process, in all problems considered
TIC	The total inventory costs in the industrial process, in all problems considered
TL	The lower bound for the temperature of a reactor in the industrial process, in all problems considered

The symbols may contain additional subscripts depending on the problem involved

TU	The upper bound for the temperature of a reactor in the industrial process, in all problems considered
u	Set of binary controls in a generic OCP formulation
$\mathbb{U}$	The domain of the set of integer decision variables in a generic mixed-integer programming problem
u_m	The set of integer decision variables in a generic mixed-integer programming problem
$unmet\_demand$	The unmet demand of product in the industrial process, in all problems considered
V	The volume of a reactor in problems considering a single reactor industrial process
v	Set of continuous controls in a generic OCP formulation
$\mathbb{V}$	The domain of the set of continuous decision variables in a generic mixed-integer programming problem
v_m	The set of continuous decision variables in a generic mixed-integer programming problem
W	Performance index in a generic OCP formulation
w	Complete set of controls in a generic OCP formulation
W_{st}	Performance index in a generic stochastic MSMIOCP formulation
X	Reactant in a generic chemical reaction
x	Set of differential state variables in a generic OCP formulation
x_0	Set of initial conditions for the differential state variables in a generic OCP formulation
Y	Reactant in a generic chemical reaction
y	Action to schedule catalyst changeover in the reactor in problems considering a single reactor industrial process

$y1$	Action to schedule catalyst changeover in reactor 1 in the problem considering an industrial process with parallel lines of reactors
$y2$	Action to schedule catalyst changeover in reactor 2 in the problem considering an industrial process with parallel lines of reactors
$y3$	Action to schedule catalyst changeover in reactor 3 in the problem considering an industrial process with parallel lines of reactors
$y4$	Action to schedule catalyst changeover in reactor 4 in the problem considering an industrial process with parallel lines of reactors
Z	Product in a generic chemical reaction
z	Set of algebraic state variables in a generic OCP formulation

Greek Symbols

β	Euler-Lagrange multipliers corresponding to the junction conditions in the generic MSMIOCP formulation
Λ	Euler-Lagrange multipliers corresponding to the differential equations in the generic MSMIOCP formulation
λ	Euler-Lagrange multipliers corresponding to the algebraic equations in the generic MSMIOCP formulation
μ	Euler-Lagrange multipliers corresponding to the constraints in the generic MSMIOCP formulation
ϕ	Point performance index in a generic OCP formulation
ϕ_1	Coefficient of linear controls in the point performance index in a generic MSMIOCP formulation
ϕ_2	Point performance index independent of linear controls in a generic MSMIOCP formulation
$\mathscr{V}$	Domain of continuous controls in a generic OCP formulation

Superscripts

The symbols may contain additional subscripts depending on the problem involved

(p) Property at stage p in a generic MSMIOCP formulation

Subscripts

k Property at iteration k in the series of problems solved when using the penalty term homotopy technique

p Property at stage p in a generic MSMIOCP formulation

pr Indicates that the term belongs to the problem involving the industrial process containing parallel lines of reactors

s Property in scenario s in the generic stochastic MSMIOCP formulation

un Indicates that the term belongs to the problem involving uncertainties in kinetic parameters

Other Symbols

$end\_var(i,j)$ Terminal conditions for variable, var, for week j of month i of the time horizon of the industrial process, in all problems considered

$init\_var(i,j)$ Initial conditions for variable, var, for week j of month i of the time horizon of the industrial process, in all problems considered

$var(i)$ Value of variable, var, in month i of the time horizon of the industrial process, in all problems considered

$var(i,j)$ Value of variable, var, in week j of month i of the time horizon of the industrial process, in all problems considered

$\widetilde{var^s}$ Value of var, which can be an uncertain parameter or state variable, in scenario, s, in the stochastic MSMIOCP formulation of the industrial process, in the problem involving uncertainties in kinetic parameters

Chapter 1

Introduction

A catalyst is a substance that increases the rate of a chemical reaction without itself being consumed in the reaction. This happens by virtue of the catalyst providing an alternative pathway for the reaction to occur, which involves a different transition state that requires a lower activation energy. The catalyst, however, does not alter the change in total free energy between the reactants and products (Figure 1.1).

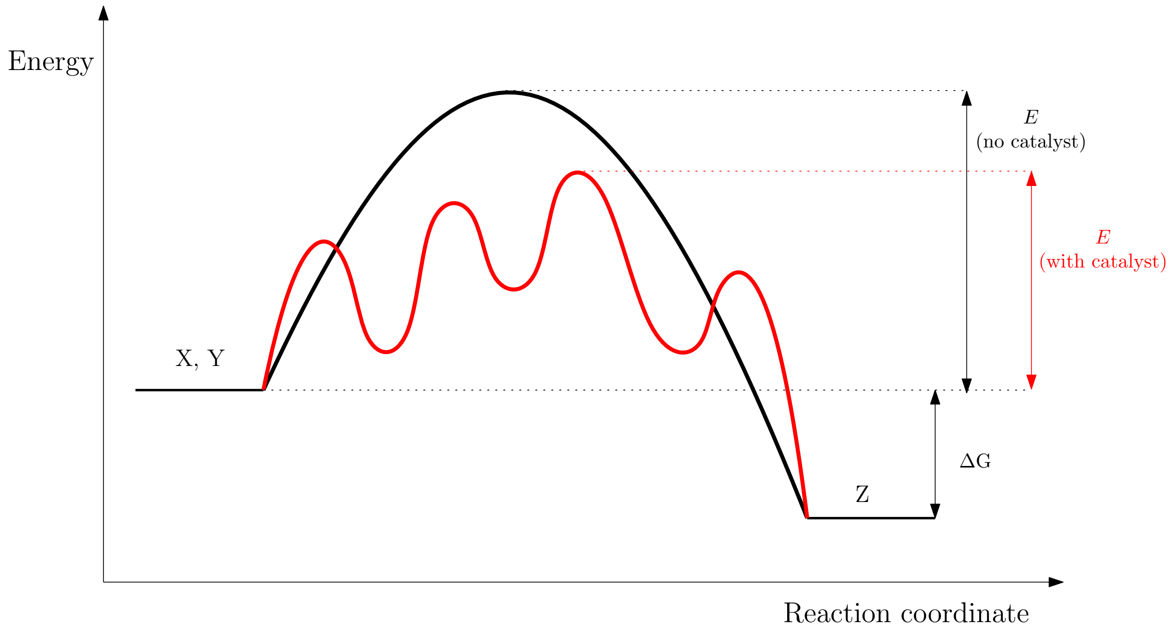


Fig. 1.1 Schematics of the reaction pathway for the reaction $X+Y \rightarrow Z$, with and without a catalyst. As can be seen, the activation energy, E, for the pathway using a catalyst is lower compared to that without the catalyst. However, the change in total free energy, ΔG, between the reactants and products is not altered if a catalyst is used.

As such, catalysis is used extensively across a wide range of industries. For example, in the bulk chemical industry, an iron catalyst is used in the Haber process to produce ammonia. Examples of catalysis in the energy processing sector include the use of zeolites in catalytic cracking, platinum in catalytic reforming and nickel in steam reforming. An example in food processing is the use of a nickel catalyst for the hydrogenation of fats to produce margarine.

The above examples constitute a very small subset of the true scale of catalysis in today's world. It is estimated that catalysts are involved at some stage in the process of manufacture of about 90% of all commercially produced chemical products (R&D Magazine, 2005). In 2019, the global demand of catalysts was estimated to be around US$ 33.9 billion and it is expected to grow at an annual rate of 4.4% from 2020 to 2027 (Grand View Research, 2020).

However, the ability of a catalyst to increase the rate of a reaction tends to decrease over time. This is expressed by saying the catalyst undergoes "deactivation" or "decay" with time. Time scales for catalyst deactivation vary considerably, from the order of seconds in some cases to the order to years in others. But the fact remains that catalyst deactivation is inevitable (Bartholomew, 2001).

Given the indispensable nature of catalysts in today's world, catalyst deactivation represents major problems. In an industrial process using a decaying catalyst, the space-time yield of the desired product decreases with the time-on-stream, thereby causing a lower production rate. Thus, if there is a continued demand for the product, the falling production rate could lead to an inability to meet this demand and hence, a loss in revenue.

In order to restore process performance and improve on low product yields, a maintenance action is required which involves shutting down the reactor using the decayed catalyst and replacing the decayed catalyst with a fresh catalyst that has full activity. Such a maintenance action is called a catalyst replacement or a catalyst changeover operation. While this maintenance action does improve product yield, there are negative impacts associated with this operation as well, such as a loss of production time because of the reactor being shut down and the energy and labour costs to replace the catalyst.

Thus, catalyst deactivation can cause significant negative economic effects to industrial processes using catalysts. While catalyst deactivation may be unavoidable, the industries can seek to minimise the associated negative effects. For this, however, a complex set of issues have to be addressed.

While frequently replacing catalyst loads can result in a high production rate, it also leads to large maintenance costs and loss in production occurring from the process shut-down for catalyst changeovers. On the other hand, a low frequency of catalyst replacements, while involving small maintenance costs and long production times, can result in a low production rate. Thus, there is a trade-off to be balanced between attaining high production rates, and having low maintenance costs and effective production times. In order to balance this trade-off optimally, a maintenance schedule is needed that specifies the optimum number of catalyst loads to use and the optimal time for each catalyst replacement.

Further, during the times when the catalyst is in operation, there is a question regarding how the operational settings such as the flow rate to and temperature of the reactor are to be managed in order to produce maximum product yield under the conditions created by catalyst deactivation. In addition, if there is a time-varying demand for the product, the product inventory levels and sales have to be managed efficiently to meet this demand. The inventory levels should be such that an adequate quantity of sales can occur to effectively meet the product demand, especially during times of catalyst replacement when there is no production occurring, while at the same time excessive storage costs are avoided. In essence, an optimal production plan is needed that specifies the operating conditions of the reactor, and the product inventory levels and sales to meet demand, and this should be obtained in tandem with the optimal maintenance schedule.

Thus, in order to minimise the negative effects of catalyst deactivation, an optimal maintenance schedule for catalyst replacements and an optimal production plan have to be obtained in an integrated manner. However, this requires solving a highly challenging modelling and optimisation problem. As such, only a limited set of publications have attempted to solve this problem and these works have obtained solutions using methodologies belonging to a category called mixed-integer optimisation techniques. However, mixed-integer optimisation techniques suffer from major drawbacks which make these techniques susceptible to face convergence difficulties and obtain inaccurate solutions.

A methodology is needed that can overcome the disadvantages of the mixed-integer methods by producing solutions to this problem that are robust, reliable and efficient in comparison to those techniques. In this thesis, a novel methodology is proposed which solves this problem as an optimal control problem and exhibits qualities suggestive of the aforementioned desired characteristics. This thesis also includes extensions of this optimal control methodology to address other related aspects of this problem, which have not been examined by existing literature.

This chapter forms the introduction of the thesis. The components of this chapter are as follows. Section 1.1 gives a brief overview of the most common mechanisms of catalyst deactivation. Section 1.2 provides details of a quantitative description of catalyst deactivation, that is relevant to this thesis. In Section 1.3, a literary review is presented of the publications concerning optimisation under catalyst deactivation. In Section 1.4 the disadvantages of using mixed-integer optimisation techniques to solve the problems under consideration are highlighted. This in turn leads to the objectives of the thesis, which are presented in Section 1.5. The chapter concludes with a brief overview of the thesis in Section 1.6.

1.1 Mechanisms of catalyst deactivation

The part of the catalyst, which may be an atom or a surface, at which one or all steps of the involved reaction occurs is called the "active" site of the catalyst. Catalyst deactivation happens when these active sites lose functionality to enable the reaction or are prevented from coming in contact with the reactants. This can happen by a variety of mechanisms. The most common mechanisms of catalyst deactivation include coking, poisoning and sintering and these are briefly outlined next.

1.1.1 Coking or Fouling

Coking usually occurs in catalytic processes involving hydrocarbons, such as catalytic cracking and catalytic reforming, when side reactions occur that form carbonaceous residues which physically deposit on the catalyst surface. For example, by accumulating within the pores of the catalyst, the residues effectively block the pores and thereby prevent the transport of reactants and products within the pores (Figure 1.2). This leads to the deactivation of the catalyst in that the number of active sites of the catalyst available for a reaction is lowered.

The chemical nature of the carbonaceous deposits depend on the mode of formation, conditions of pressure and temperature, the catalyst age and the nature of the feed and products. In general, the deposition can be distinguished as carbon and coke. Carbon is considered to be the product of disproportionation of carbon monoxide ($2CO \rightarrow C + CO_2$) while coke is formed by the cracking or condensation of hydrocarbons (Forzatti and Lietti, 1999).

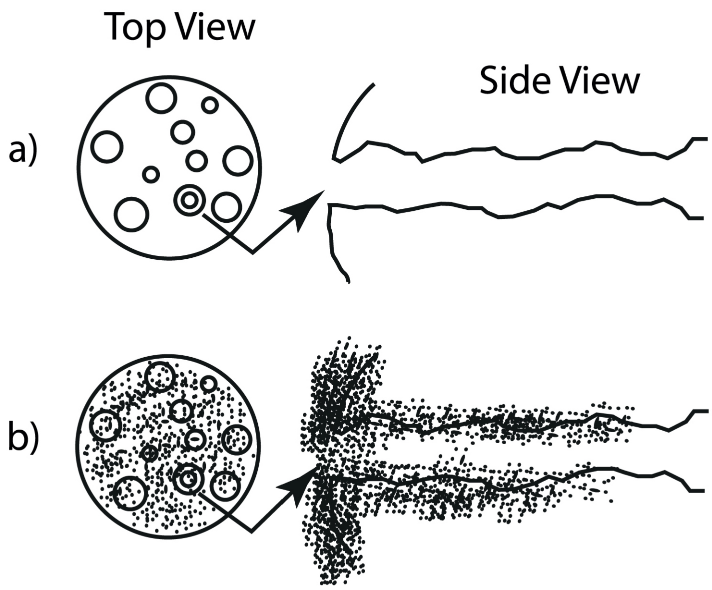


Fig. 1.2 Schematic of the top views and side views of the catalyst surface: (a) Before coking (b) After coking (Fogler et al., 1999).

Apart from coking, masking is another type of fouling wherein substances physically deposit on the outer surface of a catalyst, thereby preventing its active sites from coming in contact with reactants. An example of where masking occurs is in hydrotreating processes where metals in the feedstock, such as Nickel and Vanadium, deposit on the catalyst external surface. Another example is the deposition of Silicon compounds in automobile exhaust converters (Forzatti and Lietti, 1999).

1.1.2 Poisoning

The poisoning method of catalyst deactivation occurs when some molecules become strongly chemisorbed on the active sites of the catalyst and so, reduce the number of active sites available for the reaction to take place. In addition, the distance the reactants have to diffuse through to undergo a reaction increases (Figure 1.3).

The poisoning molecule may be an impurity in the feed stream or even a reactant or product of the reaction. Some examples of poisons on industrial catalysts are given in Table 1.1. These poisons have unoccupied orbitals, unshared electrons or multiple bonds, which serve as the driving force for bond formation with the active sites (Forzatti and Lietti, 1999).

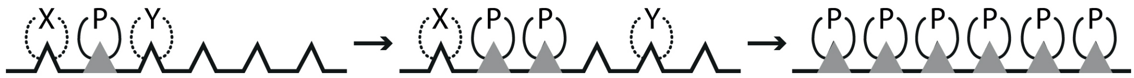


Fig. 1.3 Schematic of poisoning of a catalyst. The poisoning molecule P prevents either of the reactants, X or Y, from adsorbing onto the particular active site. X and Y are prevented in coming to close proximity of each other and so do not undergo a reaction (Fogler et al., 1999).

Table 1.1 A table of poisons of industrial catalysts (Forzatti and Lietti, 1999).

Process	**Catalyst**	**Poisons**
Catalytic cracking	Zeolites, SiO_2 - Al_2O_3	Organic bases, NH_3, Sodium
Ammonia synthesis (Haber process)	Iron (Fe)	CO, CO_2, H_2O
Catalytic converters in automobiles	Pt, Pd	Pb, P, Zn
Steam reforming	Ni / Al_2O_3	H_2S, HCl, As
Oxidation	V_2O_5	As

1.1.3 Sintering

Sintering is the process of compaction to form a solid mass of material by using heat or pressure, without melting the material. As such, for chemical reactions involving solid catalysts at high temperatures, sintering can lead to a loss of active surface area of the catalyst and hence, a loss of catalyst activity. For example, sintering can cause a narrowing of the pores of a catalyst pellet (Figure 1.4), due to which the pores are no longer available for the reaction to occur. Another example of deactivation by sintering is the loss of surface area occurring due to the atomic migration and agglomeration of small metal particles on the surface of a support into a larger site where the interior atoms are inaccessible to the reactants (Figure 1.5) (Fogler et al., 1999).

Sintering tends to occur among metal catalysts involved in high temperature gas phase reactions, such as the Nickel catalyst in steam reforming and the Platinum catalyst in catalytic converters, to name a few. The bulk mobility of the metal particles become significant and they begin to coalesce as the temperature approaches its Tamman Temperature, which is taken to be half the metal's melting point on the absolute temperature scale (Satterfield, 1991). As such, sintering will be negligible at temperatures below 40% of the melting point of the solid (Hughes, 1984).

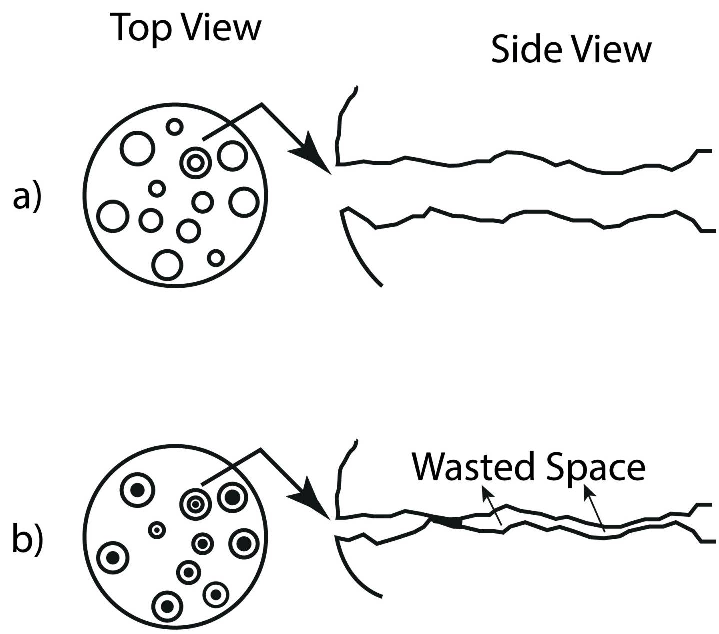


Fig. 1.4 Pore closure due to sintering. Schematic of the top views and side views of the catalyst pellet (a) Before sintering (b) After sintering (Fogler et al., 1999).

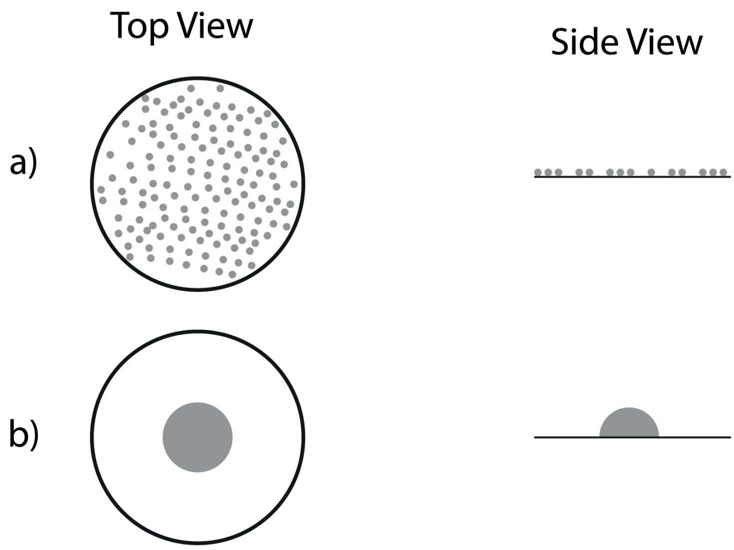


Fig. 1.5 Atomic agglomeration due to sintering. Schematic of the top views and side views (a) Before sintering (b) After sintering (Fogler et al., 1999).

While coking, poisoning and sintering are the most common mechanisms of catalyst deactivation, there are other modes by which this can occur. For instance, at high temperatures, a transformation of the solid state of the catalyst can happen, in which one crystalline phase is converted to another that has lower activity or lesser active surface area. Other mechanisms of deactivation include the loss of elements of the catalysts by attrition, erosion and volatilisation (Forzatti and Lietti, 1999).

The purpose of the preceding discussion was to give a very brief overview of the most common mechanisms of catalyst deactivation. A more comprehensive review can be found in a number of works such as those by Bartholomew (2001), Argyle and Bartholomew (2015) and Crabtree (2015), to name a few. In the next section, it is mentioned how the phenomenon of catalyst deactivation can be quantitatively described.

1.2 A quantitative description of catalyst deactivation

As mentioned previously, catalyst deactivation is inevitable and industrial processes face adverse economic effects when using decaying catalysts. A quantitative description or a measure of catalyst decay would be useful for these processes to take measures to optimise their operations and thereby, minimise the negative effects of catalyst deactivation. This quantitative description is given by the activity, a, of the catalyst, which is defined by the following equation (Forzatti and Lietti, 1999):

$$a = \frac{r}{r_0} \tag{1.1}$$

where r is the rate of the reaction after a measured time on stream while r_0 is the initial rate of the reaction, on a fresh catalyst yet to undergo deactivation. As such, a is a normalised variable $(0 \leq a \leq 1)$. The activity is a function of the catalyst history.

The rate of the reaction, in general, depends on the actual reaction conditions (such as reactant concentration (Cnc), temperature (Te) and pressure (Pr), to name a few) as well as the catalyst activity. That is:

$$r = r\left(Cnc, Te, Pr, ..., a\right) \tag{1.2}$$

However, Szépe and Levenspiel (1971) postulated that the rate of reaction could be separated into two terms: a reaction kinetics dependency and an activation dependency. This is termed separability. When separability holds, the reaction rate equations, for the product formation reactions as well as the catalyst deactivation, take the following form:

$$r = r(Cnc, Te, Pr, ...)\, r_1(a) \tag{1.3}$$

Usually, the separable factor, $r_1(a)$, is assumed to be equivalent to a. Physically, separability means that the configuration of active sites of the catalyst is not influenced by the other factors that can influence the reaction rate (e.g. concentration, temperature, pressure). For example, if the temperature is increased and the number of active sites of the catalyst does not change, this enables separability in that the terms involving temperature and activity can be written separately in mathematical equation for the reaction rate. A similar explanation applies for the separability of the other variables from the activity.

As such, the concept of separable kinetics has provided a satisfactory description to the study of deactivation by coking (Froment, 1980; Weekman Jr., 1968), and sintering (Forzatti and Lietti, 1999; Fuentes and Gamas, 1991). This is probably because the catalyst decay by these mechanisms occurs purely due to the physical coverage or loss of active sites. However, a number of other studies (Bakshi and Gavalas, 1975; Barbier et al., 1980; Onal and Butt, 1981; Weng et al., 1975) have reported difficulties with the separability of kinetics when poisoning is the mechanism of catalyst decay. This can be attributed to the chemisorption rather than purely physical coverage of poisons on the active sites. But separable kinetics can apply under certain conditions when poisoning is involved, and details regarding these conditions can be found in works by Butt et al. (1978), Forzatti et al. (1986) and Lynch and Emig (1989). In this thesis, however, only separable kinetics will be considered.

In the next section a review is carried out of the existing literature involving optimisation of processes using decaying catalysts. The shortcomings of those works considered "state-of-the-art" are highlighted, and these indicate the directions of research to be undertaken in this topic, which in turn lead to the objectives of this thesis.

1.3 Optimisation under catalyst deactivation: a literary review

While optimisation under catalyst deactivation has been attempted in the past, the literature pertaining to optimisation on an industrial scale, that identifies the optimal maintenance schedule and production plan in order to meet time-varying product demand, is limited.

Study has mainly focused on the optimal design or operation of a reactor that contains a catalyst undergoing deactivation in order to maximise conversion of the reactant. These optimum operating conditions were used until a time came when the catalyst activity fell to very low levels and had to be discarded or replaced.

Szépe and Levenspiel (1968) were the first to formulate an optimal operating policy for reactors subject to catalyst deactivation, by considering various aspects of the kinetics of the main reaction and the deactivation. They considered a batch reactor in which a single, irreversible reaction occurred under the influence of a decaying catalyst. The main reaction as well as the catalyst deactivation were considered to exhibit separable reaction kinetics, that is, their kinetic rate expressions were of the form of equation (1.3) wherein the catalyst activity was separable from the other factors influencing the reaction. In addition, the rate of catalyst deactivation was considered to be independent of composition. The following are notable results from their work:

(i) They demonstrated mathematically that if the deactivation kinetics is more sensitive to temperature than the main reaction, then it is optimal to continuously increase the temperature of operation so as to keep the product of the reaction rate constant and catalyst activity (termed effective reaction rate constant) unchanged throughout the reaction cycle.

(ii) On the other hand, if the deactivation kinetics is less sensitive to temperature than the main reaction, the optimal temperature policy is to operate at the maximum temperature limit.

(iii) Further, they applied the above conditions to flow reactors and established a policy of maintaining constant exit conversion, that is, to maintain the concentration of each reacting component exiting the reactor at a constant magnitude.

However, their analysis has been applied to determine the optimal operating policy only for a fixed reaction cycle time and a fixed final catalyst activity. They suggest to identify the optimal time to regenerate or replace the catalyst by iteratively solving for the optimal operating policy using different values of the reaction cycle time and the final catalyst activity.

Further, Crowe (1970) considered a single tubular reactor maintained at uniform temperature throughout at any time, and concluded that the optimal operational policy for such a reactor using a decaying catalyst was to end the operation at the maximum temperature limit while maintaining constant exit conversion at all times. This work was extended by Crowe and Lee (1971) to study a sequence of tubular reactors, each which used a decaying catalyst and was maintained at a uniform temperature at any time, and in this case as well, the optimal operational policy was predicted as maintaining constant exit conversions from each reactor.

Lee and Crowe (1970) further investigated concentration dependent deactivation for batch reactors and concluded that a constant effective rate coefficient was no longer an optimal policy. Crowe (1976), however, reported that for continuous stirred and plug flow reactors, even when concentration dependent deactivation is involved, if the time scale of deactivation is much larger than the time scales of the main reaction and the flow rate, constant exit conversion remains the optimal policy.

Assuming that maintaining constant exit conversion is the optimal operating policy for reactors using decaying catalysts, other studies have concentrated on obtaining a relationship between the time-on-stream (time of uninterrupted feed of reactants to the reactor) and temperature of operation. Krishnaswamy and Kittrell (1979) developed a simple mathematical model adopting power function kinetics that predicts the temperature-time relationship in order to maintain constant exit conversion of the reactant. Ho (1984) further exploited the analytical properties of the constant conversion - rising temperature policy to predict optimal operating conditions and acquire further information on the deactivation kinetics. Pacheco and Petersen (1986) derived a temperature-time relationship for a wide range of fouling data by formulating a power law model in which the order of deactivation depends on the activity. Sapre (1997) proposed another technique wherein the kinetic parameters are obtained by varying the space velocity, at constant temperature, to maintain constant exit conversion and used the information obtained to determine the time-temperature policy for maintaining constant exit concentration at a fixed space velocity. The objective behind all such studies that identified the temperature-time relationship for the optimal operating policy was to estimate the cycle length until the temperature of operation reached its upper limit or until the catalyst activity reached a minimum allowable value.

All of the aforementioned literature have focused on catalyst deactivation study at the reactor or pilot plant level, to identify the optimal operating conditions until the ‘time’ came for the catalyst to be replaced. This ‘time’ was usually when the temperature of operation reached its maximum allowable limit or the catalyst activity fell to its minimum permissible value. As such, there was no "maintenance schedule for catalyst replacements" required to be identified, as the time for catalyst replacements was already known.

On an industrial scale, however, such strategies may not constitute the optimal policy as other aspects have to be taken into consideration such as the seasonal demand figures and the storage costs. For instance, using a catalyst till its activity reaches the minimum permissible value can result in a low production rate and an inability to meet demand and hence, a loss in revenue. Or if a fresh catalyst begins to be used during a low demand season, the production rate may be far higher than that required to fulfil the demand, which might lead to large amount of unused product in the inventory and thus, excessive inventory costs.

The question then arises that, for a specified time horizon of the industrial process, how many catalyst replacements should occur and when should each catalyst replacement happen, given that replacing a catalyst, although it improves the production rate, it involves costs and a loss of production time. Further, there is the question as to how the process operating conditions, inventory levels and sales to meet demand can be optimally planned in tandem with these catalyst replacements in order to maximise the profits of the industrial process.

This leads back to the discussion at the beginning of the chapter. As mentioned in that discussion, in order to optimise the performance of the industrial process, and thereby maximise its profits, an optimal maintenance schedule for catalyst replacements and an optimal production plan have to be obtained in an integrated manner. The maintenance schedule for catalyst replacements should specify the optimum number of catalyst loads to use and the optimal time for each catalyst replacement, such that the trade-off between attaining high production rates, and having low maintenance costs and effective production times is optimally balanced. And the production plan should determine the optimal operating conditions of the reactor, as well as the optimal product inventory levels and sales, such that product demand is met effectively, while excessive inventory costs are also avoided.

Previously, a very limited set of publications have attempted to solve the problem of the integrated optimisation of the maintenance schedule for catalyst replacements and production planning in processes using decaying catalysts. These publications have obtained solutions of these problems by using formulations belonging to a category called mixed-integer programming problem formulations and solution methodologies for solving such formulations that are collectively called mixed-integer optimisation techniques. In the following subsection, brief introductions to mixed-integer programming problems and mixed-integer optimisation techniques are given, and the publications that have used these formulations and techniques to solve problems involving optimisation of maintenance scheduling and production in processes using decaying catalysts are discussed.

1.3.1 Optimisation under catalyst deactivation using mixed-integer approaches

Real world problems often involve decision variables that may be integer and/or continuous in nature. The integer decision variables can take only integer values which essentially represent a set of discrete choices. An example of a problem involving integer decision variables is deciding the number of heat exchangers to use in a set up, out of a given number available. On the other hand, a continuous decision variable can take any value within a specified range. An example of a problem involving a continuous decision variable is deciding the optimum temperature of a reactor, given the operating temperature limits, in order to maximise product yield.

When an optimisation problem involves integer as well as continuous decision variables, it is called a mixed-integer programming problem. The general form of a mixed-integer programming problem is given in equations (1.4a) – (1.4e). L_m represents the objective function, which is to be minimised by the selection of the integer decisions, u_m, and continuous decisions, v_m, when subject to the equality constraints, g_m, and inequality constraints, c_m. $\mathbb{U}$ represents the discrete set of permissible values for the integer decisions, u_m, and $\mathbb{V}$ represents the domain of the continuous decisions, v_m.

$$\min_{u_m, v_m} \quad L_m\left(u_m, v_m\right) \tag{1.4a}$$

subject to

$$g_m\left(u_m, v_m\right) = 0 \tag{1.4b}$$

$$c_m\left(u_m, v_m\right) \leq 0 \tag{1.4c}$$

$$u_m \in \mathbb{U} \tag{1.4d}$$

$$v_m \in \mathbb{V} \tag{1.4e}$$

If in equation (1.4), the objective function and the constraints are linear functions, then it is called a mixed-integer linear programming (MILP) problem. However, if in equation (1.4), the objective function and/or any of the constraints are nonlinear functions, then it is called a mixed-integer nonlinear programming (MINLP) problem.

The methodologies for solving mixed-integer programming problems are called mixed-integer optimisation techniques. Some of the common methodologies for solving MILP problems include the Branch & Bound method and Cutting Plane algorithms (Gomory, 2010).

The popular methodologies for solving MINLP problems are the Branch & Bound method, the Outer Approximation (OA) algorithm (Duran and Grossmann, 1986; Viswanathan and Grossmann, 1990) and the Generalised Benders Decomposition (GBD) method (Geoffrion, 1972). The major original results and mathematical developments in mixed-integer optimisation theory, along with a number of important application areas in chemical engineering can be found in Floudas (1995).

Now, some comments are made regarding the problem of the optimisation of maintenance scheduling and production in processes using decaying catalysts. In identifying the optimal maintenance schedule for catalyst replacements, decisions have to be made, at each time stage of the process, on whether to undertake a catalyst replacement operation or not. Thus, these decisions to schedule catalyst changeovers are of binary nature. That is, whether a catalyst replacement operation should occur or not at a given time can be indicated by values of 0 or 1, respectively, for the catalyst changeover decision variables (the reverse notation can also be used) and hence, these are integer decision variables. On the other hand, in determining the optimal production plan, the decisions to be made are those regarding the values of the process operating conditions and the product sales to meet demand, and these decisions are of continuous nature. Thus, this problem involves integer as well as continuous decision variables. Further, these decisions, especially those in the production planning part of the problem (for example: the temperature of the reactor), may appear nonlinearly in the underlying model equations. Determining the set of optimal integer and continuous decisions, in what is potentially a complex and highly nonlinear large scale problem, is challenging from the modelling as well as the optimisation stand points.

A search through literature identified only two publications that have attempted to solve problems of this type and these publications have used MINLP methods for the optimisation. The two publications, in fact, worked on a similar problem that was based on data obtained from a real-world industrial process, but used different optimisation techniques, which were based on the popular MINLP methodologies such as the OA and GBD algorithms. An overview of these publications follows next.

Houze et al. (2003) attempted to optimise the scheduling of catalyst replacements and production in an industrial plant in order to fulfil time-varying product demand. They formulated a disjunctive multiperiod model of the process, which is essentially a model that constrains a solution space with multiple sets of inequalities related by an OR statement. This disjunctive multiperiod model was then converted into an MINLP model using the big-M reformulation.

In the big-M reformulation, an arbitrarily large M is defined and using this M, each inequality of the model is reformulated such that if it is not in the set of inequalities being used, it is null, which in turn is accomplished by subtracting M from the left side of any greater than or equal to inequalities or by adding M to the right side of any less than or equal to inequalities. Their model was implemented on the GAMS software (GAMS Development Corporation, 1998), and the optimisation was performed using the software's DICOPT tool (Brooke et al., 1998), which uses the OA algorithm. They considered two time horizons of 2-years and 4-years for the industrial process, and for each of these time horizons, their model predicted how many catalyst loads to use, when to use them, the plant's monthly operating conditions of flow rate and temperature and the weekly levels of production and inventory, in order to maximise profits. However, they admit that, due to the non-convex nature of the problem and the DICOPT solver used, the solutions obtained were suboptimal. Further, the use of big-M reformulations made it difficult to obtain solutions for longer time-horizons.

Bizet et al. (2005) modified the model formulated by Houze et al. (2003), by introducing more extensive disjunctive constraints that use convex hull reformulations instead of big-M reformulations wherever possible. The convex hull reformulation involves introducing inequalities in order to form a convex hull of the solution space, which is the smallest set of points that include the full solution space and is convex. Bizet et al. (2005) further deviate from the work of Houze et al. (2003) by using two different approaches for the optimisation: a Partitioning search strategy and the GBD algorithm. The Partitioning search strategy operates by dividing the full time horizon into 1 year intervals and MINLP sub-problems were solved with DICOPT on GAMS within selected intervals. And for the GBD algorithm, which involves iteratively solving an MILP master problem and a nonlinear programming (NLP) sub-problem, the MILPs and NLPS were solved using the CPLEX code and the CONOPT3 solver, respectively, on GAMS (Brooke et al., 1998). They claim that the modifications and the new approaches enabled obtaining solutions for longer time horizons of 74-months and 9-years. They also sought to attain a global optimum using both approaches: for the Partitioning search strategy, by investigating all possible intervals, and for the GBD algorithm, by testing all possible binary combinations to initialise the first NLP. The same global optimum was obtained using the two approaches and further, this solution was superior to those obtained by just using DICOPT over the whole problem, as was done in Houze et al. (2003). But they admit that there is no rigorous proof that the solution obtained was a global optimum.

While the works of both, Houze et al. (2003) and Bizet et al. (2005), are novel with regard to the problem considered and solution methodology, neither work reveals the kinetic model or any of the parameters used in their work, due to confidentiality agreements with the industry from which the data was obtained. Thus, any possibility of reproducibility of their results is obviated.

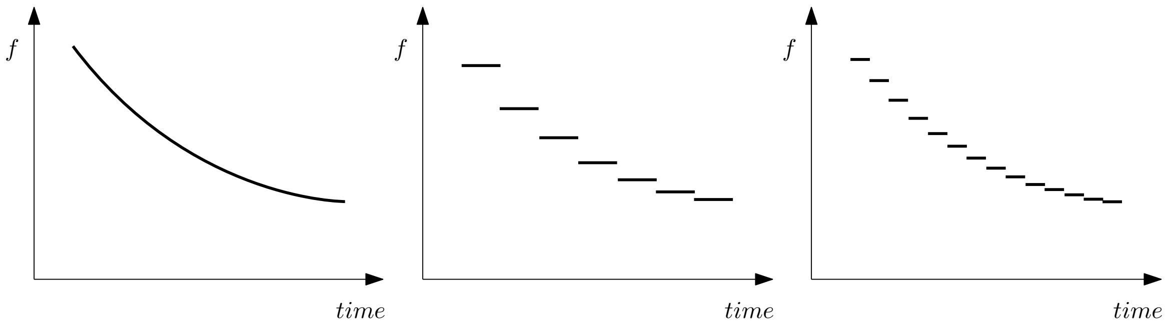
In addition, although the MINLP methods used by these works are among the most popular ones, there are considerable drawbacks involved in the use of mixed-integer optimisation techniques. These drawbacks are discussed in a detailed manner in the next section.

1.4 Disadvantages of mixed-integer optimisation techniques

First, MINLP techniques are not tailored to handle differential equations. And in processes using decaying catalysts, differential equations are commonly involved to describe, for example, the rate of catalyst decay or the rate of product formation. When MINLP techniques are used in such problems, these techniques circumvent differential equations by approximating each differential equation as a collection of steady state equations over the whole time horizon of the process. And the collection of steady state equations are imposed as equality constraints in the optimisation phase. Following such practices will cause the problem to end up containing a very large number of variables and nonlinear constraints, especially when a large number of differential equations are involved or long time horizons are considered. This could lead to the optimiser facing difficulties in converging to the optimal solutions.

Secondly, the steady state assumption, under which the differential equations are approximated, prevents an accurate description of the process dynamics within those time periods in which the differential equations are approximated as such. Thus, even if solutions are obtained, this assumption implies that the solutions may not be accurate. If greater accuracy is desired, a much larger collection of steady state equations is needed to approximate each differential equation, and this in turn would mean a larger number of variables and constraints in the optimisation phase, which would further accentuate convergence difficulties. Thus, there is a difficult compromise to be made between attaining higher accuracy and easier convergence.

The schematic in Figure 1.6 gives a visualisation of how a more accurate approximation can cause the problem to have a greater number of constraints to be fulfilled. When MINLP techniques are used, the original differential equation, f, shown in (a) is approximated by a collection of steady state equations. Examples of such approximations are shown as schematics in (b) and (c), which are labelled as Approximation 1 and Approximation 2,



(a) Original differential equation (b) Approximation 1 (c) Approximation 2

Fig. 1.6 A schematic describing how a differential equation is approximated as a collection steady state equations when MINLP methods are used. Approximation 1 in (b) is less accurate than Approximation 2 in (c), but convergence to an optimal solution would be easier in the former case compared to the latter case.

respectively. As mentioned previously, the collection of steady state equations are imposed as equality constraints in the optimisation phase. As can be seen, a larger collection of steady state equations are used to approximate f in Approximation 2 compared to Approximation 1. This implies that Approximation 2 is a more accurate approximation of f compared to Approximation 1. On the other hand, this also implies that there are a larger number of variables and constraints present in the optimisation phase when Approximation 2 is used in comparison to Approximation 1, and this means that convergence to an optimal solution would be easier in latter case compared to the former case.

It is now discussed how these two drawbacks of MINLP techniques apply in the works of Houze et al. (2003) and Bizet et al. (2005). It is noted that in these works, the model differential equations (which were not revealed due to proprietary reasons) in any week, were approximated as steady state algebraic equations that applied for the duration of the week. And the differential equations over the whole time horizon were represented by a collection of these weekly steady state equations, all of which were imposed as equality constraints in the problem. Under this practice, the number of constraints in the work of Houze et al. (2003) were of the order of thousands and that in the work of Bizet et al. (2005) were of the order of tens of thousands, and both these works contained thousands of variables.

While these works did not report any convergence difficulties, it is highlighted that because the differential equations were approximated as steady state equations over the duration of a week, it is not possible to obtain a dynamic description of the process over the duration of that week and this can be considered as a considerable loss of accuracy. For example, in these works, the differential equation for the production rate is approximated as a steady state equation over the week, and hence, a constant, steady state value of the production rate is used in place of a dynamic description over the duration of that week. This means that the value of the production rate for the week cannot be considered accurate. This also throws a big question on the accuracy of the final solution obtained by these works. Neither of these works have evaluated or even mentioned the loss of accuracy arising from this steady state approximation of the differential equations.

If, however, the differential equations were approximated on a daily rather than weekly basis, the accuracy of the solutions obtained would be considerably higher. But this in turn means that the number of variables and constraints would have increased seven-fold, which would have significantly increased the difficulty of converging to a solution. This discussion illustrates the above-mentioned two drawbacks that the use of MINLP techniques entails.

A further criticism of the works of Houze et al. (2003) and Bizet et al. (2005) is that these works do not reveal any of the underlying model equations or the parameters used, citing confidentiality reasons. Therefore, it was not possible to reproduce the results of these works or even attempt a finer approximation of the underlying equations to check if the accuracy of the results was improved or if any convergence difficulties were experienced.

There is also a third drawback in that the MINLP techniques are combinatorial in nature, which means that the computational effort to solve the problem increases exponentially with an increase in the number of the integer decision variables involved. For instance, the works of Houze et al. (2003) and Bizet et al. (2005) considered a maximum of 4 and 3 catalyst loads to be used, respectively. Since MINLP techniques such as OA and GBD, iteratively consider different combinations of binary variables in identifying solutions, the number of such combinations would increase exponentially, as would the computational effort to solve this problem, if the number of catalyst loads involved was increased. As another example, to identify the global optimum, as done in the work of Bizet et al. (2005), the number of possible intervals to examine in the Partitioning search strategy or the number of binary combinations to test in the GBD algorithm, would also increase exponentially with an increase in the number of catalyst loads involved. Thus, MINLP techniques would require enormous computational power in obtaining solutions if the number of such integer decision variables in the problem became large. This adds to a drawback that the problem size also increases as a larger number of discrete decision variables are involved, due to which the problem may become intractable. Therefore, in such cases, convergence to a solution may be difficult and even if solutions can be obtained, an enormous computational power would be required for this purpose.

The discussion till now has centred on the major drawbacks faced by MINLP methods with regard to the problem considered in the works of Houze et al. (2003) and Bizet et al. (2005). However, in comparison to the problem considered by these works, there are more realistic and complex problems in this area, which have not been considered in present literature. An overview of such problems follows next. While MINLP techniques form part of the established methodologies to solve problems of these kinds as well, it is highlighted how the discussed drawbacks would make these methods unsuitable to solve these problems.

The works by Houze et al. (2003) and Bizet et al. (2005) considered optimisation of maintenance scheduling and production in a process containing only a single reactor using decaying catalysts. However, it is common for an industrial process to have parallel lines of reactors operating simultaneously, which would improve flexibility of the process by allowing one reactor to be shut down for catalyst replacement while the remaining reactors continue to produce product to meet demand. A literature survey did not find any publications that have considered such a problem. While there has been work examining optimisation of maintenance scheduling of parallel processing lines experiencing decaying performances in other engineering applications, only a very limited set of papers address all aspects of the additional problem of production planning, such that all of the operating conditions, inventory management and sales to meet time-varying demand, are optimised. Further, even these limited set of papers admit to shortcomings, which can be traced to the mixed-integer techniques used for the optimisation. Thus, there are no encouraging signs that using MINLP techniques would produce satisfactory solutions if used for the optimisation of maintenance scheduling and production in parallel lines of reactors using decaying catalysts. Since the problem involving parallel reactor lines will be more complex than that involving a single reactor, due to the presence of a greater number of decision variables, constraints and differential equations in comparison to the latter case, the mentioned drawbacks of MINLP techniques will be aggravated if those techniques are used to obtain solutions for this problem and hence, the results obtained cannot be expected to be of good quality.

Further, the works by Houze et al. (2003) and Bizet et al. (2005) assumed that all kinetic parameters involved in the underlying model of the problem were known exactly. In reality, these kinetic parameters are rarely known exactly and there is generally an uncertainty regarding their values. This uncertainty or variation in parameter values can have a significant impact on the optimal operations of the process. A limited set of papers exist, that use online methods to include variation in kinetic parameter values in the optimisation of problems of this type. However, these online methods cannot identify the individual effect of uncertainty in each kinetic parameter or the effect of such uncertainties before the process begins, and no publication has established a technique that would enable determining these aspects. The conventional methods of determining these aspects involve using one of the popular preventive methods of handling uncertainty, such as stochastic programming, fuzzy programming, robust optimisation or parametric programming, in combination with a mixed-integer optimisation technique. However, the use of such conventional methods on a problem would require solving a problem of similar or a larger size, in comparison to when there are no uncertainty considerations, using mixed-integer optimisation techniques. Therefore, the use of conventional methods to solve this large scale problem would mean the drawbacks of the mixed-integer techniques are once again manifested or even further aggravated, and so, these methods are not expected to produce good quality results.

The preceding discussion indicates that methodologies based on mixed-integer techniques are not suitable for solving problems involving maintenance scheduling and production in large scale industrial processes using decaying catalysts. Hence, an alternative methodology to solve problems in this area is needed, which can overcome the disadvantages of mixed-integer techniques and obtain high quality solutions. A methodology is also needed that can effectively solve such problems when parametric uncertainties are involved, while overcoming the drawbacks brought about by the use of mixed-integer techniques in the established methodologies for optimisation under uncertainty. The contributions of this thesis lie in developing such methodologies, which draw from concepts in optimal control theory. The objectives of the thesis are stated in the next section.

1.5 Research objectives

The objectives of this thesis are enumerated as follows:

1. To develop a methodology that can effectively optimise the maintenance scheduling and production in a process containing a reactor using decaying catalysts, and which can overcome the drawbacks faced by mixed-integer optimisation techniques in solving this problem

2. To develop a methodology that can effectively optimise maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts,

and which can overcome the drawbacks that mixed-integer methods would face in solving problems of this kind

3. To develop a methodology that can consider uncertainties in kinetic parameters while effectively optimising the maintenance scheduling and production in a process containing a reactor using decaying catalysts, and which can overcome the disadvantages that the use of mixed-integer methods would introduce in the conventional methodologies, if such methodologies are used to solve problems of this kind

As will be seen, the methodologies developed to fulfil these objectives use concepts from optimal control theory, which enable a negation of the use of mixed-integer techniques and hence, a solution of the involved problem as a standard nonlinear optimisation problem. The optimal control methodology developed to fulfil the first objective will in fact form the base to develop the optimal control methodologies to fulfil the second and third objectives. Before proceeding, the essential components of the thesis are highlighted in a brief overview of the thesis in the next section.

1.6 Overview of thesis

The current chapter (Chapter 1) forms the introduction to the thesis. The problem of maintenance scheduling and production of industrial processes using decaying catalysts is introduced, alongside an overview of the underlying concepts and a review of the publications in this area. The drawbacks of existing methodologies to solve this problem are highlighted, and these indicate the problems to be addressed in this area, which in turn lead to the objectives of the thesis.

In Chapter 2, the focus is on fulfilling the first objective of the thesis. An optimal control methodology is developed to optimise maintenance scheduling and production in a process containing a reactor using decaying catalysts. This methodology involves formulating this problem as a multistage mixed-integer optimal control problem with the intention of using a solution procedure to solve it as a standard nonlinear optimisation problem, without mixed-integer techniques. Four case studies of the industrial process are examined which differ on the basis of the kinetics of the underlying reactions. Two solution procedures are required to be used, each which has its own relative advantages. The methodology using the second solution procedure is particularly successful in obtaining high quality solutions to this problem. Further, these solutions possess features of robustness, reliability and efficiency, which indicate the methodology's success in overcoming the drawbacks of the use of mixed-integer optimisation techniques in solving this problem.

In Chapter 3, the aim is to fulfil the second objective of the thesis. The successful optimal control methodology developed in Chapter 2 is extended to optimise maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts. The methodology when applied to a case study of this process results in high quality solutions. Further, the solutions obtained, once again, possess features of robustness, reliability and efficiency, which indicate that the methodology can overcome the drawbacks that mixed-integer optimisation techniques would face in solving problems of this kind.

In Chapter 4, the third objective of the thesis is targeted to be fulfilled. An optimal control methodology is developed to consider uncertainties in kinetic parameters in the optimisation of the maintenance scheduling and production in a process containing a reactor using decaying catalysts. This methodology is an extension of the forms of the methodologies used in Chapters 2 and 3. The methodology involves using a multiple scenario approach to consider parametric uncertainties and formulating the problem as a stochastic multistage mixed-integer optimal control problem, which is solved as a standard nonlinear optimisation problem using a procedure similar to that used in the preceding chapters. Different case studies are examined to identify the effects on the optimal operations of uncertainty in each individual parameter, of simultaneous uncertainty in all parameters and of the number of scenarios generated. The results obtained provide insights into these aspects and indicate that the methodology is capable of solving this problem. Further, the robust, reliable and efficient nature of the results obtained suggest that the methodology can overcome the disadvantages that mixed-integer methods would introduce in the conventional methodologies, if such methodologies are used to solve problems of this kind.

Chapter 5 presents the overall conclusions of the thesis and proposes future research directions.

Figure 1.7 shows a conceptual map that indicates the developmental results in each chapter and how the different chapters are connected. In this figure, a rectangular box and the text within represents a chapter. The ellipses and the text within, that are connected to a rectangle via arrows, are the developmental results from the chapter represented by that rectangle. It is seen that the ellipses themselves can be connected. It is noted that a black dot (•) at the intersection between two lines indicates a connection between the rectangles/ellipses from which the lines originate and the absence of the black dot at an intersection indicates that there is no such connection. The rectangles themselves are connected through arrows involving descriptive text, which indicate the connections between chapters.

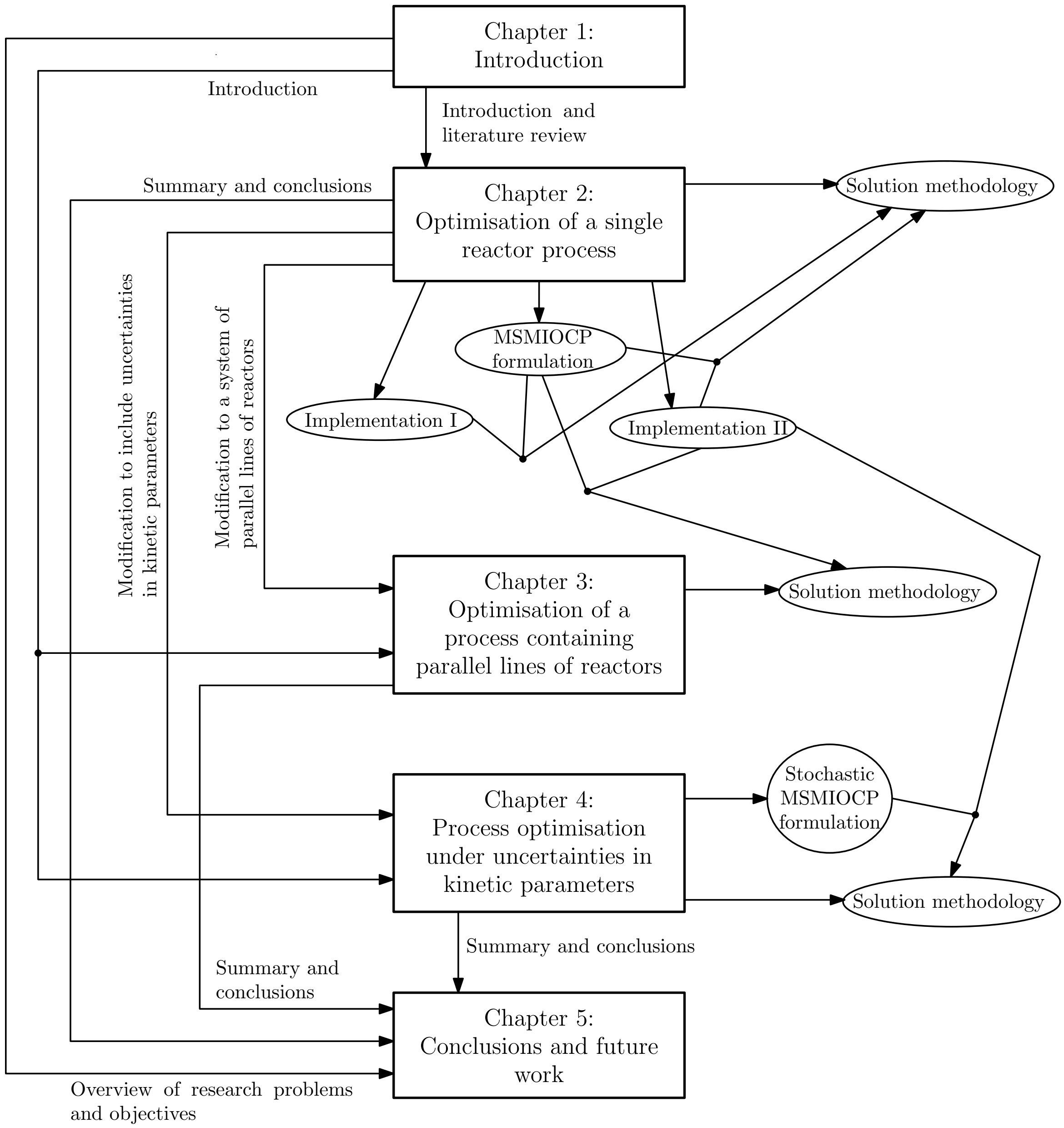


Fig. 1.7 A conceptual map that shows the developmental results in each chapter and the connections between chapters

Chapter 2

Optimisation of a single reactor process

In this chapter, an optimal control methodology is developed to optimise maintenance scheduling and production in a process containing a single reactor using decaying catalysts. This methodology involves formulating this problem as a multistage mixed-integer optimal control problem with the aim of solving it as a standard nonlinear optimisation problem, without using mixed-integer techniques. Four case studies are considered for the process, which differ depending on the kinetics of the product formation reaction or the catalyst deactivation, in the process model. As will be seen, the complex nature of the problem caused complications in obtaining solutions using the currently available solvers and two different solution implementation procedures are required to be used, each which had its own relative advantages. The methodology using the second implementation procedure is particularly successful in producing high quality solutions to this problem and indicates advantages of robustness, reliability and efficiency over mixed-integer techniques.

The structure of this chapter is as follows. In Section 2.1, an optimal control formulation of the problem is developed. In Section 2.2, the optimal control formulation is applied to case studies of an industrial process. Section 2.3 provides details of the first solution procedure attempted, Implementation I, and the results obtained using this implementation are discussed. Section 2.4 details the second solution procedure attempted, Implementation II, and discusses the results obtained using this implementation. Section 2.5 contains a summary of the chapter, a discussion comparing the results obtained with those of existing publications that considered a similar problem, and the conclusions of the chapter.

2.1 The optimal control problem formulation

Optimal control theory deals with identifying a control for a dynamic system over a period of time such that a specified objective function is optimised at the end of this time period. The applications of optimal control theory are vast, ranging from rocket science to building energy management and even economics.

An optimal control problem (OCP) is characterised by a set of control variables, state variables, differential-algebraic equations (DAEs), constraints, and a performance index (or an objective function). The controls are the decision variables in this problem, applied externally to the dynamic system in order to optimise its performance. The state variables are inherent to the system, in the sense that they represent the "state" of the system, and their values are determined from the DAEs and the values of the controls, at any time. Both, the control and state variables are usually required to fulfil some constraints. The controls, when chosen optimally, decide the optimal state variables, and these together optimise the performance index of the system.

The basic formulation of an OCP is shown in equations (2.1a) – (2.1h). The performance index consists of a point index ϕ and a continuous index L. This performance index is minimised, over a given duration beginning at time t_0 and ending at time t_F, by the selection of controls, w, and the resulting differential state variables, x and algebraic state variables, z, when subject to differential equations, f, algebraic equations, g, and constraints, c. Equations (2.1b) – (2.1c) describe an ordinary differential equation (ODE) system, given initial condition x_0. The controls w can include binary controls, u, as well as continuous controls, v, that belong to a real permissible set $\mathscr{V}$.

$$\min_{w(t)} W \quad = \quad \phi\left(x(t_F)\right) \;+ \int_{t_0}^{t_F} L\left(x(t), z(t), w(t), t\right) \mathrm{d}t \tag{2.1a}$$

subject to

$$\begin{gathered} \dot{x}(t) = f\left(x(t), z(t), w(t), t\right) \\ \forall t \in [t_0, t_F] \end{gathered} \tag{2.1b}$$

$$x(t_0) = x_0 \tag{2.1c}$$

$$\begin{gathered} 0 = g\left(x(t), z(t), w(t), t\right) \\ \forall t \in [t_0, t_F] \end{gathered} \tag{2.1d}$$

$$c\,(x(t), z(t), w(t), t) \leq \;\; 0 \qquad \forall t \in [t_0, t_F] \tag{2.1e}$$

$$w(t) = \left[[u(t)]^T \, , [v(t)]^T \right]^T \qquad \forall t \in [t_0, t_F] \tag{2.1f}$$

$$u(t) \in \{0, 1\} \qquad \forall t \in [t_0, t_F] \tag{2.1g}$$

$$v(t) \in \mathscr{V} \qquad \forall t \in [t_0, t_F] \tag{2.1h}$$

Now, the problem under consideration, that of optimising maintenance scheduling and production in a process using decaying catalyst, is developed as an optimal control problem. The dynamic system in this problem is the industrial process and the duration over which it is to be optimised is the time horizon of the process. And as mentioned in the introductory chapter, the decisions (controls) here include those of when to schedule a maintenance action to replace the catalyst in the reactor, which are binary in nature, as well as those that decide the operating conditions for the reactor and the sales, which are continuous variables. The state variables in the problem represent the "state" of the process and are determined from the appropriate DAEs that constitute the process model and the values of the controls at any time. The constraints in the problem include the operating limits of the process and the binary restrictions on those controls used to decide when to replace catalysts. And the objective function of the problem is to attain maximum profit or minimum cost for the process at the end of the time horizon.

For this problem, a ‘stage’ is regarded as a part of the time horizon of the process within which the decisions (binary as well as continuous) have to be made. Solving this problem involves making decisions at different stages within the time horizon of the industrial process. Therefore, a multistage version of equation (2.1) is used for the optimal control formulation of this problem, which is developed as follows. The whole time horizon of the process is discretised into stages (each, within which the decisions have to be made), which can be of arbitrary length. A control parametrisation approach is adopted wherein the decision variables are discretised over the whole time horizon and are taken to be piecewise constant across the times corresponding to each stage. That is, if the total number of stages is NP, the collective vectors of the controls, u and v, take up the following form:

$$u = \left[u^{(1)}, u^{(2)}, \ldots, u^{(NP)}\right]^T \tag{2.2a}$$

$$v = \left[v^{(1)}, v^{(2)}, \ldots, v^{(NP)}\right]^T \tag{2.2b}$$

The control profiles are allowed to be discontinuous at the junctions, t_p, between any two consecutive stages, p and $p+1$. Further, these stage switching times, t_p, are considered to be fixed.

In this multistage formulation, decisions of binary nature have to be made in each stage of the time horizon, that indicate whether a catalyst should be in operation or if a maintenance action should occur to replace it. For example, a value of 1 can be used to indicate that the catalyst is in operation and a value of 0 can be used to indicate that a maintenance action or a catalyst replacement operation occurs. Henceforth in this thesis, this decision will be called a "catalyst changeover control". Due to their binary nature, the catalyst changeover controls correspond to the controls, u. Further, the operating conditions of the reactor and the amount of product sales should also be decided at each stage. These are decisions of continuous form and so, correspond to the controls, v. Due to the presence of integer and continuous controls, this is a mixed-integer formulation.

The state variables, however, are retained in their continuous form, without discretisation, and are determined in each stage from the set of DAEs. The DAEs are solved to a high accuracy in the right sequential order using state-of-the-art integrators and hence, this solution methodology is called a "feasible path approach" (Vassiliadis, 1993; Vassiliadis et al., 1994a,b). The solutions of the DAEs in each stage, across the whole time horizon, are facilitated by junction conditions between any two consecutive periods, p and $p+1$, the general form of which is given by equation (2.3) (Vassiliadis, 1993):

$$\begin{aligned} J\Big(\dot{x}^{(p+1)}\left(t_p^+\right), x^{(p+1)}\left(t_p^+\right), z^{(p+1)}\left(t_p^+\right), u^{(p+1)}\left(t_p^+\right), v^{(p+1)}\left(t_p^+\right), \\ \dot{x}^{(p)}\left(t_p^-\right), x^{(p)}\left(t_p^-\right), z^{(p)}\left(t_p^-\right) u^{(p)}\left(t_p^-\right), v^{(p)}\left(t_p^-\right), t_p\Big) &= 0 \\ p = 1, 2, \ldots NP-1 \end{aligned} \tag{2.3}$$

Given that the optimal control formulation of this problem involves multiple stages and contains integer as well as continuous controls, it can be termed a multistage mixed-integer optimal control problem (MSMIOCP) formulation. The basic form of the MSMIOCP over time periods, $p = 1, 2, \ldots NP$, $t \in \left[t_{p-1}, t_p\right]$, with $t_{NP} = t_F$ is shown in equations (2.4a) – (2.4h). The terminology used in equation (2.4) is similar to the basic OCP formulation in equation (2.1), with the superscript (p) indicating that they apply to stage p. The additional terms here are the junction conditions, h, analogous to equation (2.3), that provide the initial conditions for the solution of the ODEs in stage p. An illustration of the MSMIOCP formulation is shown in Figure 2.1.

$$\min_{u,v} W = \sum_{p=1}^{NP} \left\{ \phi^{(p)} \left(x^{(p)}(t_p), z^{(p)}(t_p), u^{(p)}, v^{(p)}, t_p \right) + \int_{t_{p-1}}^{t_p} L^{(p)} \left(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t \right) \mathrm{d}t \right\} \tag{2.4a}$$

subject to

$$\begin{gathered} \dot{x}^{(p)}(t) = f^{(p)}(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t) \\ t_{p-1} \leq t \leq t_p \\ p = 1, 2, \ldots, NP \end{gathered} \tag{2.4b}$$

$$x^{(1)}(t_0) = h^{(1)} \left(u^{(1)}, v^{(1)} \right) \tag{2.4c}$$

$$\begin{gathered} x^{(p)}(t_{p-1}) = h^{(p)} \left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), u^{(p)}, v^{(p)} \right) \\ p = 2, 3, \ldots, NP \end{gathered} \tag{2.4d}$$

$$\begin{gathered} 0 = g^{(p)}(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t) \\ t_{p-1} \leq t \leq t_p \\ p = 1, 2, \ldots, NP \end{gathered} \tag{2.4e}$$

$$\begin{gathered} c^{(p)} \left(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t \right) \leq 0 \\ t_{p-1} \leq t \leq t_p \\ p = 1, 2, \ldots, NP \end{gathered} \tag{2.4f}$$

$$\begin{gathered} u^{(p)} \in \{0, 1\} \\ p = 1, 2, \ldots, NP \end{gathered} \tag{2.4g}$$

$$v^{(p)} \in \mathscr{V}$$
$$p = 1, 2, \ldots, NP \tag{2.4h}$$

Due to the presence of integer as well as continuous controls, it might seem intuitive that mixed-integer optimisation methods need to be used to solve problems of this type of formulation. However, as mentioned in Section 1.4, there are many disadvantages to using mixed-integer optimisation techniques. Therefore, a methodology is presented that attempts to solve problems of this type of formulation without the use of mixed-integer techniques and overcome the drawbacks associated with the use of those methods.

This methodology is based on the property that controls that appear linearly in an optimal control problem tend to take values at either of their bounds in the optimal solution, lending what is called a "bang-bang" behaviour to these controls (Bryson and Ho, 1975). This methodology is applied to the problem under consideration by formulating the equations such that the catalyst changeover controls, corresponding to the controls, u, appear linearly. By virtue of the bang-bang property arising from this linear occurrence, these controls can be expected to automatically take values of only 0 or 1 in the optimal solution, without the use of mixed-integer optimisation techniques. Such a methodology has previously been used by Al Ismaili et al. (2018) to optimise maintenance scheduling of heat exchanger networks, and the results indicated robust, reliable and efficient solutions in comparison to those obtained using mixed-integer techniques on the same problem.

In the following sections, a theoretical analysis is done using the basic MSMIOCP formulation in equation (2.4). In this theoretical analysis, the Pontryagin Minimum Principle (Pontryagin et al., 1962) is applied to indicate how the linearity of the binary controls in problem formulations of this kind can potentially result in a bang-bang behaviour for these controls. It is also discussed as to what advantages this methodology has the potential to offer over mixed-integer techniques.

2.1.1 A theoretical analysis leading to the bang-bang property

In this section, it is demonstrated how linearity of a control in an MSMIOCP of the form of equation (2.4) can potentially result in a bang-bang behaviour for these controls. Equation (2.4) is reformulated so that, in each stage $p = 1, 2, \ldots, NP$, all of its elements are linear with respect to the binary control, $u^{(p)}$. This reformulated equation is given by equation (2.5)

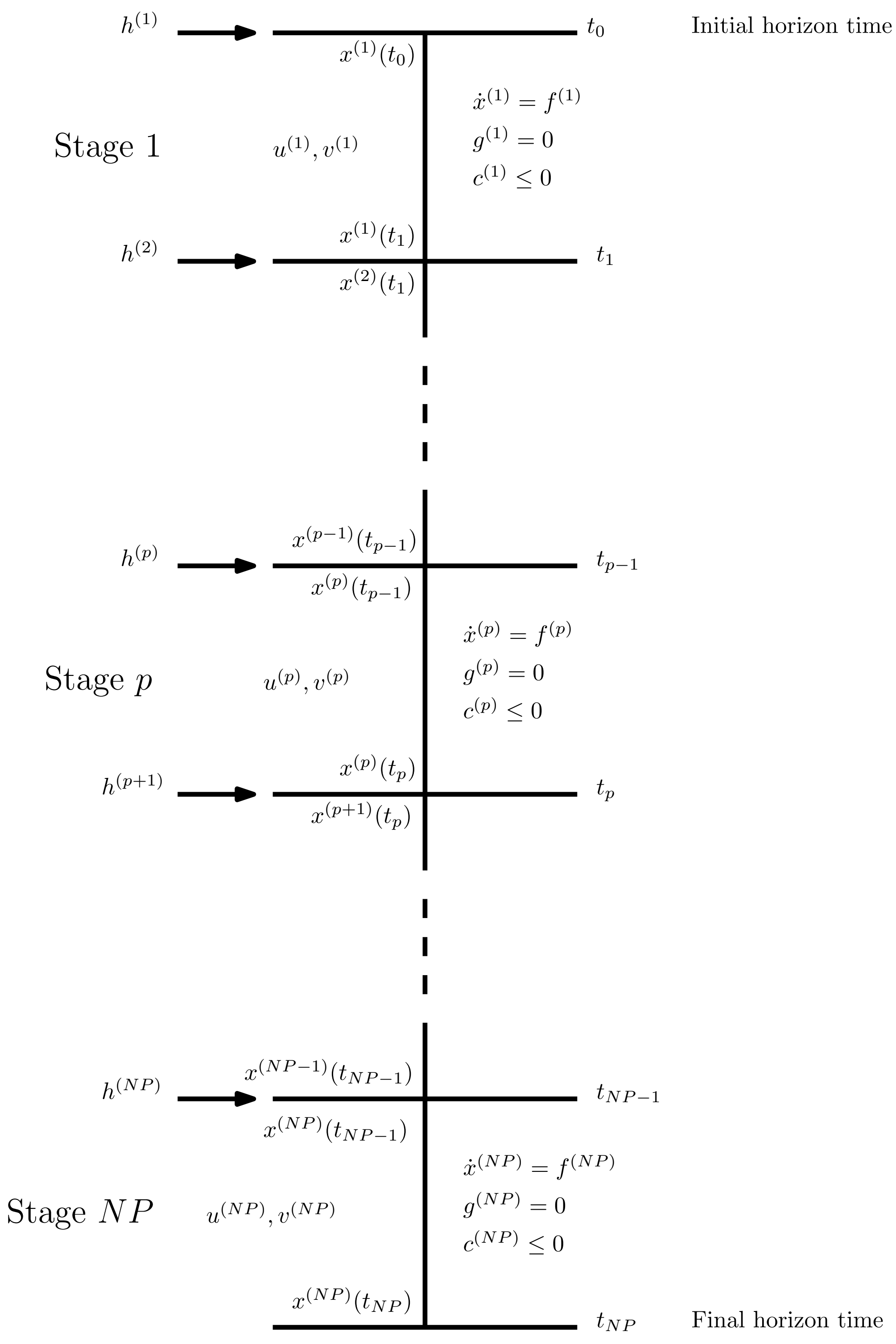


Fig. 2.1 An illustration of the MSMIOCP formulation.

and the terminology of its elements are as follows.

In equation (2.5a), for each stage $p = 1,2,\ldots,NP$, the point performance index is represented as functions of $\phi_1^{(p)}$ and $\phi_2^{(p)}$, where $\phi_1^{(p)}$ is the coefficient of the linear control, $u^{(p)}$, and both terms are themselves independent of controls, u. $L_1^{(p)}$ and $L_2^{(p)}$, $f_1^{(p)}$ and $f_2^{(p)}$, $h_1^{(p)}$ and $h_2^{(p)}$, $g_1^{(p)}$ and $g_2^{(p)}$, and $c_1^{(p)}$ and $c_2^{(p)}$ are the analogous terms for the continuous performance index in equation (2.5a), the differential equations in equation (2.5b), the junction conditions in equations (2.5c) – (2.5d), the algebraic equations in equation (2.5e) and the constraints in equation (2.5f), respectively.

$$\begin{aligned}\min_{u,v} W = \sum_{p=1}^{NP} \Bigg\{ & \left[\phi_1^{(p)}\left(x^{(p)}(t_p), z^{(p)}(t_p), v^{(p)}, t_p\right)\right]^T u^{(p)} + \phi_2^{(p)}\left(x^{(p)}(t_p), z^{(p)}(t_p), v^{(p)}, t_p\right) \\ & + \int_{t_{p-1}}^{t_p} \left[\left[L_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right]^T u^{(p)} + L_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] \mathrm{d}t \Bigg\}\end{aligned} \tag{2.5a}$$

subject to

$$\begin{gathered}\dot{x}^{(p)}(t) = \left[f_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] u^{(p)} + f_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) \\ t_{p-1} \le t \le t_p \\ p = 1,2,\ldots,NP\end{gathered} \tag{2.5b}$$

$$x^{(1)}(t_0) = \left[h_1^{(1)}\left(v^{(1)}\right)\right] u^{(1)} + h_2^{(1)}\left(v^{(1)}\right) \tag{2.5c}$$

$$\begin{gathered}x^{(p)}(t_{p-1}) = \left[h_1^{(p)}\left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), v^{(p)}\right)\right] u^{(p)} \\ + h_2^{(p)}\left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), v^{(p)}\right) \\ p = 2, 3, \ldots, NP\end{gathered} \tag{2.5d}$$

$$\begin{gathered}0 = \left[g_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] u^{(p)} + g_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) \\ t_{p-1} \le t \le t_p \\ p = 1,2,\ldots,NP\end{gathered} \tag{2.5e}$$

$$\left[c_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] u^{(p)} + c_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) \leq 0$$
$$t_{p-1} \leq t \leq t_p \tag{2.5f}$$
$$p = 1, 2, \ldots, NP$$

$$u^{(p)} \in \{0, 1\}$$
$$p = 1, 2, \ldots, NP \tag{2.5g}$$

$$v^{(p)} \in \mathscr{V}$$
$$p = 1, 2, \ldots, NP \tag{2.5h}$$

Now, a theoretical analysis is done in order to identify the necessary conditions for optimality, also termed the Euler-Lagrange equations. This theoretical analysis is similar to that done in Al Ismaili et al. (2018), with the difference being that here the controls are distinguished as occurring linearly or nonlinearly, whereas that work considered only linear controls.

The performance index in equation (2.5a) is modified so that Euler-Lagrange multipliers are introduced as shown in equation (2.6):

$$\begin{aligned}
\overline{W} = \sum_{p=2}^{NP} \Bigg\{ & \left[\phi_1^{(p)}\left(x^{(p)}(t_p), z^{(p)}(t_p), v^{(p)}, t_p\right)\right]^T u^{(p)} + \phi_2^{(p)}\left(x^{(p)}(t_p), z^{(p)}(t_p), v^{(p)}, t_p\right) \\
& + \left[\beta^{(p)}\right]^T \left[h_1^{(p)}\left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), v^{(p)}\right) u^{(p)} \right. \\
& \left. + h_2^{(p)}\left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), v^{(p)}\right) - x^{(p)}(t_{p-1})\right] \\
& + \int_{t_{p-1}}^{t_p} \left[\left[L_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right]^T u^{(p)} + L_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] \mathrm{d}t \\
& + \int_{t_{p-1}}^{t_p} \left[\Lambda^{(p)}(t)\right]^T \left[f_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) u^{(p)} \right. \\
& \left. + f_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) - \dot{x}^{(p)}(t)\right] \mathrm{d}t \\
& + \int_{t_{p-1}}^{t_p} \left[\lambda^{(p)}(t)\right]^T \left[g_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) u^{(p)} + g_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] \mathrm{d}t
\end{aligned}$$

$$
\begin{aligned}
&+\int_{t_{p-1}}^{t_p}\left[\mu^{(p)}(t)\right]^T\left[c_1^{(p)}\left(x^{(p)}(t),z^{(p)}(t),v^{(p)},t\right)u^{(p)}+c_2^{(p)}\left(x^{(p)}(t),z^{(p)}(t),v^{(p)},t\right)\right]\mathrm{d}t\Bigg\}\\
&+\left[\phi_1^{(1)}\left(x^{(1)}(t_1),z^{(1)}(t_1),v^{(1)},t_1\right)\right]^T u^{(1)}+\phi_2^{(1)}\left(x^{(1)}(t_1),z^{(1)}(t_1),v^{(1)},t_1\right)\\
&+\left[\beta^{(1)}\right]^T\left[h_1^{(1)}\left(v^{(1)}\right)u^{(1)}+h_2^{(1)}\left(v^{(1)}\right)-x^{(1)}(t_0)\right]\\
&+\int_{t_0}^{t_1}\left[\left[L_1^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)\right]^T u^{(1)}+L_2^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)\right]\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\Lambda^{(1)}(t)\right]^T\left[f_1^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)u^{(1)}\right.\\
&\left.+f_2^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)-\dot{x}^{(1)}(t)\right]\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\lambda^{(1)}(t)\right]^T\left[g_1^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)u^{(1)}+g_2^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)\right]\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\mu^{(1)}(t)\right]^T\left[c_1^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)u^{(1)}+c_2^{(1)}\left(x^{(1)}(t),z^{(1)}(t),v^{(1)},t\right)\right]\mathrm{d}t
\end{aligned}
\tag{2.6}
$$

where $\Lambda^{(p)}$, $\lambda^{(p)}$, $\mu^{(p)}$ and $\beta^{(p)}$ are the Euler-Lagrange multipliers for stage $p = 1, 2, \ldots NP$. It is noted that for the Euler-Lagrange multiplier, $\mu^{(p)}$, corresponding to the constraints in each stage, the following requirement holds:

$$
\begin{aligned}
&\mu^{(p)}\begin{cases}=0 & \text{if } c_1^{(p)}u^{(p)}+c_2^{(p)}\le 0\\ \ge 0 & \text{if } c_1^{(p)}u^{(p)}+c_2^{(p)}=0\end{cases}\\
&p=1,2,\ldots,NP
\end{aligned}
\tag{2.7}
$$

Next, the calculus of variations is applied. Variations on the parameter set of stage p' of the form $\delta u^{(p')}$ and $\delta v^{(p')}$ are considered, which result in variations in the state values at all times, as shown in equation (2.8). For the sake of convenience, the arguments within the parentheses for each term are neglected. Clearly, the state vector of stage p, where $p < p'$, will not be influenced and so $\delta x^{(p)}(t) = 0$ and $\delta z^{(p)}(t) = 0$ for those stages.

$$
\delta\overline{W} \;=\; \sum_{p=2}^{NP}\left\{\left[\left[u^{(p)}\right]^T\frac{\partial\phi_1^{(p)}}{\partial x^{(p)}(t_p)}+\frac{\partial\phi_2^{(p)}}{\partial x^{(p)}(t_p)}\right]\delta x^{(p)}(t_p)\right.
$$

$$
\begin{aligned}
&+\left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial z^{(p)}(t_p)}+\frac{\partial \phi_2^{(p)}}{\partial z^{(p)}(t_p)}\right]\delta z^{(p)}(t_p)\\
&+\left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial v^{(p)}}+\frac{\partial \phi_2^{(p)}}{\partial v^{(p)}}\right]\delta v^{(p)}+\left[\phi_1^{(p)}\right]^T\delta u^{(p)}\\
&+\left[\beta^{(p)}\right]^T\left[\left(\frac{\partial h_1^{(p)}}{\partial x^{(p-1)}(t_{p-1})}u^{(p)}+\frac{\partial h_2^{(p)}}{\partial x^{(p-1)}(t_{p-1})}\right)\delta x^{(p-1)}(t_{p-1})\right.\\
&+\left(\frac{\partial h_1^{(p)}}{\partial z^{(p-1)}(t_{p-1})}u^{(p)}+\frac{\partial h_2^{(p)}}{\partial z^{(p-1)}(t_{p-1})}\right)\delta z^{(p-1)}(t_{p-1})\\
&\left.+\left(\frac{\partial h_1^{(p)}}{\partial v^{(p)}}u^{(p)}+\frac{\partial h_2^{(p)}}{\partial v^{(p)}}\right)\delta v^{(p)}+h_1^{(p)}\delta u^{(p)}-\delta x^{(p)}(t_{p-1})\right]\\
&+\int_{t_{p-1}}^{t_p}\left[\left(\left[u^{(p)}\right]^T\frac{\partial L_1^{(p)}}{\partial x^{(p)}}(t)+\frac{\partial L_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\right.\\
&+\left(\left[u^{(p)}\right]^T\frac{\partial L_1^{(p)}}{\partial z^{(p)}}(t)+\frac{\partial L_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&\left.+\left(\left[u^{(p)}\right]^T\frac{\partial L_1^{(p)}}{\partial v^{(p)}}(t)+\frac{\partial L_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}+\left[L_1^{(p)}\right]^T\delta u^{(p)}\right]\mathrm{d}t\\
&+\int_{t_{p-1}}^{t_p}\left[\Lambda^{(p)}(t)\right]^T\left[\left(\frac{\partial f_1^{(p)}}{\partial x^{(p)}}(t)\,u^{(p)}+\frac{\partial f_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\right.\\
&+\left(\frac{\partial f_1^{(p)}}{\partial z^{(p)}}(t)\,u^{(p)}+\frac{\partial f_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&\left.+\left(\frac{\partial f_1^{(p)}}{\partial v^{(p)}}(t)\,u^{(p)}+\frac{\partial f_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}+f_1^{(p)}\delta u^{(p)}-\delta\dot{x}^{(p)}(t)\right]\mathrm{d}t\\
&+\int_{t_{p-1}}^{t_p}\left[\lambda^{(p)}(t)\right]^T\left[\left(\frac{\partial g_1^{(p)}}{\partial x^{(p)}}(t)\,u^{(p)}+\frac{\partial g_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\right.\\
&+\left(\frac{\partial g_1^{(p)}}{\partial z^{(p)}}(t)\,u^{(p)}+\frac{\partial g_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&\left.+\left(\frac{\partial g_1^{(p)}}{\partial v^{(p)}}(t)\,u^{(p)}+\frac{\partial g_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}(t)+g_1^{(p)}\delta u^{(p)}\right]\mathrm{d}t\\
&+\int_{t_{p-1}}^{t_p}\left[\mu^{(p)}(t)\right]^T\left[\left(\frac{\partial c_1^{(p)}}{\partial x^{(p)}}(t)\,u^{(p)}+\frac{\partial c_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\right.
\end{aligned}
$$

$$
\begin{aligned}
&+\left(\frac{\partial c_1^{(p)}}{\partial z^{(p)}}(t)\,u^{(p)}+\frac{\partial c_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&+\left(\frac{\partial c_1^{(p)}}{\partial v^{(p)}}(t)\,u^{(p)}+\frac{\partial c_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}+c_1^{(p)}\delta u^{(p)}\Bigg]\,\mathrm{d}t\;\Bigg\}\\
&+\Bigg[\left(\left[u^{(1)}\right]^T\frac{\partial \phi_1^{(1)}}{\partial x^{(1)}(t_1)}+\frac{\partial \phi_2^{(1)}}{\partial x^{(1)}(t_1)}\right)\delta x^{(1)}(t_1)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial \phi_1^{(1)}}{\partial z^{(1)}(t_1)}+\frac{\partial \phi_2^{(1)}}{\partial z^{(1)}(t_1)}\right)\delta z^{(1)}(t_1)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial \phi_1^{(1)}}{\partial v^{(1)}}+\frac{\partial \phi_2^{(1)}}{\partial v^{(1)}}\right)\delta v^{(1)}+\left[\phi_1^{(1)}\right]^T\delta u^{(1)}\Bigg]\\
&+\left[\beta^{(1)}\right]^T\left[\left(\frac{\partial h_1^{(1)}}{\partial v^{(1)}}u^{(1)}+\frac{\partial h_2^{(1)}}{\partial v^{(1)}}\right)\delta v^{(1)}+h_1^{(1)}\delta u^{(1)}-\delta x^{(1)}(t_0)\right]\\
&+\int_{t_0}^{t_1}\Bigg[\left(\left[u^{(1)}\right]^T\frac{\partial L_1^{(1)}}{\partial x^{(1)}}(t)+\frac{\partial L_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial L_1^{(1)}}{\partial z^{(1)}}(t)+\frac{\partial L_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial L_1^{(1)}}{\partial v^{(1)}}(t)+\frac{\partial L_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+\left[L_1^{(1)}\right]^T\delta u^{(1)}\Bigg]\,\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\Lambda^{(1)}(t)\right]^T\Bigg[\left(\frac{\partial f_1^{(1)}}{\partial x^{(1)}}(t)\,u^{(1)}+\frac{\partial f_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\frac{\partial f_1^{(1)}}{\partial z^{(1)}}(t)\,u^{(1)}+\frac{\partial f_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&+\left(\frac{\partial f_1^{(1)}}{\partial v^{(1)}}(t)\,u^{(1)}+\frac{\partial f_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+f_1^{(1)}\delta u^{(1)}-\delta\dot{x}^{(1)}(t)\Bigg]\,\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\lambda^{(1)}(t)\right]^T\Bigg[\left(\frac{\partial g_1^{(1)}}{\partial x^{(1)}}(t)\,u^{(1)}+\frac{\partial g_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\frac{\partial g_1^{(1)}}{\partial z^{(1)}}(t)\,u^{(1)}+\frac{\partial g_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&+\left(\frac{\partial g_1^{(1)}}{\partial v^{(1)}}(t)\,u^{(1)}+\frac{\partial g_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+g_1^{(1)}\delta u^{(1)}\Bigg]\,\mathrm{d}t
\end{aligned}
$$

$$
\begin{aligned}
&+\int_{t_0}^{t_1}\left[\mu^{(1)}(t)\right]^T\left[\left(\frac{\partial c_1^{(1)}}{\partial x^{(1)}}(t)\,u^{(1)}+\frac{\partial c_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\right.\\
&+\left(\frac{\partial c_1^{(1)}}{\partial z^{(1)}}(t)\,u^{(1)}+\frac{\partial c_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&\left.+\left(\frac{\partial c_1^{(1)}}{\partial v^{(1)}}(t)\,u^{(1)}+\frac{\partial c_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+c_1^{(1)}\delta u^{(1)}\right]\mathrm{d}t \qquad (2.8)
\end{aligned}
$$

Integration by parts for the term involving $\delta\dot{x}^{(p)}(t)$ is used to obtain equation (2.9), wherein the term $\dot{\Lambda}^{(p)}(t)$ is the time differential of $\Lambda^{(p)}(t)$.

$$
\begin{aligned}
\delta\overline{W} \;=\; &\sum_{p=2}^{NP}\left\{\left[\left[u^{(p)}\right]^T\frac{\partial\phi_1^{(p)}}{\partial x^{(p)}(t_p)}+\frac{\partial\phi_2^{(p)}}{\partial x^{(p)}(t_p)}\right]\delta x^{(p)}(t_p)\right.\\
&+\left[\left[u^{(p)}\right]^T\frac{\partial\phi_1^{(p)}}{\partial z^{(p)}(t_p)}+\frac{\partial\phi_2^{(p)}}{\partial z^{(p)}(t_p)}\right]\delta z^{(p)}(t_p)\\
&+\left[\left[u^{(p)}\right]^T\frac{\partial\phi_1^{(p)}}{\partial v^{(p)}}+\frac{\partial\phi_2^{(p)}}{\partial v^{(p)}}\right]\delta v^{(p)}+\left[\phi_1^{(p)}\right]^T\delta u^{(p)}\\
&+\left[\beta^{(p)}\right]^T\left[\left(\frac{\partial h_1^{(p)}}{\partial x^{(p-1)}(t_{p-1})}u^{(p)}+\frac{\partial h_2^{(p)}}{\partial x^{(p-1)}(t_{p-1})}\right)\delta x^{(p-1)}(t_{p-1})\right.\\
&+\left(\frac{\partial h_1^{(p)}}{\partial z^{(p-1)}(t_{p-1})}u^{(p)}+\frac{\partial h_2^{(p)}}{\partial z^{(p-1)}(t_{p-1})}\right)\delta z^{(p-1)}(t_{p-1})\\
&\left.+\left(\frac{\partial h_1^{(p)}}{\partial v^{(p)}}u^{(p)}+\frac{\partial h_2^{(p)}}{\partial v^{(p)}}\right)\delta v^{(p)}+h_1^{(p)}\delta u^{(p)}-\delta x^{(p)}(t_{p-1})\right]\\
&+\int_{t_{p-1}}^{t_p}\left[\left(\left[u^{(p)}\right]^T\frac{\partial L_1^{(p)}}{\partial x^{(p)}}(t)+\frac{\partial L_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\right.\\
&+\left(\left[u^{(p)}\right]^T\frac{\partial L_1^{(p)}}{\partial z^{(p)}}(t)+\frac{\partial L_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&\left.+\left(\left[u^{(p)}\right]^T\frac{\partial L_1^{(p)}}{\partial v^{(p)}}(t)+\frac{\partial L_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}+\left[L_1^{(p)}\right]^T\delta u^{(p)}\right]\mathrm{d}t\\
&+\int_{t_{p-1}}^{t_p}\left[\Lambda^{(p)}(t)\right]^T\left[\left(\frac{\partial f_1^{(p)}}{\partial x^{(p)}}(t)\,u^{(p)}+\frac{\partial f_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\right.
\end{aligned}
$$

$$
\begin{aligned}
&+\left(\frac{\partial f_1^{(p)}}{\partial z^{(p)}}(t)\,u^{(p)}+\frac{\partial f_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&+\left(\frac{\partial f_1^{(p)}}{\partial v^{(p)}}(t)\,u^{(p)}+\frac{\partial f_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}+f_1^{(p)}\delta u^{(p)}\Bigg]\,\mathrm{d}t\\
&+\int_{t_{p-1}}^{t_p}\left[\left[\dot{\Lambda}^{(p)}(t)\right]^T\delta x^{(p)}(t)\right]\mathrm{d}t\\
&+\left[\Lambda^{(p)}(t_{p-1})\right]^T\delta x^{(p)}(t_{p-1})-\left[\Lambda^{(p)}(t_p)\right]^T\delta x^{(p)}(t_p)\\
&+\int_{t_{p-1}}^{t_p}\left[\lambda^{(p)}(t)\right]^T\Bigg[\left(\frac{\partial g_1^{(p)}}{\partial x^{(p)}}(t)\,u^{(p)}+\frac{\partial g_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\\
&+\left(\frac{\partial g_1^{(p)}}{\partial z^{(p)}}(t)\,u^{(p)}+\frac{\partial g_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&+\left(\frac{\partial g_1^{(p)}}{\partial v^{(p)}}(t)\,u^{(p)}+\frac{\partial g_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}(t)+g_1^{(p)}\delta u^{(p)}\Bigg]\,\mathrm{d}t\\
&+\int_{t_{p-1}}^{t_p}\left[\mu^{(p)}(t)\right]^T\Bigg[\left(\frac{\partial c_1^{(p)}}{\partial x^{(p)}}(t)\,u^{(p)}+\frac{\partial c_2^{(p)}}{\partial x^{(p)}}(t)\right)\delta x^{(p)}(t)\\
&+\left(\frac{\partial c_1^{(p)}}{\partial z^{(p)}}(t)\,u^{(p)}+\frac{\partial c_2^{(p)}}{\partial z^{(p)}}(t)\right)\delta z^{(p)}(t)\\
&+\left(\frac{\partial c_1^{(p)}}{\partial v^{(p)}}(t)\,u^{(p)}+\frac{\partial c_2^{(p)}}{\partial v^{(p)}}(t)\right)\delta v^{(p)}+c_1^{(p)}\delta u^{(p)}\Bigg]\,\mathrm{d}t\;\Bigg\}\\
&+\Bigg[\left(\left[u^{(1)}\right]^T\frac{\partial \phi_1^{(1)}}{\partial x^{(1)}(t_1)}+\frac{\partial \phi_2^{(1)}}{\partial x^{(1)}(t_1)}\right)\delta x^{(1)}(t_1)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial \phi_1^{(1)}}{\partial z^{(1)}(t_1)}+\frac{\partial \phi_2^{(1)}}{\partial z^{(1)}(t_1)}\right)\delta z^{(1)}(t_1)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial \phi_1^{(1)}}{\partial v^{(1)}}+\frac{\partial \phi_2^{(1)}}{\partial v^{(1)}}\right)\delta v^{(1)}+\left[\phi_1^{(1)}\right]^T\delta u^{(1)}\Bigg]\\
&+\left[\beta^{(1)}\right]^T\left[\left(\frac{\partial h_1^{(1)}}{\partial v^{(1)}}\,u^{(1)}+\frac{\partial h_2^{(1)}}{\partial v^{(1)}}\right)\delta v^{(1)}+h_1^{(1)}\delta u^{(1)}-\delta x^{(1)}(t_0)\right]\\
&+\int_{t_0}^{t_1}\Bigg[\left(\left[u^{(1)}\right]^T\frac{\partial L_1^{(1)}}{\partial x^{(1)}}(t)+\frac{\partial L_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\left[u^{(1)}\right]^T\frac{\partial L_1^{(1)}}{\partial z^{(1)}}(t)+\frac{\partial L_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)
\end{aligned}
$$

$$
\begin{aligned}
&+\left(\left[u^{(1)}\right]^T \frac{\partial L_1^{(1)}}{\partial v^{(1)}}(t)+\frac{\partial L_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+\left[L_1^{(1)}\right]^T \delta u^{(1)}\Bigg]\,\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\Lambda^{(1)}(t)\right]^T\Bigg[\left(\frac{\partial f_1^{(1)}}{\partial x^{(1)}}(t)\,u^{(1)}+\frac{\partial f_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\frac{\partial f_1^{(1)}}{\partial z^{(1)}}(t)\,u^{(1)}+\frac{\partial f_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&+\left(\frac{\partial f_1^{(1)}}{\partial v^{(1)}}(t)\,u^{(1)}+\frac{\partial f_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+f_1^{(1)}\delta u^{(1)}\Bigg]\,\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\left[\dot{\Lambda}^{(1)}(t)\right]^T\delta x^{(1)}(t)\right]\mathrm{d}t\\
&+\left[\Lambda^{(1)}(t_0)\right]^T\delta x^{(1)}(t_0)-\left[\Lambda^{(1)}(t_1)\right]^T\delta x^{(1)}(t_1)\\
&+\int_{t_0}^{t_1}\left[\lambda^{(1)}(t)\right]^T\Bigg[\left(\frac{\partial g_1^{(1)}}{\partial x^{(1)}}(t)\,u^{(1)}+\frac{\partial g_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\frac{\partial g_1^{(1)}}{\partial z^{(1)}}(t)\,u^{(1)}+\frac{\partial g_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&+\left(\frac{\partial g_1^{(1)}}{\partial v^{(1)}}(t)\,u^{(1)}+\frac{\partial g_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+g_1^{(1)}\delta u^{(1)}\Bigg]\,\mathrm{d}t\\
&+\int_{t_0}^{t_1}\left[\mu^{(1)}(t)\right]^T\Bigg[\left(\frac{\partial c_1^{(1)}}{\partial x^{(1)}}(t)\,u^{(1)}+\frac{\partial c_2^{(1)}}{\partial x^{(1)}}(t)\right)\delta x^{(1)}(t)\\
&+\left(\frac{\partial c_1^{(1)}}{\partial z^{(1)}}(t)\,u^{(1)}+\frac{\partial c_2^{(1)}}{\partial z^{(1)}}(t)\right)\delta z^{(1)}(t)\\
&+\left(\frac{\partial c_1^{(1)}}{\partial v^{(1)}}(t)\,u^{(1)}+\frac{\partial c_2^{(1)}}{\partial v^{(1)}}(t)\right)\delta v^{(1)}+c_1^{(1)}\delta u^{(1)}\Bigg]\,\mathrm{d}t
\end{aligned}
\tag{2.9}
$$

For a stationary point, infinitesimal variations in the right hand side should yield no change to the performance index, i.e. $\delta\overline{W}=0$, and hence related terms must be chosen so that they always guarantee this. This leads to the following set of Euler-Lagrange equations and the Pontryagin Minimum Principle (Pontryagin et al., 1962).

The $\delta x^{(p)}(t)$, $\delta x^{(p)}(t_p)$ and $\delta x^{(p)}\left(t_{p-1}\right)$ terms are cancelled through the condition that the following differential equations and point conditions given by equations (2.10), (2.11) and (2.12)), respectively, hold:

$$\dot{\Lambda}^{(p)}(t) = -\left[\left[u^{(p)}\right]^T \frac{\partial L_1^{(p)}}{\partial x^{(p)}}(t) + \frac{\partial L_2^{(p)}}{\partial x^{(p)}}(t)\right]^T - \left[\frac{\partial f_1^{(p)}}{\partial x^{(p)}}(t)\, u^{(p)} + \frac{\partial f_2^{(p)}}{\partial x^{(p)}}(t)\right]^T \left[\Lambda^{(p)}(t)\right]$$
$$- \left[\frac{\partial g_1^{(p)}}{\partial x^{(p)}}(t)\, u^{(1)} + \frac{\partial g_2^{(p)}}{\partial x^{(p)}}(t)\right]^T \left[\lambda^{(p)}(t)\right] - \left[\frac{\partial c_1^{(p)}}{\partial x^{(p)}}(t)\, u^{(1)} + \frac{\partial c_2^{(p)}}{\partial x^{(p)}}(t)\right]^T \left[\mu^{(p)}(t)\right]$$

$$t_{p-1} \leq t \leq t_p \qquad p = 1, 2, \ldots, NP \tag{2.10}$$

$$\Lambda^{(p)}(t_p) = \left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial x^{(p)}(t_p)} + \frac{\partial \phi_2^{(p)}}{\partial x^{(p)}(t_p)}\right]^T + \left[\frac{\partial h_1^{(p+1)}}{\partial x^{(p)}(t_p)}\, u^{(p+1)} + \frac{\partial h_2^{(p+1)}}{\partial x^{(p)}(t_p)}\right]^T \beta^{(p+1)}$$

$$p = 1, 2, \ldots, NP - 1 \tag{2.11a}$$

$$\Lambda^{(p)}(t_p) = \left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial x^{(p)}(t_p)} + \frac{\partial \phi_2^{(p)}}{\partial x^{(p)}(t_p)}\right]^T \qquad p = NP \tag{2.11b}$$

$$\beta^{(p)} = \Lambda^{(p)}(t_{p-1}) \qquad p = 1, 2, \ldots, NP \tag{2.12}$$

Algebraic equations and point condition equations (2.13) and (2.14) must hold in order to cancel the $\delta z^{(p)}$ and $\delta z^{(p)}(t_p)$ terms, respectively.

$$\left[\left[u^{(p)}\right]^T \frac{\partial L_1^{(p)}}{\partial z^{(p)}}(t) + \frac{\partial L_2^{(p)}}{\partial z^{(p)}}(t)\right]^T + \left[\frac{\partial f_1^{(p)}}{\partial z^{(p)}}(t)\, u^{(p)} + \frac{\partial f_2^{(p)}}{\partial z^{(p)}}(t)\right]^T \left[\Lambda^{(p)}(t)\right]$$
$$+ \left[\frac{\partial g_1^{(p)}}{\partial z^{(p)}}(t)\, u^{(p)} + \frac{\partial g_2^{(p)}}{\partial z^{(p)}}(t)\right]^T \left[\lambda^{(p)}(t)\right] + \left[\frac{\partial c_1^{(p)}}{\partial z^{(p)}}(t)\, u^{(p)} + \frac{\partial c_2^{(p)}}{\partial z^{(p)}}(t)\right]^T \left[\mu^{(p)}(t)\right] = 0$$

$$t_{p-1} \leq t \leq t_p \qquad p = 1, 2, \ldots, NP \tag{2.13}$$

$$\left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial z^{(p)}(t_p)} + \frac{\partial \phi_2^{(p)}}{\partial z^{(p)}(t_p)}\right]^T + \left[\frac{\partial h_1^{(p+1)}}{\partial z^{(p)}(t_p)} u^{(p+1)} + \frac{\partial h_2^{(p+1)}}{\partial z^{(p)}(t_p)}\right]^T \beta^{(p+1)} = 0$$

$$p = 1, 2, \ldots, NP-1 \tag{2.14a}$$

$$\left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial z^{(p)}(t_p)} + \frac{\partial \phi_2^{(p)}}{\partial z^{(p)}(t_p)}\right]^T = 0 \qquad p = NP \tag{2.14b}$$

As per the Pontryagin Minimum Principle, the decision variables of the problem should be chosen to minimise the Hamiltonian. The Hamiltonian gradient conditions, taken from the coefficients of $\delta v^{(p)}$ and $\delta u^{(p)}$, are given by equations (2.15) to (2.16).

$$\begin{aligned}
\nabla_{v^{(p)}} H^{(p)} = & \left[\left[u^{(p)}\right]^T \frac{\partial \phi_1^{(p)}}{\partial v^{(p)}} + \frac{\partial \phi_2^{(p)}}{\partial v^{(p)}}\right]^T + \left[\frac{\partial h_1^{(p)}}{\partial v^{(p)}} u^{(p)} + \frac{\partial h_2^{(p)}}{\partial v^{(p)}}\right]^T \beta^{(p)} \\
& + \int_{t_{p-1}}^{t_p} \left[\left[\left[u^{(p)}\right]^T \frac{\partial L_1^{(p)}(t)}{\partial v^{(p)}} + \frac{\partial L_2^{(p)}(t)}{\partial v^{(p)}}\right]^T \right. \\
& + \left[\frac{\partial f_1^{(p)}(t)}{\partial v^{(p)}} u^{(p)} + \frac{\partial f_2^{(p)}(t)}{\partial v^{(p)}}\right]^T \Lambda^{(p)}(t) + \left[\frac{\partial g_1^{(p)}(t)}{\partial v^{(p)}} u^{(p)} + \frac{\partial g_2^{(p)}(t)}{\partial v^{(p)}}\right]^T \lambda^{(p)}(t) \\
& \left. + \left[\frac{\partial c_1^{(p)}(t)}{\partial v^{(p)}} u^{(p)} + \frac{\partial c_2^{(p)}(t)}{\partial v^{(p)}}\right]^T \mu^{(p)}(t)\right] \mathrm{d}t \\
& = 0
\end{aligned}$$

$$\begin{gathered} t_{p-1} \le t \le t_p \\ p = 1, 2, \ldots, NP \end{gathered} \tag{2.15}$$

$$\begin{aligned}
\nabla_{u^{(p)}} H^{(p)} = & \ \phi_1^{(p)} + \left[h_1^{(p)}\right]^T \beta^{(p)} \\
& + \int_{t_{p-1}}^{t_p} \left[L_1^{(p)} + \left[f_1^{(p)}\right]^T \Lambda^{(p)}(t) + \left[g_1^{(p)}\right]^T \lambda^{(p)}(t) + \left[c_1^{(p)}\right]^T \mu^{(p)}(t)\right] \mathrm{d}t \\
& = 0
\end{aligned}$$

$$
\begin{gathered}
t_{p-1} \leq t \leq t_p \\
p = 1, 2, \ldots, NP
\end{gathered}
\tag{2.16}
$$

As can be seen in equation (2.16), despite the interaction between the linear and nonlinear controls within the problem, the Hamiltonian gradient, in stage p, with respect to the linear control, $u^{(p)}$, in that stage, is independent of that control. This expression can be termed a "switching function" in the sense that it can cause the values of the controls in the set, u, to switch between stages, in order to minimise the Hamiltonian in each stage. This is elaborated as follows:

1. If, in a stage, p, the switching function is positive, the Hamiltonian is minimised when the control $u^{(p)}$ is at its lower bound, 0. On the other hand, if the switching function is negative in a stage, p, the Hamiltonian is minimised when the control $u^{(p)}$ is at its upper bound, 1. Thus, in a stage, p, the Hamiltonian is minimised when the linear control, $u^{(p)}$, takes values at either of its bounds, 0 or 1, depending on the sign of the switching function involved. This phenomenon of an optimal control action for a linear control occurring at either bound of its feasible region is called a "bang-bang" control behaviour (Bryson and Ho, 1975).

2. If, in a stage, p, the switching function becomes zero, the Hamiltonian gradient in that stage becomes insensitive to variations in the linear control, $u^{(p)}$, in that stage. In such cases, a bang-bang behaviour may not be observed for the linear control and the stage is called a singular arc.

Thus, when the controls, u, appear linearly in an MSMIOCP of the form of equation (2.4), these controls can be expected to exhibit a bang-bang behaviour with potential singular arcs.

However, as can be seen in equation (2.15), the Hamiltonian gradient, in stage p, with respect to the control $v^{(p)}$ in that stage, which appears nonlinearly in the problem, is not independent of that control. Hence, the controls, v, are not expected to exhibit a bang-bang behaviour.

From the above theoretical analysis, it is seen that in an MSMIOCP of the form of equation (2.4), when the binary controls, u, appear linearly, these controls can be expected to exhibit bang-bang behaviour and therefore can be expected to take values at either of their bounds in the optimal solution, apart from in those stages where singular arcs can occur. Thus, even if the controls, u, were considered continuous variables in the range $[0,1]$, rather than just discrete variables in the set $\{0,1\}$, due to the bang-bang behaviour, these controls can still be expected to take values of only 0 or 1 (that is, at their bounds) in the optimal solution. This has a major advantage in that, by considering these controls as continuous rather than discrete variables, the problem can be solved as a standard nonlinear optimisation problem, using the feasible path approach, without using mixed-integer optimisation methods, and yet optimum values of only binary nature (0 or 1) can be expected to be obtained for these controls.

Therefore, the integer restrictions on the controls, u, in equation (2.5g) can be relaxed and instead, these controls can be considered as continuous variables in the range $[0,1]$. A modified form of equation (2.5), that assumes such a relaxation is shown in equation (2.17). As can be seen, the binary restrictions on the controls, u, in equation (2.5g), have been replaced by the condition stating that these controls are continuous variables in the range $[0,1]$, in equation (2.17g). Problems of the form of equation (2.17) will be referred to as an "MSMIOCP with linear and relaxed binary controls" in this thesis. These are essentially standard nonlinear optimisation problems, as no discrete controls are involved and only an optimal set of continuous controls have to be identified, while using the feasible path approach to solve the differential equations.

$$\begin{aligned}\min_{u,v} W = \sum_{p=1}^{NP} \Bigg\{ & \left[\phi_1^{(p)}\left(x^{(p)}(t_p), z^{(p)}(t_p), v^{(p)}, t_p\right)\right]^T u^{(p)} + \phi_2^{(p)}\left(x^{(p)}(t_p), z^{(p)}(t_p), v^{(p)}, t_p\right) \\ & + \int_{t_{p-1}}^{t_p} \left[\left[L_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right]^T u^{(p)} + L_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] \mathrm{d}t \Bigg\}\end{aligned} \tag{2.17a}$$

subject to

$$\begin{gathered}\dot{x}^{(p)}(t) = \left[f_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right] u^{(p)} + f_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) \\ t_{p-1} \le t \le t_p \\ p = 1, 2, \ldots, NP\end{gathered} \tag{2.17b}$$

$$x^{(1)}(t_0) = \left[h_1^{(1)}\left(v^{(1)}\right)\right] u^{(1)} + h_2^{(1)}\left(v^{(1)}\right) \tag{2.17c}$$

$$
\begin{aligned}
x^{(p)}(t_{p-1}) = {} & \left[h_1^{(p)}\left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), v^{(p)}\right)\right]u^{(p)} \\
& + h_2^{(p)}\left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), v^{(p)}\right) \\
& p = 2, 3, \ldots, NP
\end{aligned}
\tag{2.17d}
$$

$$
\begin{aligned}
0 = \left[g_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right]u^{(p)} + g_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) \\
t_{p-1} \leq t \leq t_p \\
p = 1, 2, \ldots, NP
\end{aligned}
\tag{2.17e}
$$

$$
\begin{aligned}
\left[c_1^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right)\right]u^{(p)} + c_2^{(p)}\left(x^{(p)}(t), z^{(p)}(t), v^{(p)}, t\right) \leq 0 \\
t_{p-1} \leq t \leq t_p \\
p = 1, 2, \ldots, NP
\end{aligned}
\tag{2.17f}
$$

$$
\begin{aligned}
u^{(p)} \in [0, 1] \\
p = 1, 2, \ldots, NP
\end{aligned}
\tag{2.17g}
$$

$$
\begin{aligned}
v^{(p)} \in \mathscr{V} \\
p = 1, 2, \ldots, NP
\end{aligned}
\tag{2.17h}
$$

The optimal solutions for the problem given by equation (2.17) can be expected to exhibit a bang-bang behaviour for the controls, u, with potential singular arcs.

Bang-bang optimal control has previously been utilised in other applications. A brief overview of the publications in this area follows next.

Pure bang-bang optimal control problems (without singular arcs) have been demonstrated in minimum time problems for linear systems by Bellman et al. (1956) and for bilinear systems by Mohler (1973). Blakemore and Aris (1962) have shown that the control of the rate of cooling in a batch reactor, to obtain a given conversion in least time, exhibits bang-bang behaviour. Belghith et al. (1986) have demonstrated bang-bang behaviour for a component of control in building energy management. The optimal drug administration in cancer chemotheraphy has shown to result in bang-bang behaviour by Ledzewicz and Schättler (2002), wherein it is recommended to alternate periods of administering drugs at full dosage with complete rest periods without chemotherapy in between.

Zandvliet et al. (2007), however, have shown that for nonlinear optimisation problems, the bang-bang principle does not always hold. They have shown that when controls come linearly in relation to the continuous state variables, if the only constraints on the controls are upper and lower bounds, then bang-bang solutions can occur in combination with singular arcs. They have demonstrated this for reservoir flooding problems wherein the controls decide the operation of the valves of producer and injector wells. Thus, the predictions of the theoretical analysis carried out here is consistent with the results of Zandvliet et al. (2007).

Sager (2009) has presented a methodology to handle nonlinear dynamic systems involving discrete and continuous controls. Techniques are presented to reformulate the problem to avoid nonlinearities and enforce discrete controls via auxiliary binary controls that occur linearly in the system dynamics and exhibit a bang-bang behaviour. Heuristics, e.g. rounding or sum up rounding strategies or algorithms such as Branch & Bound are used to ensure integer solutions when singular arcs appear. This methodology has been used in the energy optimal operation mode of subway trains (Sager et al., 2009), the gear choices in time optimal car driving (Kirches et al., 2010) and the control of the tail deflection angle for the time optimal control of an F-8 aircraft (Sager, 2005), to name a few applications. In this thesis, there is no need for any such reformulation because the discrete controls already occur linearly in the system equations. It is worth mentioning, however, that the theoretical analysis' predictions of bang-bang behaviour for the linear controls, even when in combination with other continuous controls, are consistent with those of Sager (2009).

In this chapter, it is proposed to solve the problem of optimising maintenance scheduling and production in a process containing a single reactor using decaying catalysts, as an MSMIOCP with linear and relaxed binary controls, using the feasible path approach. This methodology can potentially offer a number of advantages over using mixed-integer methods to solve problems of this kind. These potential advantages are detailed next.

2.1.2 Potential advantages over mixed-integer methods

In Section 1.4, the drawbacks of using mixed-integer optimisation methods were presented as three major points. In this section, it is discussed how the use of the proposed solution methodology can potentially overcome those drawbacks and thereby be advantageous over the use of mixed-integer techniques in attempting to solve the problem under consideration here.

1. When mixed-integer methods are used, all differential equations present in the problem are approximated as a collection of steady state equations, which are then imposed as equality constraints in the optimisation phase. Following such practices can cause the problem to contain a very large number of potentially highly nonlinear constraint equations. In such cases, the optimiser could face difficulties in converging to a solution, which is further accentuated when a large number of differential equations are involved, longer time horizons are considered or higher accuracy is required. In addition, if a larger number catalyst loads are available to be used, the problem size becomes larger and this can lead to further difficulties in converging to a solution.

   However, in the methodology proposed here, the differential equations will be solved using the feasible path approach, without being considered as constraints in the optimisation step.
   Further, in this methodology, the binary controls to schedule catalyst changeover are going to be considered as continuous controls that appear linearly in the problem equations and therefore, are expected to exhibit a bang-bang behaviour. By virtue of this bang-bang behaviour, the 0 or 1 values for these controls will be obtained by just solving this problem as a standard nonlinear optimisation problem, without the use of mixed-integer optimisation methods. Hence, even if an infinite number of catalyst loads are available, the problem size will not increase as the bang-bang behaviour will decide how many catalyst loads to use and when to schedule catalyst changeovers.

   Thus, the resulting problem is expected to be of a much smaller size compared to when mixed-integer techniques are used and optimal solutions can be be obtained from random starting points, even when a large number of differential equations are involved, long time horizons are considered and a large number of catalyst loads are available.

   Hence, the proposed methodology is potentially more robust in converging to optimal solutions, in comparison to mixed-integer techniques.

2. As mentioned in the previous point, the mixed-integer techniques approximate the differential equations as a collection of steady state equations. This negates an accurate description of the dynamics of the problem within the time period in which those differential equations are approximated as such.

However, in the proposed methodology, a feasible path approach will be used, wherein state-of-the-art integrators are employed to solve the DAEs present in the problem. These integrators can usually solve even complicated DAEs to a very high accuracy.

Therefore, the solutions obtained using the proposed methodology are expected to be of greater accuracy and hence, potentially more reliable than those obtained using mixed-integer methodologies.

3. Mixed-integer techniques are combinatorial in nature, meaning that the computational effort to solve a problem using these methods increases exponentially with the number of integer decision variables involved in the problem. If mixed-integer techniques are used in the problem under consideration here, when a large number of catalyst loads are present, the computational effort involved in optimising the scheduling of catalyst changeovers will become huge and so, an enormous amount of computational power will be needed to solve the problem.

   However, as mentioned previously, in the proposed methodology, by virtue of a bang-bang behaviour for the catalyst changeover controls, the 0 or 1 values for these controls will be obtained by just solving this problem as a standard nonlinear optimisation problem, without the use of mixed-integer optimisation methods. Hence, no computational effort will have to be spent in deciding when to schedule catalyst changeovers or how many catalyst loads to use, as the bang-bang behaviour will take care of this. And this will be the case regardless of the number of catalyst loads involved, thereby making it possible for the methodology to obtain optimal maintenance schedules for catalyst replacements even if an infinite number of catalyst loads are available to be used.

   This feature of the proposed methodology makes it potentially more efficient in comparison to mixed-integer methods.

Thus, the proposed methodology of solving this problem as an MSMIOCP with linear and relaxed binary controls, using the feasible path approach, has great potential for providing robust, reliable and efficient solutions in comparison to mixed-integer techniques. A further potential advantage is that the feasible path approach can enable avoiding having to make the difficult compromise between accuracy and ease of convergence, which is faced by mixed-integer methods.

However, the use of the feasible path approach by this methodology implies that a large amount of computational effort is spent in solving the DAEs to a high accuracy at each iteration of the optimisation, even at those iterations away from the optimal solution. This could mean that the computational time to obtain solutions using this methodology could be quite high, far higher than the times required by mixed-integer methods to obtain solutions. While long computation times can be perceived as a drawback of this methodology, this drawback is outweighed by the robust, reliable and efficient solutions the methodology can potentially provide in comparison to the mixed-integer techniques. In addition, with the advent of high performance computing and parallel computing facilities in today's world, each DAE can be simulated entirely on a separate computer and further, any required gradient evaluations can also be parallelised within the computer on which each simulation occurs. The use of such facilities can significantly reduce computational time to obtain solutions when using this methodology.

Another limitation is that, since the problem is non-convex, only local solutions can be obtained by this methodology. However, even the use of mixed-integer techniques face this shortcoming. If the global optimum is to be identified while using the proposed methodology, several runs using different start points have to be performed. The high performance and parallel computing facilities would make such a task feasible as well.

This section describes the features of the optimal control methodology proposed to solve the problem of optimising maintenance scheduling and production in a process containing a single reactor using decaying catalysts. The advantages that this methodology can potentially offer over mixed-integer techniques to solve this problem have also been discussed. In the next section, different case studies of an industrial process are developed as per the proposed optimal control formulation. The sections following this describe the implementation procedures to solve these case studies and discuss the results obtained.

2.2 Case studies: Problem formulation

In this section, the problem of optimising maintenance scheduling and production in an industrial process containing a single reactor using decaying catalysts, is formulated as an MSMIOCP with linear and relaxed binary controls of the form of equation (2.17). A schematic of this industrial process is shown in Figure 2.2. Different case studies of the industrial process are considered, which differ either in the kinetics of the product formation reaction or the catalyst deactivation. The essential elements of the formulation are presented

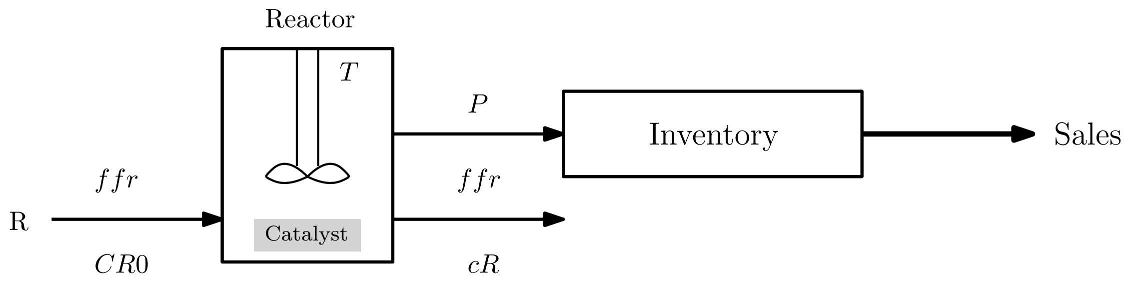


Fig. 2.2 A schematic of the process containing a single reactor using decaying catalysts.

in this section, and the solution implementation procedures and results obtained for each case study are discussed in the sections following this.

In the industrial problem addressed, the following assumptions apply:

1. The industrial process operates over a fixed time horizon, in the order of years. Each year is constituted by 12 months and there are a total of NM months, wherein each month is constituted by 4 weeks.

2. The industrial process functions according to a certain process model and is subject to operating constraints.

3. The reactor containing the deactivating catalyst is a Continuous Stirred Tank Reactor (CSTR) that is of known and fixed volume.

4. The catalyst performance decays with time and has to be replaced before it crosses a certain maximum age. Various forms of catalyst deactivation kinetics will be investigated in the different case studies.

5. The catalyst deactivation rate constant is taken to be independent of the temperature of operation.

6. There is a maximum number of catalyst loads that can be used over the given time horizon.

7. All available catalysts exhibit identical functioning and performance.

8. The time required to shut down the process, replace the catalyst and restart the process is taken to be one month, during which time no production occurs.

9. The main reaction is assumed to be of the form:

$$R \rightarrow Q \tag{2.18}$$

where R is the reactant and Q is the desired product. The different case studies will examine first and second order kinetics with respect to the reactant's concentration. Further, in each case study, the reaction rate will be considered separable from the catalyst activity.

10. The reaction rate constant exhibits an Arrhenius form of temperature dependence.
11. The feed inlet concentration is taken to be known and constant.
12. The flow rate of raw material to the reactor has to be specified on a weekly basis.
13. The flow rate of raw material to the reactor has an upper limit during catalyst operation and is stopped when the catalyst is being replaced.
14. The temperature of the reactor has to be specified on a weekly basis.
15. The temperature of the reactor can be operated only within fixed bounds during catalyst operation and is set to its lower bound during catalyst replacement.
16. The reactor is operated isothermally. No energy balances are considered.
17. During catalyst operation, the product produced is stored continuously as inventory.
18. The product is sold on a weekly basis.
19. The seasonal demand figures for the product are given.
20. The sales for each week is less than or equal to the customer demand for the product in that week.
21. There is a penalty corresponding to the unmet demand in each period.
22. The costs involved in the process are known and are subject to a known value of annual inflation. These include the sales price of the product, the cost of inventory, the cost of feed of raw material, the cost of catalyst changeover and the penalty for unmet demand. There are, however, no costs related to heating and cooling procedures.
23. There is no uncertainty regarding the values of any of the parameters involved [1].

Given the above assumptions, the optimisation model must determine the following values, which constitute the controls of the MSMIOCP:

[1] The consideration of uncertainty is presented in Chapter 4

(i) The catalyst changeover decision variable, $y(i)$, for each month, i, which determines whether a catalyst is in operation ($y(i) = 1$) or being replaced ($y(i) = 0$) during that month.

(ii) The feed flow rate to the reactor, $ffr(i, j)$, during each week, j, of each month, i.

(iii) The temperature of operation of the reactor, $T(i, j)$, during each week, j, of each month, i.

(iv) The amount of product sold, $sales(i, j)$, at the end of each week, j, of each month, i.

In the above list, $j \in \{1,2,3,4\}$ and $i \in \{1,2,...,NM\}$. The catalyst changeover decisions correspond to the binary controls, u, in equation (2.17g) while the other decision variables correspond to continuous controls, v, in equation (2.17h).

The state variables that characterise the MSMIOCP formulation of this industrial process include the following:

(i) The catalyst age, cat_age

(ii) The catalyst activity, cat_act

(iii) The concentration of the reactant at the exit of the reactor, cR

(iv) The inventory level, inl

(v) The cumulative inventory costs, cum_inc.

These state variables are determined by the decision variables' values at any time using a set of ordinary differential equations (ODEs) which constitute the process model. In the following, process models to describe different case studies of the industrial process are formulated. These ODEs apply for week $j \in \{1,2,3,4\}$ of month $i \in \{1,2,...,NM\}$ of the process and are of the form of equation (2.17b). Unless specified, a particular model equation applies to all case studies:

1. The catalyst age varies linearly with time when the catalyst is in operation $(y(i) = 1)$ but does not increase at times of catalyst replacement $(y(i) = 0)$. Hence, the differential equation describing the catalyst age at all times, that takes into account both scenarios, is given by:

$$\frac{\mathrm{d}\,(cat\_age)}{\mathrm{d}t} = y(i) \tag{2.19}$$

2. The catalyst activity decays according to a deactivation rate law during times of catalyst operation $(y(i) = 1)$ but experiences no change during times of catalyst replacement $(y(i) = 0)$, when there is no production occurring. Thus, the differential equation for the catalyst activity, that takes into account both scenarios, takes the form:

$$\frac{\mathrm{d}\,(cat\_act)}{\mathrm{d}t} = y(i) \times rD \tag{2.20}$$

where rD is the rate of catalyst deactivation. Different models of catalyst deactivation kinetics are considered as separate case studies:

<u>Case Study A</u>: Composition independent catalyst deactivation

$$rD = -Kd \times cat\_act \tag{2.21}$$

<u>Case Study B</u>: Reactant concentration dependent catalyst deactivation

$$rD = -Kd \times cat\_act \times cR \tag{2.22}$$

<u>Case Studies C and D</u>: Product concentration dependent catalyst deactivation

$$rD = -Kd \times cat\_act \times (CR0 - cR) \tag{2.23}$$

where Kd is the deactivation rate constant and $CR0$ is the reactant entry concentration.

3. The reactor is assumed to be completely stirred and so the reactant exit concentration (cR) is obtained from the generic mass balance equation of a CSTR during times of catalyst operation $(y(i) = 1)$. However, during catalyst replacement $(y(i) = 0)$, no reaction occurs and the reactor is assumed to be filled with fresh, unreacted reactant at the entry concentration $(CR0)$, to be used by the new catalyst after replacement. The differential equation that accounts for both scenarios is given by:

$$\frac{\mathrm{d}\,(V \times cR)}{\mathrm{d}t} = [ffr(i,j) \times (CR0 - cR)] - [y(i) \times (V \times rR)] \tag{2.24}$$

where V is the volume of the reactor and rR is the rate of reaction (2.18). The case studies consider different forms of rR:

Case Studies A, B and C: First order kinetics for reaction (2.18)

$$rR = Kr \times cat\_act \times cR \tag{2.25}$$

Case Study D: Second order kinetics for reaction (2.18)

$$rR = Kr \times cat\_act \times cR^2 \tag{2.26}$$

where Kr is the rate constant. For all case studies, Kr is assumed to exhibit an Arrhenius form of temperature dependence, of the form:

$$Kr = Ar \times \exp\left(-\frac{Ea}{R_g \times T(i,j)}\right) \tag{2.27}$$

where Ar is the pre-exponential factor, Ea is the activation energy for the reaction and R_g is the universal gas constant.

4. It is assumed that whatever product is produced is stored as inventory before being sold at the end of the week. During catalyst operation $(y(i) = 1)$, the increase in inventory level at any time depends on the rate of production ($= V \times rR$) of the product chemical, but during catalyst replacement $(y(i) = 0)$, there is no increase in inventory level. Hence, the differential equation that provides a description of the inventory level (inl) for both scenarios is given by:

$$\frac{\mathrm{d}(inl)}{\mathrm{d}t} = y(i) \times (V \times rR) \tag{2.28}$$

where the expression for rR depends on the case study.

5. Finally, the increase in the cumulative inventory cost (cum_inc) at any time depends on the inventory level at that time and the Inventory Cost Factor (icf) (adjusted for inflation), which stipulates the cost per unit product per unit time:

$$\frac{\mathrm{d}(cum\_inc)}{\mathrm{d}t} = inl \times icf \tag{2.29}$$

The icf at any time is given by the following equation:

$$icf = base\_icf \times (1 + inflation)^{\lfloor i/12 \rfloor} \tag{2.30}$$

where $base\_icf$ is the inventory cost factor before inflation, $inflation$ is the annual inflation rate and $\lfloor\cdot\rfloor$ is the greatest integer function.

For each case study, the process model is solved repeatedly over a weekly time span, which corresponds to one stage of the MSMIOCP. In order to solve these ODEs, for each stage, suitable initial conditions have to be provided. The initial conditions for week 1 of month 1 are assumed to be known and are of the form of equation (2.17c). The initial conditions for the other stages are obtained using junction conditions between two successive stages of the process, of the form of equation (2.17d).

The initial conditions corresponding to week 1 of month 1, represented as $init\_var(1,1)$ for variable var, are as follows:

1. The initial catalyst age is that of a fresh catalyst, which is zero:

$$init\_cat\_age\,(1,1) \quad = \quad 0 \tag{2.31}$$

2. The initial catalyst activity is that of a fresh catalyst $(start\_cat\_act)$:

$$init\_cat\_act\,(1,1) \quad = \quad start\_cat\_act \tag{2.32}$$

3. At the start of the process, the reactor is filled with the reactant R at its entry concentration $CR0$. Hence, the initial exit concentration is given by:

$$init\_cR\,(1,1) \quad = \quad CR0 \tag{2.33}$$

4. There is no inventory at the beginning of the process, and so:

$$init\_inl\,(1,1) \quad = \quad 0 \tag{2.34}$$

5. There is no inventory at the start of the process and so the initial cumulative inventory cost is given by:

$$init\_cum\_inc\,(1,1) \quad = \quad 0 \tag{2.35}$$

The junction conditions are described next. For each state variable, the junction condition is used to provide the initial condition to solve the respective differential equation in the weeks other than week 1 of month 1 of the time horizon, by providing a relationship between the state variable value at the beginning of that week and the end of the previous week. As such, these junction conditions differ depending on whether the catalyst is in operation $(y(i)=1)$ or is being replaced $(y(i)=0)$ during that month. In the following text, the expressions $init\_var(i,\, j)$ and $end\_var(i,\, j)$ indicate the initial and end conditions, respectively for the variable *var*, for week j of month i:

1. During months of catalyst operation $(y(i)=1)$, the initial catalyst age for a week corresponds to the catalyst age at the end of the previous week. But during months of catalyst replacement $(y(i)=0)$, the catalyst age has to be set to zero, the age of a new catalyst. The junction conditions that describe both scenarios is given by:

$$\begin{aligned} init\_cat\_age(i,\, j+1) &= end\_cat\_age(i,\, j) \\ j = 1,2,3 \qquad & i = 1,2,\ldots,NM \end{aligned} \tag{2.36a}$$

$$\begin{aligned} init\_cat\_age(i,\, 1) &= y(i) \times end\_cat\_age(i-1,\, 4) \\ & i = 2,3,\ldots,NM \end{aligned} \tag{2.36b}$$

2. During months of catalyst operation $(y(i)=1)$, the initial catalyst activity for the week corresponds to the catalyst activity at the end of the previous week. However, during months of catalyst replacement $(y(i)=0)$, the catalyst activity has to be reset to the activity corresponding to that of a fresh catalyst, which remains the same throughout the duration of month i. The junction conditions that describe both scenarios are given by:

$$\begin{aligned} init\_cat\_act(i,\, j+1) &= end\_cat\_act(i,\, j) \\ j = 1,2,3 \qquad & i = 1,2,\ldots,NM \end{aligned} \tag{2.37a}$$

$$\begin{aligned} init\_cat\_act(i,\, 1) &= [y(i) \times end\_cat\_act(i-1,\, 4)] + [(1-y(i)) \times start\_cat\_act] \\ & i = 2,3,\ldots,NM \end{aligned} \tag{2.37b}$$

3. During months of catalyst operation $(y(i)=1)$, the exit concentration at the beginning of a week corresponds to the exit concentration at the end of the previous week. And during months of catalyst replacement $(y(i)=0)$, an artificial condition is imposed wherein the reactor is filled with reactant at entry concentration $CR0$, ready to be used by the fresh catalyst at the beginning of the next month. So, the junction conditions take the form:

$$\begin{aligned} init\_cR(i,\, j+1) &= end\_cR(i,\, j) \\ j = 1,2,3 \qquad & i = 1,2,\ldots,NM \end{aligned} \tag{2.38a}$$

$$init\_cR(i,\,1) = [y(i) \times end\_cR(i-1,\,4)] + [(1-y(i)) \times CR0] \quad i = 2,3,\ldots,NM \tag{2.38b}$$

4. At the end of a week, an amount, $sales(i,\,j)$ of the stored product is sold. Thus, the initial inventory level for the week corresponds to the inventory present after the sales at the end of the previous week. The following junction conditions apply during months of catalyst operation as well as catalyst replacement, as the sales do not cease at any time:

$$init\_inl\,(i,\,j+1) \;=\; end\_inl(i,\,j) - sales(i,\,j) \quad j = 1,2,3 \qquad i = 1,2,\ldots,NM \tag{2.39a}$$

$$init\_inl\,(i,\,1) \;=\; end\_inl(i-1,\,4) - sales\,(i-1,\,4) \quad i = 2,3,\ldots,NM \tag{2.39b}$$

5. The inventory cost accumulated until the beginning of a week is equal to the value of the inventory cost accumulated until the end of the previous week and the following junction conditions apply regardless of whether the catalyst is being used or replaced:

$$init\_cum\_inc\,(i,\,j+1) \;=\; end\_cum\_inc(i,\,j) \quad j = 1,2,3 \qquad i = 1,2,\ldots,NM \tag{2.40a}$$

$$init\_cum\_inc\,(i,\,1) \;=\; end\_cum\_inc(i-1,\,4) \quad i = 2,3,\ldots,NM \tag{2.40b}$$

The initial conditions (2.31) – (2.35) and junction conditions (2.37) – (2.40) enable a solution for the ODEs for all stages, and thereby obtain the values of the state variables for the entire time horizon.

The obtained state variables, along with the control variables, are required to fulfil some constraints, which represent the operational limits of the process and restrictions on the values of the controls to be chosen. The constraints, of the form of equation (2.17f), that apply in this formulation of the industrial process, for week $j \in \{1,2,3,4\}$ of month $i \in \{1,2,...,NM\}$, are as follows:

1. In the context of the formulation as an MSMIOCP with linear and relaxed binary controls, the catalyst changeover decision variables $y\,(i)$, for a month i, are considered

continuous variables that vary between 0 and 1 (but are expected to take only 0 or 1 values due to the bang-bang nature of the formulation), and so the following bounds are imposed:

$$0 \leq y(i) \leq 1 \tag{2.41}$$

2. The flow rate of raw material to the reactor has an upper limit (FU) at which it can operate. Hence, the following bounds are set on the feed flow rate for each week:

$$0 \leq ffr(i,j) \leq FU \tag{2.42}$$

3. The sales in each week are assumed to be less than or equal to the demand for the product in that week $(demand(i,j))$. Hence, the following bounds on the sales at the end of each week are imposed:

$$0 \leq sales(i,j) \leq demand(i,j) \tag{2.43}$$

4. The temperature of the reactor operates between known, fixed lower and upper bounds, TL and TU, respectively. Hence, the following bounds are set on the weekly temperature of operation of the reactor:

$$TL \leq T(i,j) \leq TU \tag{2.44}$$

5. During times of catalyst replacement, the process is shut down and so the flow of raw material to the reactor stops. The following constraint ensures that the weekly feed flow rate remains below the upper bound during times of catalyst operation $(y(i) = 1)$ and drops to zero when there is catalyst replacement $(y(i) = 0)$.

$$ffr(i,j) - [FU \times y(i)] \leq 0 \tag{2.45}$$

6. When the process is shut down for catalyst replacement, the temperature of the reactor is required to drop to its lower bound. This condition is imposed using the following constraint which ensures that the temperature for the week remains between its bounds during times of catalyst operation $(y(i) = 1)$ and drops to the lower bound when there is catalyst replacement $(y(i) = 0)$:

$$TL \leq T(i,j) \leq [(TU - TL) \times y(i)] + TL \tag{2.46}$$

7. There is only a certain number of catalysts available to be used by the process. The limit on the maximum number of catalyst changeovers (n) allowed is imposed using the following constraint:
$$\sum_{i=1}^{NM} y(i) \geq NM - n \tag{2.47}$$

8. In order to ensure that more product than available is not sold, the inventory level at the end of each week should be greater than the sales for the week. This is imposed using the following constraint:
$$end\_inl(i,\, j) - sales(i, j) \quad \geq \quad 0 \tag{2.48}$$

9. The catalyst undergoes deactivation over time and has to be replaced before it crosses a certain maximum age (max_cat_age). As the the decision on whether to replace a catalyst or not is made on a monthly basis, it is sufficient to ensure that the catalyst age does not cross this limit at the end of each month i:
$$end\_cat\_age\,(i, 4) \quad \leq \quad max\_cat\_age \tag{2.49}$$

   It is noted that a limit on the maximum duration of catalyst use can be imposed by using a catalyst activity based constraint in place of a catalyst age based constraint. In that case, the constraint would be of the form, $end\_cat\_act\,(i, 4) \geq min\_cat\_act$, where min_cat_act is the minimum allowable catalyst activity.

The aim is to maximise the profits or minimise the costs of the process under the influence of these ODEs, initial conditions, junction conditions and constraints. The net costs of the process are represented by the objective function of this problem, of the form of equation (2.17a), and comprises the following elements:

1. The Gross Revenue from Sales (GRS)
   This term represents the revenue for the process from the net sales of the product chemical over the whole time horizon:
$$GRS \quad = \quad \sum_{i=1}^{NM} \sum_{j=1}^{4} psp(i, j) \times sales(i, j) \tag{2.50}$$

   where $psp(i, j)$ is the sales price per unit product for week j of month i, adjusted for inflation at that time:

$$psp(i,j) = base\_psp \times (1 + inflation)^{\lfloor i/12 \rfloor} \tag{2.51}$$

where $base\_psp$ is the unit product sales price before inflation.

2. The Total Inventory Costs (TIC)
   This term represents the net storage costs for the product over the whole time horizon and is obtained from the solution of the ODEs for the state variable cum_inc at the end of the final week of the process:

$$TIC = end\_cum\_inc(NM, 4) \tag{2.52}$$

3. The Total Costs of Catalyst Changeovers ($TCCC$)
   The total expenditure for the catalyst changeover operations is:

$$TCCC = \sum_{i=1}^{NM} crc(i) \times (1 - y(i)) \tag{2.53}$$

   where $crc(i)$ is the cost of the catalyst replacement operation for month i, adjusted for inflation at that time:

$$crc(i) = base\_crc \times (1 + inflation)^{\lfloor i/12 \rfloor} \tag{2.54}$$

   where $base\_crc$ is the cost of a catalyst changeover operation before inflation. It is highlighted that the terms within the summation remain non-zero only during the times of catalyst replacement ($y(i) = 0$) and only these terms contribute to the total costs.

4. The Net Penalty for Unmet Demand ($NPUD$)
   The unmet demand in each week ($unmet\_demand(i,j)$) is the quantity of product by which the sales falls short of the demand in that week:

$$\begin{aligned} unmet\_demand(i,j) &= demand(i,j) - sales(i,j) \\ j = 1,2,3,4 \quad & \quad i = 1,2,\ldots,NM \end{aligned} \tag{2.55}$$

   There is a penalty associated with this unmet demand and the net penalty costs over the entire time horizon is given by:

$$NPUD = \sum_{i=1}^{NM} \sum_{j=1}^{4} pen(i,j) \times unmet\_demand(i,j) \tag{2.56}$$

where $pen(i,j)$ is the penalty cost of unmet demand per unit product for week j of month i, adjusted for inflation at that time:

$$pen(i,j) \quad = \quad base\_pen \times (1 + inflation)^{\lfloor i/12 \rfloor} \tag{2.57}$$

where $base\_pen$ is the penalty cost of unmet demand per unit product before inflation.

5. <u>The Total Flow Costs (TFC)</u>
   This term represents the net expenditure on the feed of raw material to the reactor and is given by:

$$TFC = \sum_{i=1}^{NM} \sum_{j=1}^{4} cof(i,j) \times ffr(i,j) \tag{2.58}$$

   where $cof(i,j)$ is the cost of raw material per unit volume per week for week j of month i, adjusted for inflation at that time:

$$cof(i,j) \quad = \quad base\_cof \times (1 + inflation)^{\lfloor i/12 \rfloor} \tag{2.59}$$

   where $base\_cof$ is the cost of raw material per unit volume per week before inflation.

If the Net Costs are represented by NC, the objective function for this optimisation problem takes the form:

$$\min NC \quad = \quad -GRS + TIC + TCCC + NPUD + TFC \tag{2.60}$$

The essential elements of the problem formulation have now been described in detail. The aim is to make the appropriate decisions in order to minimise the net costs (or maximise the net profit) of the industrial process, when subject to the process model, initial and junction conditions and the constraints. It is highlighted that the catalyst changeover decision variables (y) occur linearly in all elements of the problem formulation. Thus, these variables are expected to exhibit a bang-bang behaviour in the optimal solution and the constraint, $y(i) \in [0,1]$ is equivalent to $y(i) = \{0,1\}$.

The elements of the problem set up here are similar to that in Houze et al. (2003) and Bizet et al. (2005). However, those publications did not reveal any parameters used in their studies, citing confidentiality reasons. So, in this article, case studies were created using a set of constructed parameter values, which have been mentioned in Table 2.1. The time horizon chosen here is 3 years, which is more realistic in present day industries compared to the much longer durations studied in Bizet et al. (2005).

The problem size details for the chosen time horizon, applicable for all case studies, are shown in Table 2.2. It is important to note that the number of variables and constraints in this formulation are much smaller than if MINLP approaches were used, in which case there would have been thousands of decision variables and tens of thousands of constraints present.

Table 2.1 List of parameters.

Parameter Symbol	**Value**
Ar	885 (1/day)
$base\_cof$	\$ 210 /week
$base\_crc$	\$ 10^7
$base\_icf$	\$ 0.01 /(kmol day)
$base\_pen$	\$ 1250 /kmol
$base\_psp$	\$ 1000 /kmol
$CR0$	1 kmol/m^3
$demand$	1st quarter of year: 8000 kmol/week
	2nd quarter of year: 7200 kmol/week
	3rd quarter of year: 3300 kmol/week
	4th quarter of year: 4500 kmol/week
Ea	30,000 J/gmol
FU	9600 m^3/day
$inflation$	5%
Kd	Case Study A: 0.0024 (1/day)
	Case Study B: 0.0024 (1/(day . kmol/m^3))
	Case Studies C, D: 0.024 (1/(day . kmol/m^3))
max_cat_age	504 days (= 1.5 years)
n	5
NM	36 months (= 3 years)

Table 2.1 List of parameters.

Parameter Symbol	**Value**
R_g	8.314 J/(gmol.K)
$start\_cat\_act$	1.0
TL	400 K
TU	1000 K
V	50 m^3

Table 2.2 Problem size specifications, applicable for each case study.

Property		**Size**
Ordinary differential equations		720
Decision variables	Catalyst changeover actions (y)	36
	Feed flow rate (ffr)	144
	Sales ($sales$)	144
	Temperature (T)	144
	Total	*468*
Constraints	Constraints (2.41)	72
	Constraints (2.42)	288
	Constraints (2.43)	288
	Constraints (2.44)	288
	Constraints (2.45)	144
	Constraints (2.46)	288
	Constraint (2.47)	1
	Constraints (2.48)	144
	Constraints (2.49)	36
	Total	*1549*

In the next sections, the problem solution implementation details will be discussed and the results obtained will be presented. As will be seen, the complex nature of the problem caused complications in obtaining solutions using the solvers currently available. Different solution implementations were attempted on different solvers: Implementation I was performed on MATLAB and Implementation II was carried out in Python, each of which had their own relative advantages.

2.3 Implementation I: Details, results and discussions

2.3.1 Implementation I details

Implementation I was performed on MATLAB® R2018a with its Optimisation Toolbox™ (MATLAB and Optimisation Toolbox, 2018), as a code that solves a standard multistage optimal control problem using the feasible path approach, by linking an ODE solver with the optimiser $fmincon$. Two types of ODE solvers were tried: the *ode15s* solver available on MATLAB® R2018a (Shampine and Reichelt, 1997) and the *IDAS* solver of sundialsTB, a MATLAB interface to the open-source set of differential-algebraic equation solvers, SUNDIALS (Serban, 2009). In both cases, the solver was designated to have an absolute tolerance of 10^{-6} and a relative tolerance of 10^{-4}. The Jacobian was provided to the solvers to improve its reliability and efficiency. It was found that *IDAS* of sundialsTB was faster in computation compared to *ode15s* and so was preferred for this implementation.

The optimisation on $fmincon$ was performed using the Sequential Quadratic Programming (SQP) algorithm (Nocedal and Wright, 2006) with the following convergence criteria: constraint tolerance of 10^{-3}, step tolerance of 10^{-3} and optimality tolerance of 10^{-4}. A forward finite difference scheme was used for the estimation of gradients. Given the wide variation in the magnitude of the different decision variables (*e.g.* $y \in [0,1]$, but $sales \sim 10^3$), the starting points to the optimiser were scaled down using the respective upper bounds of each decision variable to avoid scaling problems in the optimisation. Further, in order to accelerate convergence, constraint (2.48) was scaled down by a factor of 10^3 and the objective function value was scaled down by a factor of 10^6.

In order to demonstrate the robustness of the developed methodology, it was desired to obtain a solution from a set of random values for the initial guesses of the decision variables to the optimiser. However, it was important to ensure that the set of random starting points were a set of 'feasible' points. Using highly infeasible starting points in this problem of complex nature could cause great difficulties to the optimiser in converging to a solution.

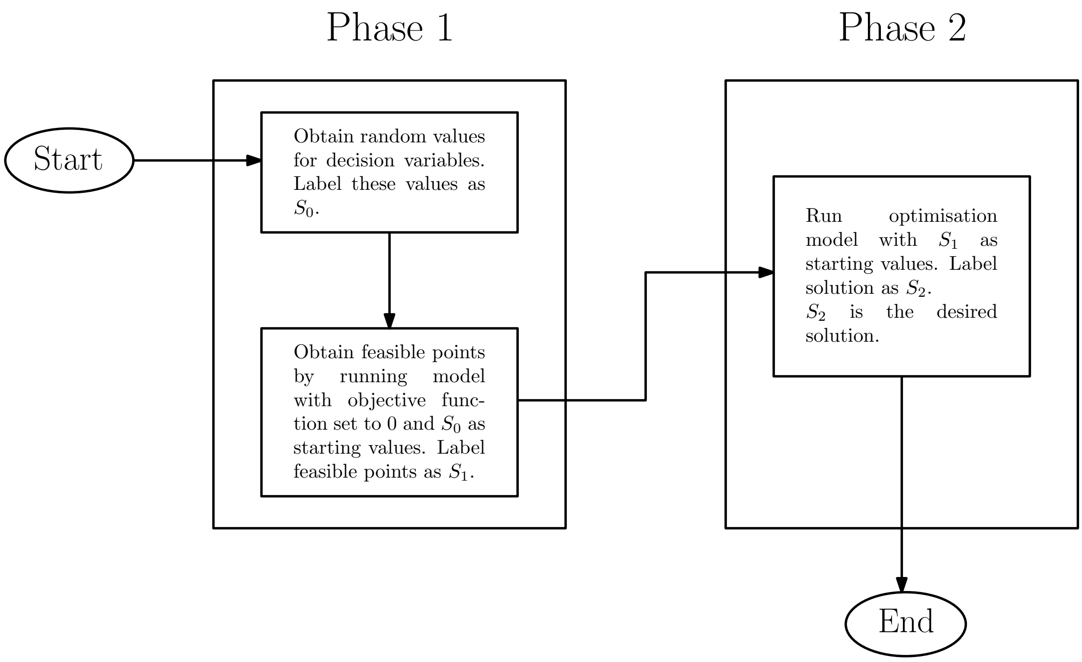


Fig. 2.3 An algorithmic flowchart for the procedure of Implementation I.

So in the initial part of Implementation I called Phase 1, a set of feasible start points for the decision variables was obtained by first generating a set of random points using the *rand* function in MATLAB® and running the optimisation model with the objective function set to zero. These feasible points were then used as the starting values for the actual optimisation problem in Phase 2 of the implementation. An algorithmic flowchart for the procedure of Implementation I is shown in Figure 2.3.

The implementation was performed on a 3.2 GHz Intel Core i5, 16 GB RAM, Windows machine running on Microsoft Windows 7 Enterprise. Since the problem is non-convex, multiple runs were performed with different starting points. Test runs were performed using the Parallel Computing Toolbox™ on MATLAB® to compare the computational times between parallelising the gradient evaluations versus parallelisation of a loop of multiple start points using a $parfor$ loop, and the latter was found to be faster. So, using the $parfor$ loop for parallelisation, 50 runs were attempted for each case study.

Table 2.3 Implementation I performance details.

Case Study	**Number of runs converging successfully (out of 50 runs)**	**Number of runs converging prematurely (out of 50 runs)**	**Number of runs crashing due to integration problems (out of 50 runs)**
Case Study A	13	28	9
Case Study B	22	23	5

2.3.2 Implementation I: General performance discussion

It was found that Implementation I had limited success when applied to Case Studies A and B whereas for Case Studies C and D, the technique failed completely.

With regard to Case Studies A and B, while some runs exhibited a very good bang-bang behaviour for the catalyst changeover controls, in many other simulations, the runs either converged prematurely to poor solutions or crashed due to the integrator failing (Table 2.3). In addition, even in the set of successful runs, the $fmincon$ optimiser experienced considerable "numerical noise" in identifying the optimal values of the feed flow rates in that there were significant distortions in the obtained profiles of these controls. It is unclear as to why such disturbances were observed only for these controls, but the presence of this numerical noise suggests inadequacies of the $fmincon$ optimiser.

Statistics regarding the solutions obtained and the computational effort involved, for the successful runs of Case Studies A and B are given in Tables 2.4 and 2.5, respectively. In Table 2.4, in the sub-column titled, ‘Max’, under the column titled, ‘Profit (Million $)’, the two rows indicate the maximum among the 13 profits obtained from the 13 successful runs of Case Study A and the maximum among the 22 profits obtained from the 22 successful runs of Case Study B. In the sub-columns titled, ‘Min’ and ‘Mean’, the two rows indicate the minimum and mean profits among the aforementioned values for the two case studies. Analogous explanations hold for the values presented in the two rows under the sub-columns titled, ‘Max’, ‘Min’, ‘Mean’ and ‘Mode’ within the columns titled, ‘Number of catalyst replacements’ and ‘Catalyst age (days)’ in Table 2.4 and under the columns titled ‘Phase 1’ and ‘Phase 2’ within the columns titled ‘Number of SQP iterations’ and ‘CPU time (seconds)’ in Table 2.5.

Table 2.4 Implementation I solution statistics.

Case Study	**Profit (Million $)**			**Number of catalyst replacements**			**Catalyst Age (days)**		
	Max	**Min**	**Mean**	**Max**	**Min**	**Mode**	**Max**	**Min**	**Mean**
Case Study A (For 13 successful runs)	438.08	345.6470	392.719	5	2	4	420	28	189.2
Case Study B (For 22 successful runs)	475.225	408.3674	443.0515	5	2	4	392	28	197.6

Table 2.5 Implementation I size statistics.

Case Study	**Number of SQP Iterations**						**CPU time (seconds)**					
	Phase 1			**Phase 2**			**Phase 1**			**Phase 2**		
	Max	**Min**	**Mean**	**Max**	**Min**	**Mean**	**Max**	**Min**	**Mean**	**Max**	**Min**	**Mean**
Case Study A (For 13 successful runs)	4	3	4	130	59	90	495.5	262.8	407.1	20383	6738	12009
Case Study B (For 22 successful runs)	4	3	4	188	52	93	489.3	333.1	440	34855	6134	13394

Further insights regarding the distribution of the solutions presented in Table 2.4 are provided in Figures 2.4 – 2.6. Following are comments regarding these figures:

- Figure 2.4 shows the distributions of the profits obtained from the successful runs of Case Studies A and B, when using Implementation I. In the histogram presented in subplot (a) of this figure, the height of each bin represents the number of runs out of the 13 successful runs of Case Study A that result in profit values within the range specified by the horizontal edges of that bin. A similar explanation holds for the histogram relating to Case Study B in subplot (b) of the figure.

- Figure 2.5 shows the distribution of the number of catalyst replacements obtained in the successful runs of Case Studies A and B, when using Implementation I. In the histogram presented in subplot (a) of this figure, the height of each bin represents the number of runs out of the 13 successful runs of Case Study A that involved the number of catalyst replacements given by the midpoint of the horizontal width of the bin. A similar explanation holds for the histogram relating to Case Study B in subplot (b) of the figure. It is noted that the maximum allowable number of catalyst replacements, as per the invented set of parameters used, is 5.

- Figure 2.6 shows the distribution of the ages of all catalyst used from all the successful runs of Case Studies A and B, when using Implementation I. In the histogram presented in subplot (a) of this figure, the height of each bin represents the number of catalysts, out of all the catalysts used from all the 13 successful runs of Case Study A, that were used up to the age given by the midpoint of the horizontal width of the bin. A similar explanation holds for the histogram relating to Case Study B in subplot (b) of the figure. It is noted that since catalysts can be replaced only at the end of a month, all catalyst ages are multiples of 28 and the maximum allowable age of each catalyst, as per the invented set of parameters used, is 504 days.

A presentation of the distribution of the number of SQP iterations and CPU times in Table 2.5, however, has not been done because these are dependent on the computer used to obtain solutions, and hence, cannot be generalised.

With regard to Case Studies C and D, however, every single run crashed showing an error with the integration. These unexpected integration problems were experienced by both sets of ODE solvers which were tried. These problems suggest inadequacies of the available MATLAB ODE integrator suites in integrating the more nonlinear differential equations, such as those of Case Studies C and D.

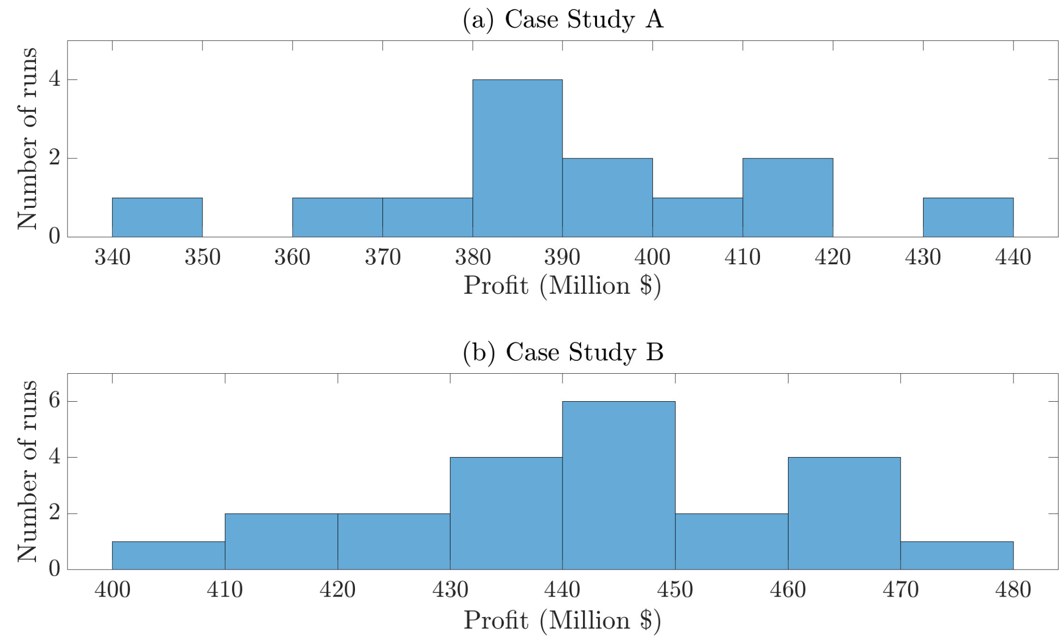


Fig. 2.4 The distribution of the profits obtained using Implementation I for (a) the 13 successful runs of Case Study A and (b) the 22 successful runs of Case Study B

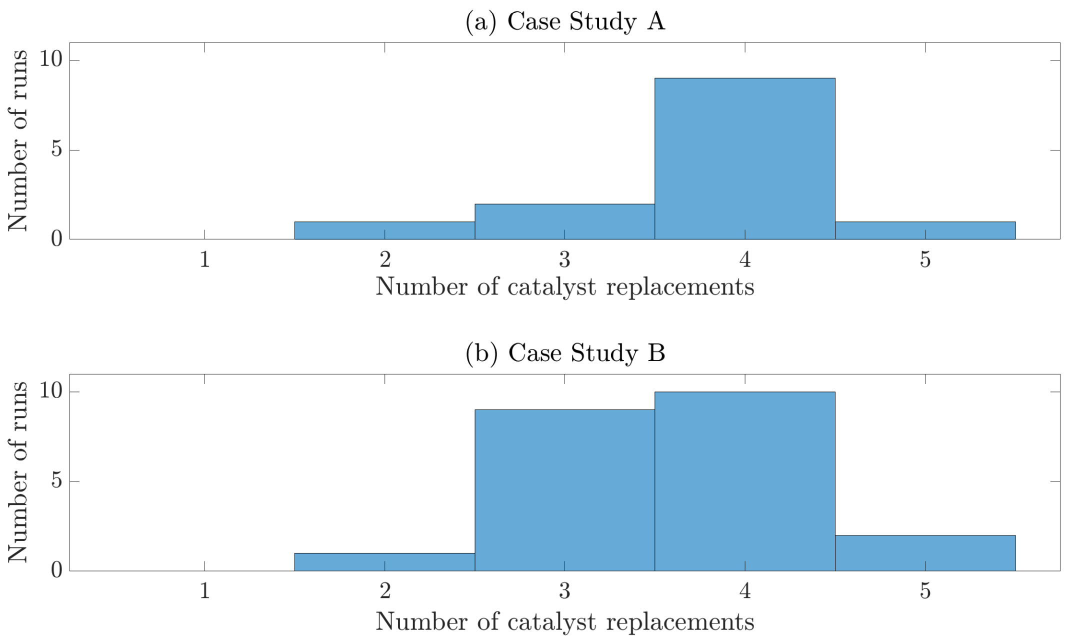


Fig. 2.5 The distribution of the number of catalyst replacements obtained using Implementation I for (a) the 13 successful runs of Case Study A and (b) the 22 successful runs of Case Study B

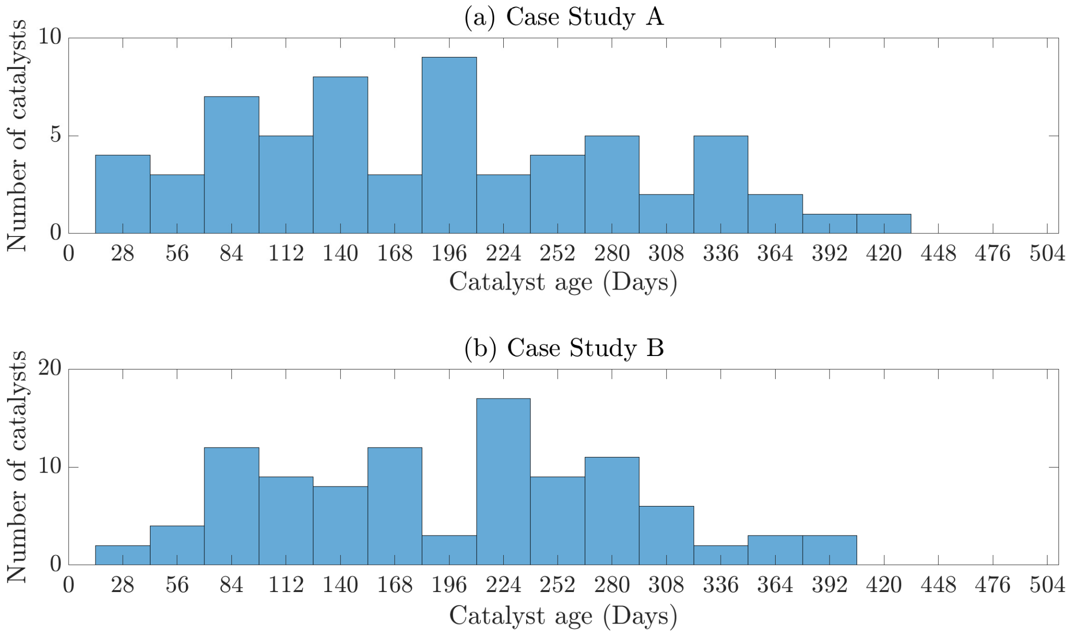


Fig. 2.6 The distribution of the catalyst ages obtained using Implementation I for (a) the 13 successful runs of Case Study A and (b) the 22 successful runs of Case Study B

It was seen if using other optimisation algorithms available to *fmincon*, such as 'interior point' and 'active-set' methods, enabled the integrator to overcome such problems or prevented a premature convergence. However, runs with the interior point method led to poor, non bang-bang answers for the catalyst changeover controls. The active-set method also faced premature convergence and integration problems, and produced solutions similar to those produced by the SQP method, although at a slower computational time compared to the latter. So the SQP algorithm was the most effective amongst those available to *fmincon* and was used as the preferred algorithm for Implementation I.

Overall, the performance of Implementation I was unsatisfactory in providing solutions to all case studies. Despite this, there is a very good reason for reporting this solution procedure in this thesis: it is observed that a bang-bang behaviour is exhibited by the catalyst changeover controls, even when those linear controls occur in combination with other process control variables that occur nonlinearly in the system equations. This is consistent with the predictions of the theoretical analysis done in Section 2.1.1. In the ensuing text, the optimal control and state variables of the most profitable run from the set of 50 different, random starting points for each of Case Studies A and B are reported, along with relevant economic statistics. In the section following this, an alternative solution procedure, Implementation II, is presented, which is superior to Implementation I in terms of producing high quality solutions for all case studies, but does not demonstrate the bang-bang property of the catalyst changeover controls.

2.3.3 Case Study A: Results and discussions

Figures 2.7 – 2.10 and Table 2.6 report the features of the best local optimum among the 13 successful runs for Case Study A, in which the main reaction is of first order kinetics with respect to the reactant (equation (2.25)) and the catalyst deactivation kinetics is independent of the species' concentrations (equation (2.21)).

Figure 2.7 illustrates the variation of the monthly catalyst changeover controls over the whole time horizon. It can be seen that these controls take values of either 0 or 1, thus exhibiting a bang-bang behaviour, consistent with the prediction for linear controls from the analysis in Section 2.1.1. The graph indicates that the optimal policy for the industrial process is to use 4 of the 6 available catalysts over the 3-year horizon, with the 3 replacements ($y = 0$) occurring on the 8$^{\text{th}}$, 17$^{\text{th}}$ and 24$^{\text{th}}$ months. The first replacement occurs during the quarter of lowest demand in order to minimise losses. The other replacements occur only when a sufficient inventory level (Figure 2.10) is present to meet the demand during process shut-down.

Figure 2.8 plots the weekly flow rates to the reactor (ffr) and temperatures of operation (T), made dimensionless by their respective upper bounds and the exit concentration of the reactant from the reactor (cR), over the whole time horizon of the process. Some notable points regarding these trends:

- The model's optimal policy during catalyst operation is to maintain a constant exit conversion by reducing the flow rate to compensate for the catalyst deactivation and operate temperature at its upper bound. This is consistent with the work of Szépe and Levenspiel (1968) for continuous reactors, which predicted similar policies when the main reaction is more sensitive to temperature than the catalyst deactivation and the latter is independent of the species' concentration.

- During the operation of the last catalyst, the sharp drop in the flow rate causes a corresponding effect in the exit concentration and this occurs to bring the production rate to a value that exactly fulfils the demand for the remainder of the time horizon. This ensures efficient operation, as excessive raw material is prevented from being used, thereby minimising the costs and maximising the profits of the process.

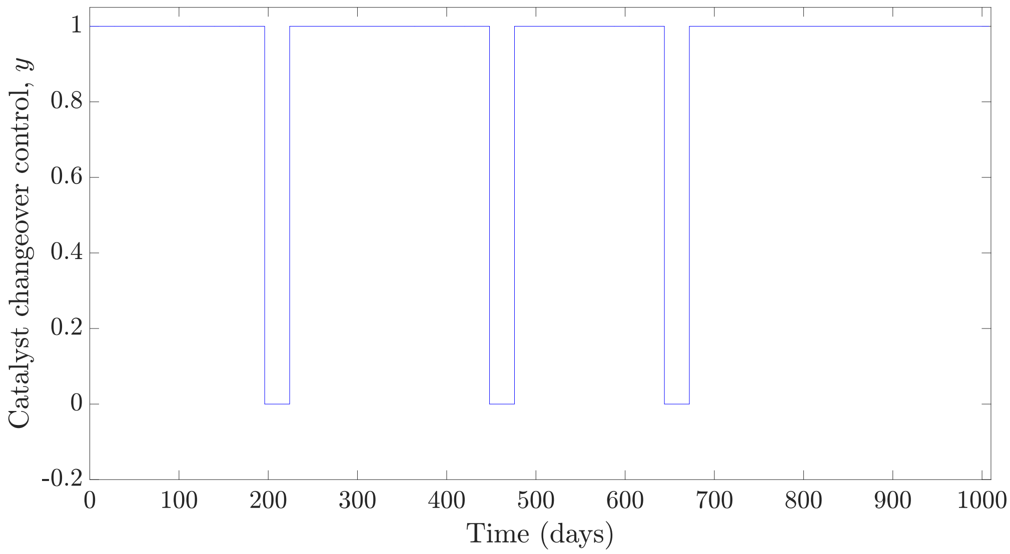


Fig. 2.7 The variation of the catalyst changeover controls over the time horizon in the best solution of Case Study A, obtained using Implementation I.

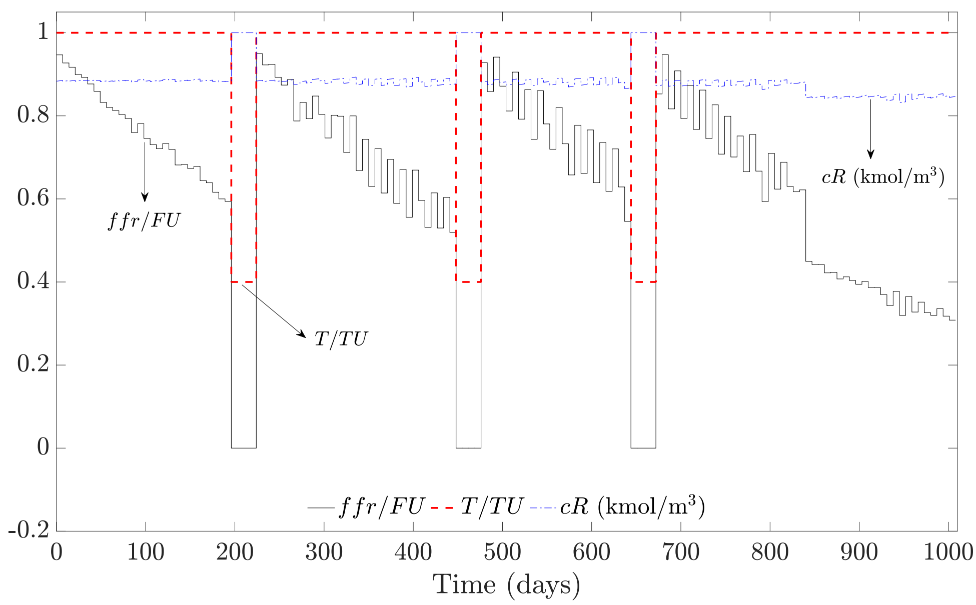


Fig. 2.8 The variation of the feed flow rate, temperature and reactant exit concentration over the time horizon in the best solution of Case Study A, obtained using Implementation I.

- During times the catalyst is being replaced in the reactor, the feed flow rate is set to zero, the temperature of operation is set to its lower bound (TL) and the reactant exit concentration is set to its entry value ($CR0$), as per constraints (2.45) and (2.46) and junction conditions (2.38), respectively.
- It is highlighted that the flow rate does not exhibit a bang-bang behaviour as these controls appear nonlinearly in the system equations, consistent with the prediction from Section 2.1.1. It is interesting to note that the temperature controls only take values at their upper or lower bounds, and this follows from the nature of the problem and the constraints imposed, without a correlation to their nonlinear occurrence in the system equations.

The numerical noise experienced by $fmincon$ in identifying the optimal ffr values are evident in this figure, especially during the the operation of the last three catalysts: the decrease of the feed flow rate is not continuous and considerable disturbances are present.

A comparison of the optimal quantity of product sales with the corresponding product demand and unmet demand for each week over the whole time horizon, is shown in Figure 2.9. While a considerable amount of unmet demand exists during the first year of the process, it is nil for the remainder years. The explanation for this is as follows:

- At the beginning of the time horizon, there is no prior inventory of the product present. What is produced by the process is what is used to meet the product demand. And it so happens that the production capacity of the process is unable to fully meet the high demand at the beginning of the time horizon. While there is relatively less product demand towards the end of the first year, there is still a substantial amount of unmet demand at that time, even though there is considerable product inventory present. This occurs because the model prefers to hoard the product in order to enable greater sales thereby nil unmet demand during the second and third years.
- Why there is no unmet demand during the second and third years can be attributed to the annual inflation, which causes an increase in all prices involved every year. Due to inflation, the product sales price and penalty for unmet demand are higher in the second and third years compared to the first year. Therefore, if a larger amount of sales and thereby, nil unmet demand, occurs during the second and third years, a greater amount of profit can be obtained.

It is also highlighted that the sales continue throughout the time horizon, even at times of process shut down for catalyst replacement.

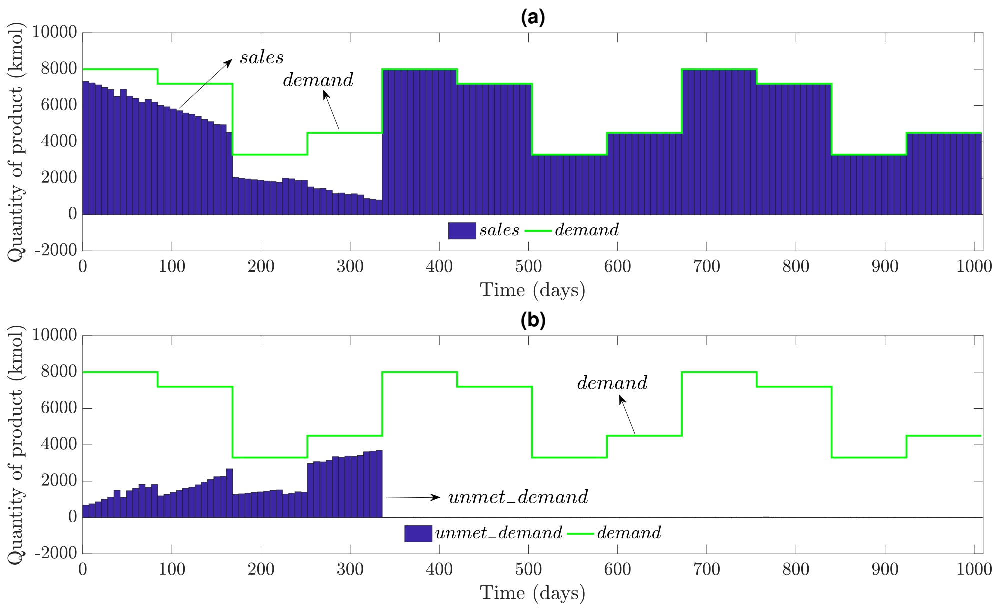


Fig. 2.9 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the time horizon in the best solution of Case Study A, obtained using Implementation I.

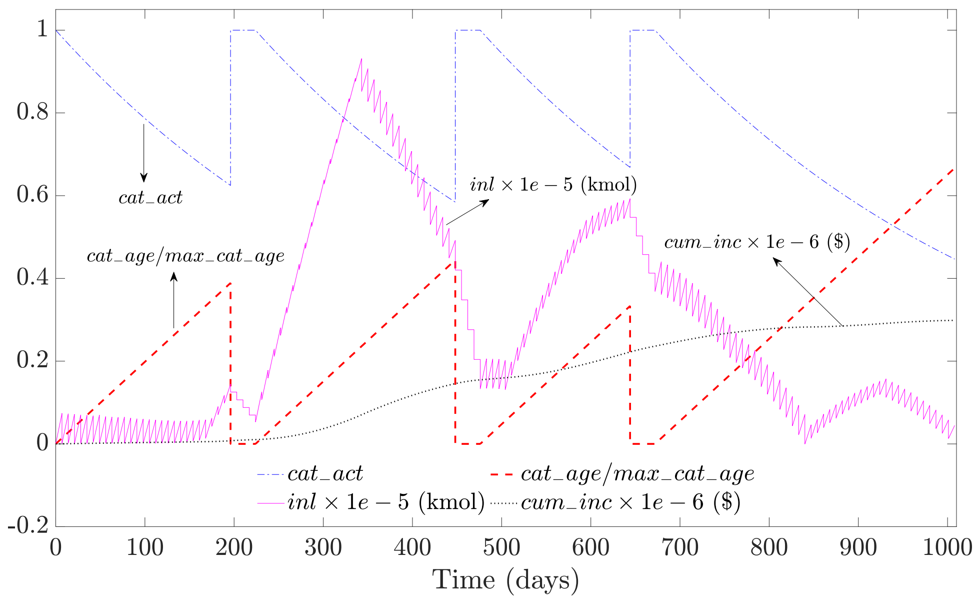


Fig. 2.10 The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory cost over the time horizon in the best solution of Case Study A, obtained using Implementation I.

The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory costs over the time horizon are shown in Figure 2.10. An explanation for these trends are as follows:

- The catalyst age increases linearly with time when the catalyst is in operation. But during catalyst replacement, the age remains at zero as the catalyst is unused during this time. These trends follow directly from differential equation (2.19) and junction condition (2.36).
- An exponential decrease in the activity is seen during times of catalyst operation, due to the deactivation kinetics being first order. During catalyst replacement, the catalyst activity remains constant at the value of the activity of a fresh catalyst, as there is no reaction occurring. These trends follow directly from differential equation (2.20), deactivation rate expression (2.21) and junction condition (2.37).
- Some notable points regarding the plot of the inventory level:
  - When the catalyst is in operation, an oscillating behaviour for the inventory level arises from the build up of storage from production during the week and a reduction due to sales at the end of the week. However, in the months corresponding to catalyst replacement, as there is no production, the inventory level remains constant over the course of a week, at the end of which it drops by a value equivalent to the sales of the product for the week. These trends follow directly from differential equation (2.28) and junction condition (2.39).
  - In the beginning of the process, the inventory level barely increases as most of the stored product is sold to meet the high demand at that time. However, towards the end of the first year, the inventory level shows a significant increase, despite there being a considerable amount of unmet demand at that time. This happens in order to enable greater amount of sales and thereby eliminate the unmet demand during the later years when the product sales price and the penalty for unmet demand has increased due to inflation, thus enlarging the profit obtained. Thereafter, the optimal decisions enable the right amount of inventory to be maintained to exactly meet the demand.

  Thus, this optimal management of the inventory level ensures increased profits as well as sufficient product to meet the demand, while also preventing wastage of product and high inventory costs.

- The profile of the cumulative inventory cost follows from differential equation (2.29) and junction condition (2.40). The slope of the curve is higher when the inventory level is higher and the curve seems to stagnate when the inventory is relatively low.

The magnitudes of the various economic aspects that form the elements of the objective function are given in Table 2.6. The table indicates that the cost of flow and raw material constitutes more than half of the total expenses with the net penalty for unmet demand also forming a significant proportion. The cost of catalyst changeovers contributes relatively less while the inventory costs form the lowest percentage of the total expenditure. It is also seen that the costs of operation take away about 43.6% of the revenue generated by the product sales.

Table 2.6 Details of the economic aspects of the best solution of Case Study A, obtained using Implementation I.

Economic aspect		**Symbol**	**Value ($ Millions)**
Gross Revenue from Sales		GRS	776.422
Costs	Total Inventory Costs	TIC	0.299
	Total Costs of Catalyst Changeovers	$TCCC$	30.999
	Net Penalty for Unmet Demand	$NPUD$	117.089
	Total Flow Costs	TFC	189.955
Profit		$-NC$	438.08

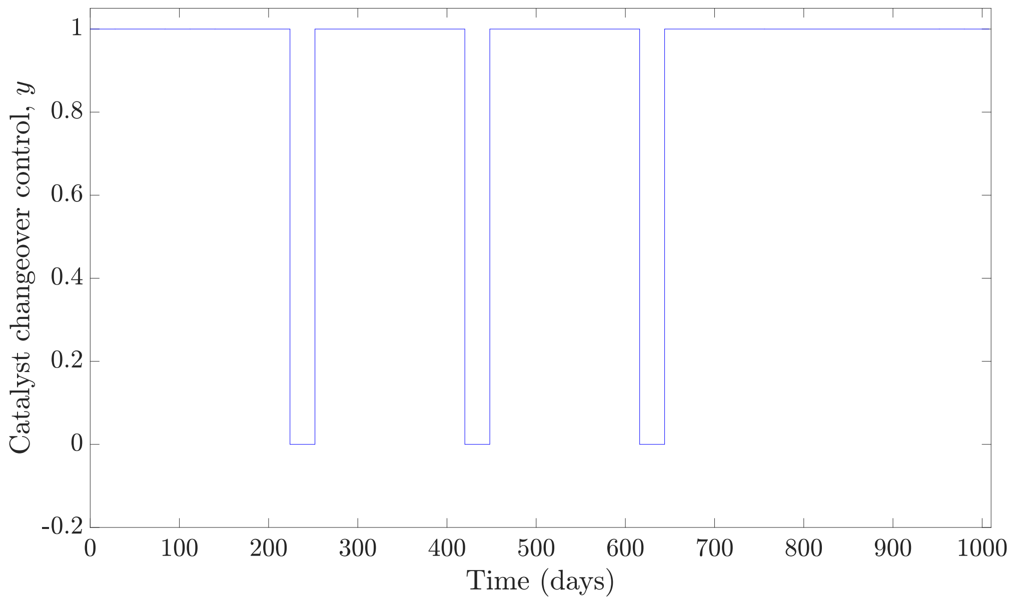


Fig. 2.11 The variation of the catalyst changeover controls over the time horizon in the best solution of Case Study B, obtained using Implementation I.

2.3.4 Case Study B: Results and discussions

Figures 2.11 – 2.14 and Table 2.7 report the features of the best local optimum among the 22 successful runs for Case Study B, in which the main reaction is of first order kinetics with respect to the reactant (equation (2.25)) and the catalyst deactivation kinetics is proportional to the reactant concentration (equation (2.22)).

Figure 2.11 shows the variation of the monthly catalyst changeover controls over the time horizon. Once again, a bang-bang behaviour is exhibited, consistent with the analysis in Section 2.1.1. The recommendation is to use 4 of the 6 available catalysts over the 3-year horizon, with the 3 replacements $(y = 0)$ occurring on the 9$^{\text{th}}$, 16$^{\text{th}}$ and 23$^{\text{rd}}$ months. Once again, the first replacement occurs at a time to minimise losses and the other changeovers occur only when there is sufficient inventory to meet the demand.

Figure 2.12 is the analogue of Figure 2.8 in Case Study A. Once again, the numerical noise experienced by the optimiser in identifying the optimal values of ffr are evident here: there are considerable distortions in the profiles of this control. However, the trends of ffr and cR during catalyst operation are different from in Case Study A: the decrease in ffr is such that its rate of decrease is slower than the rate of catalyst deactivation and this causes cR to show a roughly linear increase in magnitude.

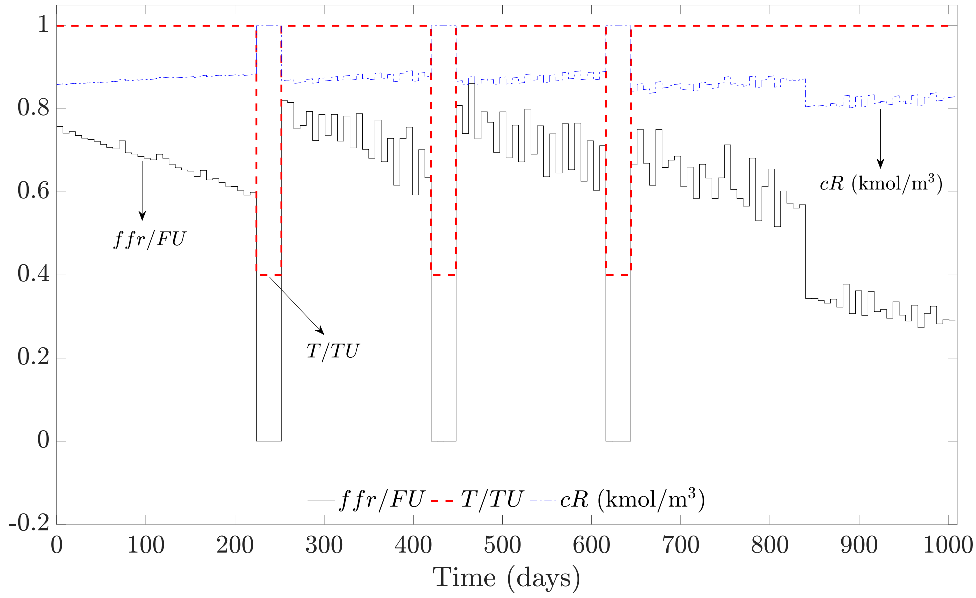


Fig. 2.12 The variation of the feed flow rate, temperature and reactant exit concentration over the time horizon in the best solution of Case Study B, obtained using Implementation I.

In Section 1.3, a work by Crowe (1976) was mentioned, which predicted that even when the rate of catalyst deactivation is dependent on the reacting species' concentration, the optimal policy at the reactor level is to maintain a constant exit conversion, provided the time scale for the deactivation is much larger than the time scales of the main reaction and the flow rate. The choice of parameters for this case study are such that the time scale for the catalyst deactivation (Kd = 0.0024 (1/day)) is certainly much larger than that of the main reaction (about 24 (1/day) for temperatures used during catalyst operation) or the flow rate (thousands of cubic metres a day) and yet cR does not remain at a constant value during times of catalyst operation. Thus, the behaviour of cR observed here is not consistent with the predictions of the work of Crowe (1976).

An explanation for the profile of cR in Figure 2.12 is offered using the following points:

- A larger magnitude of cR implies a faster deactivation of the catalyst, following from equation (2.22), and this is unfavourable for the process.
- A larger magnitude of cR means a larger reaction rate, following from equation (2.25), and this is favourable for the process.

Thus, there is a trade-off to be balanced in maintaining a particular magnitude of cR. The flow rate is chosen such that at the beginning of operation of a new catalyst, a relatively low value of cR occurs, which although lowers the reaction rate, it prevents the fresh catalyst from deactivating too fast. However, as the catalyst deactivates, the focus shifts to maintaining a higher reaction rate and this is done by the appropriate reduction of ffr to raise cR. This linearly increasing trend enables to optimally balance the positive and negative effects of maintaining a particular magnitude of cR.

Figures 2.13, 2.14 and Table 2.7 are the analogues in Case Study B of Figures 2.9, 2.10 and Table 2.6, respectively, in Case Study A. In Figure 2.13, the reasons for the distribution of the sales such that the unmet demand is present in a considerable magnitude throughout the first year but is nil in the second and third years, can be explained using logic similar to that used for the explanation of the trends of the analogous variables in Figure 2.9 in Case Study A. In Figure 2.14, the profile for the catalyst activity during catalyst operation follows from deactivation expression (2.22). The trends of the other variables in Figure 2.14, namely the catalyst age, the inventory level and the cumulative inventory cost, can be explained using reasons similar to that used for the explanation of the profiles of the analogous variables in Figure 2.10 in Case Study A. Table 2.7 shows that the costs of operation take away about 39.5% of the revenue generated by the product sales.

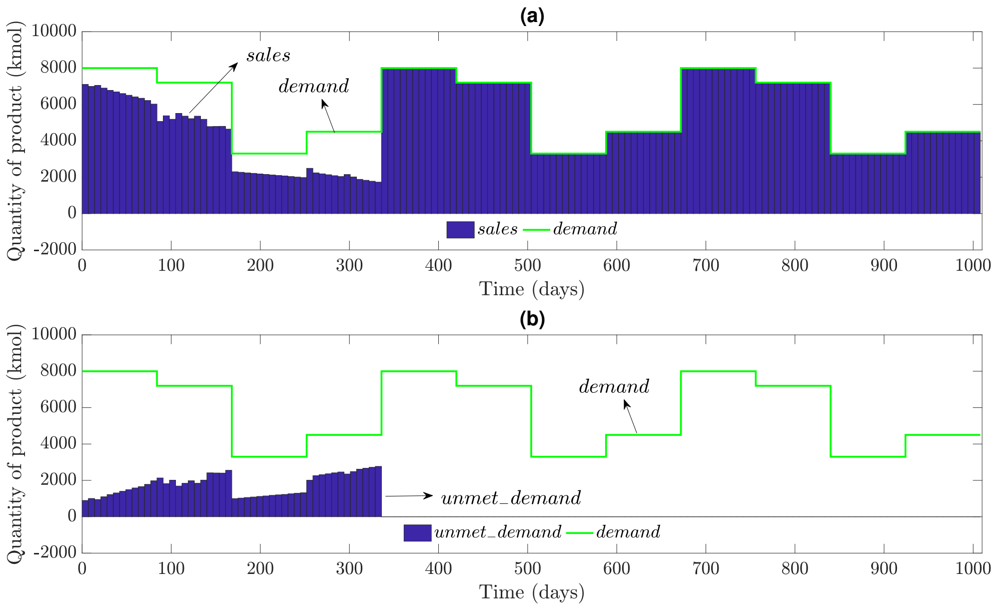


Fig. 2.13 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the time horizon in the best solution of Case Study B, obtained using Implementation I.

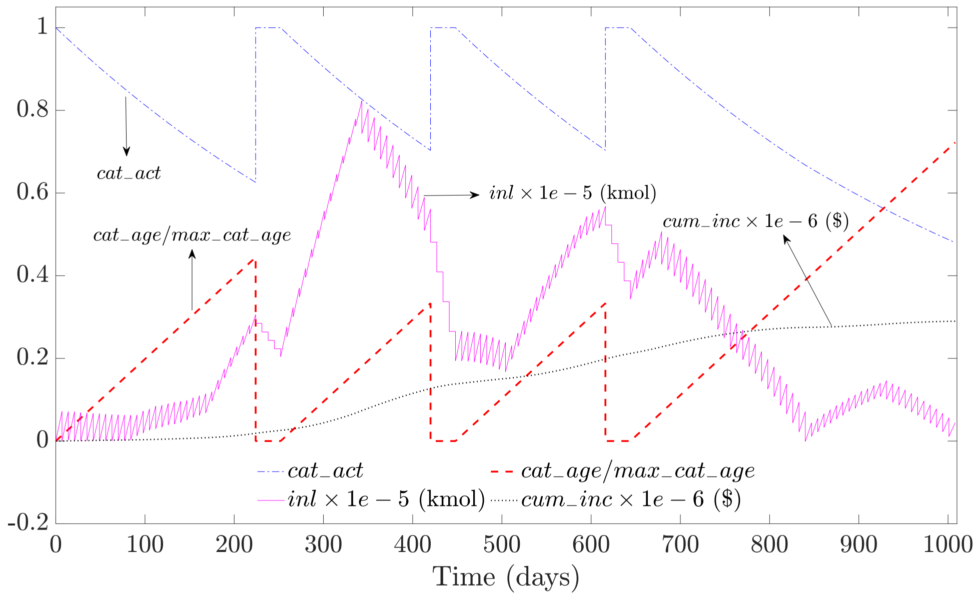


Fig. 2.14 The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory cost over the time horizon in the best solution of Case Study B, obtained using Implementation I.

Table 2.7 Details of the economic aspects of the best solution of Case Study B, obtained using Implementation I.

Economic aspect		Symbol	Value ($ Millions)
Gross Revenue from Sales		GRS	785.245
Costs	Total Inventory Costs	TIC	0.290
	Total Costs of Catalyst Changeovers	$TCCC$	30.999
	Net Penalty for Unmet Demand	$NPUD$	106.061
	Total Flow Costs	TFC	172.67
Profit		$-NC$	475.225

2.4 Implementation II: Details, results and discussions

Given the inadequacies of Implementation I, it was decided to attempt an alternate implementation in Python$^{\text{TM}}$ 3.7.1 under PyCharm 2018.2.4 (Community Edition). This section discusses the details and performances of a preliminary implementation called Implementation IIA, before doing the same for Implementation II, a modification of the former. Subsequently, the results of all case studies obtained using Implementation II are presented.

2.4.1 Implementation IIA details

Implementation IIA was carried out as a Python code that solved a standard multistage optimal control problem using the feasible path approach, of the form similar to that in Implementation I. The code was written using CasADi, an open source software that enables a symbolic framework for numerical optimisation (Andersson, 2013). The elements of the problem, as given in Section 2.2, were defined as symbolic expressions using CasADi v3.4.5. The automatic differentiation (AD) feature of CasADi enabled constructions of symbolic expressions of the derivatives of all predefined functions, thereby maintaining differentiability to an arbitrary order. This allowed for an efficient calculation of gradients, that did not suffer from round-off and truncation errors, unlike gradient calculation using finite differences.

CasADi contains plug-ins to the open source SUNDIALS suite (Hindmarsh et al., 2005) and IPOPT by COIN-OR (Wächter and Biegler, 2006), which were used for the integration of ODEs and optimisation, respectively. The *IDAS* solver of SUNDIALS was used for the integration of the ODEs with the following termination criteria: an absolute tolerance of 10^{-6} and a relative tolerance of 10^{-6}. For the optimisation by IPOPT, Table 2.8 presents the termination and 'acceptable' termination criteria, wherein the 'acceptable' number of iterations concerning the latter was set at 15. The logic behind the use of acceptable termination criteria is as follows: if the algorithm encounters 15 iterations in a row that fulfil the acceptable termination criteria, it will terminate before the termination criteria is met. This would be useful in cases where the algorithm might not be able to attain the termination criteria.

The above implementation procedure was run on the same hardware and operating system used for Implementation I. A set of random starting guesses for the decision variables were provided using the *rand* method of the *random* class within the *numpy* module.

Table 2.8 Criteria for termination of optimisation by IPOPT

Property	Termination tolerance	Acceptable termination tolerance
Optimality error	10^{-4}	10^{-4}
Dual infeasibility	1	10^{6}
Constraint violation	10^{-4}	10^{-2}
Complementarity	10^{-4}	10^{-2}

2.4.2 Implementation IIA: General performance discussion

For multiple test runs, it was found that the catalyst changeover actions did not exhibit a bang-bang behaviour when this implementation methodology was used. Other adjustments such as tighter optimality tolerances, scaling of the objective functions and constraints or providing feasible starting guesses to the decision variables made little difference and there remained non-integral catalyst changeover control values in the final solution. The reason for the lack of bang-bang behaviour for the catalyst changeover controls, while not clear, is probably due to a shortcoming of the IPOPT tool.

CasADi plug-ins to other optimisation algorithms such as ‘sqpmethod’, ‘stabilizedsqp’, ‘knitro’ and ‘snopt’ were attempted to check if a bang-bang behaviour for the catalyst changeover controls could be obtained. However, these other algorithms did not produce good results.

Thus, the analysis done in Section 2.1.1 is not applicable here and further modifications were needed to Implementation IIA in order to attain the desired results and this led to Implementation II.

2.4.3 Implementation II details

Implementation II is composed of executing Implementation IIA with a penalty term homotopy, a technique similar to that suggested by Sager (2005, 2009).

Since the basic MSMIOCP formulation, given by equation (2.4), was used as basis for formulating the problem under consideration here, that equation will be used to explain the principle of this implementation. First, the binary restrictions for the controls, u, in equation (2.4g) are relaxed and instead, these controls are considered as continuous variables in the range $[0,1]$. A modified form of equation (2.4), that assumes such a relaxation is shown in equation (2.61). As can be seen, the binary restrictions on the controls, u, in equation (2.4g), have been replaced by the condition stating that these controls are continuous variables in the range $[0,1]$, in equation (2.61g). Problems of the form of equation (2.61) will be referred to as an "MSMIOCP with relaxed binary controls" in this thesis.

$$\begin{aligned}\min_{u,v} W = \sum_{p=1}^{NP} \Big\{ \phi^{(p)} \left(x^{(p)}(t_p), z^{(p)}(t_p), u^{(p)}, v^{(p)}, t_p \right) \\ + \int_{t_{p-1}}^{t_p} L^{(p)} \left(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t \right) \mathrm{d}t \Big\}\end{aligned} \tag{2.61a}$$

subject to

$$\begin{gathered}\dot{x}^{(p)}(t) = f^{(p)}(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t) \\ t_{p-1} \le t \le t_p \\ p = 1,2,\dots,NP\end{gathered} \tag{2.61b}$$

$$x^{(1)}(t_0) = h^{(1)} \left(u^{(1)}, v^{(1)} \right) \tag{2.61c}$$

$$\begin{gathered}x^{(p)}(t_{p-1}) = h^{(p)} \left(x^{(p-1)}(t_{p-1}), z^{(p-1)}(t_{p-1}), u^{(p)}, v^{(p)} \right) \\ p = 2, 3, \dots, NP\end{gathered} \tag{2.61d}$$

$$\begin{gathered}0 = g^{(p)}(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t) \\ t_{p-1} \le t \le t_p \\ p = 1,2,\dots,NP\end{gathered} \tag{2.61e}$$

$$\begin{gathered}c^{(p)} \left(x^{(p)}(t), z^{(p)}(t), u^{(p)}, v^{(p)}, t \right) \le \quad 0 \\ t_{p-1} \le t \le t_p \\ p = 1,2,\dots,NP\end{gathered} \tag{2.61f}$$

$$\begin{gathered}u^{(p)} \in [0,1] \\ p = 1, 2, \dots, NP\end{gathered} \tag{2.61g}$$

$$\begin{gathered}v^{(p)} \in \mathscr{V} \\ p = 1, 2, \dots, NP\end{gathered} \tag{2.61h}$$

Problems of the form of equation (2.61) are essentially standard multistage optimal control problems and hence, standard nonlinear optimisation problems, as no discrete controls are involved and only an optimal set of continuous controls have to be identified, while using the feasible path approach to solve the differential equations. However, in order to obtain solutions equivalent to that of the original problem of the form of equation (2.4), wherein the controls, $u^{(p)}$, for each stage $p = 1, 2, \ldots NP$, take values of only 0 or 1, a penalty term homotopy technique, similar to that suggested by Sager (2005, 2009), is used. In this technique, a monotonically increasing penalty term is added to the objective function in equation (2.61a) and a series of standard multistage optimal control problems of the following generic form are solved:

$$F_k : \min \left\{ W + M_k \sum_{p=1}^{NP} u^{(p)} \left[1 - u^{(p)} \right] \right\} \tag{2.62}$$

subject to equations (2.61b) – (2.61h), for

$$k = 1, 2, 3 \ldots$$
$$M_1 = 0$$

Every iteration, k, is referred to as 'major iteration'. The problem, F_1, in the first major iteration $(k = 1)$ of the series, is designated a weight of $M_1 = 0$ and this problem is equivalent to the problem given by equation (2.61). If solving problem F_1 does not produce binary values for controls u, the second major iteration occurs in which a weight $M_2 > 0$ is chosen and problem F_2 is solved using the solution of F_1 as initial guesses. This procedure is repeated in an iterative manner, by choosing a weight $M_{k+1} > M_k$ and solving problem F_{k+1} with the solution of F_k as initial guesses, until iteration K $(K \geq 1)$ such that all controls in u, in the solution of problem F_K, are forced by weight M_K to take values of either 0 or 1. This solution procedure is presented as an algorithmic flowchart in Figure 2.15.

The progression for the increase of weights, M_k, is chosen arbitrarily, by trial and error, and is dependent on the parameters of the problem. It should be remembered that if the weight is increased too slowly, the computational time becomes large, while if it is increased too fast, the optimiser can fail to recognise a solution and continue iterations indefinitely.

Since Implementation IIA did not obtain integer values for the catalyst changeover controls, the above solution procedure involving the penalty term homotopy technique is applied to that implementation and this forms Implementation II. In applying Implementation II to the problem under consideration here, it is highlighted that the elements of the problem in

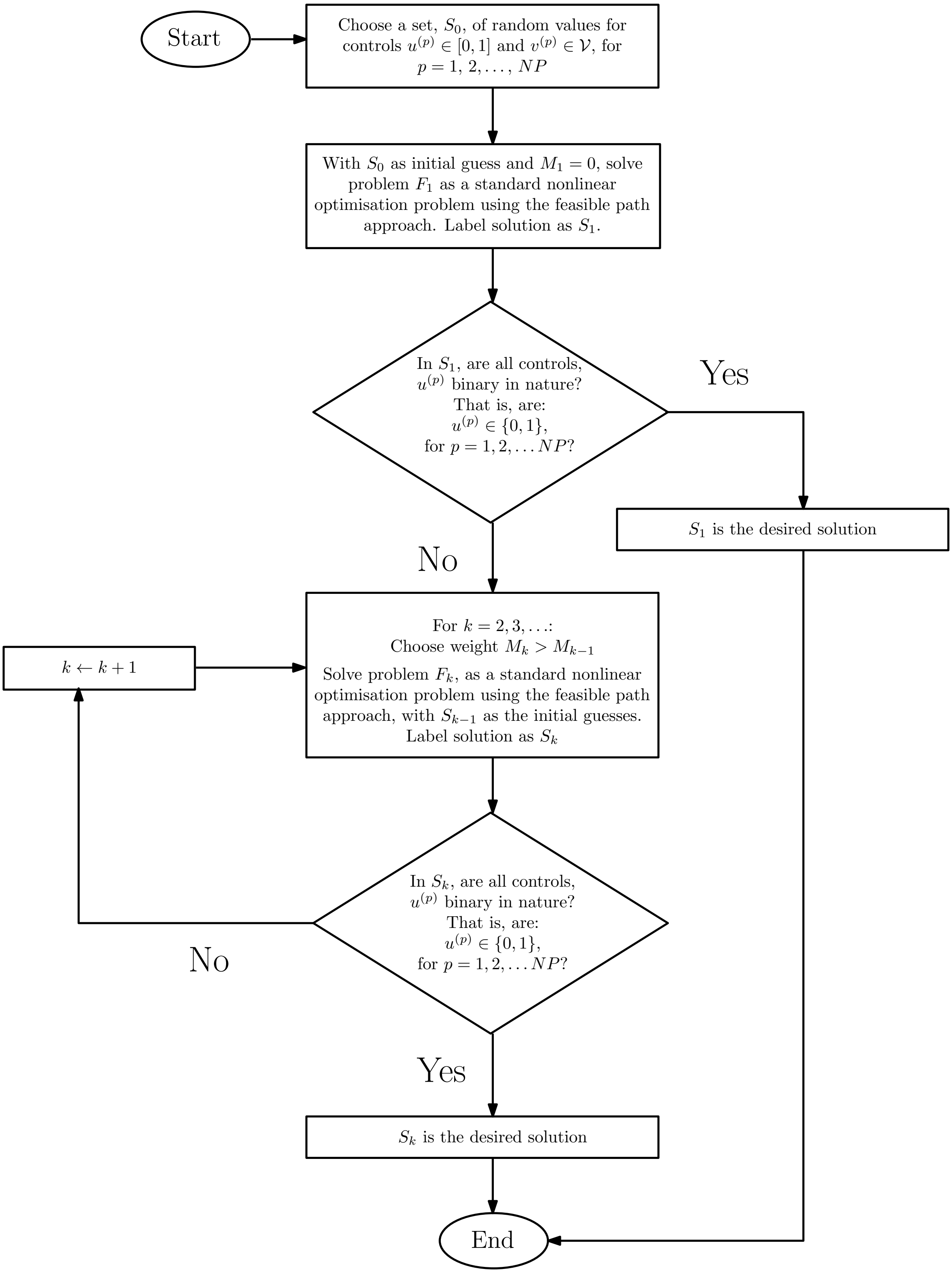


Fig. 2.15 An algorithmic flowchart for the principle of Implementation II.

Section 2.2 were formulated as an MSMIOCP with linear and relaxed binary controls, as per equation (2.17), which is also a form of the MSMIOCP with relaxed binary controls given by equation (2.61). What was done in Implementation IIA was essentially solving the first major iteration ($k = 1$) of the series of optimisation problems to be solved (wherein the weight $M_1 = 0$) in Implementation II. The code for this implementation, is essentially an extension of the code written on CasADi in Python in Implementation IIA, in order to solve the following series of standard multistage optimal control problems:

$$G_k : \min \left\{ NC + M_k \sum_{i=1}^{NM} y(i)\left[1 - y(i)\right] \right\} \tag{2.63}$$

subject to the appropriate differential equations for each case study, the initial conditions, the junction conditions and constraints presented in Section 2.2, for

$$\begin{aligned} k &= 1,2,3\ldots \\ M_1 &= 0 \end{aligned}$$

In accordance with this procedure, if in the solution of problem G_k, the condition, $y(i) \in \{0,1\}$ for $i = 1,2,\ldots NM$, does not apply, then problem G_{k+1} is solved using the solution of G_k as initial guesses, with weight $M_{k+1} > M_k$. For the choice of parameters used in these case studies, the weight term is increased as per the arithmetic progression in equation (2.64), which was chosen by trial and error:

$$\begin{aligned} M_{k+1} &= (2 \times M_k) + \left(5 \times 10^7\right) \\ M_1 &= 0 \\ k &= 1,2,3\ldots \end{aligned} \tag{2.64}$$

As in Implementation IIA, *IDAS* of SUNDIALS was used for the integration and IPOPT was used for the optimisation in each problem (major iteration) of the series. And the termination criteria for the integration and optimisation in each problem of the series were also set similar to that in Implementation IIA.

The implementation was performed on the same hardware as for Implementations I and IIA. Once again, multiple runs were performed with different starting points due to the non-convex nature of the problem. Test runs using the *multiprocessing* module in Python, to parallelise a loop of multiple start points, executed slower than when the runs were done serially. So for each case study, 50 runs were executed in a serial manner.

2.4.4 Implementation II: General performance discussion

It was found that Implementation II produced high quality solutions for all case studies. Not in a single run for any case study, regardless of the degree of nonlinearity of the process model, was any integration or convergence problem encountered. Unlike Implementation I, there was no need for supplying feasible starting guesses or scaling of the controls, objective functions or constraints in order to accelerate convergence to a solution. In addition, the solutions obtained did not experience any ‘numerical noise’ as was the case in Implementation I.

Statistics regarding the solutions, regarding the profits, the number of catalyst replacements and the catalyst ages, obtained from the 50 runs for all case studies using Implementation II are given in Table 2.9. Under the column titled, "Profit (Million $)", the values under the sub-columns titled ‘Max’, ‘Min’, ‘Mean’ for the row labelled, ’Case Study A’ indicate the maximum, minimum and mean profit values, respectively, among the 50 runs of Case Study A. Similar explanations hold for those values under this column for the rows labelled ’Case Study B’, ’Case Study C’ and ’Case Study D’. Analogous explanations hold for the sub-columns titled, ‘Max’, ‘Min’, ‘Mean’ and ‘Mode’ within the columns of "Number of catalyst replacements" and "Catalyst age (days)", for the rows pertaining to each case study. Some notable points regarding this table are as follows:

- The range of optimal profit values obtained for Case Studies A and B are comparable to those obtained from the limited set of successful runs for the same case studies using Implementation I, which can be considered optimal solutions as the bang-bang behaviour was exhibited for the catalyst changeover controls in those solutions. But such comparisons are not possible for the runs of Case Studies C and D as Implementation I failed to produce solutions for those case studies. However, the good correlation between the optimal profits obtained using Implementation I and Implementation II for Case Studies A and B, suggests that Implementation II is indeed capable of attaining the optimal solution and that the results obtained for Case Studies C and D using Implementation II are indeed optimal.

- Another notable point to be highlighted from the data presented in this table is that in no run for any case study, are all 5 available catalyst replacements used, with the maximum number being either 3 or 4 and the mode number being either 2 or 3.

Further insights regarding the distribution of the solutions presented in Table 2.9 are provided in Figures 2.16 – 2.18. Following are comments regarding these figures:

- Figure 2.16 shows the distributions of the profits obtained from all runs of all case studies, when using Implementation II. In the histogram presented in subplot (a) of this

Table 2.9 Implementation II solution statistics.

Case Study	Profit (Million $)			Number of catalyst replacements			Catalyst Age (days)		
	Max	**Min**	**Mean**	**Max**	**Min**	**Mode**	**Max**	**Min**	**Mean**
Case Study A	449.946	353.347	424.119	4	2	3	448	112	229.7
Case Study B	480.135	411.704	463.562	3	2	3	448	112	239
Case Study C	430.493	326.327	407.282	3	2	3	476	140	247.5
Case Study D	325.089	260.277	295.367	3	2	2	504	140	275

Table 2.10 Implementation II size statistics.

Case Study	CPU time (seconds)			Number of major iterations		
	Max	**Min**	**Mean**	**Max**	**Min**	**Mode**
Case Study A	27438	9826	17440	5	2	3
Case Study B	48808	9498	18848	6	2	3
Case Study C	48285	13484	24419	5	2	3
Case Study D	38033	16877	28323	5	2	4

Table 2.11 Statistics for each major iteration of Implementation II. The sub-column titled ‘Runs’ indicates the number of runs out of 50 which progressed until that major iteration. The sub-columns titled ‘Max’, ‘Min’ and ‘Mean’ indicate the maximum, minimum and mean number of IPOPT iterations within that major iteration, respectively.

Case Study	**Major iteration 1**				**Major iteration 2**				**Major iteration 3**			
	Runs	**Max**	**Min**	**Mean**	**Runs**	**Max**	**Min**	**Mean**	**Runs**	**Max**	**Min**	**Mean**
Case Study A	50	376	144	236	50	101	56	72	46	95	52	63
Case Study B	50	471	130	224	50	114	64	77	43	96	59	65
Case Study C	50	842	179	307	50	124	66	88	44	134	57	68
Case Study D	50	418	179	279	50	142	68	87	48	87	54	62

Case Study	**Major iteration 4**				**Major iteration 5**				**Major iteration 6**			
	Runs	**Max**	**Min**	**Mean**	**Runs**	**Max**	**Min**	**Mean**	**Runs**	**Max**	**Min**	**Mean**
Case Study A	19	68	56	60	8	83	58	65	N/A	-	-	-
Case Study B	17	94	57	62	7	767	59	184	1	60	60	60
Case Study C	20	71	52	62	6	90	58	75	N/A	-	-	-
Case Study D	32	91	49	60	15	143	54	79	N/A	-	-	-

figure, the height of each bin represents the number of runs out of the 50 runs of Case Study A that result in profit values within the range specified by the horizontal edges of that bin. Similar explanations hold for the histograms relating to Case Studies B, C and D in subplots (b), (c) and (d), respectively, of the figure.

- Figure 2.17 shows the distribution of the number of catalyst replacements obtained from all runs of all case studies, when using Implementation II. In the histogram presented in subplot (a) of this figure, the height of each bin represents the number of runs out of the 50 runs of Case Study A that involved the number of catalyst replacements given by the midpoint of the horizontal width of the bin. Similar explanations hold for the histograms relating to Case Studies B, C and D in subplots (b), (c) and (d), respectively, of the figure. It is noted that the maximum allowable number of catalyst replacements, as per the invented set of parameters used, is 5.

- Figure 2.18 shows the distribution of the ages of all catalyst used in all runs of all case studies, when using Implementation II. In the histogram presented in subplot (a) of this figure, the height of each bin represents the number of catalysts, out of all the catalysts used in all the 50 runs of Case Study A, that were used up to the age given by the midpoint of the horizontal width of the bin. Similar explanations hold for the histograms relating to Case Studies B, C and D in subplots (b), (c) and (d), respectively, of the figure. It is noted that since catalysts can be replaced only at the end of a month, all catalyst ages are multiples of 28 and the maximum allowable age of each catalyst, as per the invented set of parameters used, is 504 days.

Statistics regarding the computational effort involved are given in Tables 2.10 and 2.11. In Table 2.10, under the column titled, "CPU time (seconds)", the values under the sub-columns titled ‘Max’, ‘Min’ and ‘Mean’ for the row labelled, ‘Case Study A’ indicate the maximum, minimum and mean solution times, respectively, among the 50 runs of Case Study A. Similar explanations hold for those values under this column for the rows labelled ‘Case Study B’, ‘Case Study C’ and ‘Case Study D’. Analogous explanations hold for the sub-columns titled, ‘Max’, ‘Min’ and ‘Mode’ within the column of "Number of major iterations" for the rows pertaining to each case study. Table 2.11 provides details regarding the number of runs out of 50 that progressed up to a certain major iteration and the maximum, minimum and mean of the number of IPOPT iterations within each major iteration, for each case study. Some notable comments regarding these tables are as follows:

- From Table 2.10, it is seen that a minimum of 2 major iterations and a mode of 3 or 4 major iterations was needed to obtain integer values for the catalyst changeover

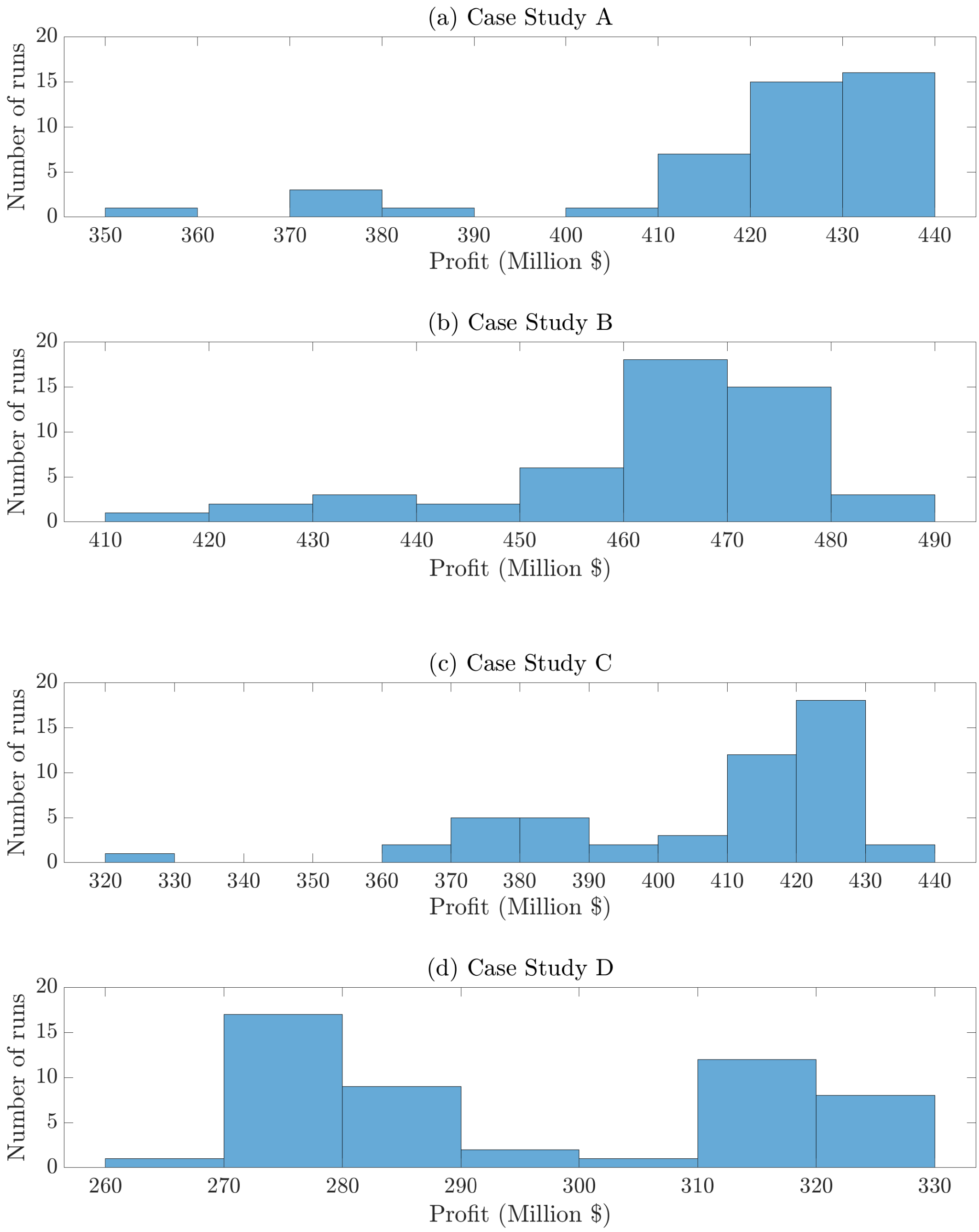


Fig. 2.16 The distribution of the profits obtained over all runs using Implementation II for (a) Case Study A (b) Case Study B (c) Case Study C (d) Case Study D

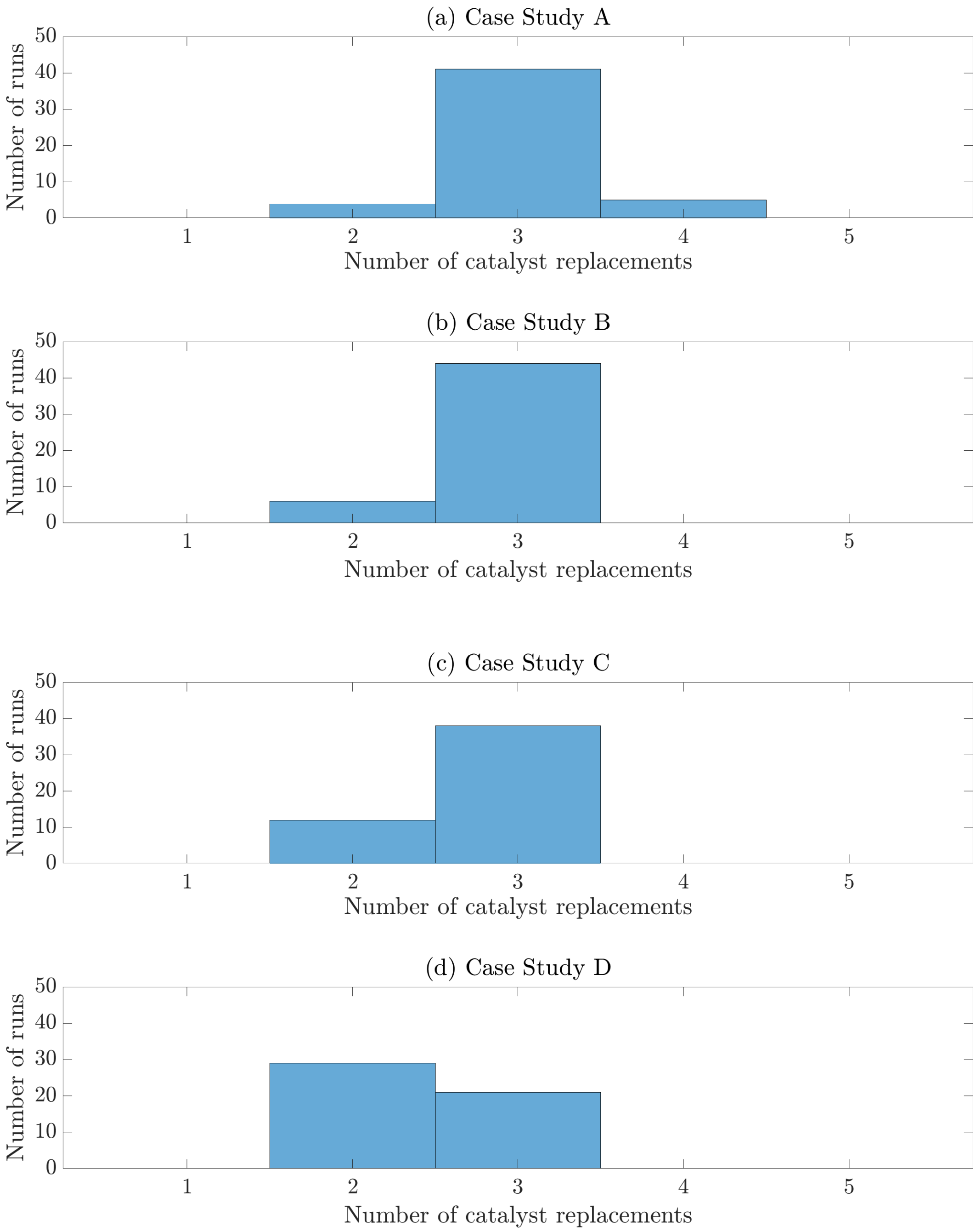


Fig. 2.17 The distribution of the number of catalyst replacements obtained over all runs using Implementation II for (a) Case Study A (b) Case Study B (c) Case Study C (d) Case Study D

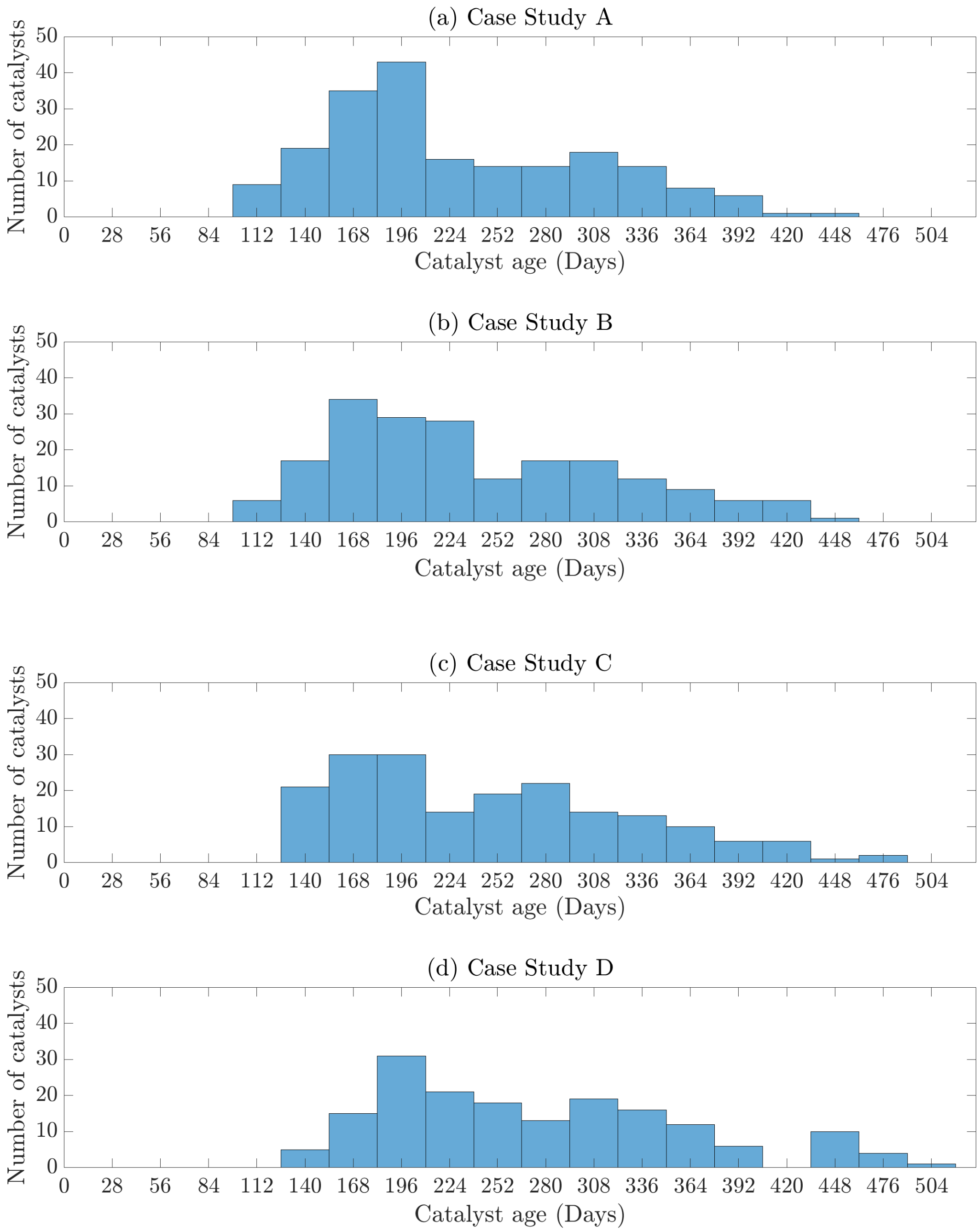


Fig. 2.18 The distribution of the ages of all catalysts used over all runs when using Implementation II for (a) Case Study A (b) Case Study B (c) Case Study C (d) Case Study D

Table 2.12 Comparison of solution times of Implementation I and II

Case Study	Phase 2 of Implementation I CPU time (seconds)			Implementation II CPU time (seconds)		
	Max	Min	Mean	Max	Min	Mean
Case Study A	20383	6738	12009	27438	9826	17440
Case Study B	34855	6134	13394	48808	9498	18848

controls. This underlines the inability of this implementation to obtain the bang-bang behaviour for the catalyst changeover controls.

- With regard to the solution times for Case Studies A and B at least, the minimum, maximum and mean solution times for Implementation II are longer, about 1.35 – 1.5 times their counterparts among the successful runs in Phase 2 of Implementation I. To facilitate such a comparison, these details have been provided in Table 2.12. It should be remembered that this comparison holds only for the specific computer used to obtain these solutions and the comparison may well be different if a different type of computer is used.
- A presentation of the distribution of the solution details presented in Tables 2.10 and 2.11 has not been done because these are dependent on the computer used to obtain solutions, and hence, cannot be generalised.

Overall, Implementation II was more robust, compared to Implementation I, in producing high quality solutions. Next, the results obtained using this implementation, of the best solution from the set of 50 runs, for each of the case studies, are discussed.

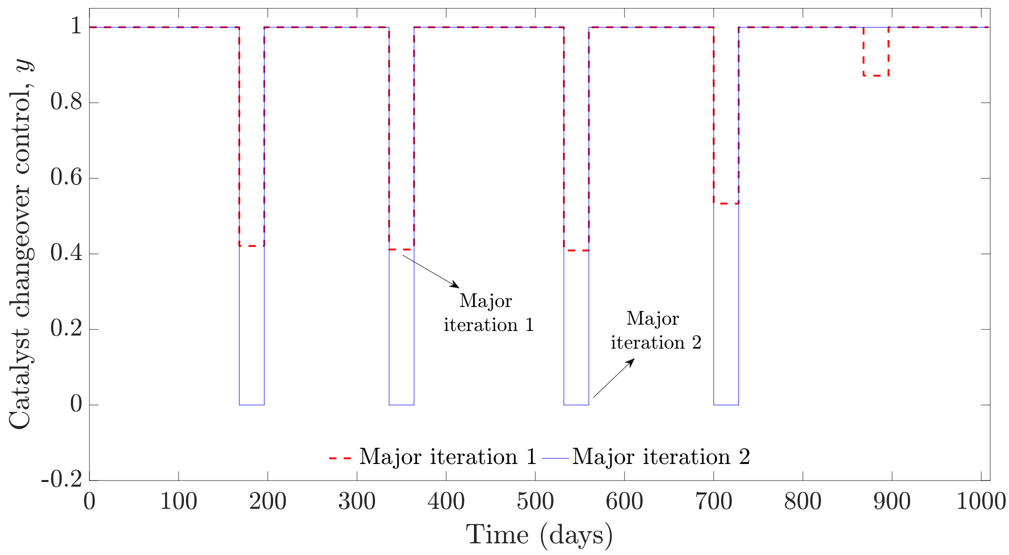


Fig. 2.19 The variation of the catalyst changeover controls over the time horizon in the best solution of Case Study A, obtained using Implementation II.

2.4.5 Case Study A: Results and discussions

Figures 2.19–2.22 and Table 2.13 report the features of the best local optimum among the 50 runs for Case Study A using Implementation II. These are the analogues of Figures 2.7 – 2.10 and Table 2.6, respectively, obtained using Implementation I in Section 2.3.3.

Figure 2.19 shows the variation of the monthly catalyst changeover controls over the time horizon, across different major iterations. It is seen that the solution of the first major iteration is not of bang-bang form, while in the second iteration, integer values are obtained for these controls. The recommendation is to use 5 of the 6 available catalysts over the 3-year horizon, with the 4 replacements $(y = 0)$ occurring on the 7^{th}, 13^{th}, 20^{th} and 26^{th} months. Similar to Figure 2.7, the first replacement occurs at a time to minimise losses and the other replacements occur only when there is sufficient inventory to meet the demand. The other results presented are those obtained as solutions of the second major iteration.

The explanations for the trends of the variables in Figures 2.20 – 2.22 are similar to those of their analogues in Section 2.3.3. Once again, the optimal policies suggested at the reactor level by Szépe and Levenspiel (1968) for continuous reactors are followed here for cR and T. Table 2.13 shows that the profit here is comparable to that in Table 2.6.

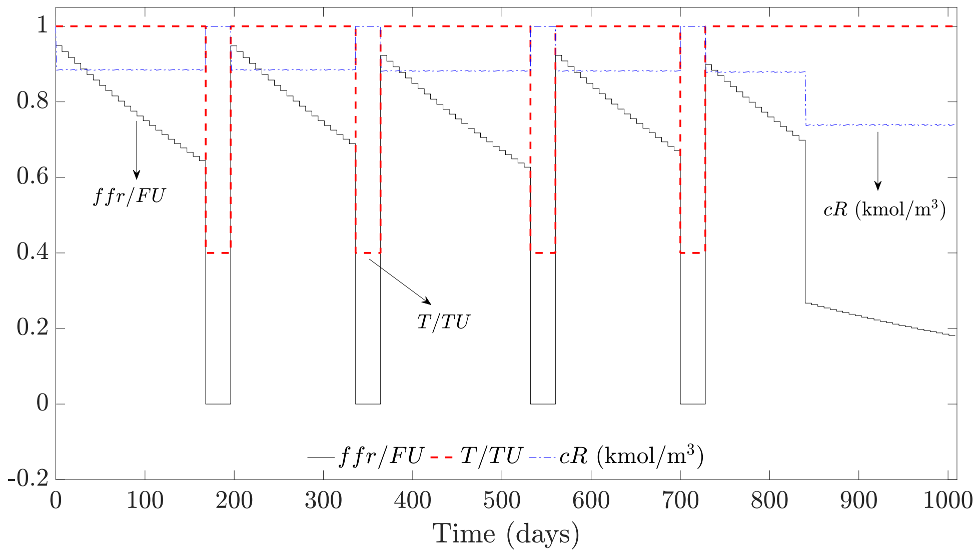


Fig. 2.20 The variation of the feed flow rate, temperature and reactant exit concentration over the time horizon in the best solution of Case Study A, obtained using Implementation II.

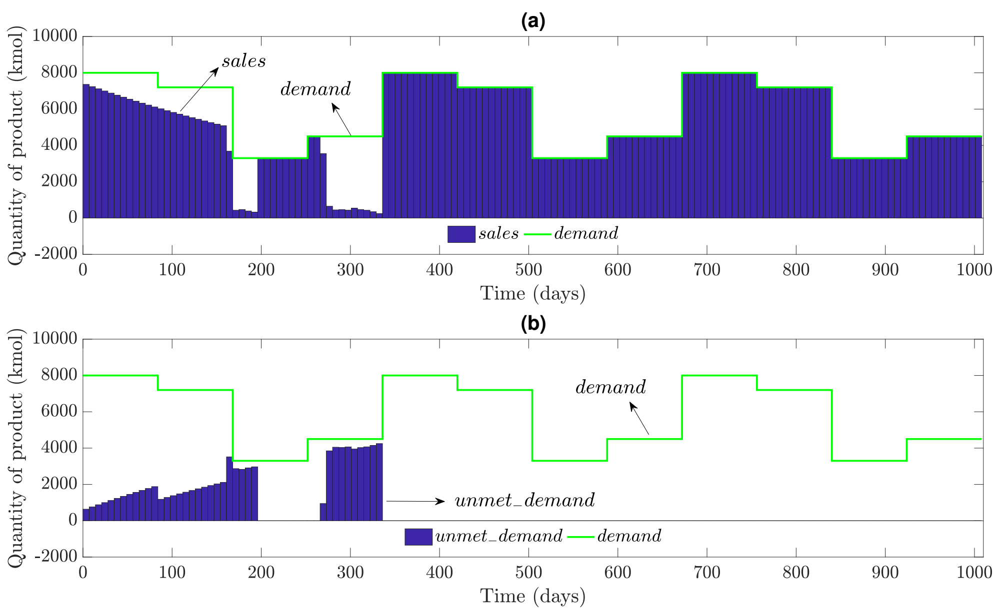


Fig. 2.21 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the time horizon in the best solution of Case Study A, obtained using Implementation II.

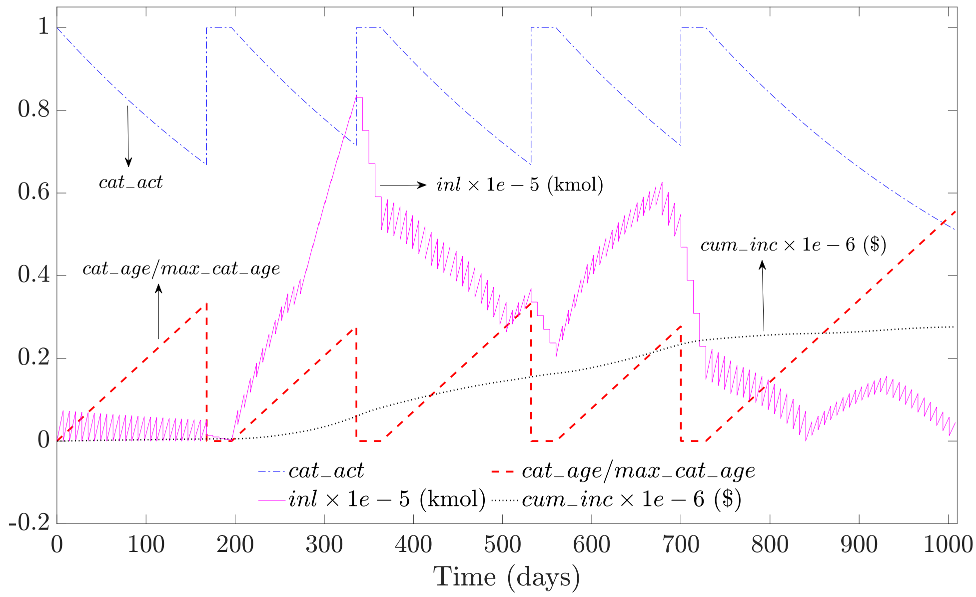


Fig. 2.22 The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory cost over the time horizon in the best solution of Case Study A, obtained using Implementation II.

Table 2.13 Details of the economic aspects of the best solution of Case Study A, obtained using Implementation II.

Economic aspect		**Symbol**	**Value ($ Millions)**
Gross Revenue from Sales		GRS	783.722
Costs	Total Inventory Costs	TIC	0.276
	Total Costs of Catalyst Changeovers	$TCCC$	42.025
	Net Penalty for Unmet Demand	$NPUD$	107.96
	Total Flow Costs	TFC	183.515
Profit		$-NC$	449.946

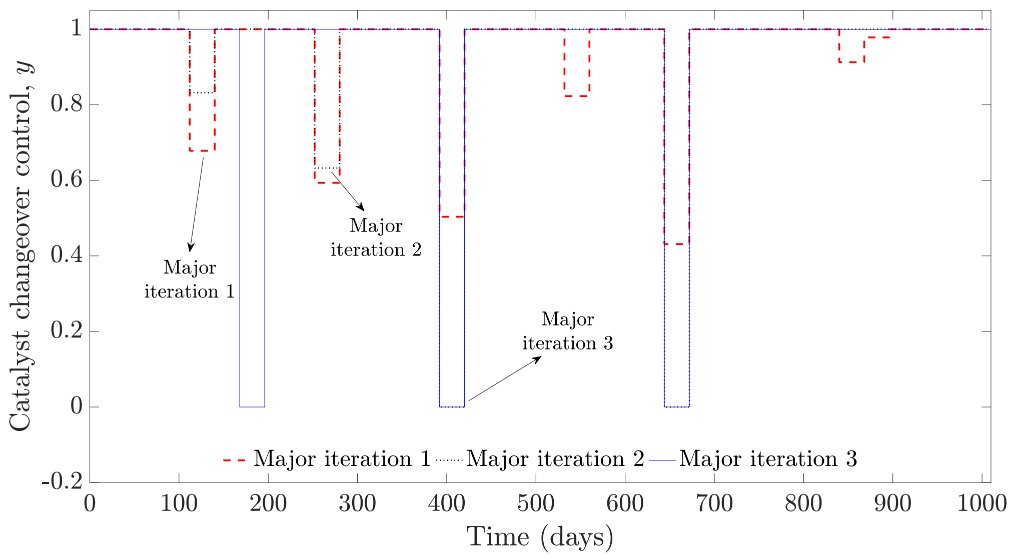


Fig. 2.23 The variation of the catalyst changeover controls over the time horizon in the best solution of Case Study B, obtained using Implementation II.

2.4.6 Case Study B: Results and discussions

Figures 2.23 - 2.26 and Table 2.14 report the features of the best local optimum among the 50 runs for Case Study B using Implementation II. These are the analogues of Figures 2.11 – 2.14 and Table 2.7, respectively, obtained using Implementation I in Section 2.3.4.

In this case, three major iterations are needed to force the catalyst changeover controls to take integer values (Figure 2.23) and the other results presented in this section correspond to the solution of the third major iteration. The explanations of the trends for all variables, and the final profit and costs values are very similar to those in Section 2.3.4.

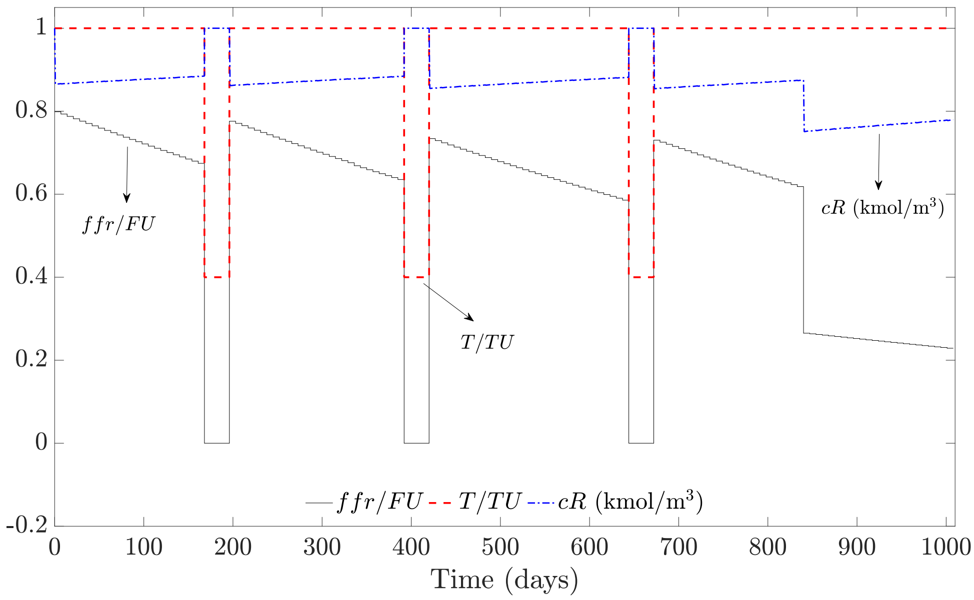


Fig. 2.24 The variation of the feed flow rate, temperature and reactant exit concentration over the time horizon in the best solution of Case Study B, obtained using Implementation II.

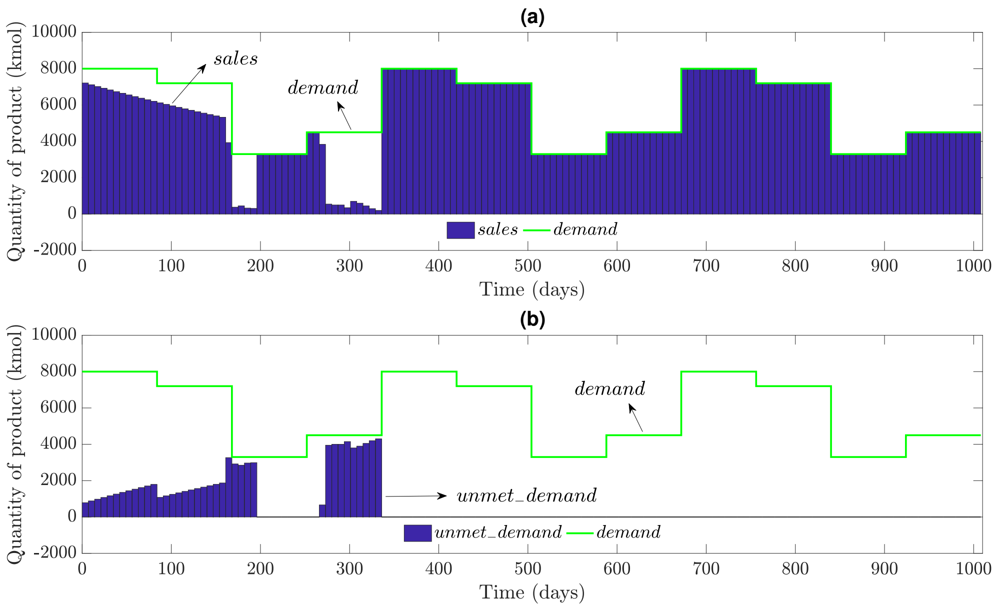


Fig. 2.25 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the time horizon in the best solution of Case Study B, obtained using Implementation II.

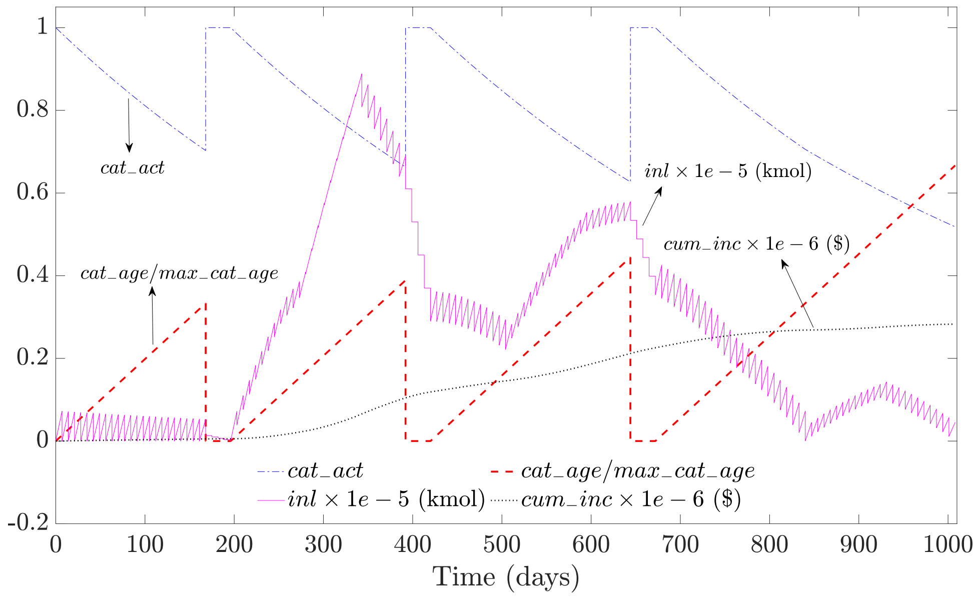


Fig. 2.26 The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory cost over the time horizon in the best solution of Case Study B, obtained using Implementation II.

Table 2.14 Details of the economic aspects of the best solution of Case Study B, obtained using Implementation II.

Economic aspect		**Symbol**	**Value ($ Millions)**
Gross Revenue from Sales		GRS	785.902
Costs	Total Inventory Costs	TIC	0.282
	Total Costs of Catalyst Changeovers	$TCCC$	30.999
	Net Penalty for Unmet Demand	$NPUD$	105.235
	Total Flow Costs	TFC	169.251
Profit		$-NC$	480.135

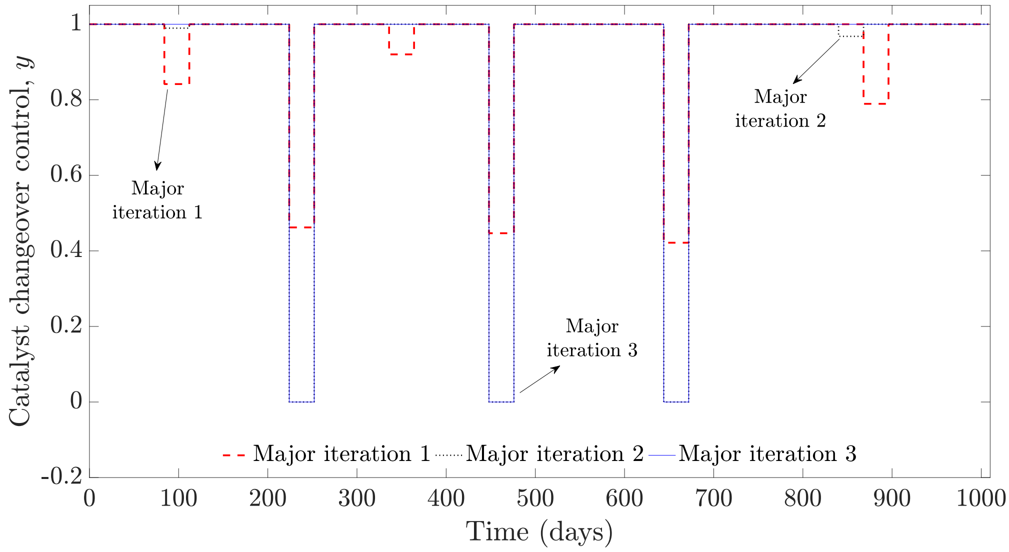


Fig. 2.27 The variation of the catalyst changeover controls over the time horizon in the best solution of Case Study C, obtained using Implementation II.

2.4.7 Case Study C: Results and discussions

Figures 2.27 – 2.30 and Table 2.15 report the features of the best local optimum among the 50 runs for Case Study C using Implementation II. Here the main reaction is of first order kinetics with respect to the reactant (equation (2.25)) and the catalyst deactivation kinetics is dependent on the product concentration (equation (2.23)). Implementation I failed to obtain results for this case study, due to problems in integrating the highly nonlinear system of ODEs.

Figure 2.27 shows the variation of the monthly catalyst changeover controls over the time horizon, across different major iterations. In this case, three major iterations are needed to force the catalyst changeover controls to take integer values. 4 of the 6 available catalysts are used, with the changeovers occurring on the 9^{th}, 17^{th} and 24^{th} months, which are times when a sufficient inventory level is present to meet the demand. All other results presented here are those obtained at the end of the third major iteration.

Figure 2.28 shows that the profiles of ffr and cR during times of catalyst operation are different from other case studies. Once again, although the time scale for the catalyst deactivation (Kd = 0.024 (1/day)) is much larger than that of the main reaction (about 24 (1/day) for temperatures used during catalyst operation) or the flow rate (thousands of cubic metres a day), cR does not remain at a constant value during catalyst operation. Therefore, the trend for cR here is not consistent with the work of Crowe (1976) at the reactor level, which predicted constant exit conversion as the optimal policy under similar conditions. The scenarios are:

- The ffr is constant at its maximum value during when the deactivation of the catalyst causes cR to increase with time.
- The ffr decreases at a rate that causes cR to decrease.

The flow costs are high in the former scenario while they are considerably lower in the latter. However, a higher value of cR in the former scenario is favourable economically as this leads to a slower rate of catalyst deactivation and a larger reaction rate, following from equations (2.23) and (2.25), respectively, while the reverse is true in the latter scenario.

Thus, it can be said that there is an interplay between the elements of the process economics, which affect the variation of ffr and cR during catalyst operation. The following interpretations are offered:

- The flow rate remains constant at its upper bound during the time the catalyst activity is relatively high. This is because the revenue from higher production and lesser unmet demand outweigh the flow costs for this time. Eventually, the catalyst activity falls low enough and causes this balance to shift. At this point, the ffr begins to decrease.
- When ffr begins to decrease, cR begins to decrease from its maximum value. Overall, a large production rate is preferred but at the same time, ffr has to be reduced in order to lower the flow costs. This compromise is attained by decreasing ffr at a rate that minimises the rate of change of cR away from its maximum value and thereby keeps the production rate as large as possible.
- During the operation of the final catalyst, the ffr experiences a sharp drop and exhibits a rate of decrease to result in a production rate that exactly fulfils the demand for the remainder of the time horizon.

Figures 2.29 - 2.30 and Table 2.15 are the analogues of Case Study C to Figures 2.21 - 2.22 and Table 2.13 in Case Study A. The profile for the catalyst activity during catalyst operation in Figure 2.30 follows from equation (2.23). The explanations for the trends of all variables in Figures 2.29 and 2.30 are similar to those of their Case Study A analogues. Table 2.15 reveals that the costs of operation take away about 45.9% of the revenue generated by the product sales, with the flow costs take up a larger proportion of the total expenses here compared to previous case studies.

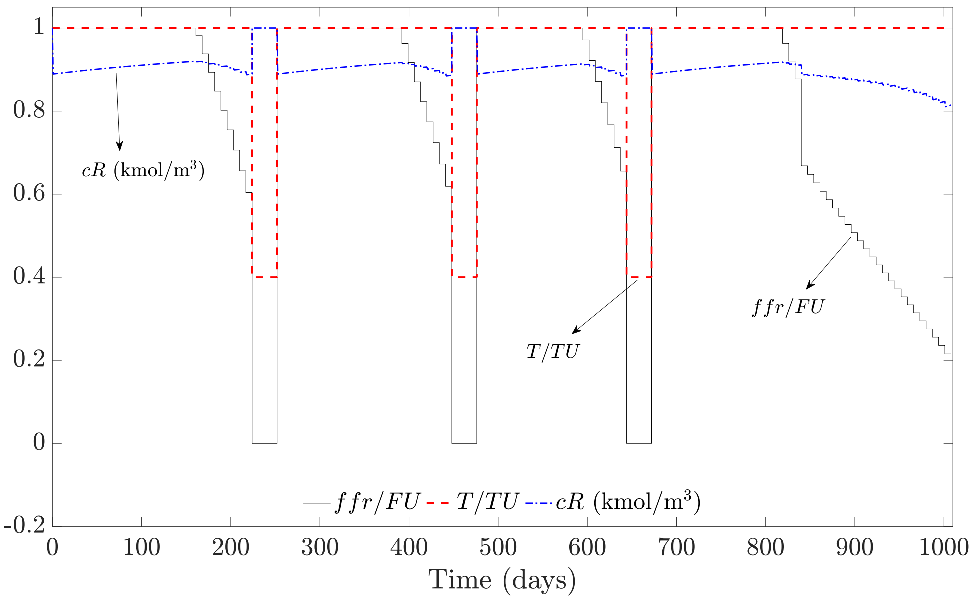


Fig. 2.28 The variation of the feed flow rate, temperature and reactant exit concentration over the time horizon in the best solution of Case Study C, obtained using Implementation II.

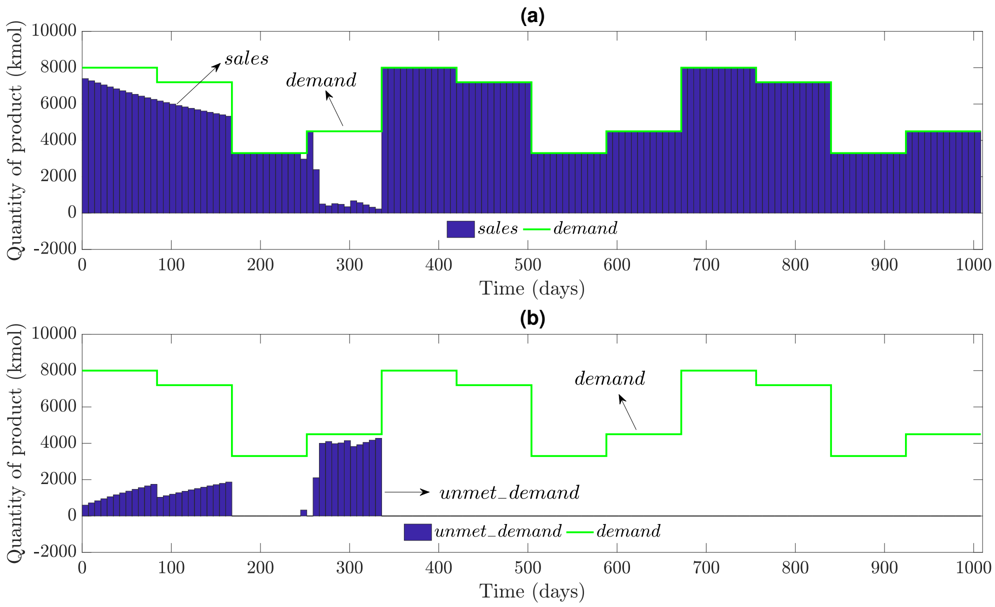


Fig. 2.29 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the time horizon in the best solution of Case Study C, obtained using Implementation II.

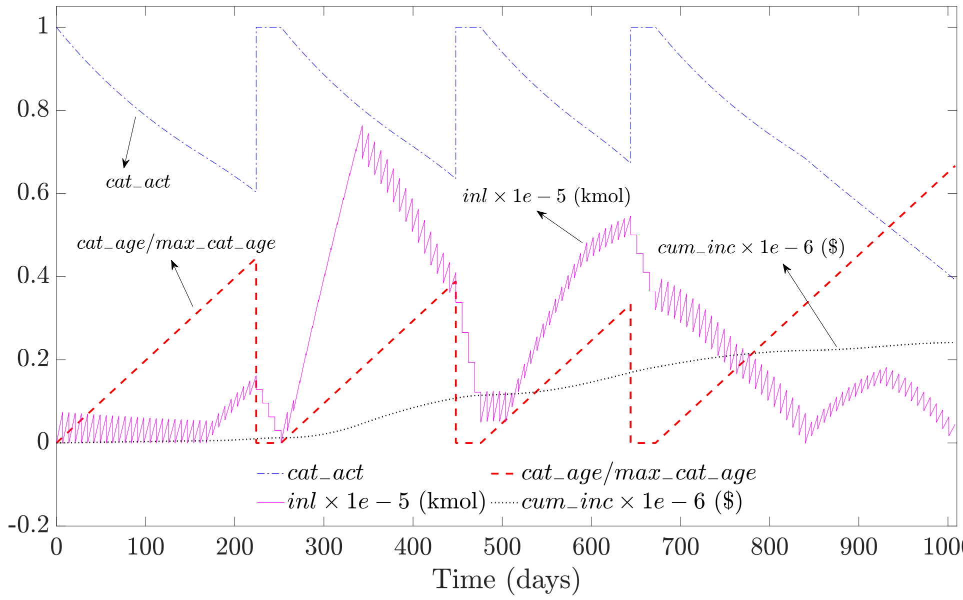


Fig. 2.30 The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory cost over the time horizon in the best solution of Case Study C, obtained using Implementation II.

Table 2.15 Details of the economic aspects of the best solution of Case Study C, obtained using Implementation II.

Economic aspect		Symbol	Value ($ Millions)
Gross Revenue from Sales		GRS	795.192
Costs	Total Inventory Costs	TIC	0.241
	Total Costs of Catalyst Changeovers	$TCCC$	30.999
	Net Penalty for Unmet Demand	$NPUD$	93.623
	Total Flow Costs	TFC	239.836
Profit		$-NC$	430.493

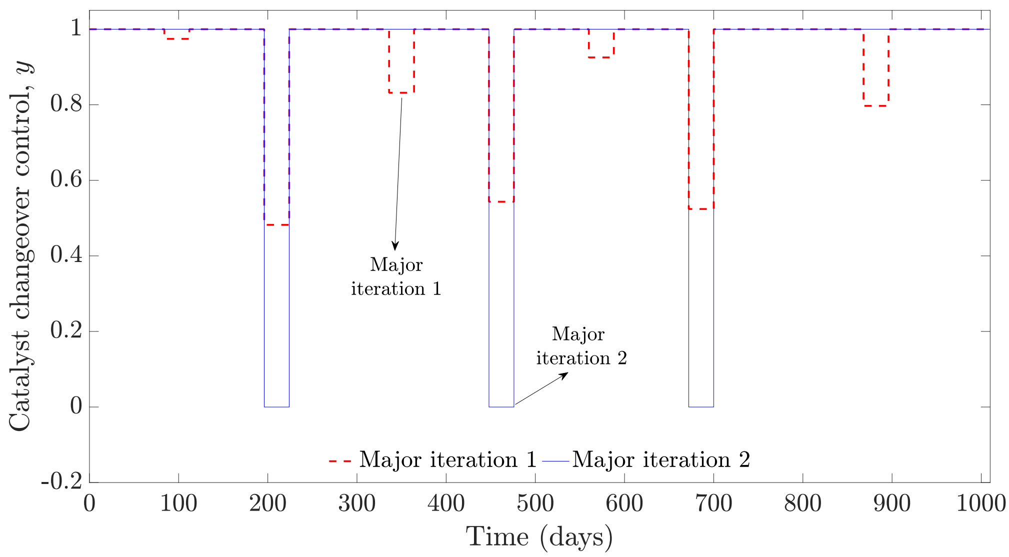


Fig. 2.31 The variation of the catalyst changeover controls over the time horizon in the best solution of Case Study D, obtained using Implementation II.

2.4.8 Case Study D: Results and discussions

Figures 2.31 - 2.34 and Table 2.16 report the features of the best local optimum among the 50 runs for Case Study D using Implementation II. Here the main reaction is of second order kinetics with respect to the reactant (equation (2.26)) and the catalyst deactivation kinetics is dependent on the product concentration (equation (2.23)). Such solutions could not be obtained by Implementation I, once again, due to problems in integrating the highly nonlinear system of ODEs.

As seen in Figure 2.31, this solution required two major iterations to force the catalyst changeover controls to take integer values. The suggestion is to use 4 of the 6 available catalysts, with the replacements occurring on the 8^{th}, 17^{th} and 25^{th} months. Similar to the previous case studies, the timing of these replacements is such that losses are minimised or sufficient inventory is present to meet demand. All other results discussed here are from the solutions of the second major iteration.

The profiles of ffr and cR in Figure 2.32 are similar to those in Figure 2.28. Only here, the ffr remains at its maximum value for a longer duration than in Case Study C because a higher value of cR is needed to compensate for the lower reaction rate.

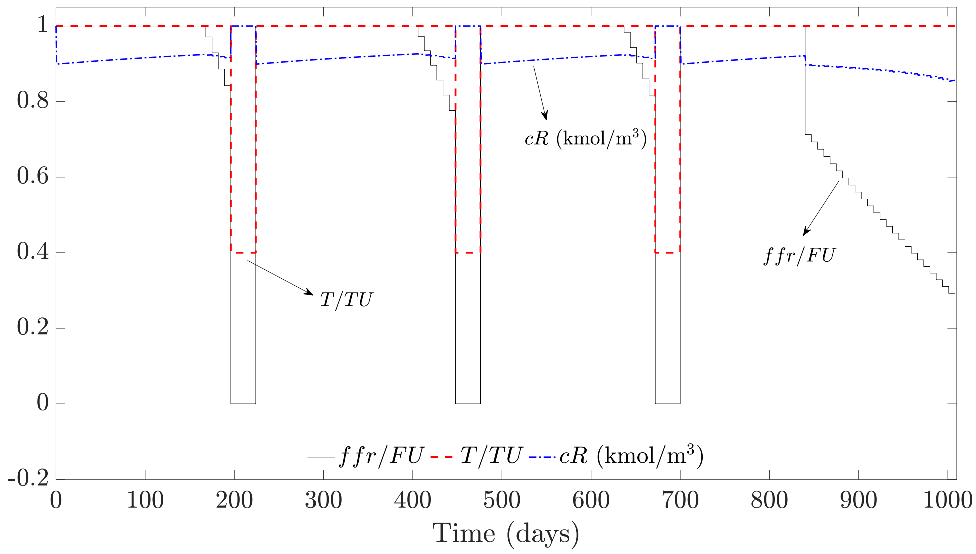


Fig. 2.32 The variation of the feed flow rate, temperature and reactant exit concentration over the time horizon in the best solution of Case Study D, obtained using Implementation II.

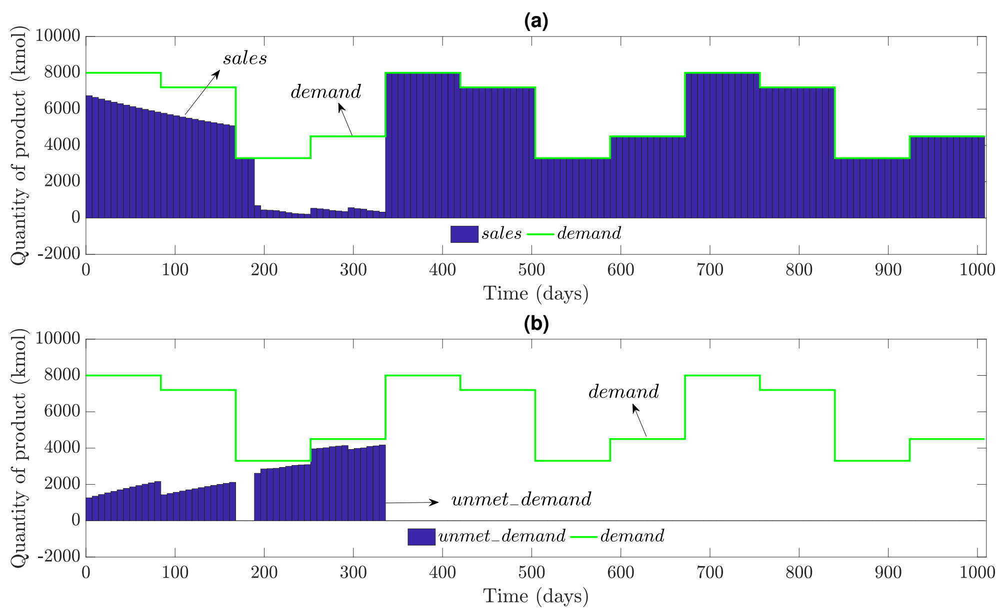


Fig. 2.33 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the time horizon in the best solution of Case Study D, obtained using Implementation II.

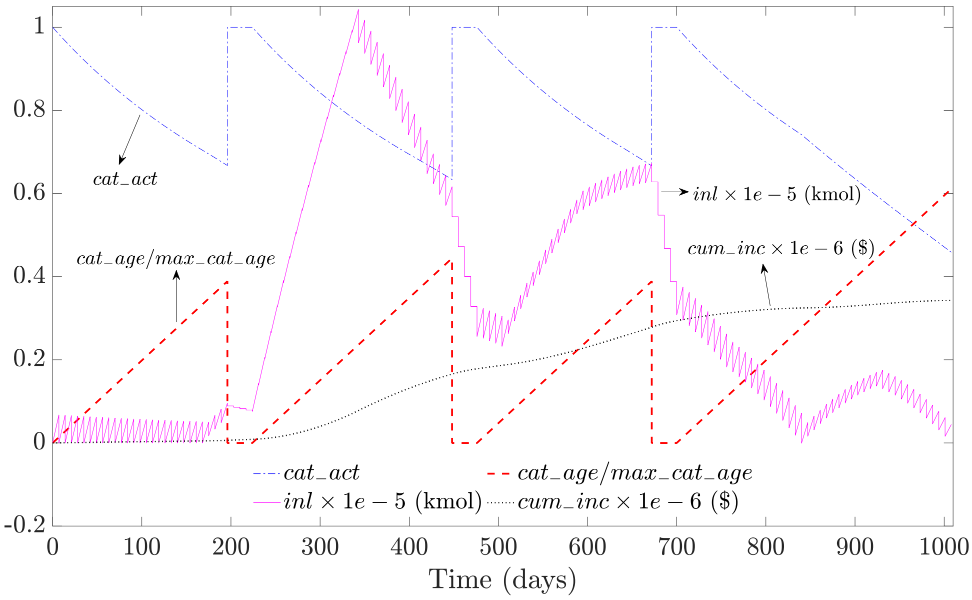


Fig. 2.34 The variation of the catalyst activity, catalyst age, inventory level and cumulative inventory cost over the time horizon in the best solution of Case Study D, obtained using Implementation II.

Table 2.16 Details of the economic aspects of the best solution of Case Study D, obtained using Implementation II.

Economic aspect		**Symbol**	**Value ($ Millions)**
Gross Revenue from Sales		*GRS*	752.937
Costs	Total Inventory Costs	*TIC*	0.343
	Total Costs of Catalyst Changeovers	*TCCC*	31.525
	Net Penalty for Unmet Demand	*NPUD*	146.441
	Total Flow Costs	*TFC*	249.539
Profit		$-NC$	325.089

The explanations for the trends of the variables in all other figures are similar to those of their Case Study C analogues. Table 2.16 reveals that the costs of operation take away about 56.8% of the revenue generated by the product sales.

2.4.9 General comments

In this section, some general comments are offered regarding the results obtained by Implementation II for all case studies.

The results presented include the trends of the decision variables, and the state variables that follow accordingly, over the time horizon of only the best solution, or the local optima that resulted in maximum profit, of each case study. While it is possible to present a distribution of each decision variable over the 50 runs for each case study in a manner similar to that done for the profit, for example, in Figures 2.4 and 2.16, these are not informative enough to justify the volume of the thesis that would be consumed by such a presentation, given that there are hundreds of decision variables involved. However, there are some notable points to be stated regarding decision variables obtained in all the other local optima of each case study:

- Within a particular case study, the local optima differed considerably in the obtained values of the catalyst changeover decision variables that decided the number and timing of catalyst replacements. This was the major factor leading to differences in profits between the different local optima obtained as the different catalyst replacement schedules impacted the decision variables of feed flow rate and sales.
- Within a particular case study, the decision variables of the feed flow rate differed between different local optima in the values obtained during the operation of the final catalyst. In each case study, in the best solution presented, there was a sharp drop observed in the feed flow rate near the end of the time horizon which occurred in order to bring the exit concentration, and hence the production rate, to a value that exactly fulfilled demand for the remainder of the time horizon and thereby maximise the profits of the process by preventing the use of excess raw material. In the other local optima of each case study, while the trend of the feed flow rate was similar to in the best solution and the demand near the end of the time horizon was fulfilled, the sharp drop in the feed flow rate was either not present or was of a smaller magnitude in comparison to that observed in the best solution. This led to the total flow costs in the other local optima being higher compared to that in the best solution and this could be attributed to the catalyst replacement schedules in the other local optima being less efficient than

in the best solution. However, it is highlighted that for all optima obtained within each case study, during the times of operation of all catalysts except the final one, the feed flow rates were similar in terms of magnitudes as well as trends, which led to similar magnitudes and trends of the exit concentration during these times as well.

- For all optima in all case studies, the temperature was set to its upper bound during times of catalyst operation. This followed from the fact that in all case studies, the catalyst deactivation rate constant was independent of temperature and the maximum rate of the product formation reaction could be obtained by operating at the maximum allowable temperature.

- Within each case study, the decision variables of the sales differed between the obtained optima in terms of the distribution and quantity occurring in the first year of the time horizon. In comparison to the best solution, the distribution of sales in the first year in the other local optima was such that the quantity of sales was either similar or lower in comparison to the former. This led to the gross revenue from the sales in the first year in the other local optima to be less than or at most similar to that in the best solution and this could be attributed to the catalyst replacement schedules in the other local optima being less efficient than in the best solution. It is highlighted that in all optima obtained in all case studies, the sales completely fulfilled the product demand in the second and third years of the time horizon and this could be attributed to the inflation causing the product sales price and penalty for unmet demand to be higher in those years compared to the first.

The different case studies examined differ on the basis of the kinetics of either the catalyst deactivation or the product formation reaction. A comparison of the solutions of Case Studies C and D, which involved similar expressions of the rate of catalyst deactivation but different expressions for the rate of the product formation reaction, indicated that the profiles of the decision and state variables of these case studies over the time horizon were quite similar. However, a comparison of the solutions of Case Studies A, B and C, which involved similar expressions for the rate of the product formation reaction but different expressions of the rate of catalyst deactivation, indicated similarities in the profiles of some of the variables over the time horizon, but considerable differences among other variables of these case studies. The following comments can be made regarding the effect of the different deactivation rate expressions on the different variables involved:

- Depending on the expression of the rate of deactivation, the profile of the decision variable of the feed flow rate over the time horizon differed considerably during times of catalyst operation. This led to the profile of the state variable of the reactant exit concentration during those times also varying considerably depending on the expression of the rate of deactivation used. Clearly, the profile of the state variable of the catalyst activity over the time horizon during catalyst operation also varied depending on the deactivation rate expression considered.

- Regarding the values obtained by the catalyst changeover controls, for all the different expressions of the rate of deactivation examined, the number of catalyst replacements varied between 2 and 4. No patterns could be observed in the timing of catalyst replacements with regard to the deactivation rate expression used.

- The explanations for the profiles over the time horizon of the decision variables of the temperature and the sales, and the state variables of the catalyst age, the inventory level and the cumulative inventory cost did not vary when different expressions of the rate of deactivation were used.

In terms of the economics of the best solutions of all case studies, it is observed that the total flow costs always form the largest contribution, followed by the penalty for the unmet demand. This merely follows from the values of parameters used which, as mentioned previously, were invented, as the previous publications in this area did not reveal any such data citing confidentiality reasons. It may well be that for a different set of parameter values, the order of decreasing contribution of the various costs is different. As such, no other comments can be made regarding the magnitudes of the economic components or the order of contribution of the various costs involved.

2.5 Summary, further discussions and conclusions

In this chapter, a novel, optimal control methodology has been developed to optimise maintenance scheduling and production in a process containing a single reactor using decaying catalysts. This methodology is based on a formulation of this problem as a multistage mixed integer optimal control problem (MSMIOCP). The integer controls in this formulation are the binary decisions to decide when to schedule catalyst changeovers, the continuous controls include the operating conditions of the reactor and the quantity of product sales, and the state variables characterise the state of the process. The DAEs of the formulation represent the process model, the constraints represent the operating limits of the process and the binary restrictions on the catalyst changeover controls, and the objective function represent the net costs of the process.

The elements of the process considered in this chapter are of similar structure to the process studied in the works of Houze et al. (2003) and Bizet et al. (2005). However, since those works did not reveal the process model or any of the parameters used, citing confidentiality reasons, in this chapter, the process model was constructed and an invented set of parameters were used. Four case studies were examined for the process, which differed depending on the kinetics of the product formation reaction or the catalyst deactivation, in the process model.

The solution methodology attempted was to formulate this process as an MSMIOCP in which the binary controls that scheduled catalyst changeovers appeared linearly in the model equations. A theoretical analysis suggested that, by virtue of the linear occurrence of these binary controls, these controls could be expected to exhibit a bang-bang behaviour in the optimal solution. Hence, these controls were considered continuous rather than discrete in the MSMIOCP formulation of the process, and this problem was attempted be solved as a standard nonlinear optimisation problem, without using mixed-integer optimisation methods. Due to the non-convex nature of the problem, 50 optimisation runs, each which used different initial guesses, were attempted for each case study.

However, the solution implementation faced complications due to the complex nature of the problem and required using two different implementation methodologies, Implementation I and Implementation II, each of which had their own relative advantages:

1. Implementation I was favourable from a theoretical point of view, as its solutions exhibited the bang-bang property for the catalyst changeover controls, consistent with predictions of the theoretical analysis, and so, the solutions produced by this implementation can be considered to be optimal. However, this implementation's performance was sensitive to the initial guesses used and even for the relatively less nonlinear process models of Case Studies A and B, only a limited set of successful runs could be obtained, and the vast majority of runs either converged prematurely or crashed due to problems in integration. For the more nonlinear process models of Case Studies C and D, every single run crashed due to problems in the integration. The integration problems could probably be attributed to inadequacies of the available MATLAB ODE integrator suites.

2. Implementation II did not exhibit the bang-bang property for the catalyst changeover controls and required using a penalty term homotopy technique in order to force these controls to take values of 0 or 1. But this implementation was robust in providing high quality solutions for all case studies, regardless of the initial guesses used. The lack of

bang-bang behaviour is probably due to a shortcoming of the IPOPT tool. The range of profit values obtained for Case Studies A and B using Implementation II compared well with those of the successful runs of Implementation I, thereby suggesting that these solutions obtained by Implementation II were indeed optimal. Further, this also suggests that the solutions obtained by Implementation II for Case Studies C and D were optimal as well.

Overall, Implementation II was more robust in comparison to Implementation I in the sense that unlike the latter, the former was not sensitive to the initial guesses used, in obtaining solutions within the stipulated tolerances. This suggests that the optimal control methodology to be used to solve problems of this kind is to use an MSMIOCP formulation of the problem combined with a solution procedure of the principle of Implementation II.

For the best solution among the successful runs for Implementation I for each of Case Studies A and B, and for the best solution among the 50 runs carried out using Implementation II for each case study, the variation of all control and state variables were plotted over the time horizon and the economics of the process was presented in a table. Explanations were provided for the trends of all variables, which were mainly focused on increasing profit while efficiently managing all costs in order to balance the trade-offs involved.

A notable result was in Case Study A wherein the policies for the reactant exit concentration and the temperature of operation correlated well with that of published literature (Szépe and Levenspiel, 1968) at the reactor level. However, the policy for the reactant exit concentration in the obtained solutions of the other case studies was not consistent with the related work (Crowe, 1976) at the reactor level, indicating that that policy may not hold when inventory, sales and demand considerations come into play.

The elements of the industrial process considered in this chapter are similar to that considered in the works of Houze et al. (2003) and Bizet et al. (2005) and the aim of developing this optimal control methodology was to improve on the quality of solutions obtained for that process in those works, which used mixed-integer optimisation methods. An ideal evaluation of the quality of this methodology would have been by applying this methodology to the problem considered in those works and comparing the solutions obtained with the solutions of those works. However, this was not possible, as those works did not reveal the model or any parameters used, citing confidentiality reasons and therefore, in this chapter, the process model was constructed and an invented set of parameters were used. But a comparison between the methodology developed in this chapter and the mixed-integer methods used in those works can be drawn using a number of other properties, and these are described next.

Details of the size of the two problems in the work by Houze et al. (2003), which considered time horizons of 24 months and 48 months, while using a total of 4 catalyst loads for each of these time horizons, are shown in Table 2.17. Details of the size of the two problems in the work by Bizet et al. (2005), which considered 2 catalyst loads to be used over a time horizons of 74 months and 3 catalyst loads to be used over a time horizon of 108 months, are shown in Table 2.18. Table 2.19 shows how the problem sizes for these time horizons would come out to be if the proposed methodology was applied for the problem formulations.

As can be seen from Table 2.19, for a given time horizon, the number of discrete variables, continuous variables and constraints involved, when the proposed methodology is applied, are considerably smaller in comparison to the corresponding time horizon in Table 2.17 or Table 2.18. This is because, while mixed-integer methodologies approximate differential equations as a collection of steady state equations, which creates additional variables and constraints in the optimisation phase, in the proposed methodology, these differential equations are solved by the feasible path approach, which prevents that from happening. And since, in the proposed methodology, catalyst replacements occur inherently during the optimisation, without requiring mixed-integer methods, the problem sizes in Table 2.19 for each time horizon apply regardless of the number of catalyst loads available to be used.

Thus, the features of the proposed methodology are such that, for a give time horizon, the size of the problem formulated using the proposed methodology is much smaller compared to when mixed-integer formulations are used. The proposed methodology's characteristic of enabling smaller problem sizes implies that convergence to optimal solutions is facilitated when using this methodology and this characteristic provides the methodology with the advantage of robustness over mixed-integer methodologies.

Table 2.17 Details of the size of the problems considered in the work of Houze et al. (2003).

Number of months	**Number of catalyst loads**	**Number of discrete variables**	**Number of continuous variables**	**Number of constraints**
24	4	299	1503	3960
48	4	587	2943	8576

Table 2.18 Details of the size of the problems considered in the work of Bizet et al. (2005).

Number of months	**Number of catalyst loads**	**Number of discrete variables**	**Number of continuous variables**	**Number of constraints**
74	2	334	3717	12596
108	3	816	8013	33230

Table 2.19 Details of the size of the problems obtained when using the proposed optimal control methodology for the time horizons considered in the works of Houze et al. (2003) and Bizet et al. (2005).

Property size	**Number of months**			
	24	**48**	**74**	**108**
Number of ODEs (for all state variables)	480	960	1480	2160
Number of discrete variables	24	48	74	108
Number of continuous variables	288	576	888	1296
Number of constraints	1033	2065	3183	4645

In addition, the following points of comparison can be drawn between the methodology presented in this chapter and the works of Houze et al. (2003) and Bizet et al. (2005):

1. The number of catalyst loads considered in the work of Houze et al. (2003) was 4 and that in the work of Bizet et al. (2005) was either 2 or 3. If that number was increased, the number of combinations involved in their solution methodology would increase exponentially and so, obtaining good solutions would be difficult and require a very large amount of computational effort. On the other hand, in the proposed methodology, catalyst replacements are scheduled inherently during the optimisation, without mixed-integer methods. Therefore, as demonstrated in this chapter, good solutions can be obtained in a reasonable amount of time even if the number of available catalyst loads is 6, and this would be the case even if an infinite number of catalyst loads are available to be used. This highlights the efficiency of the methodology over the mixed-integer optimisation methods.

2. In the works of Houze et al. (2003) and Bizet et al. (2005), the flow rate, temperature and sales are decisions to be taken on a monthly basis, whereas here, those controls

are optimised on a weekly basis. The smaller problem size enabled by the proposed methodology facilitates producing solutions which are more informative compared to those works. If in those works, these decisions were taken on a weekly rather than monthly basis, the problem sizes, as shown in Tables 2.17 and 2.18, would have increased almost 4-fold, and the mixed-integer methodologies used by those works would have faced great difficulties in obtaining solutions.

3. The use of the feasible path approach in this methodology, wherein state-of-the-art integrators solve the differential equations, enables an accurate description of the dynamics of the process and hence, accurate solutions. However, in the works of Houze et al. (2003) and Bizet et al. (2005), the differential equations present are approximated as a collection of weekly steady state equations, which means the results obtained in those works cannot be considered accurate. Thus, the solutions obtained by the proposed methodology are more accurate and hence, more reliable in comparison to the results obtained in those works.

4. The solution times in the works of Houze et al. (2003) and Bizet et al. (2005) are in the order of seconds. However, the solution times for the methodology proposed here are in the order of hours. This is due to the large computational effort spent in solving the differential equations to a high accuracy at each iteration of the optimisation. However, this additional computational effort is outweighed by the robust, reliable and efficient solutions obtained. Though not done here, the solution times can be greatly reduced, by the use of high performance and parallel computing facilities.

The preceding discussion indicates the high quality of solutions obtained by the proposed methodology to solve the problem of optimising maintenance scheduling and production in a process containing a single reactor using decaying catalysts, in comparison to previous publications that used mixed-integer optimisation techniques to solve this problem. It is intended to conclude this chapter by highlighting how the proposed methodology overcomes the drawbacks of, and is therefore advantageous over, mixed-integer optimisation techniques.

While Section 2.1.2 mentions the advantages the proposed methodology could potentially offer over mixed-integer techniques, this section cites the bang-bang behaviour of the catalyst scheduling controls as the cause for the efficiency of the methodology, without knowledge of the complications that the solution procedures would face. As was seen, the solution procedure of Implementation I, while demonstrating the bang-bang behaviour, did not perform satisfactorily and it was the solution procedure of Implementation II, which did not exhibit the bang-bang behaviour, that produced high quality solutions and is suggested to solve problems of this kind.

Therefore, a modified form of Section 2.1.2 that reflects the features of Implementation II is presented in the next section, which form the conclusions of this chapter. This section will also serve as a reference for the future chapters of the thesis.

2.5.1 Advantages over mixed-integer methods

The advantages offered by the proposed methodology, of using an MSMIOCP formulation in combination with the solution procedure of Implementation II, over mixed-integer optimisation techniques are highlighted using the following points:

1. When mixed-integer methods are used, all differential equations present in the problem are approximated as a collection of steady state equations, which are then imposed as equality constraints in the optimisation phase. Following such practices causes the problem to contain a very large number of variables and a large number of potentially highly nonlinear constraint equations. In such cases, the optimiser could face difficulties in converging to a solution, which is further accentuated when a large number of differential equations are involved, longer time horizons are considered or higher accuracy is required. In addition, if a larger number catalyst loads are available to be used, the problem size becomes larger and this can lead to further difficulties in converging to a solution.

   However, in the methodology proposed here, the differential equations are solved using the feasible path approach, without being considered as constraints in the optimisation step.
   Further, in the proposed solution procedure, the binary controls to schedule catalyst changeover are considered as continuous controls which are forced to take values of 0 or 1 by means of a penalty term homotopy technique. By virtue of this technique, the 0 or 1 values for these controls are obtained inherently during the solution of this problem as a standard nonlinear optimisation problem, without the use of mixed-integer optimisation methods. Hence, even if an infinite number of catalyst loads are available, the problem size will not increase as the decisions on how many catalyst loads to use and when to schedule catalyst changeovers are taken inherently during the optimisation.

   Thus, when using the proposed methodology, the problem involved is of a much smaller size compared to when mixed-integer techniques are used and optimal solutions can be obtained from random starting points, even when a large number of differential equations are involved, long time horizons are considered and a large number of catalyst loads are available.

Hence, the proposed methodology is more robust in converging to optimal solutions, in comparison to mixed-integer techniques.

2. As mentioned in the previous point, the mixed-integer techniques approximate the differential equations as a collection of steady state equations. This negates an accurate description of the dynamics of the problem within the time period in which those differential equations are approximated as such.

   However, in the proposed methodology, a feasible path approach is used, wherein state-of-the-art integrators are employed to solve the DAEs present in the problem. These integrators solve even complicated DAEs to a very high accuracy.

   Therefore, the solutions obtained using the proposed methodology are of greater accuracy and hence, more reliable than those obtained using mixed-integer methodologies.

   By enabling the advantages of robustness and reliability, the feasible path approach also enables the methodology to avoid making the difficult compromise between accuracy and ease of convergence, which is faced by mixed-integer techniques.

3. Mixed-integer techniques are combinatorial in nature, meaning that the computational effort to solve a problem using these methods increases exponentially with the number of integer decision variables involved in the problem. If mixed-integer techniques are used in the problem under consideration here, when a large number of catalyst loads are present, the computational effort involved in optimising the scheduling of catalyst changeovers will become huge and so, an enormous amount of computational power will be needed to solve the problem.

   However, as mentioned previously, in the proposed solution procedure, by means of the penalty term homotopy technique, the 0 or 1 values for the catalyst changeover controls, assumed continuous, are obtained inherently during the solution of this problem as a standard nonlinear optimisation problem, without the use of mixed-integer optimisation methods.

Hence, no computational effort is spent in deciding when to schedule catalyst changeovers or how many catalyst loads to use as the penalty term homotopy technique takes care of this. And this will be the case regardless of the number of catalyst loads involved, thereby making it possible for the methodology to obtain optimal maintenance schedules for catalyst replacements even if an infinite number of catalyst loads are available to be used.

This feature of the proposed methodology makes it more efficient in comparison to mixed-integer methods.

Thus, in this chapter, in line with the first objective of the thesis, a methodology has been developed that can effectively optimise the maintenance scheduling and production in a process containing a reactor using decaying catalysts, and which can overcome the drawbacks faced by mixed-integer optimisation techniques in solving this problem. In the next chapter, the optimal control methodology developed in this chapter is extended to optimise maintenance scheduling and production in parallel lines of reactors using decaying catalysts.

Chapter 3

Optimisation of a process containing parallel lines of reactors

In this chapter, an optimal control methodology is developed for optimising maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts. This methodology is an extension of the methodology developed in the previous chapter, which was used to optimise similar aspects in a process containing a single reactor using decaying catalysts. Following from the previous chapter, this methodology involves a multistage mixed-integer optimal control problem (MSMIOCP) formulation in combination with a solution procedure of the principle of Implementation II to solve this problem. The methodology is applied to the case study of an industrial process that operates a single feed over a set of 4 reactors which operate in parallel and produce the same product. The solution procedure is successful in producing high quality solutions for this case study. Further, the results obtained indicate that the methodology can provide potential advantages of robustness, reliability and efficiency over mixed-integer optimisation methods in solving problems of this kind.

The structure of this chapter is as follows. Section 3.1 contains an introduction to the problem and a literature review of the publications that have examined topics similar to this problem. In Section 3.2, the proposed optimal control methodology is applied to the case study of an industrial process, beginning with an optimal control formulation of the problem, which is followed by details of the implementation of the solution procedure and a discussion of the results obtained. Section 3.3 contains a summary and the conclusions of the chapter.

3.1 Introduction and literature review

In the previous chapter, the process examined involved only a single reactor using decaying catalysts. However, given that the maintenance operation to replace the catalyst involves shutting down the reactor and hence, a loss of production time, it is common for industries to use parallel lines of reactors. A parallel set up can avoid a complete stop of production and greatly improve the flexibility of the process, by allowing one reactor to be shut down for catalyst replacement, while the remaining reactors continue to produce product to meet the demand.

However, in order to minimise the negative effects of catalyst deactivation and ensure efficient operation of such a set up, an optimal operational plan for the process is needed, which is similar to what was required to optimise the performance of the process containing a single reactor using decaying catalysts. That is, the optimal operational plan should specify an optimal maintenance schedule for catalyst replacements in each reactor and an optimal production plan for the whole process in an integrated manner.

The optimal maintenance schedule for catalyst replacements should specify the optimum number of catalyst loads to use and the optimal time for catalyst replacements in each reactor of the parallel set up, such that, the trade-off between attaining a high production rate, and having low maintenance costs and effective production times for each reactor is optimally balanced in view of the whole process. The maintenance schedule may also be required to fulfil a constraint that no two reactors undergo catalyst replacement at the same time due to production requirements or the maintenance labour and equipment availability.

The optimal production plan should specify the optimal operating conditions of each reactor as well as the optimal management of product inventory and sales to meet seasonal demand. The operating conditions to be specified include the flow rate to and temperature of operation of each reactor during the times the catalyst is in operation, such that the product yield is maximised under the conditions created by the catalyst deactivation. These operating conditions should also be tailored to produce an adequate inventory of product such that the sales can occur to effectively meet varying demand across the time horizon, while excessively high storage costs are also avoided.

Identifying such an optimal maintenance schedule and an optimal production plan in an integrated manner requires solving a highly challenging modelling and optimisation problem containing a very large number of variables and constraints. This problem is of significantly higher complexity compared to the problem of optimising such aspects in a process containing a single reactor, which was considered in the previous chapter.

A thorough literature survey did not result in finding any work that has explicitly claimed to optimise maintenance scheduling, operating conditions, inventory management and sales to meet time-varying demand in a process involving parallel lines of reactors using decaying catalysts. Most existing literature investigate the optimisation of maintenance scheduling and a subset of the other mentioned decisions in different industrial applications that use parallel processing lines and experience decaying performances. Further, these contributions are not related to reactions systems. And it is mixed-integer methods that have mainly been used for the optimisation.

For example, Castro et al. (2014) have presented a maintenance scheduling model for a gas engine plant with parallel units that aims to maximise revenue from electricity sales while considering seasonal variation in electricity pricing. While no decay models were explicitly involved, the maintenance operations were of a preventive nature, in order to avoid premature aging and failure of the gas engine units which could lead to unplanned and costly power outages.

In the bio-pharmaceutical industry, Liu et al. (2014) have optimised the scheduling of product manufacture on parallel suites, as well as inventory management and sales to meet varying demand, while considering decaying performance of the chromatography resin in the downstream purification process. The decaying performance of the chromatography resin was obtained from a known table that provided information on the resin yields for the number of batches manufactured and time slots operated.

But the two works mentioned above did not consider the optimisation of operating conditions, and so the underlying equations were mostly free from the occurrence of nonlinearities. Thus, less computationally intensive MILP models could be used for the solution of these problems.

For a parallel network of compressors experiencing decaying performance due to fouling and degradation by fluid particles, Kopanos et al. (2015) and Xenos et al. (2016) have optimised the maintenance scheduling, the operating mass flow rate and product inventory management to meet varying demand. The decaying performances of the compressors in Kopanos et al. (2015) were modelled using the power consumption in the compressors, which were expressed by regression functions that were derived using technical and historical data. In Xenos et al. (2016), the decaying performance of each compressor was modelled using a linear function between the extra power consumption due to degradation and cumulative operation time. But the authors of these works admit to seeking to avoid hard MINLP formulations by linearising the underlying equations to form MILP models and concede that such linear approximations can cause errors in the results.

Heluane et al. (2007, 2004) have developed MINLP formulations to optimise the maintenance scheduling and operating mass flow rate conditions of evaporator systems involving parallel lines that decay in performance due to heat transfer induced fouling. For each evaporator, the decaying performance was modelled using an experimentally obtained expression for the decrease of the heat transfer coefficient with time, which was a function of temperature and concentration. However, these works focused on obtaining cyclic schedules and did not consider the problem of inventory management or sales to meet seasonal demand.

In ethylene plants, a cracking furnace is used to break long-chain hydrocarbons into valuable products and these plants have multiple such furnaces operating in parallel. Coke depositions on the walls reduce the efficiency of the furnaces and shut downs for decoking operations are necessary to restore performance. Because of the importance of ethylene in the petrochemical industry and the nonlinearity of the equations involved, a number of strategies involving MINLP formulations have been proposed for optimising the decoking maintenance scheduling in tandem with the operating conditions.

To optimise the scheduling of decoking and operating flow rates in parallel cracking furnaces of an ethylene plant, Jain and Grossmann (1998) proposed an MINLP model and used a Branch & Bound method for its solution. The decaying performance was modelled by assuming that the yield of ethylene decreased exponentially with time. The MINLP model of Jain and Grossmann (1998) was improved by the MINLP framework devised by Liu et al. (2010) which removed the assumption of identical feed processing times and also ensured that decoking of any two furnaces did not occur simultaneously. Zhao et al. (2010) extended the problem to consider secondary cracking of recycled ethane in a separate furnace and obtained solutions using a framework similar to Liu et al. (2010). But in all these works, the operating profiles of flow rate and temperature were constant over a feed run length.

In order to provide a dynamic description of the optimal operation profiles of flow rate and temperature in each furnace, Jin et al. (2015) presented a mixed-integer dynamic optimisation

(MIDO) formulation. In this formulation, the state variables involved, including the coke deposition thickness that quantified the decay in furnace efficiency, were predicted using a surrogate model, which in turn was formulated by applying an artificial neural network to data obtained from a furnace simulator. The MIDO formulation was then discretised and converted into a large scale MINLP to be solved using standard solvers. They claim to significantly improve economic performance compared to traditional methods that keep the operating variables constant over a run length. But a major limitation of this work is that simultaneous decoking of furnaces was allowed.

Given the difficulties faced by the traditional gradient based optimisation methods in handling the highly nonlinear constraints of MINLP models, Yu et al. (2017) have proposed an alternative methodology. They have used a population-based Diversity Learning Teaching Learning Based Optimisation (DLTLBO) algorithm on a problem set up similar to that of Jin et al. (2015) and have produced better solutions compared to the latter, without the assumption of simultaneous decoking. However, as a metaheuristic approach, a theoretical convergence to optimality is not guaranteed.

Lin and Du (2018) have proposed another methodology, which combines deterministic and population based optimisation methods. A MIDO formulation is discretised into an MINLP and a two-level nested optimisation problem is solved. A Genetic Algorithm is used for the outer level MILP problem that solves for the scheduling of parallel cracking furnaces and a Sequential Quadratic Programming algorithm solves the inner level NLP problem, which optimises the operating conditions. The formulation here also modelled the coke thickness deposition that quantified the decay in furnace efficiency, alongside other state variables, using a surrogate model that was formulated by applying an artificial neural network to data obtained from a furnace simulator. The methodology enabled a drastic reduction in problem size and produced better profits in smaller computational times compared to standard MINLP methods. But the authors themselves admit that treating the integer and continuous variables separately may be inadequate if these variables are highly interdependent.

Different from the above works which obtained cyclic schedules, Lim et al. (2006) developed an MINLP formulation for a fixed time horizon. The work aimed to optimise the decoking schedule for a set of parallel furnaces while ensuring non-simultaneous decoking and also deciding the distribution of the inlet naphtha feed flow among them for a naphta cracking furnace system. The model for the decay in furnace efficiency in this work as well was a part of a surrogate model that was formulated by applying an artificial neural network to data obtained from a furnace simulator. To handle the computationally intractable problem size, three solution strategies were proposed to improve solution quality and computational time. The best of these strategies involved solving the decoking scheduling as a master MILP problem and the flow rate optimisation as a subproblem. But as mentioned previously, treating the integer and continuous variable separately may be inadequate.

Further, all of the aforementioned papers assume a constant demand over an infinite time horizon and do not consider the problem of inventory management or sales to meet time-varying demand. Only a limited number of publications consider this additional problem and these are discussed next.

Schulz et al. (2006a,b) developed a multistage MINLP model for scheduling shutdowns of 8 parallel furnaces in an ethylene plant, while also determining the operational profiles and inventory management to satisfy time-varying product demand. However, this work did not reveal the underlying model equations and had restrictive assumptions such as identical cycle times for all furnaces and a linear coking rate to represent the decay in performance.

Other works have focused on developing new strategies to overcome the drawbacks of large computational times and intractable sizes of large scale MINLP problems. For example, Su et al. (2015) presented strategies such as multi-generational cuts, hybrid methods and partial surrogate cuts which, when used with MINLP methodologies such as the Generalised Benders Decomposition and Outer Approximation can enhance convergence in large MINLPs. Su et al. (2016) demonstrated that one such algorithm produced faster convergence in a problem of optimising operation and cleanup scheduling of parallel furnaces while also managing inventory to meet demand requirements in an ethylene cracking process with feedstocks and energy constraints. However, the model involved simplistic assumptions such as constant operating flow rates and a linear coking rate to represent the decay in performance. More complex models that could cause greater intractability of the MINLPs have not been investigated.

Finally, for a large scale MINLP of decoking scheduling and operational optimisation in an ethylene plant to meet known product demand, wherein the decay is modelled as an exponential decrease in the yield of ethylene over time, Wang et al. (2016) have proposed a Lagrangian decomposition method. In this method, the problem is decomposed into planning and scheduling problems and Lagrange multipliers are used to communicate information between the two. While the algorithm has produced better objective function values in smaller CPU times in comparison to standard MINLP solvers, the authors admit that due to the complexity of the formulation, they face difficulties in converging to optimality. Further, this method can mainly be applied only in cases where the underlying model exhibits a block angular structure.

The preceding literature review indicates that few papers address the entirety of the problem of optimising the maintenance scheduling of parallel processing lines that experience decaying performance in combination with operational planning, inventory management and sales to meet time-varying demand. These articles report difficulties in attaining optimality, even after applying significant approximation and decomposition techniques, thereby indicating the complexity of the problem. Even the other publications that address only a subset of these decisions exhibit such shortcomings.

These difficulties can be traced to the mixed-integer formulations of these problems. The drawbacks of mixed-integer optimisation techniques, mentioned in Section 1.4, form the roots of these difficulties. As mentioned in that section, the mixed-integer methods are combinatorial in nature and these methods approximate the differential equations present in the problem as a collection of steady state equations, which are imposed as additional constraints in the problem.

It is these features of mixed-integer methods which cause some of the aforementioned works that attempt to solve large scale problems using these methods to report intractable problem sizes and difficulties in converging to optimal solutions. In order to avoid solving hard MINLP problems, some of the other aforementioned works admit to approximating nonlinear terms present in the problem as linear terms. The approximations induced by the use of mixed-integer methods, such as linearisation of nonlinear terms and representation of differential equations as a collection of steady state equations, imply that the solutions obtained by these methods cannot be considered accurate. More complex models or an increase in the scale of the problem can further accentuate these difficulties.

The preceding discussion indicates that there are no encouraging signs that the use of mixed-integer optimisation methods to solve the problem of maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts will produce satisfactory results. Therefore, another methodology is needed that can effectively solve this problem and can overcome the drawbacks that the use of mixed-integer optimisation methods would face. In the previous chapter, an optimal control methodology was developed for optimising maintenance scheduling and production in a process containing a single reactors using decaying catalysts and this methodology was successful in obtaining robust, reliable and efficient solutions and overcoming the drawbacks faced by using mixed-integer techniques to solve that problem. That optimal control methodology will be extended in this chapter to optimise similar aspects in a process containing parallel lines of reactors using decaying catalysts.

This optimal control methodology involves a multistage mixed-integer optimal control problem (MSMIOCP) formulation in combination with a solution procedure of the principle of Implementation II, which was detailed in Section 2.4.3 of the previous chapter. As per this methodology, the problem under consideration is first formulated as an MSMIOCP with relaxed binary controls, of the form of equation (2.61), which is essentially a standard nonlinear optimisation problem. The elements of this MSMIOCP represent the various elements of the process in an analogous manner to that in Chapter 1. Next, a series of such standard nonlinear optimisation problems are solved as per a penalty term homotopy technique wherein a monotonically increasing penalty term is added to the objective function to force the the binary controls, considered continuous in this formulation, to attain values of 0 or 1.

In the next section, this optimal control methodology is applied to the case study of an industrial process. The problem formulation is presented first, followed by the implementation details and a discussion of the results obtained.

3.2 Case study: Problem formulation, implementation, results and discussion

In this section, the optimal control methodology is applied in a case study to optimise maintenance scheduling and production in an industrial process wherein a single feed is split over a set of parallel reactors using decaying catalysts to produce a single product. As mentioned in Section 3.1, currently no publication explicitly addresses such a problem. Hence, there was no process in any publication that could be used as a base to develop this formulation. Instead, the process examined in the previous chapter, for a single reactor, is modified to consider 4 parallel reactors.

A schematic of the process is shown in Figure 3.1. Here, a maintenance schedule is required that specifies, for the set of 4 parallel reactors, how many catalyst loads to use in each reactor as well as when the maintenance action to replace each of the used catalyst should occur in each reactor. Further, the maintenance schedule should ensure that no two reactors undergo catalyst replacement at the same time. The decisions to be made in this regard include whether to replace a catalyst or not, in each reactor, at regular intervals throughout the time horizon of the process. In addition, a production plan is needed which involves decisions that specify the flow rate to and temperature of operation of each of the 4 reactors, as well as decisions on the quantity of product sales to meet time-varying demand, at regular intervals over the process time horizon. This production plan should be managed in tandem with the maintenance schedule and while taking catalyst deactivation in each reactor into account. An integrated optimisation of the maintenance scheduling and production operations in the set of parallel reactors will enable attaining the objective of maximum profits for this process.

First, as per the methodology, the case study of this process is formulated as an MSMIOCP with relaxed binary controls, of the form of equation (2.61). In the case study considered in this chapter, the kinetics for the product formation reaction and the catalyst deactivation are similar to that of Case Study A in the previous chapter. The elements of the formulation here are developed as per principles largely similar to that of the single reactor process considered in the preceding chapter, but also contain some additional features compared to the formulation in the latter. The terminology used in this formulation is also similar to that in the previous chapter, with the exception that a subscript, *pr*, is added to each symbol used, in order to highlight the application to a "parallel reactor" set up, as well as to differentiate each term from the analogous term in that chapter. The sections following that of the problem formulation, present the implementation details and a discussion of the results obtained.

3.2.1 Problem formulation

In the problem addressed, the following assumptions apply:

1. The industrial process operates over a fixed time horizon, in the order of years. Each year is constituted by 12 months and there are a total of NM_{pr} months, wherein each month is constituted by 4 weeks.

2. The industrial process functions according to a certain process model and is subject to operating constraints.

3. The process has 4 Continuous Stirred Tank Reactors (CSTRs) of equal volumes, that operate in parallel.

4. There is a single feed to the process which is to be divided among the 4 reactors.

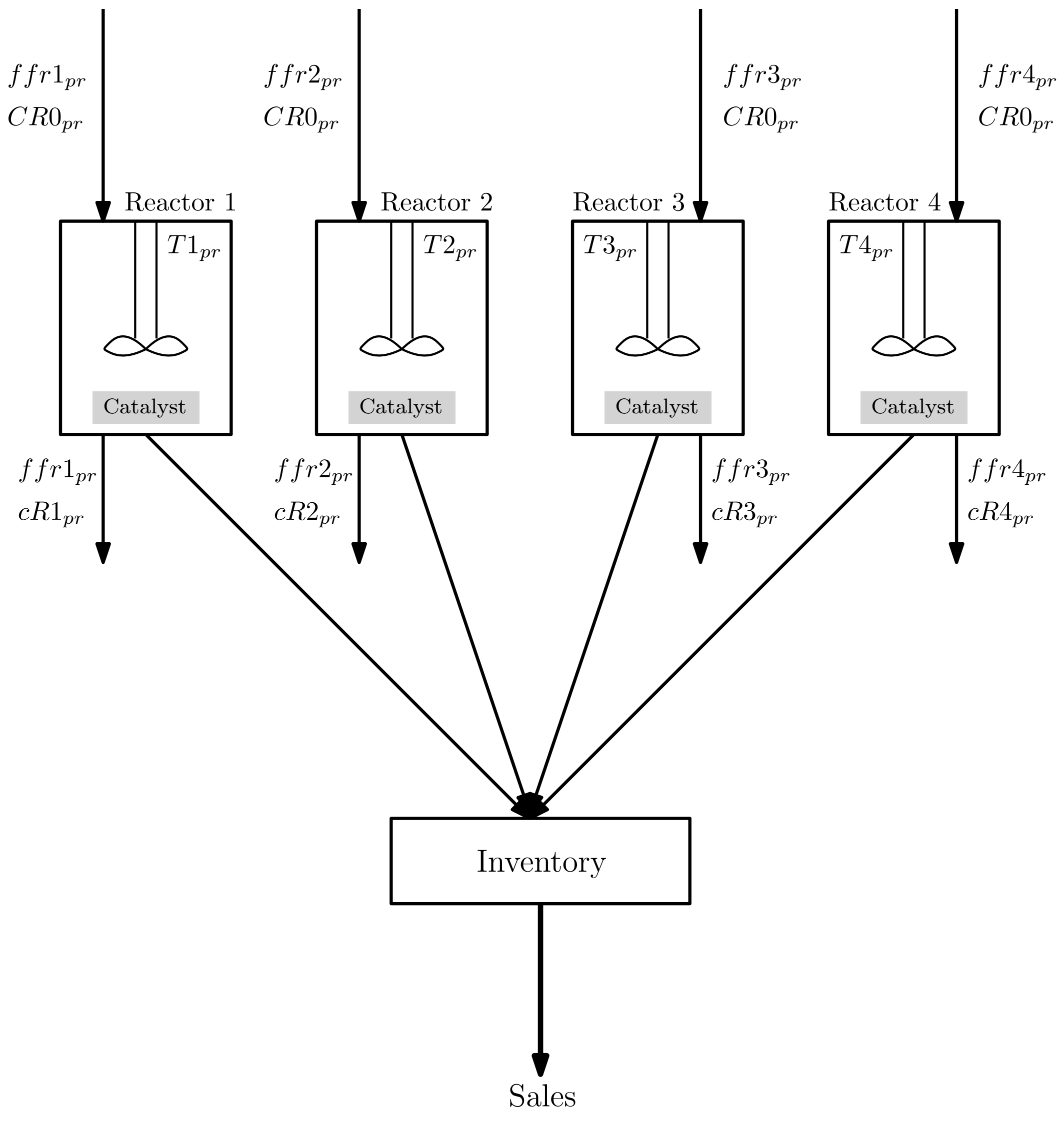


Fig. 3.1 A schematic of the process.

5. Each reactor processes the inlet feed using a catalyst to manufacture the same type of product.

6. In all reactors, the catalyst performance decays with time and has to be replaced before it crosses a certain maximum age.

7. The catalyst deactivation kinetics is first order with respect to the catalyst activity and is independent of the concentration of the reacting species. That is, the general form of the deactivation rate equation is given by equation (2.21) of the previous chapter:

$$rD \quad = \quad -Kd \times cat\_act$$

8. The rate constant in the deactivation kinetics is taken to be independent of the temperature of operation.

9. All reactors use similar types of catalysts, that are identical in functioning and performance. That is, all used catalysts have an identical value of the deactivation rate constant.

10. For each reactor, there is a maximum number of catalyst loads that can be used over the given time horizon. This number is the same for all reactors.

11. For each reactor, the time required for the maintenance action of shutting down the reactor, replacing the catalyst and restarting operation, is taken to be one month, during which time no production occurs.

12. The availability of labour and equipment in the process is such that in any one month, only one reactor can undergo catalyst replacement.

13. The main reaction is assumed to be of the same form as in the previous chapter, as given by equation (2.18):

$$R \rightarrow Q$$

    The reaction rate is considered separable from the catalyst activity and is first order with respect to the concentration of the reactant, R. That is, the general form of the reaction rate equation is given by equation (2.25) of the previous chapter:

$$rR \quad = \quad Kr \times cat\_act \times cR$$

14. The reaction rate constant is taken to exhibit an Arrhenius form of temperature dependence, of general form given by equation (2.27) of the previous chapter:

$$Kr = Ar \times \exp\left(-\frac{Ea}{R_g \times T(i,j)}\right)$$

15. The type of expressions to describe the kinetics of the catalyst deactivation and product formation reaction are the same in all reactors.

16. The flow rate of feed to the process has an upper limit. That is, the sum of feed flow rates to all reactors cannot exceed this limit.

17. The feed flow rate to each reactor has to be specified on a weekly basis.

18. For each reactor, the flow of feed is stopped during the maintenance action of catalyst replacement.

19. The concentration of reactant in the feed to the process is known and constant.

20. The temperature of operation of each reactor has to be specified on a weekly basis.

21. The temperature of each reactor can be operated only within fixed bounds during catalyst operation and is set to its lower bound during catalyst replacement.

22. Each reactor is operated isothermally. No energy balances are considered for any of the reactors.

23. The product produced by all reactors is stored continuously as inventory.

24. The weekly product demand is known for the whole time horizon.

25. The amount of product sales from the inventory present has to be specified on a weekly basis.

26. The product sales for each week is less than or equal to the demand in that week.

27. There is a penalty corresponding to the unmet demand in each week.

28. The costs involved in the process are known and are subject to a known value of annual inflation. These include the sales price of the product, the cost of inventory, the cost of feed and raw material, the cost of catalyst changeover and the penalty for unmet demand. There are, however, no costs related to heating and cooling procedures.

29. There is no uncertainty regarding the values of any of the parameters involved.

Given the above assumptions, the optimisation model must determine the following sets of values, which constitute the controls of the MSMIOCP:

(i) The catalyst changeover decision variables, for each month i, for reactors 1, 2, 3 and 4, represented by symbols $y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$ and $y4_{pr}(i)$, respectively. For reactor 1, $y1_{pr}(i) = 1$ indicates that a catalyst is in operation and $y1_{pr}(i) = 0$ indicates that the catalyst is being replaced, during month i. An analogous description applies for variables $y2_{pr}(i)$, $y3_{pr}(i)$ and $y4_{pr}(i)$ in reactors 2, 3 and 4, respectively

(ii) The feed flow rate to reactors 1, 2, 3 and 4, during each week, j, of each month, i, represented by symbols $ffr1_{pr}(i,j)$, $ffr2_{pr}(i,j)$, $ffr3_{pr}(i,j)$ and $ffr4_{pr}(i,j)$, respectively

(iii) The amount of product sold, at the end of each week, j, of each month, i, represented by $sales_{pr}(i,j)$

(iv) The temperature of operation of reactors 1, 2, 3 and 4, during each week, j, of each month, i, represented by symbols $T1_{pr}(i,j)$, $T2_{pr}(i,j)$, $T3_{pr}(i,j)$ and $T4_{pr}(i,j)$, respectively

In the above list, $j \in \{1,2,3,4\}$ and $i \in \{1,2,...,NM_{pr}\}$. The catalyst changeover decisions correspond to the binary controls, u, in equation (2.61g) while the other decision variables correspond to continuous controls, v, in equation (2.61h).

The state variables that characterise the MSMIOCP formulation of this industrial process include the following sets of variables:

(i) The ages of the catalysts in reactors 1, 2, 3 and 4, represented by symbols cat_age1_{pr}, cat_age2_{pr}, cat_age3_{pr} and cat_age4_{pr}, respectively

(ii) The activities of the catalysts in reactors 1, 2, 3 and 4, represented by symbols cat_act1_{pr}, cat_act2_{pr}, cat_act3_{pr} and cat_act4_{pr}, respectively

(iii) The concentration of the reactant at the exit of the reactors 1, 2, 3 and 4, represented by symbols $cR1_{pr}$, $cR2_{pr}$, $cR3_{pr}$ and $cR4_{pr}$, respectively

(iv) The product inventory level, inl_{pr}

(v) The cumulative inventory costs, cum_inc_{pr}

These state variables are determined by the decision variables' values at any time using a set of ODEs and under the influence of constraints. Next, ODEs of the form of equation (2.61b), that apply for week $j \in \{1,2,3,4\}$ of month $i \in \{1,2,...,NM_{pr}\}$ of the process are formulated.

1. In all reactors, the catalyst age varies linearly with time when the catalyst is in operation $(y1_{pr}(i),\ y2_{pr}(i),\ y3_{pr}(i),\ y4_{pr}(i) = 1)$ but does not increase at times of catalyst replacement $(y1_{pr}(i),\ y2_{pr}(i),\ y3_{pr}(i),\ y4_{pr}(i) = 0)$. Hence, the differential equations describing the catalyst age in reactors 1, 2, 3 and 4, accounting for both scenarios, are given by equations, (3.1a), (3.1b), (3.1c) and (3.1d), respectively:

$$\frac{\mathrm{d}\left(cat\_age1_{pr}\right)}{\mathrm{d}t} = y1_{pr}(i) \tag{3.1a}$$

$$\frac{\mathrm{d}\left(cat\_age2_{pr}\right)}{\mathrm{d}t} = y2_{pr}(i) \tag{3.1b}$$

$$\frac{\mathrm{d}\left(cat\_age3_{pr}\right)}{\mathrm{d}t} = y3_{pr}(i) \tag{3.1c}$$

$$\frac{\mathrm{d}\left(cat\_age4_{pr}\right)}{\mathrm{d}t} = y4_{pr}(i) \tag{3.1d}$$

2. In all reactors, the catalyst activity decays on a first order basis during times of catalyst operation $(y1_{pr}(i),\ y2_{pr}(i),\ y3_{pr}(i),\ y4_{pr}(i) = 1)$ but experiences no change during times of catalyst replacement $(y1_{pr}(i),\ y2_{pr}(i),\ y3_{pr}(i),\ y4_{pr}(i) = 0)$, as there is no production occurring. Thus, the differential equations for the catalyst activity in reactors 1, 2, 3 and 4, accounting for both scenarios, are given by equations, (3.2a), (3.2b), (3.2c) and (3.2d), respectively:

$$\frac{\mathrm{d}\left(cat\_act1_{pr}\right)}{\mathrm{d}t} = y1_{pr}(i) \times \left[-Kd_{pr} \times cat\_act1_{pr}\right] \tag{3.2a}$$

$$\frac{\mathrm{d}\left(cat\_act2_{pr}\right)}{\mathrm{d}t} = y2_{pr}(i) \times \left[-Kd_{pr} \times cat\_act2_{pr}\right] \tag{3.2b}$$

$$\frac{\mathrm{d}\left(cat\_act3_{pr}\right)}{\mathrm{d}t} = y3_{pr}(i) \times \left[-Kd_{pr} \times cat\_act3_{pr}\right] \tag{3.2c}$$

$$\frac{\mathrm{d}\left(cat\_act4_{pr}\right)}{\mathrm{d}t} = y4_{pr}(i) \times \left[-Kd_{pr} \times cat\_act4_{pr}\right] \tag{3.2d}$$

   where Kd_{pr} is the catalyst deactivation rate constant.

3. Since all reactors are assumed to be completely stirred, the concentration of reactant exiting each reactor $(cR1_{pr},\ cR2_{pr},\ cR3_{pr},\ cR4_{pr})$ is obtained from the generic mass

balance equation of a CSTR during times of catalyst operation ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 1$). However, during times of catalyst replacement ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$), no reaction occurs and an artificial condition is imposed wherein the reactor is assumed to be filled with fresh, unreacted reactant at the entry concentration ($CR0_{pr}$), to be used by the new catalyst after replacement. The differential equations that account for both scenarios, for reactors 1, 2, 3 and 4 are given by equations, (3.3a), (3.3b), (3.3c) and (3.3d), respectively:

$$\begin{aligned}\frac{\mathrm{d}\left(V_{pr} \times cR1_{pr}\right)}{\mathrm{d}t} =& ffr1_{pr}(i,j) \times (CR0_{pr} - cR1_{pr}) \\ & - y1_{pr}(i) \times [V_{pr} \times Kr1_{pr} \times cat\_act1_{pr} \times cR1_{pr}]\end{aligned} \tag{3.3a}$$

$$\begin{aligned}\frac{\mathrm{d}\left(V_{pr} \times cR2_{pr}\right)}{\mathrm{d}t} =& ffr2_{pr}(i,j) \times (CR0_{pr} - cR2_{pr}) \\ & - y2_{pr}(i) \times [V_{pr} \times Kr2_{pr} \times cat\_act2_{pr} \times cR2_{pr}]\end{aligned} \tag{3.3b}$$

$$\begin{aligned}\frac{\mathrm{d}\left(V_{pr} \times cR3_{pr}\right)}{\mathrm{d}t} =& ffr3_{pr}(i,j) \times (CR0_{pr} - cR3_{pr}) \\ & - y3_{pr}(i) \times [V_{pr} \times Kr3_{pr} \times cat\_act3_{pr} \times cR3_{pr}]\end{aligned} \tag{3.3c}$$

$$\begin{aligned}\frac{\mathrm{d}\left(V_{pr} \times cR4_{pr}\right)}{\mathrm{d}t} =& ffr4_{pr}(i,j) \times (CR0_{pr} - cR4_{pr}) \\ & - y4_{pr}(i) \times [V_{pr} \times Kr4_{pr} \times cat\_act4_{pr} \times cR4_{pr}]\end{aligned} \tag{3.3d}$$

Here V_{pr} is the volume, considered equal for all reactors. $Kr1_{pr}$, $Kr2_{pr}$, $Kr3_{pr}$ and $Kr4_{pr}$ are the rate constants for the reactions occurring in reactors 1, 2, 3 and 4, respectively, and they exhibit an Arrhenius form of dependence on the temperature of operation of the respective reactors, of the following form:

$$Kr1_{pr} = Ar_{pr} \times \exp\left(-\frac{Ea_{pr}}{R_g \times T1_{pr}(i,j)}\right) \tag{3.4a}$$

$$Kr2_{pr} = Ar_{pr} \times \exp\left(-\frac{Ea_{pr}}{R_g \times T2_{pr}(i,j)}\right) \tag{3.4b}$$

$$Kr3_{pr} = Ar_{pr} \times \exp\left(-\frac{Ea_{pr}}{R_g \times T3_{pr}(i,j)}\right) \tag{3.4c}$$

$$Kr4_{pr} = Ar_{pr} \times \exp\left(-\frac{Ea_{pr}}{R_g \times T4_{pr}(i,j)}\right) \tag{3.4d}$$

Here Ar_{pr} and Ea_{pr} are the pre-exponential factor and the activation energy, respectively, of reaction of the main reaction and R_g is the universal gas constant.

4. It is assumed that product produced by all reactors is stored as inventory before being sold at the end of the week. During times of catalyst operation ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 1$) in a reactor, the inventory level increases equivalent to the production rate (volume times reaction rate) of that reactor. But when catalyst replacement occurs ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$) in a reactor, the reactor does not contribute to an increase in inventory level as there is no production occurring. So, the differential equation describing the inventory level (inl_{pr}), that takes into account production from all reactors, while considering scenarios of catalyst operation as well as replacement, is given by:

$$\begin{aligned} \frac{\mathrm{d}(inl_{pr})}{\mathrm{d}t} = \quad & y1_{pr}(i) \times [V_{pr} \times Kr1_{pr} \times cat\_act1_{pr} \times cR1_{pr}] \\ & + y2_{pr}(i) \times [V_{pr} \times Kr2_{pr} \times cat\_act2_{pr} \times cR2_{pr}] \\ & + y3_{pr}(i) \times [V_{pr} \times Kr3_{pr} \times cat\_act3_{pr} \times cR3_{pr}] \\ & + y4_{pr}(i) \times [V_{pr} \times Kr4_{pr} \times cat\_act4_{pr} \times cR4_{pr}] \end{aligned} \tag{3.5}$$

5. Lastly, the increase in the cumulative inventory cost (cum_inc_{pr}) at any time depends on the inventory level at that time and the Inventory Cost Factor (icf_{pr}) (adjusted for inflation), which stipulates the cost per unit product per unit time:

$$\frac{\mathrm{d}(cum\_inc_{pr})}{\mathrm{d}t} = inl_{pr} \times icf_{pr} \tag{3.6}$$

The icf_{pr} at any time is given by the following equation:

$$icf_{pr} = base\_icf_{pr} \times (1 + inflation_{pr})^{\lfloor i/12 \rfloor} \tag{3.7}$$

where $base\_icf_{pr}$ is the inventory cost factor before inflation, $inflation_{pr}$ is the annual inflation rate and $\lfloor \cdot \rfloor$ is the greatest integer function.

The set of ODEs are solved repeatedly over a weekly time span, which corresponds to one stage of the MSMIOCP. In order to solve these ODEs, for each stage, suitable initial conditions have to be provided. The initial conditions for week 1 of month 1 are assumed to be known and are of the form of equation (2.61c). The initial conditions for the other stages are obtained using junction conditions between two successive stages of the process, of the form of equation (2.61d).

The initial conditions corresponding to week 1 of month 1, represented as $init\_var(1,1)$ for variable var, are as follows:

1. In all reactors, a new catalyst is used at the beginning of the process and so the initial catalyst age in all reactors is set to zero:

$$init\_cat\_age1_{pr}(1,1) = 0 \tag{3.8a}$$

$$init\_cat\_age2_{pr}(1,1) = 0 \tag{3.8b}$$

$$init\_cat\_age3_{pr}(1,1) = 0 \tag{3.8c}$$

$$init\_cat\_age4_{pr}(1,1) = 0 \tag{3.8d}$$

2. Since, in all reactors, a new catalyst is used at the beginning of the process, the initial catalyst activity for the catalysts in all reactors is set to that of a fresh catalyst ($start\_cat\_act_{pr}$):

$$init\_cat\_act1_{pr}(1,1) = start\_cat\_act_{pr} \tag{3.9a}$$

$$init\_cat\_act2_{pr}(1,1) = start\_cat\_act_{pr} \tag{3.9b}$$

$$init\_cat\_act3_{pr}(1,1) = start\_cat\_act_{pr} \tag{3.9c}$$

$$init\_cat\_act4_{pr}(1,1) = start\_cat\_act_{pr} \tag{3.9d}$$

3. At the start of the process, all reactors are filled with the reactant, R, at its entry concentration $CR0_{pr}$. Hence, the initial exit concentration in all reactors is given by:

$$init\_cR1_{pr}(1,1) = CR0_{pr} \tag{3.10a}$$

$$init\_cR2_{pr}(1,1) = CR0_{pr} \tag{3.10b}$$

$$init\_cR3_{pr}(1,1) = CR0_{pr} \tag{3.10c}$$

$$init\_cR4_{pr}(1,1) = CR0_{pr} \tag{3.10d}$$

4. There is no inventory at the beginning of the process, and so:

$$init\_inl_{pr}(1,1) \quad = \quad 0 \tag{3.11}$$

5. There is no inventory at the start of the process and so the initial cumulative inventory cost is nil at that time:

$$init\_cum\_inc_{pr}(1,1) \quad = \quad 0 \tag{3.12}$$

The junction conditions are described next. For all reactors, the junction conditions differ depending on whether the catalyst is in operation ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 1$) or is being replaced ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$) during that month. In the following text, the expressions $init\_var\,(i,\, j)$ and $end\_var\,(i,\, j)$ indicate the initial and end conditions, respectively for the variable *var*, for week j of month i:

1. During months of catalyst operation ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 1$), in all reactors, the initial catalyst age for a week corresponds to the catalyst age at the end of the previous week. But during months of catalyst replacement ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$), the catalyst age has to be set to zero, the age of a new catalyst. The junction conditions that describe both scenarios for all reactors, are as follows.

$$init\_cat\_age1_{pr}\,(i,\, j+1) \;=\; end\_cat\_age1_{pr}(i,\, j) \tag{3.13a}$$

$$init\_cat\_age2_{pr}\,(i,\, j+1) \;=\; end\_cat\_age2_{pr}(i,\, j) \tag{3.13b}$$

$$init\_cat\_age3_{pr}\,(i,\, j+1) \;=\; end\_cat\_age3_{pr}(i,\, j) \tag{3.13c}$$

$$init\_cat\_age4_{pr}\,(i,\, j+1) \;=\; end\_cat\_age4_{pr}(i,\, j) \tag{3.13d}$$

$$j = 1,2,3 \qquad i = 1,2,\ldots,NM_{pr}$$

$$init\_cat\_age1_{pr}\,(i,\, 1) \;=\; y1_{pr}(i) \times end\_cat\_age1_{pr}(i-1,\, 4) \tag{3.13e}$$

$$init\_cat\_age2_{pr}\,(i,\, 1) \;=\; y2_{pr}(i) \times end\_cat\_age2_{pr}(i-1,\, 4) \tag{3.13f}$$

$$init\_cat\_age3_{pr}\,(i,\, 1) \;=\; y3_{pr}(i) \times end\_cat\_age3_{pr}(i-1,\, 4) \tag{3.13g}$$

$$init\_cat\_age4_{pr}\,(i,\, 1) \;=\; y4_{pr}(i) \times end\_cat\_age4_{pr}(i-1,\, 4) \tag{3.13h}$$

$$i = 2,3,\ldots,NM_{pr}$$

2. During months of catalyst operation ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 1$), in all reactors, the initial catalyst activity for the week corresponds to the catalyst activity at the end of the previous week. However, during months of catalyst replacement ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$), in all reactors, the catalyst activity has to be reset to the activity corresponding to that of a fresh catalyst, which remains the same throughout the duration of month i. The junction conditions that describe both scenarios for all reactors is given as follows.

$$init\_cat\_act1_{pr}\,(i,\, j+1) \;=\; end\_cat\_act1_{pr}(i,\, j) \tag{3.14a}$$

$$init\_cat\_act2_{pr}(i,\, j+1) \;=\; end\_cat\_act2_{pr}(i,\, j) \tag{3.14b}$$

$$init\_cat\_act3_{pr}(i,\, j+1) \;=\; end\_cat\_act3_{pr}(i,\, j) \tag{3.14c}$$

$$init\_cat\_act4_{pr}(i,\, j+1) \;=\; end\_cat\_act4_{pr}(i,\, j) \tag{3.14d}$$

$$j = 1,2,3 \qquad i = 1,2,\ldots,NM_{pr}$$

$$\begin{aligned} init\_cat\_act1_{pr}(i,\, 1) = {} & [y1_{pr}(i) \times end\_cat\_act1_{pr}(i-1,\, 4)] \\ & + [(1-y1_{pr}(i)) \times start\_cat\_act_{pr}] \end{aligned} \tag{3.14e}$$

$$\begin{aligned} init\_cat\_act2_{pr}(i,\, 1) = {} & [y2_{pr}(i) \times end\_cat\_act2_{pr}(i-1,\, 4)] \\ & + [(1-y2_{pr}(i)) \times start\_cat\_act_{pr}] \end{aligned} \tag{3.14f}$$

$$\begin{aligned} init\_cat\_act3_{pr}(i,\, 1) = {} & [y3_{pr}(i) \times end\_cat\_act3_{pr}(i-1,\, 4)] \\ & + [(1-y3_{pr}(i)) \times start\_cat\_act_{pr}] \end{aligned} \tag{3.14g}$$

$$\begin{aligned} init\_cat\_act4_{pr}(i,\, 1) = {} & [y4_{pr}(i) \times end\_cat\_act4_{pr}(i-1,\, 4)] \\ & + [(1-y4_{pr}(i)) \times start\_cat\_act_{pr}] \end{aligned} \tag{3.14h}$$

$$i = 2,3,\ldots,NM_{pr}$$

3. During months of catalyst operation ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 1$), in all reactors, the exit concentration for the beginning of a week corresponds to the exit concentration at the end of the previous week. And when the catalyst is being replaced ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$) in a reactor, an artificial condition is imposed wherein the reactor is filled with reactant at entry concentration $CR0_{pr}$, ready to be used by the fresh catalyst at the beginning of the next month. The junction conditions that describe both scenarios for all reactors is given as follows.

$$init\_cR1_{pr}(i,\, j+1) \;=\; end\_cR1_{pr}(i,\, j) \tag{3.15a}$$

$$init\_cR2_{pr}(i,\, j+1) \;=\; end\_cR2_{pr}(i,\, j) \tag{3.15b}$$

$$init\_cR3_{pr}(i,\, j+1) \;=\; end\_cR3_{pr}(i,\, j) \tag{3.15c}$$

$$init\_cR4_{pr}(i,\, j+1) \;=\; end\_cR4_{pr}(i,\, j) \tag{3.15d}$$

$$j = 1,2,3 \qquad i = 1,2,\ldots,NM_{pr}$$

$$init\_cR1_{pr}(i,\, 1) = [y1_{pr}(i) \times end\_cR1_{pr}(i-1,\, 4)] + [(1-y1_{pr}(i)) \times CR0_{pr}] \tag{3.15e}$$

$$init\_cR2_{pr}(i,\, 1) = [y2_{pr}(i) \times end\_cR2_{pr}(i-1,\, 4)] + [(1-y2_{pr}(i)) \times CR0_{pr}] \tag{3.15f}$$

$$init\_cR3_{pr}(i,\,1) = [y3_{pr}(i) \times end\_cR3_{pr}(i-1,\,4)] + [(1-y3_{pr}(i)) \times CR0_{pr}] \tag{3.15g}$$

$$init\_cR4_{pr}(i,\,1) = [y4_{pr}(i) \times end\_cR4_{pr}(i-1,\,4)] + [(1-y4_{pr}(i)) \times CR0_{pr}] \tag{3.15h}$$

$$i = 2,3,\ldots,NM_{pr}$$

4. At the end of a week, an amount, $sales_{pr}(i,\,j)$ of the stored product is sold. Thus, the initial inventory level for the week corresponds to the inventory present after the sales at the end of the previous week. The following junction conditions apply during months of catalyst operation as well as catalyst replacement, as the sales do not cease at any time:

$$\begin{aligned} init\_inl_{pr}(i,\,j+1) &= end\_inl_{pr}(i,\,j) - sales_{pr}(i,\,j) \\ j = 1,2,3 &\qquad i = 1,2,\ldots,NM_{pr} \end{aligned} \tag{3.16a}$$

$$\begin{aligned} init\_inl_{pr}(i,\,1) &= end\_inl_{pr}(i-1,\,4) - sales_{pr}(i-1,\,4) \\ i &= 2,3,\ldots,NM_{pr} \end{aligned} \tag{3.16b}$$

5. The inventory cost accumulated until the beginning of a week is equal to the value of the inventory cost accumulated until the end of the previous week. So the following junction conditions apply regardless of whether the catalyst is being used or replaced:

$$\begin{aligned} init\_cum\_inc_{pr}(i,\,j+1) &= end\_cum\_inc_{pr}(i,\,j) \\ j = 1,2,3 &\qquad i = 1,2,\ldots,NM_{pr} \end{aligned} \tag{3.17a}$$

$$\begin{aligned} init\_cum\_inc_{pr}(i,\,1) &= end\_cum\_inc_{pr}(i-1,\,4) \\ i &= 2,3,\ldots,NM_{pr} \end{aligned} \tag{3.17b}$$

The initial conditions (3.8) – (3.12) and junction conditions (3.13) – (3.17) enable a solution for the ODEs for all stages and thereby obtain the values of the state variables. These obtained state variables, along with the control variables, are required to fulfil some constraints, which include the operational limits of the process and restrictions on the values of the controls to be chosen.

The constraints, of the form of equation (2.61f), that apply to this industrial process for week $j \in \{1,2,3,4\}$ of month $i \in \{1,2,...,NM_{pr}\}$ are as follows:

1. In the context of the formulation as an MSMIOCP with relaxed binary controls, the catalyst changeover decision variables for all reactors ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$,

$y4_{pr}(i)$), for a month i, are considered continuous variables that vary between 0 and 1, and so the following bounds are imposed:

$$0 \leq y1_{pr}(i) \leq 1 \tag{3.18a}$$

$$0 \leq y2_{pr}(i) \leq 1 \tag{3.18b}$$

$$0 \leq y3_{pr}(i) \leq 1 \tag{3.18c}$$

$$0 \leq y4_{pr}(i) \leq 1 \tag{3.18d}$$

2. The net flow rate of feed to the process has an upper limit (FU_{pr}). That is, the sum of the flow rates of feeds to all reactors has to remain within this limit and so, the following bounds are imposed, for each week:

$$0 \leq ffr1_{pr}(i,j) + ffr2_{pr}(i,j) + ffr3_{pr}(i,j) + ffr4_{pr}(i,j) \leq FU_{pr} \tag{3.19}$$

3. The sales in each week are assumed to be less than or equal to the demand for the product in that week $(demand_{pr}(i,j))$. Hence, the following bounds on the sales at the end of each week are imposed:

$$0 \leq sales_{pr}(i,j) \leq demand_{pr}(i,j) \tag{3.20}$$

4. The temperature of each reactor operates between known, fixed lower and upper bounds, given by TL_{pr} and TU_{pr}, respectively. Hence, the following bounds are set on the weekly temperature of operation of each reactor:

$$TL_{pr} \leq T1_{pr}(i,j) \leq TU_{pr} \tag{3.21a}$$

$$TL_{pr} \leq T2_{pr}(i,j) \leq TU_{pr} \tag{3.21b}$$

$$TL_{pr} \leq T3_{pr}(i,j) \leq TU_{pr} \tag{3.21c}$$

$$TL_{pr} \leq T4_{pr}(i,j) \leq TU_{pr} \tag{3.21d}$$

5. When a catalyst is being replaced in a reactor $(y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) = 0)$, the flow of raw material to that reactor stops. The following constraints ensure that the weekly feed flow rate to each reactor remains below the upper bound during times of catalyst operation $(y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) = 1)$ and drops to zero when

catalyst replacement occurs $(y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) = 0)$:

$$ffr1_{pr}(i,j) - [FU_{pr} \times y1_{pr}(i)] \leq 0 \tag{3.22a}$$

$$ffr2_{pr}(i,j) - [FU_{pr} \times y2_{pr}(i)] \leq 0 \tag{3.22b}$$

$$ffr3_{pr}(i,j) - [FU_{pr} \times y3_{pr}(i)] \leq 0 \tag{3.22c}$$

$$ffr4_{pr}(i,j) - [FU_{pr} \times y4_{pr}(i)] \leq 0 \tag{3.22d}$$

6. When a catalyst is being replaced in a reactor $(y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) = 0)$, the temperature of the reactor is required to drop to its lower bound. This condition is imposed using the following constraints which ensure that the temperature of each reactor, for each week, remains between its bounds during times of catalyst operation $(y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) = 1)$ but drops to the lower bound when catalyst replacement occurs $(y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) = 0)$:

$$TL_{pr} \leq T1_{pr}(i,j) \leq [(TU_{pr} - TL_{pr}) \times y1_{pr}(i)] + TL_{pr} \tag{3.23a}$$

$$TL_{pr} \leq T2_{pr}(i,j) \leq [(TU_{pr} - TL_{pr}) \times y2_{pr}(i)] + TL_{pr} \tag{3.23b}$$

$$TL_{pr} \leq T3_{pr}(i,j) \leq [(TU_{pr} - TL_{pr}) \times y3_{pr}(i)] + TL_{pr} \tag{3.23c}$$

$$TL_{pr} \leq T4_{pr}(i,j) \leq [(TU_{pr} - TL_{pr}) \times y4_{pr}(i)] + TL_{pr} \tag{3.23d}$$

7. For each reactor, there is a maximum number of catalyst replacements that can occur. In this case study, it is assumed that this maximum number is the same for all reactors and this number is designated as n_{pr}. The limit on the maximum number of catalyst changeovers allowed for each reactor is imposed using the following set of constraints:

$$\sum_{i=1}^{NM_{pr}} y1_{pr}(i) \geq NM_{pr} - n_{pr} \tag{3.24a}$$

$$\sum_{i=1}^{NM_{pr}} y2_{pr}(i) \geq NM_{pr} - n_{pr} \tag{3.24b}$$

$$\sum_{i=1}^{NM_{pr}} y3_{pr}(i) \geq NM_{pr} - n_{pr} \tag{3.24c}$$

$$\sum_{i=1}^{NM_{pr}} y4_{pr}(i) \geq NM_{pr} - n_{pr} \tag{3.24d}$$

8. The availability of equipment and labour in the process is such that only one reactor can undergo catalyst replacement ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$) during any month. Mathematically, this means that among the catalyst changeover controls for all reactors ($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i)$), at most one control can take a value of 0 (or at least three controls should have values of 1) during any month, i. This condition of non-simultaneous catalyst replacement is enforced using the following constraint:

$$y1_{pr}(i) + y2_{pr}(i) + y3_{pr}(i) + y4_{pr}(i) \quad \geq \quad 3 \tag{3.25}$$

9. In order to ensure that more product than available is not sold, the inventory level at the end of each week should be greater than the sales for the week. This is imposed using the following constraint:

$$end\_inl_{pr}(i, j) - sales_{pr}(i, j) \quad \geq \quad 0 \tag{3.26}$$

10. The catalysts in all reactors undergo deactivation over time and have to be replaced before crossing a certain maximum age. Since all reactors use catalysts that are identical in functioning and performance, a common for all maximum catalyst age, designated as $max\_cat\_age_{pr}$, is used. As the decision on whether to replace a catalyst or not is made on a monthly basis, it is sufficient to ensure that the catalyst age in each reactor does not cross this limit at the end of each month, i:

$$end\_cat\_age1_{pr}(i, 4) \quad \leq \quad max\_cat\_age_{pr} \tag{3.27a}$$

$$end\_cat\_age2_{pr}(i, 4) \quad \leq \quad max\_cat\_age_{pr} \tag{3.27b}$$

$$end\_cat\_age3_{pr}(i, 4) \quad \leq \quad max\_cat\_age_{pr} \tag{3.27c}$$

$$end\_cat\_age4_{pr}(i, 4) \quad \leq \quad max\_cat\_age_{pr} \tag{3.27d}$$

The aim is to maximise the profits or minimise the costs of the process under the influence of these ODEs, initial conditions, junction conditions and constraints. The net costs of the process are represented by the objective function, of the form of equation (2.61a), and comprises of the following elements:

1. The Gross Revenue from Sales (GRS_{pr})
   This term represents the revenue for the process from the net sales of the product

chemical over the whole time horizon:

$$GRS_{pr} = \sum_{i=1}^{NM_{pr}} \sum_{j=1}^{4} psp_{pr}(i,j) \times sales_{pr}(i,j) \tag{3.28}$$

where $psp_{pr}(i,j)$ is the sales price per unit product for week j of month i, adjusted for inflation at that time:

$$psp_{pr}(i,j) = base\_psp_{pr} \times (1+inflation_{pr})^{\lfloor i/12 \rfloor}$$

where $base\_psp_{pr}$ is the unit product sales price before inflation.

2. <u>The Total Inventory Costs (TIC_{pr})</u>
   This term represents the net storage costs for the product over the whole time horizon and is obtained from the solution of the ODEs for the state variable cum_inc_{pr} at the end of the final week of the process:

$$TIC_{pr} = end\_cum\_inc_{pr}(NM_{pr}, 4) \tag{3.29}$$

3. <u>The Total Costs of Catalyst Changeovers $(TCCC_{pr})$</u>
   The total expenditure for the catalyst changeover operations is given by the sum of the catalyst changeover costs for all 4 reactors. Since these costs are incurred only during months of catalyst replacement($y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0$), $TCCC_{pr}$ is obtained in the following manner:

$$\begin{aligned} TCCC_{pr} = &\sum_{i=1}^{NM_{pr}} crc_{pr}(i) \times [1 - y1_{pr}(i)] + \sum_{i=1}^{NM_{pr}} crc_{pr}(i) \times [1 - y2_{pr}(i)] \\ &+ \sum_{i=1}^{NM_{pr}} crc_{pr}(i) \times [1 - y3_{pr}(i)] + \sum_{i=1}^{NM_{pr}} crc_{pr}(i) \times [1 - y4_{pr}(i)] \end{aligned} \tag{3.30}$$

It is highlighted that the terms within the summations remain non-zero only during the times of catalyst replacement $(y1_{pr}(i)$, $y2_{pr}(i)$, $y3_{pr}(i)$, $y4_{pr}(i) = 0)$ and only these terms contribute to the total costs. Here $crc_{pr}(i)$ is the cost of the catalyst replacement operation, considered the same for all reactors, for month i, adjusted for inflation at that time:

$$crc_{pr}(i) = base\_crc_{pr} \times (1+inflation_{pr})^{\lfloor i/12 \rfloor}$$

where $base\_crc_{pr}$ is the cost of a catalyst changeover operation before inflation.

4. The Net Penalty for Unmet Demand $(NPUD_{pr})$
The unmet demand in each week $(unmet\_demand_{pr}(i,j))$ is the quantity of product by which the sales falls short of the demand in that week:

$$\begin{aligned} unmet\_demand_{pr}(i,j) &= demand_{pr}(i,j) - sales_{pr}(i,j) \\ j = 1,2,3,4 &\qquad i = 1,2,\ldots,NM_{pr} \end{aligned} \tag{3.31}$$

There is a penalty associated with this unmet demand and the net penalty costs over the entire time horizon is given by:

$$NPUD_{pr} = \sum_{i=1}^{NM_{pr}} \sum_{j=1}^{4} pen_{pr}(i,j) \times unmet\_demand_{pr}(i,j) \tag{3.32}$$

where $pen_{pr}(i,j)$ is the penalty cost of unmet demand per unit product for week j of month i, adjusted for inflation at that time:

$$pen_{pr}(i,j) = base\_pen_{pr} \times (1 + inflation_{pr})^{\lfloor i/12 \rfloor}$$

where $base\_pen_{pr}$ is the penalty cost of unmet demand per unit product before inflation.

5. The Total Flow Costs (TFC_{pr})
This term represents the net expenditure on the feed of raw material to all reactors in the process and is given by:

$$TFC_{pr} = \sum_{i=1}^{NM_{pr}} \sum_{j=1}^{4} cof_{pr}(i,j) \times [ffr1_{pr}(i,j) + ffr2_{pr}(i,j) + ffr3_{pr}(i,j) + ffr4_{pr}(i,j)] \tag{3.33}$$

where $cof_{pr}(i,j)$ is the cost of raw material per unit volume per week for week j of month i, adjusted for inflation at that time:

$$cof_{pr}(i,j) = base\_cof_{pr} \times (1 + inflation_{pr})^{\lfloor i/12 \rfloor}$$

where $base\_cof_{pr}$ is the cost of raw material per unit volume per week before inflation.

If the Net Costs are represented by NC_{pr}, the objective function for this optimisation problem takes the form:

$$\min NC_{pr} = -GRS_{pr} + TIC_{pr} + TCCC_{pr} + NPUD_{pr} + TFC_{pr} \tag{3.34}$$

The essential elements of the problem formulation have now been described in detail. The aim is to make the appropriate decisions in order to minimise the net costs (or maximise the profit) of the industrial process, when subject to the ODEs, initial and junction conditions and the constraints.

As in the previous chapter, the set of parameters used for this problem are constructed and their values are given in Table 3.1. These parameters are mostly similar to those used in the previous chapter, except here the volumes of the 4 reactors used add up to the volume of the single reactor in that chapter and the base cost of catalyst replacement ($base\_crc_{pr}$) is adjusted accordingly to be quarter of the cost ($base\_crc$) in that chapter. A 3-year time horizon is considered here as well and the details of the problem size under this formulation for this horizon length are given in Table 3.2. In the next section, the implementation details of the solution procedure are discussed.

Table 3.1 List of parameters.

Parameter Symbol	**Value**
Ar_{pr}	885 (1/day)
$base\_cof_{pr}$	\$ 210 /week
$base\_crc_{pr}$	\$ 25×10^5
$base\_icf_{pr}$	\$ 0.01 /(kmol day)
$base\_pen_{pr}$	\$ 1250 /kmol
$base\_psp_{pr}$	\$ 1000 /kmol
$CR0_{pr}$	1 kmol/m^3
$demand_{pr}$	1st quarter of year: 8000 kmol/week
	2nd quarter of year: 7200 kmol/week
	3rd quarter of year: 3300 kmol/week
	4th quarter of year: 4500 kmol/week
Ea_{pr}	30,000 J/gmol
FU_{pr}	9600 m^3/day
$inflation_{pr}$	5%
Kd_{pr}	0.0024 (1/day)
$max\_cat\_age_{pr}$	504 days (= 1.5 years)
n_{pr}	5
NM_{pr}	36 months (= 3 years)
R_g	8.314 J/(gmol.K)
$start\_cat\_act_{pr}$	1.0
TL_{pr}	400 K
TU_{pr}	1000 K
V_{pr}	12.5 m^3

Table 3.2 Problem size specifications.

Property		**Size**
Ordinary Differential Equations		2016
Decision variables	Catalyst changeover actions $(y1_{pr}, y2_{pr}, y3_{pr}, y4_{pr})$	144
	Feed flow rates $(ffr1_{pr}, ffr2_{pr}, ffr3_{pr}, ffr4_{pr})$	576
	Sales $(sales_{pr})$	144
	Temperatures $(T1_{pr}, T2_{pr}, T3_{pr}, T4_{pr})$	576
	Total	*1440*
Constraints	Constraints (3.18)	288
	Constraints (3.19)	288
	Constraints (3.20)	288
	Constraints (3.21)	1152
	Constraints (3.22)	576
	Constraints (3.23)	1152
	Constraints (3.24)	4
	Constraints (3.25)	36
	Constraints (3.26)	144
	Constraints (3.27)	144
	Total	*4072*

3.2.2 Implementation details

In the previous section, the industrial process was developed as an MSMIOCP with relaxed binary controls, which is a standard multistage optimal control problem and hence, a standard nonlinear optimisation problem. As per the proposed optimal control methodology to optimise this industrial process, the formulation developed in the previous section is solved using a solution procedure of the principle of Implementation II, which was detailed in Section 2.4.3. This involves applying a penalty term homotopy technique, of the form of equation (2.62), to the developed problem formulation and solving the following series of standard multistage optimal control problems:

$$
\begin{aligned}
Gp_k : \min \Bigg\{ NC_{pr} + Mp_k \Bigg[& \sum_{i=1}^{NM_{pr}} y1_{pr}(i)\,[1 - y1_{pr}(i)] + \sum_{i=1}^{NM_{pr}} y2_{pr}(i)\,[1 - y2_{pr}(i)] \\
& + \sum_{i=1}^{NM_{pr}} y3_{pr}(i)\,[1 - y3_{pr}(i)] + \sum_{i=1}^{NM_{pr}} y4_{pr}(i)\,[1 - y4_{pr}(i)] \Bigg] \Bigg\}
\end{aligned} \tag{3.35}
$$

subject to the differential equations, the initial conditions, the junction conditions and constraints of the developed formulation, which were presented in Section 3.2.1, for

$$
\begin{aligned}
k &= 1, 2, 3\ldots \\
Mp_1 &= 0
\end{aligned}
$$

In accordance with the proposed solution procedure, if in the solution of problem Gp_k, the condition, $y1_{pr}(i), y2_{pr}(i), y3_{pr}(i), y4_{pr}(i) \in \{0,1\}$ for $i = 1, 2, \ldots NM_{pr}$, does not apply, then problem Gp_{k+1} is solved using the solution of Gp_k as initial guesses, with weight $Mp_{k+1} > Mp_k$. The weight term, Mp_k, is increased as per the arithmetic progression given by equation (3.36). This progression is similar to the progression of increase of weights in equation (2.64) for the single reactor problem in the previous chapter, and is chosen as such because it produced satisfactory results in this problem as well.

$$
\begin{aligned}
Mp_{k+1} &= (2 \times Mp_k) + \left(5 \times 10^7\right) \\
Mp_1 &= 0 \\
k &= 1, 2, 3\ldots
\end{aligned} \tag{3.36}
$$

As in Section 2.4.3, the implementation was performed using Python™ 3.7.1 in PyCharm 2019.3.3 (Community Edition). Once again, CasADi v3.4.5 was used to define as symbolic expressions, the elements of the series of problems given by equation (3.35).

CasADi plug-ins to the *IDAS* solver of the open source SUNDIALS suite and IPOPT by COIN-OR were once again used for the integration of ODEs and optimisation, respectively, with similar termination criteria to that used in Section 2.4.3. That is, the integration by *IDAS* had the following termination criteria: an absolute tolerance of 10^{-6} and a relative tolerance of 10^{-6}. For the optimisation by IPOPT, Table 3.3 presents the termination and ‘acceptable’ termination criteria, wherein the ’acceptable’ number of iterations concerning the latter was set at 15, as was done in Chapter 2.

Table 3.3 Criteria for termination of optimisation by IPOPT

Property	Termination tolerance	Acceptable termination tolerance
Optimality error	10^{-4}	10^{-4}
Dual infeasibility	1	10^{6}
Constraint violation	10^{-4}	10^{-2}
Complementarity	10^{-4}	10^{-2}

The implementation was once again performed on a 3.2 GHz Intel Core i5, 16 GB RAM, Windows machine running on Microsoft Windows 10 Enterprise. Since the problem is non-convex in this case as well, 50 runs were performed in a serial manner with different random initial guesses for the decision variables, as was done in the previous chapter. In this case, though, the random initial guesses were generated using Latin Hypercube Sampling (McKay et al., 1979), obtained in Python using the *lhs* method of the *pyDOE* module (version 0.3.8).

In the next section, the results obtained are discussed. Statistics describing the essential solution features for the 50 runs are provided in the form of tables, and figures of the trends of the decision and state variables over the time horizon for the most profitable run are examined.

3.2.3 Results and discussions

As in the previous chapter, this methodology produced high quality solutions. Regardless of the initial guesses used, each of the 50 runs successfully converged to a local optimum within the specified optimality tolerance, thereby indicating the robustness of the procedure. The nonlinear differential equations were solved to a high accuracy using state-of-the-art integrators, without any approximation techniques, thus underscoring the reliability of the obtained solutions. The penalty term homotopy technique was successful, not only in obtaining binary values for the catalyst changeover controls in all reactors but also in enforcing the condition of non-simultaneous catalyst replacement, without the use of mixed-integer programming methods, and this underlines the efficiency of the methodology.

Statistics regarding the ranges of the profits, computation times and number of major iterations involved in the set of 50 runs are shown in Table 3.4. For the row labelled, "Profit (Million \$)", the columns titled ‘Maximum’, ‘Minimum’ and ‘Mean/Mode’ represent the maximum, minimum and mean profits among the set of 50 runs. Analogous explanations hold with regard to the maximum, minimum and mean solution times for those columns, for the row labelled, "CPU time (seconds)". From the row labelled, "Number of Major Iterations", it is seen that a minimum of 2 and a maximum of 4 major iterations are needed to obtain binary values for the catalyst changeover controls, with the mode being 3. The statistics regarding the number of IPOPT iterations within each major iteration are given in Table 3.5.

Further insights into the distribution of the profits over the 50 runs is shown in Figure 3.2. In the histogram in this figure, the height of each bin represents the number of runs out of 50 that result in profit values within the range specified by the horizontal edges of that bin. Given that the other properties discussed in Tables 3.4 and 3.5, namely the CPU time, the number of major iterations and the number of IPOPT iterations within each major iteration, are dependent on the computer used to obtains solutions, similar distributions for these properties have not been presented as these cannot be generalised.

Table 3.4 Solution statistics over the 50 multi-start runs.

Property	**Maximum**	**Minimum**	**Mean/Mode**
Profit (Million $)	435.595	378.817	411.855
CPU time (seconds)	330269	174663	238882
Number of Major Iterations	4	2	3

Table 3.5 Statistics for each major iteration. The column titled 'Runs' indicates the number of runs out of 50 that progressed until that major iteration. The columns titled 'Max', 'Min' and 'Mean' indicate the maximum, minimum and mean number of IPOPT iterations within that major iteration, respectively.

Major iteration	**Runs**	**Max**	**Min**	**Mean**
Major iteration 1	50	400	206	331
Major iteration 2	50	200	79	121
Major iteration 3	44	200	52	76
Major iteration 4	2	63	58	60.5

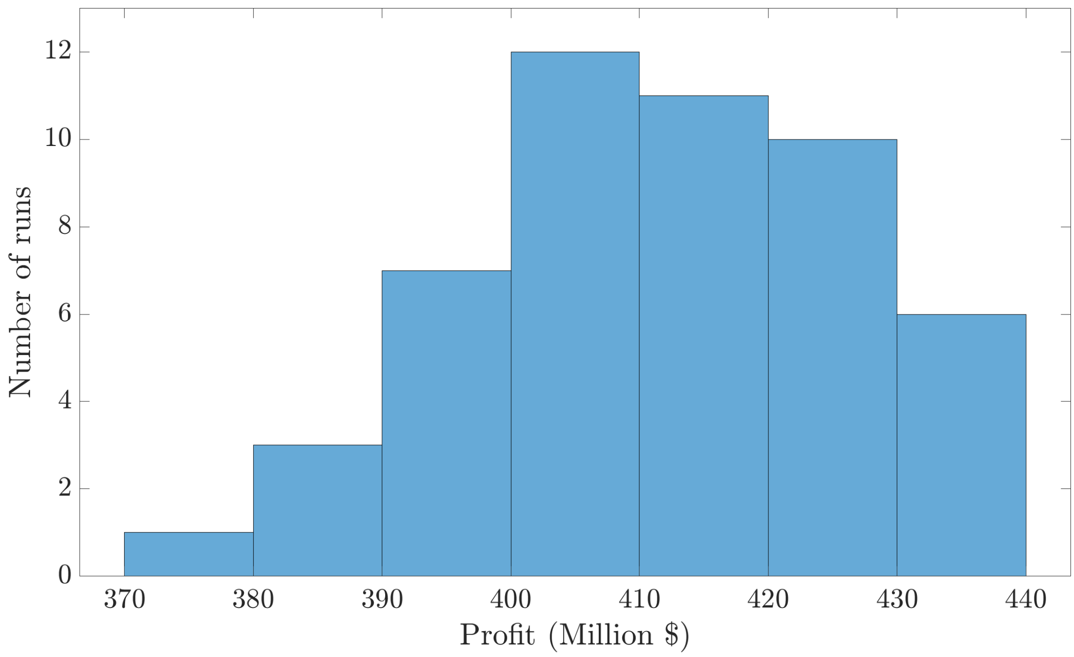


Fig. 3.2 The distribution of the profits obtained over all runs

Table 3.6 A comparison of the problem size and solution times between the single and parallel reactor studies

Property	**Single reactor study**	**Parallel reactor study**
Number of differential equations	720	2016
Number of decision variables	468	1440
Number of constraints	1549	4072
Maximum CPU time among 50 runs (seconds)	27438	330269
Minimum CPU time among 50 runs (seconds)	9826	174663
Mean CPU time among 50 runs (seconds)	17440	238882

As mentioned previously, the kinetics of the product formation reaction and catalyst deactivation in this case study are similar to that of Case Study A in the single reactor study of the previous chapter. Table 3.6 shows a comparison between the size of the optimal control problem formulations for the single reactor and parallel reactor studies, as well as a comparison between the CPU times for obtaining solutions using Implementation II for Case Study A of the single reactor study and for the parallel reactor study. As can be seen from the table, the number of differential equations, decision variables and constraints for the parallel reactor study are about 2.6–3.1 times those in the single reactor study. With regard to the solution times for the 50 runs for Case Study A of the single reactor study and the parallel reactor study, each which used a different set of initial guesses for the decision variables, the maximum, minimum and mean solution times for the latter study are about 12-18 times that in the former study. This provides insights into how the computation time scales with the number of variables, when using a solution procedure of the principle of Implementation II, for the type of computer used to obtain solutions. This correlation might well vary when a different type of computer is used.

Table 3.7 presents statistics regarding the catalyst ages and the number of catalyst replacements. It is noted that among all runs, for all reactors, only a maximum of 4 catalyst replacements and not all of the available 5 are used, with the mode being 3. It is also seen that the mean catalyst age for all reactors is about half the maximum age of 504 days and in no run, for any reactor, is a catalyst recommended to be used up to that maximum age.

Table 3.7 Statistics regarding the catalyst replacements in each reactor.

Reactor	**Number of catalyst replacements**			**Catalyst age (Days)**		
	Max	**Min**	**Mode**	**Max**	**Min**	**Mean**
Reactor 1	4	2	3	476	112	243.2
Reactor 2	4	2	3	476	112	252
Reactor 3	4	2	3	476	112	246
Reactor 4	4	2	3	476	112	234.9

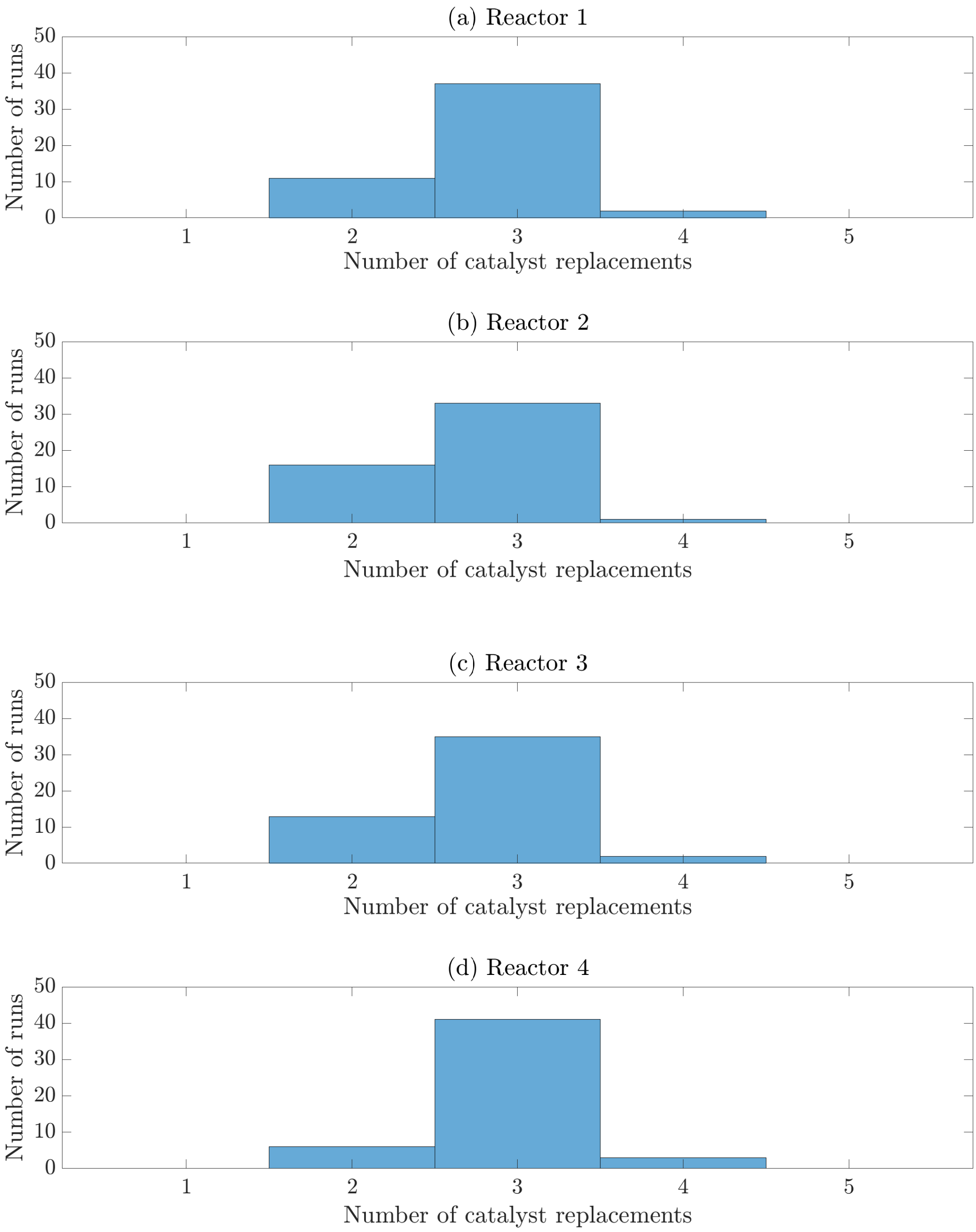


Fig. 3.3 The distribution of the number of catalyst replacements over all runs for (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4

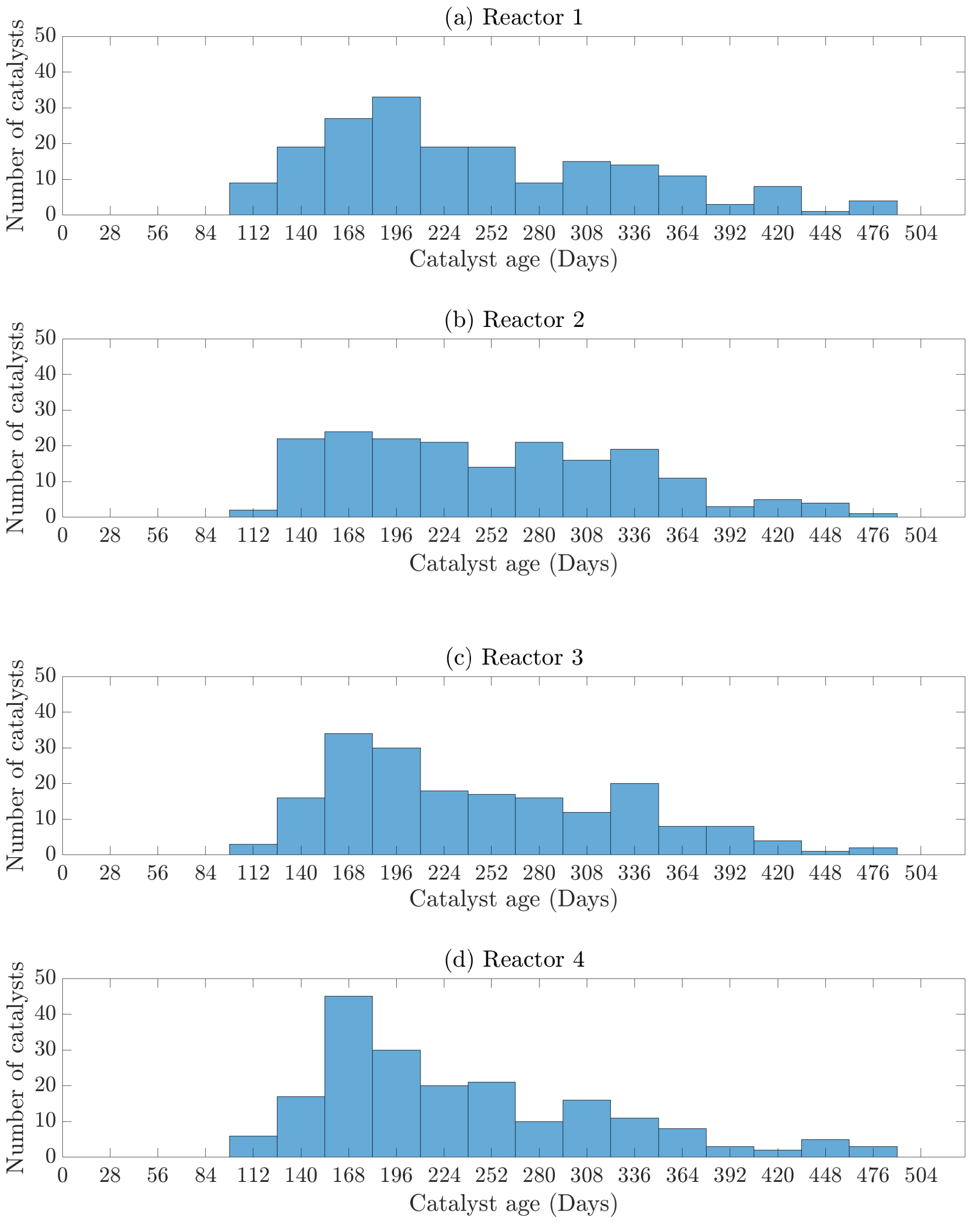


Fig. 3.4 The distribution of the ages of the catalyst used over all runs for (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4

As the statistics in Table 3.7 regarding the number of catalyst replacements and catalyst ages in all reactors are very similar, further insights into the distributions of these properties have been provided in Figures 3.3 and 3.4. Some comments regarding these figures:

- Figure 3.3 shows the distribution of the number of catalyst replacements obtained from all runs for all reactors. The histogram presented in subplot (a) of this figure pertains to Reactor 1 wherein the height of each bin represents the number of runs out of 50 that involved the number of catalyst replacements given by the midpoint of the horizontal width of the bin. Similar explanations hold for the histograms relating to Reactors 2, 3 and 4 in subplots (b), (c) and (d), respectively, of the figure. It is noted that the maximum allowable number of catalyst replacements for each reactor, as per the invented set of parameters used, is 5.

- Figure 3.4 shows the distribution of the ages of all catalyst used in all runs for all reactors. The histogram presented in subplot (a) of this figure pertains to Reactor 1 wherein the height of each bin represents the number of catalysts, out of all the catalysts used in all the 50 runs, that were used up to the age given by the midpoint of the horizontal width of the bin. Similar explanations hold for the histograms relating to Reactors 2, 3 and 4 in subplots (b), (c) and (d), respectively, of the figure. It is noted that since catalysts can be replaced only at the end of a month, all catalyst ages are multiples of 28 and the maximum allowable age of each catalyst, as per the invented set of parameters used, is 504 days.

The trends of the decision and state variables over the time horizon for the most profitable solution among the set of 50 runs are given in Figures 3.5 – 3.13. It was found that the trends over the time horizon, of the variables corresponding to each reactor in this parallel set up came out to be very similar to those of the single reactor in the results presented for Case Study A in Section 2.3.3 and Section 2.4.5 of the previous chapter, wherein the kinetics of the main reaction and catalyst deactivation were of a similar form to what is considered here. Therefore, the explanations for the trends of these variables are also similar to the explanations presented for the trends of the corresponding variables in those sections. A discussion of the obtained trends follows next.

Figure 3.5 illustrates the variation of the monthly catalyst changeover controls over the whole time horizon for all 4 reactors ($y1_{pr}$, $y2_{pr}$, $y3_{pr}$, $y4_{pr}$). In this case, two major iterations are needed to force the catalyst changeover controls for all reactors to take integer values. From the graph, it is seen that the model indicates that it is optimal for the process to replace the catalyst three times in Reactors 1, 2 and 3 and four times in Reactor 4, during those months corresponding to when their respective catalyst changeover controls become 0. It is highlighted that the months when catalyst replacements occur for the four reactors do not overlap, thereby fulfilling constraint (3.25) for non-simultaneous catalyst replacement. The other graphs presented are those obtained as solutions of the second major iteration.

The profiles of the feed flow rates to each reactor ($ffr1_{pr}$, $ffr2_{pr}$, $ffr3_{pr}$, $ffr4_{pr}$) and the total feed flow rate to the process ($ffr1_{pr} + ffr2_{pr} + ffr3_{pr} + ffr4_{pr}$) over the whole time horizon, are shown in Figures 3.6 and 3.7, respectively. The trends of the feed flow rates to all reactors in Figure 3.6 are related to the corresponding profiles of the temperature of operation ($T1_{pr}$, $T2_{pr}$, $T3_{pr}$, $T4_{pr}$), shown in Figure 3.9, and the reactant exit concentrations from the reactors ($cR1_{pr}$, $cR2_{pr}$, $cR3_{pr}$, $cR4_{pr}$), shown in Figure 3.12. The explanations of the trends of all these variables are as follows:

- In all reactors, during times of catalyst operation, the feed flow rate is decreased at a rate matching that of the reactor's catalyst deactivation and the temperature of the reactor is maintained at its upper bound. Such an operation causes the reactant exit concentration to maintain a constant value. This operational policy is consistent with the work of Szépe and Levenspiel (1968) for continuous reactors at the reactor level, which predicted similar operational strategies when the main reaction is more sensitive to temperature than the catalyst deactivation and the latter is independent of the species' concentration, as is the consideration in this case study.

- During times of catalyst replacement in all reactors, the feed flow rate is set to zero, the temperature of operation is set to its lower bound (TL_{pr}) and the reactant exit concentration is set to its entry value ($CR0_{pr}$), as per constraints (3.22) and (3.23) and junction conditions (3.15), respectively.

- In all reactors, during the operation of the last catalyst, a sharp drop in the flow rate occurs that causes a corresponding effect in the exit concentration. This occurs to bring the production rate to a value that exactly fulfils the demand for the remainder of the time horizon and thereby minimise the costs of flow and raw material.

In Figure 3.7, the profile of the net feed flow rate to the process over the time horizon, does not follow a regular trend, due to the variations in the feed flow rates to each reactor, that operate independently of each other. It is noted from the graph that the net flow rate does not reach its upper bound (FU_{pr}) at any point throughout the time horizon, indicating that the optimal policy does not require the maximum feed to the process.

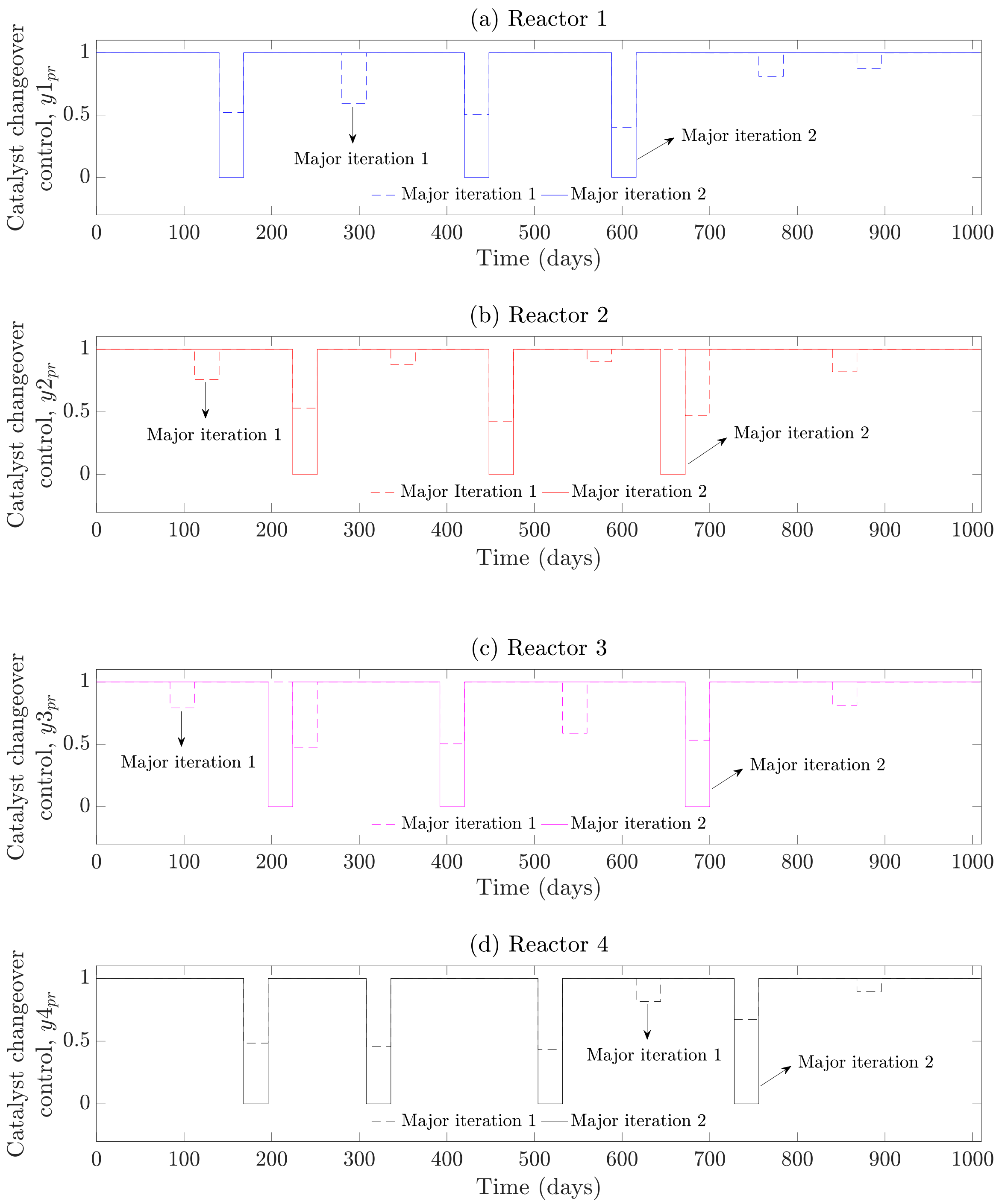


Fig. 3.5 The variation of the catalyst changeover controls over the time horizon in (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4, in the best solution.

Figure 3.8 shows a comparison of the optimal quantity of product sales ($sales_{pr}$) with the corresponding product demand ($demand_{pr}$) and unmet demand ($unmet\_demand_{pr}$) over the whole time horizon and it is seen that the trends are similar to that in the analogous graph of the corresponding single reactor case study. There is a considerable amount of unmet demand during the first year of the process, but it is nil for the remainder years. Within the first year, the unmet demand at the beginning occurs because there is no prior product inventory present and the production capacity of the process is unable to meet the high demand at that time. Towards the end of the first year, there is considerable unmet demand in order to enable a hoarding of product which in turn enables a greater amount of sales and nil unmet demand in the second and third years. This can be attributed to the increase in the sales price and the penalty for unmet demand due to annual inflation, which implies that a greater amount of sales and no unmet demand in the later years can enable attaining a larger profit. It is highlighted that the use of parallel reactor lines and the condition that only one reactor can undergo catalyst replacement at any time, enables the sales to occur continuously throughout the time horizon.

The variations of the catalyst ages over the time horizon in the 4 reactors (cat_age1_{pr}, cat_age2_{pr}, cat_age3_{pr} and cat_age4_{pr}) are shown in Figure 3.10. The trends of a linear increase with time during catalyst operation and a constant value of 0 during catalyst replacement follow directly from differential equations (3.1) and junction conditions (3.13). An analogous graph for the catalyst activities in the 4 reactors (cat_act1_{pr}, cat_act2_{pr}, cat_act3_{pr} and cat_act4_{pr}) is shown in Figure 3.11. In this figure, the trends of an exponential decrease during catalyst operation and a constant value at the starting catalyst activity during catalyst replacement follow directly from differential equations (3.2) and junction conditions (3.14).

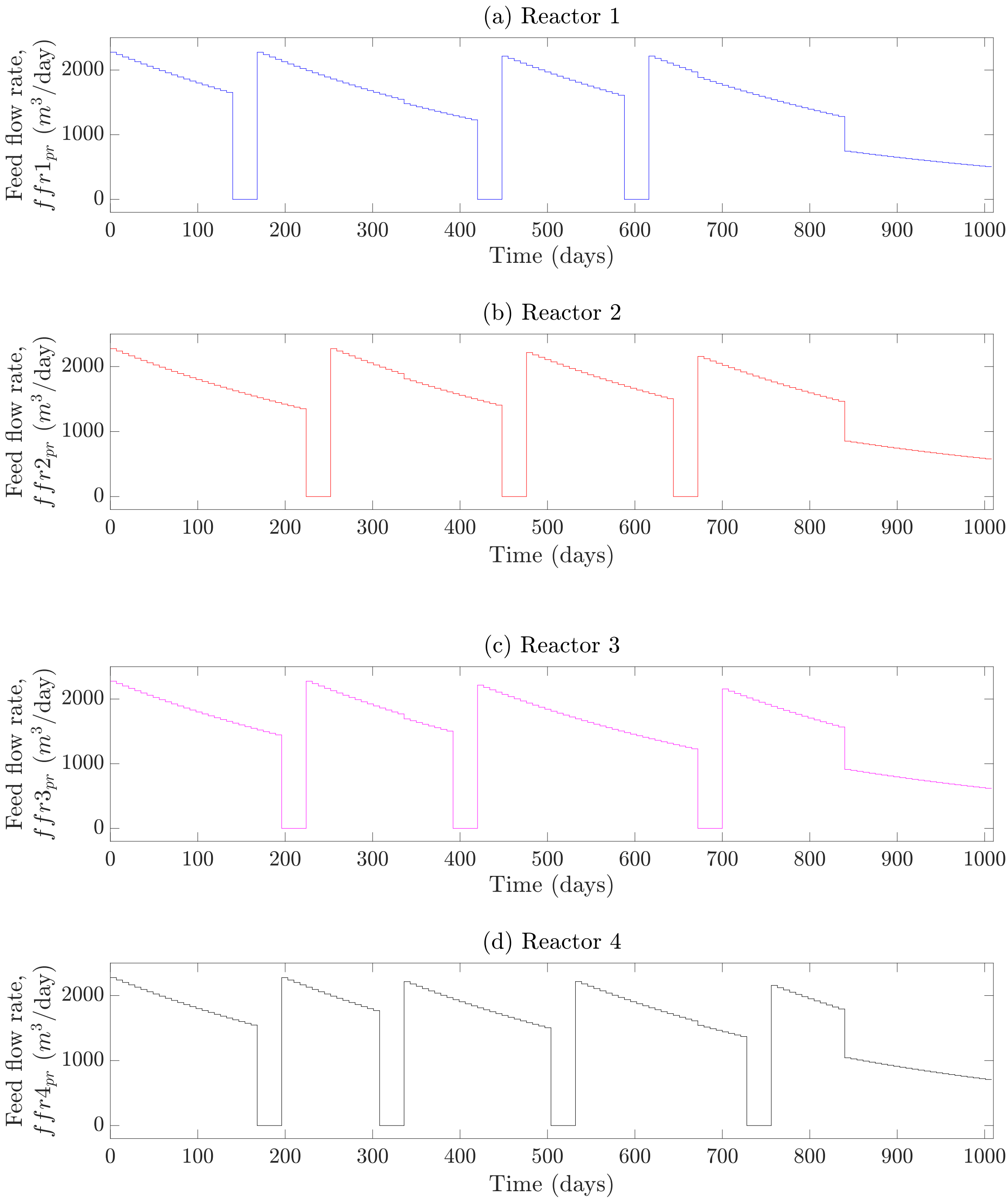


Fig. 3.6 The variation of the feed flow rate over the time horizon in (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4, in the best solution.

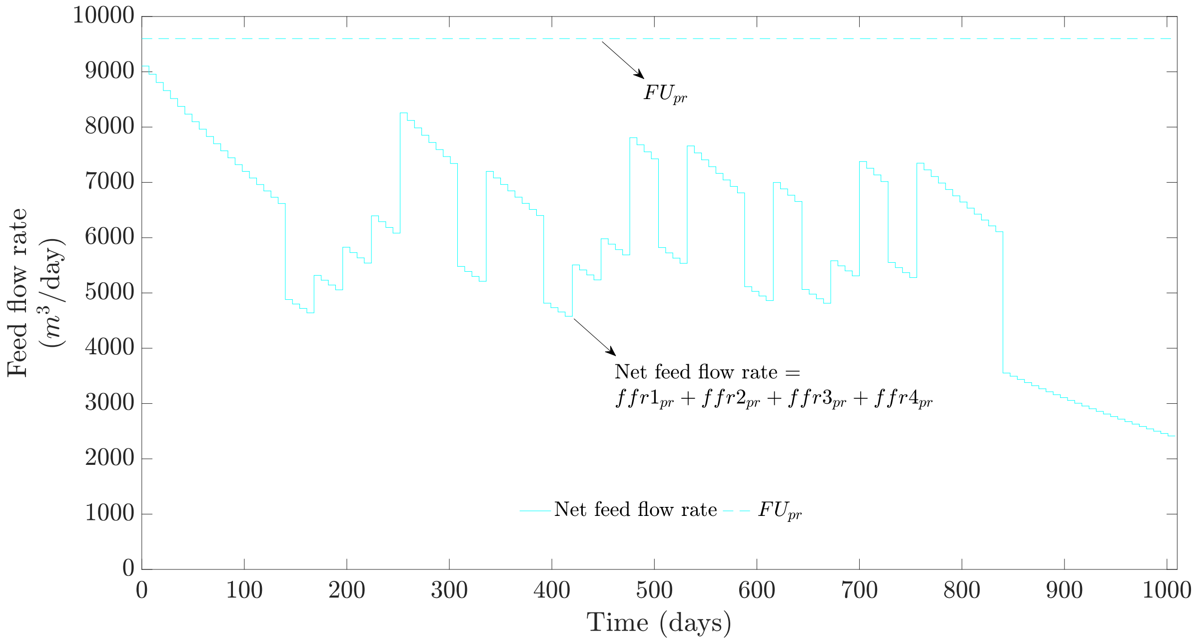


Fig. 3.7 The variation of the net feed flow rate to the process over the time horizon in the best solution.

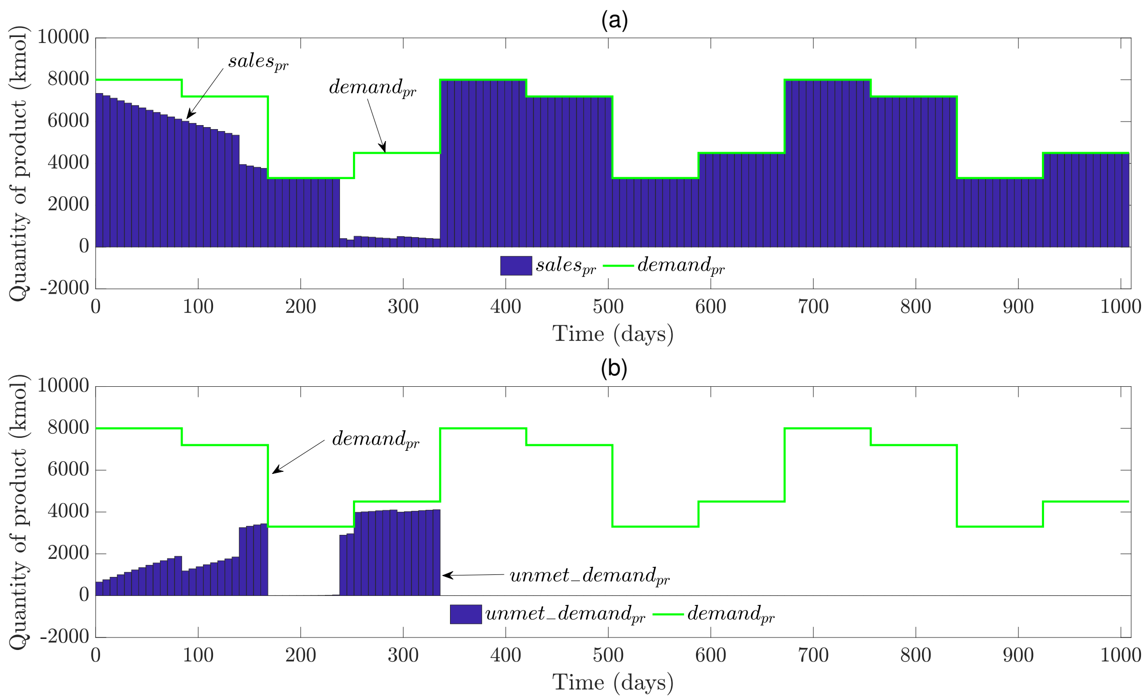


Fig. 3.8 The variation of (a) sales and (b) unmet demand, in comparison to the demand over the whole time horizon, in the best solution.

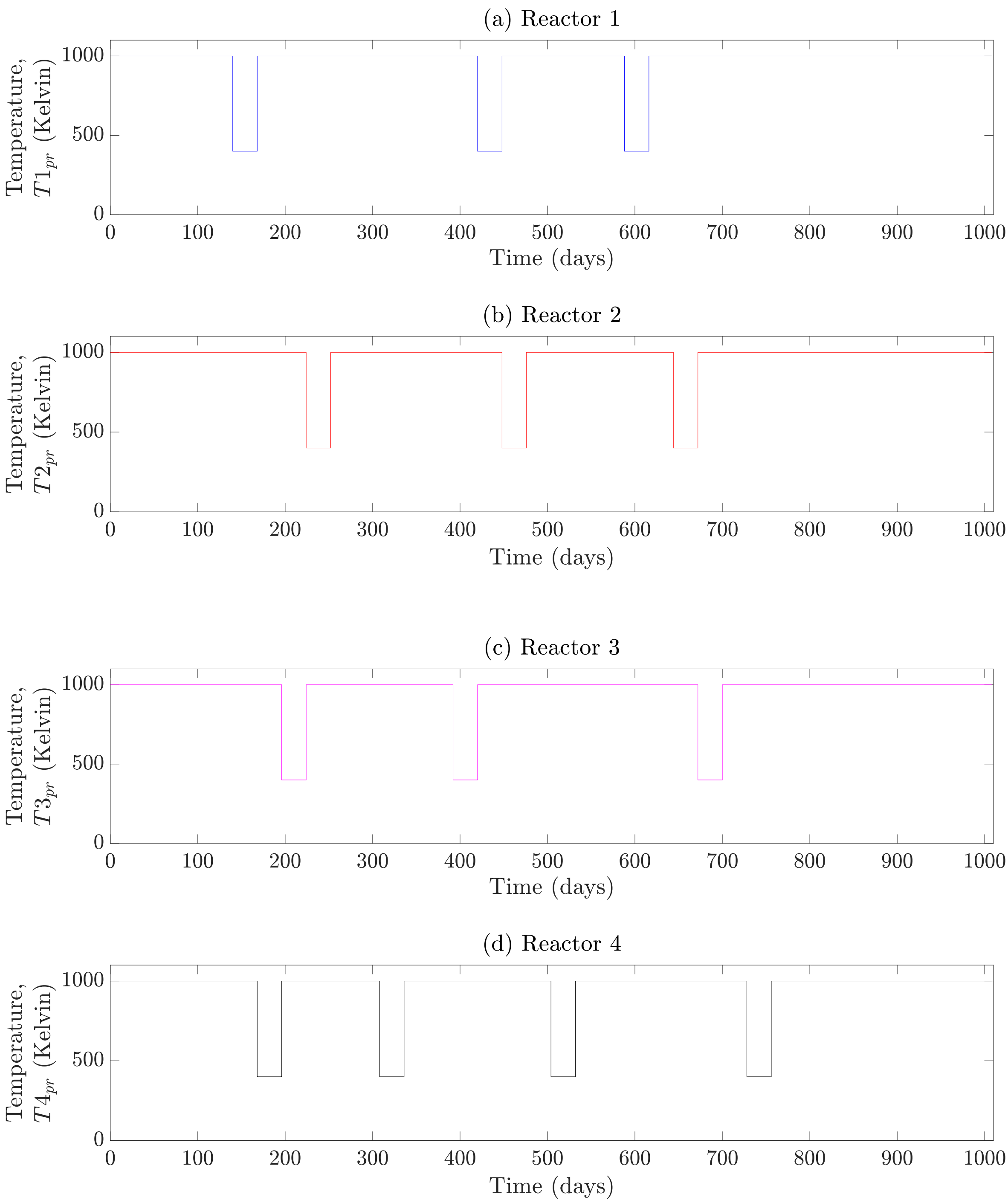


Fig. 3.9 The variation of the temperature of operation over the time horizon in (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4, in the best solution.

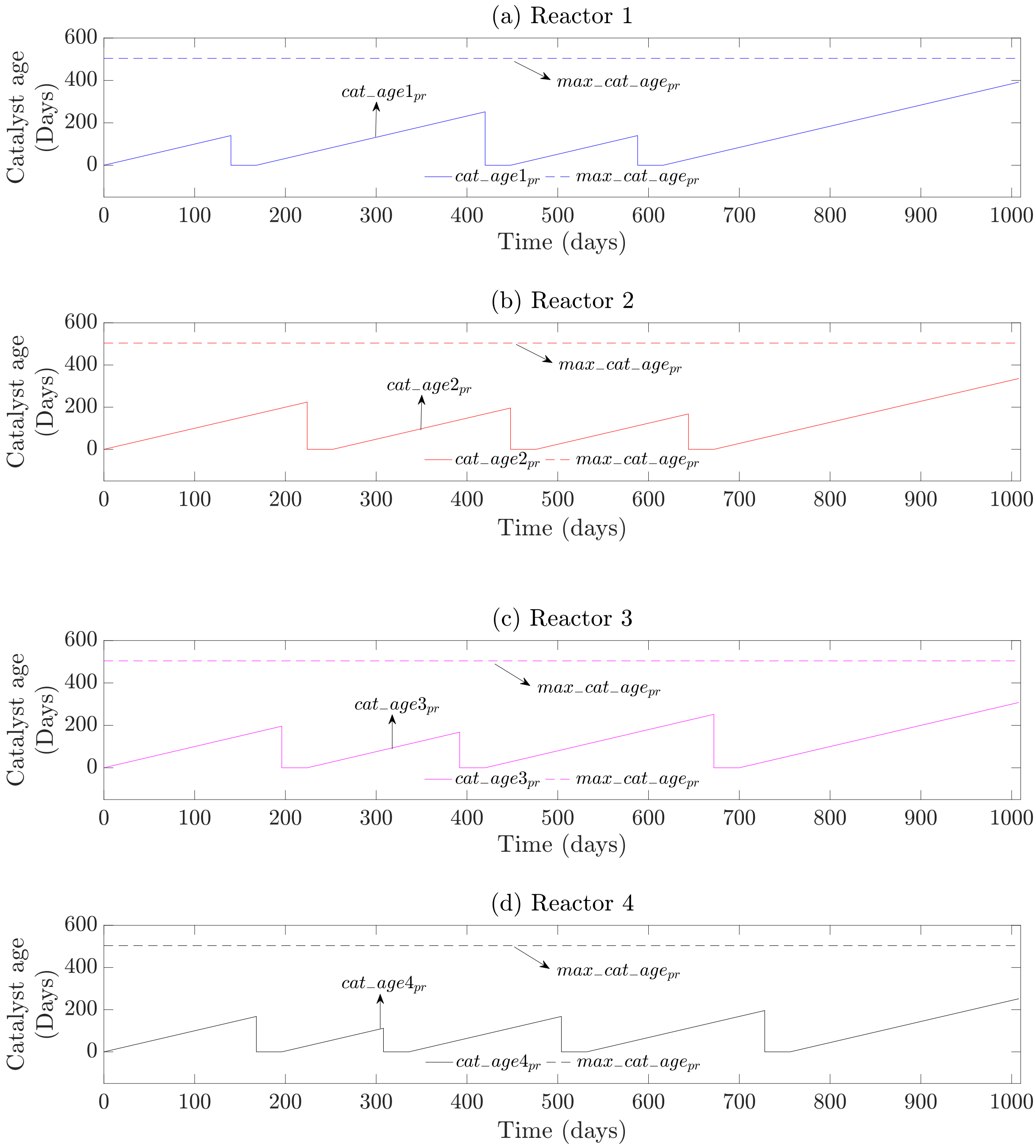


Fig. 3.10 The variation of the catalyst age over the time horizon in (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4, in the best solution.

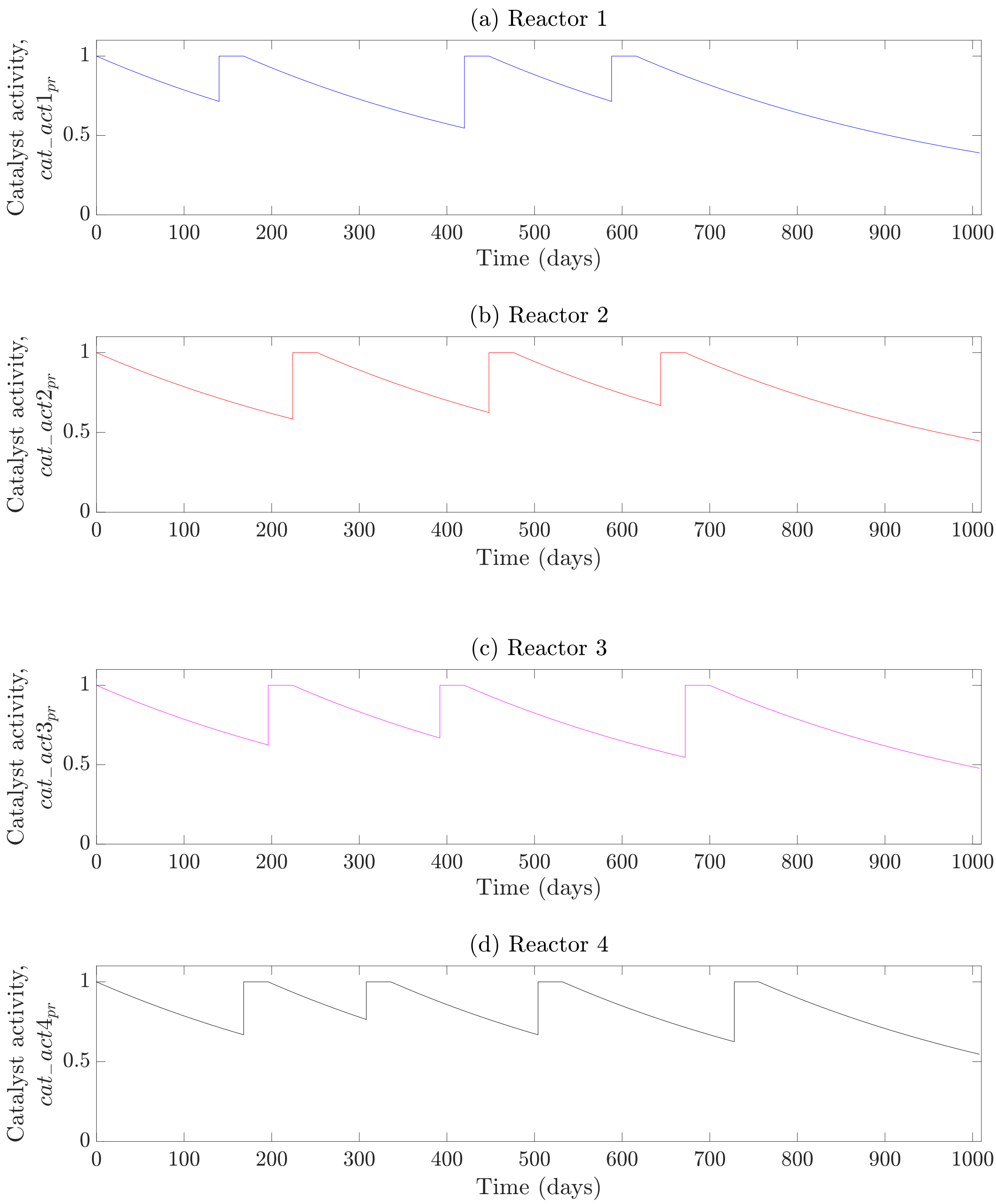


Fig. 3.11 The variation of the catalyst activity over the time horizon in (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4, in the best solution.

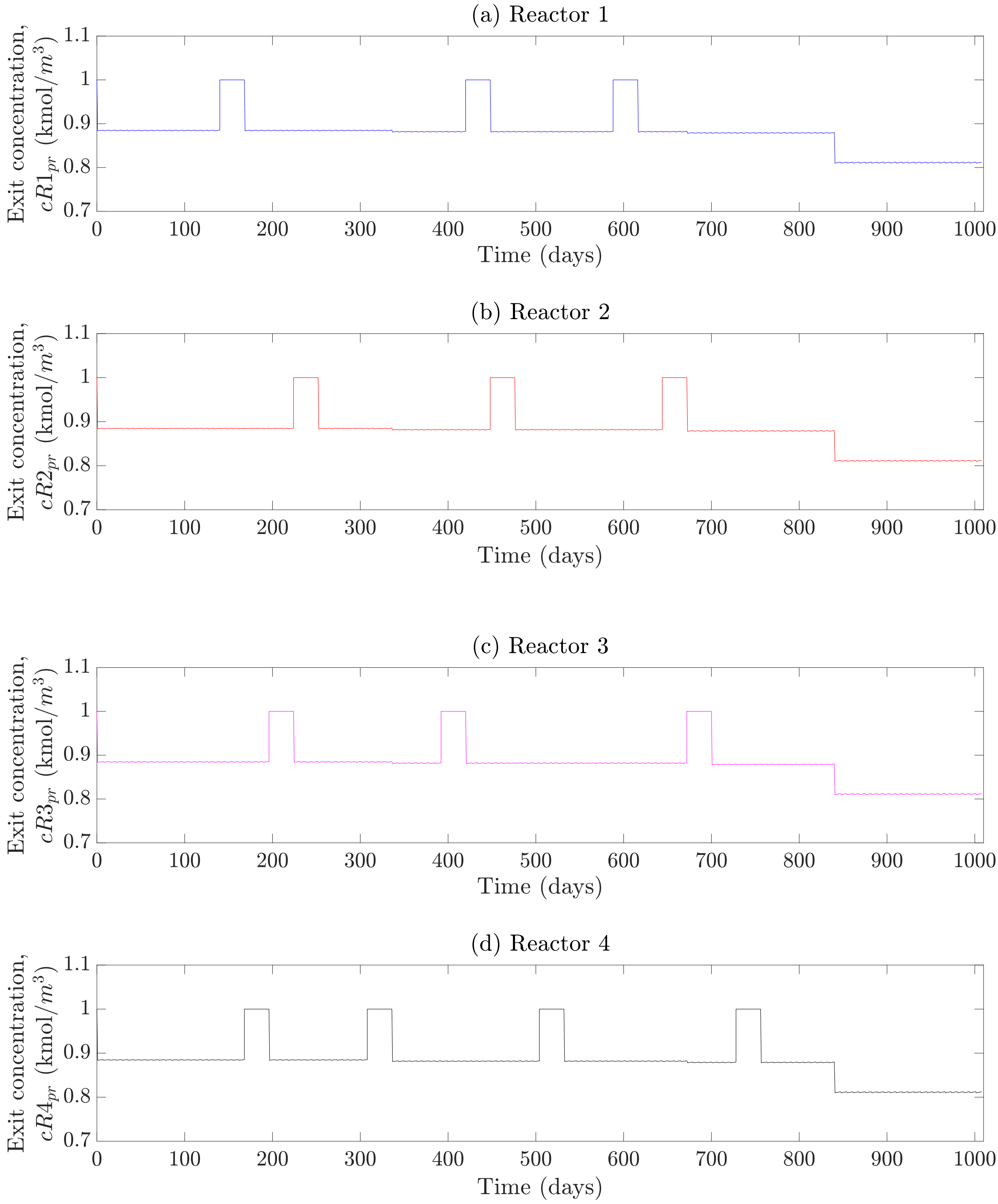


Fig. 3.12 The variation of the reactant exit concentration over the time horizon in (a) Reactor 1 (b) Reactor 2 (c) Reactor 3 (d) Reactor 4, in the best solution.

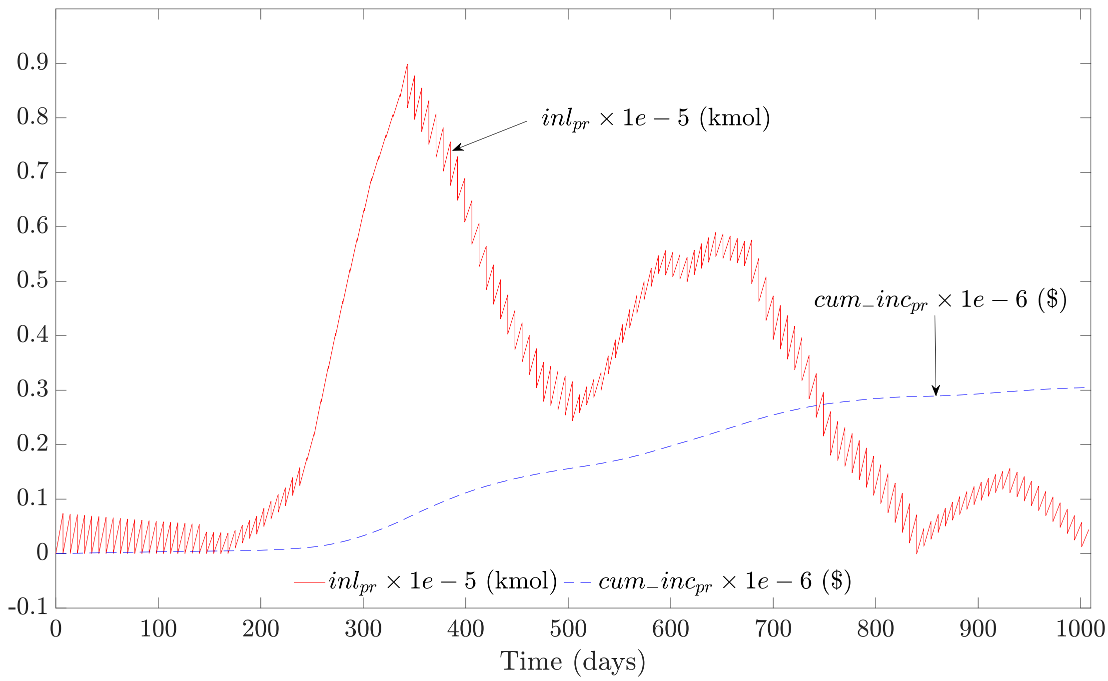


Fig. 3.13 The variation of the product inventory level and cumulative inventory costs over the time horizon, in the best solution.

Table 3.8 Details of the economic aspects of the best solution.

Economic aspect		**Symbol**	**Value ($ Millions)**
Gross Revenue from Sales		GRS_{pr}	774.672
Costs	Total Inventory Costs	TIC_{pr}	0.305
	Total Costs of Catalyst Changeovers	$TCCC_{pr}$	33.762
	Net Penalty for Unmet Demand	$NPUD_{pr}$	119.273
	Total Flow Costs	TFC_{pr}	185.737
Profit		$-NC_{pr}$	435.595

The variation of the inventory level (inl_{pr}) and cumulative inventory costs (cum_inc_{pr}) over the time horizon are shown in Figure 3.13. The oscillating behaviour of the inventory level follows from the interplay between the increase in inventory due to production from all reactors (differential equation (3.5)) and the decrease in inventory due to the sales (junction condition (3.16)). It is highlighted that towards the end of the first year, the inventory level shows a significant increase, despite there being a considerable amount of unmet demand at that time. This happens in order to enable greater amount of sales and thereby eliminate the unmet demand during the later years when the product sales price and the penalty for unmet demand have increased due to inflation, thus enlarging the profit obtained. The trend for the cumulative inventory costs follows directly from differential equation (3.6) and junction condition (3.17).

The magnitudes of the various economic aspects that form the elements of the objective function are given in Table 3.8. The table indicates that the cost of flow and raw material, and the net penalty for unmet demand form the biggest proportions of the costs. The cost of catalyst changeovers contributes relatively less while the inventory costs form a very low percentage of the total expenditure. The costs of operation take away about 43.77% of the revenue generated by the product sales. It is highlighted that no comment can be made regarding the magnitudes of the economic components or the order of contribution of the various costs involved, apart from that these follow from the values of the set of invented parameters used. This is similar to what was mentioned in Section 2.4.9 of the previous chapter regarding the economics of the various case studied examined in the single reactor problem.

The graphs presented above showcase the trends of the decision variables and of the state variables that follow accordingly, of only the best solution or the local optima that obtained the best profit. While it is possible to present a distribution of each decision variable over the 50 runs in a manner similar to that done for the profit, for example, in Figure 3.2, these are not informative enough to justify the volume of the thesis that would be consumed by such a presentation, given that there are hundreds of decision variables involved. However, further comments can be made regarding the decision variables obtained in the other local optima. These comments are similar to those mentioned in Section 2.4.9 of the previous chapter and are stated as follows:

- The local optima differed in the obtained values of the catalyst changeover decision variables of at least one reactor. The different catalyst replacement schedules, which decided the number and timing of catalyst replacements for each reactor, impacted the decision variables of feed flow rate of each reactor and sales of the process, and as such, was the major factor leading to differences in profits between the different local optima obtained.

- Between different local optima, the decision variables of the feed flow rate of at least one reactor differed in the values obtained during the operation of the final catalyst in the respective reactor. In the other local optima, towards the end of the time horizon, while the trend of the feed flow rate in each reactor was similar to those in the best solution and the demand was fulfilled, the sharp drop in the feed flow rate for each reactor was either not present or was of a smaller magnitude in comparison to those observed in the best solution. This led to the total flow costs in the other local optima being higher compared to that in the best solution and this could be attributed to the combined catalyst replacement schedules for all reactors in the other local optima being less efficient than in the best solution. However, it is highlighted that for all optima obtained, in each reactor, during the times of operation of all catalysts except the final one, the feed flow rates were similar in terms of magnitudes as well as trends, which led to constant values of the exit concentration of similar magnitudes during these times as well.

- For all optima, in all reactors, the temperature was set to its upper bound during times of catalyst operation. This followed from the fact that in all reactors, the catalyst deactivation rate constant was independent of temperature and the maximum rate of product formation could be obtained by operating at the maximum allowable temperature.

- In comparison to the best solution, the distribution of the sales in the first year differed from that in the other local optima in that the quantity of sales was either similar or lower in comparison to the former. This led to the gross revenue from the sales in that first year in the other local optima to be less than or at most similar to that in the best solution and this could be attributed to the combined catalyst replacement schedules for all reactors in the other local optima being less efficient than in the best solution. However, in all optima obtained, the sales completely fulfilled the product demand in the second and third years and this could be attributed to the inflation causing the product sales price and penalty for unmet demand to be higher in those years compared to the first.

3.3 Summary and conclusions

In this chapter, an optimal control methodology has been developed for optimising maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts. This methodology is an extension of the optimal control methodology developed in the previous chapter for optimising similar aspects in a process containing a single reactor using decaying catalysts.

This methodology involves using an MSMIOCP formulation of the problem in combination with a solution procedure of the principle of Implementation II, which was developed in the previous chapter. The elements of this MSMIOCP formulation represent the various elements of the process in an analogous manner to that in the previous chapter. As per this methodology, the binary restrictions on the catalyst changeover controls in the MSMIOCP formulation of the problem are relaxed and a penalty term homotopy technique is applied wherein a series of standard multistage optimal control problems are solved using a feasible path approach, with a monotonically increasing penalty term added to the objective function to enforce binary values for those controls. The highlights of this methodology are that the penalty term homotopy technique obviates the need for mixed-integer optimisation methods to schedule catalyst changeovers and the feasible path approach guarantees accuracy, and both these features enable a smaller problem size which facilitates an easier convergence to solutions.

No previous publication has explicitly addressed such a problem and so there was no process that could be used as a base to apply this methodology to. Therefore, a case study of the process containing a single reactor, examined in the previous chapter, was modified into a case study of a process in which a single feed is split over 4 parallel reactors, and the proposed optimal control methodology was applied to optimise maintenance scheduling and production in such a set up.

Due to the non-convex nature of the problem, the optimisation was performed using 50 different initial guesses and each of these runs successfully converged to a local optimum without any difficulties. For the best solution among these runs, the profiles of the decision and state variables over the time horizon and the economics of the industrial process were presented. A notable result was that the optimal operating policies for the temperature of each reactor and the reactant exit concentration from each reactor correlated well with that of published literature (Szépe and Levenspiel, 1968) at the reactor level, and this is similar to the observation in the related case study in the single reactor problem considered in the previous chapter.

In conclusion, the use of the proposed optimal control methodology to optimise maintenance scheduling and production in parallel lines of reactors using decaying catalysts has resulted in high quality solutions. The nature of the methodology and the solutions obtained indicate a number of potential advantages of the use of this methodology over mixed-integer methods to optimise problems of this kind. These potential advantages are similar to the points presented in Section 2.5.1, in the conclusions of the previous chapter. However, for the sake of completeness of this chapter, the potential advantages of the use of this optimal control methodology over mixed-integer methods to solve this problem are elaborated briefly:

1. It is robust because the smaller problem size enabled facilitates convergence to optimal solutions from any random initial guess.

2. It is reliable because solutions can be obtained to a high degree of accuracy using state-of-the-art integrators, without the use of any approximation techniques. Thus, by using this approach, the dynamics of the parallel system of reactors can be described very accurately.

3. It is efficient because a maintenance schedule for catalyst replacements in the parallel reactor set up, that fulfils the condition of non-simultaneous catalyst replacements, can be obtained inherently during the optimisation, without using any additional computational effort in deciding when to schedule catalyst changeovers.

Thus, in this chapter, in line with the second objective of the thesis, a methodology has been developed that can effectively optimise maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts, and which can overcome the drawbacks that mixed-integer methods would face in solving problems of this kind. In the next chapter, the principle of the optimal control methodology used in this and the preceding chapter is applied as part of a methodology to optimise similar operations while considering uncertainties in the kinetic parameters of the underlying process model.

Chapter 4

Process optimisation under uncertainties in kinetic parameters

In this chapter, an optimal control methodology is developed for considering uncertainties in kinetic parameters in the optimisation of maintenance scheduling and production in a process containing a reactor using decaying catalysts. This methodology is an extension of the forms of methodologies used in the previous chapters to solve problems which did not consider parametric uncertainty. Using a multiple scenario approach to consider parametric uncertainty, an initial multistage mixed-integer optimal control formulation of the problem is converted into its stochastic counterpart, which is then solved as a standard nonlinear optimisation problem as per a procedure of the principle of Implementation II, which was developed in Chapter 2. Different case studies are examined in order to identify the effects on the optimal operations of uncertainty in each individual parameter, of simultaneous uncertainty in all parameters and of the number of scenarios generated. The results obtained provide insights into such aspects and indicate that the methodology is capable of solving this problem. Further, the results obtained possess features of robustness, reliability and efficiency which suggest that the methodology can overcome the drawbacks that mixed-integer methods would introduce in the conventional methodologies, if such methodologies are used to solve problems of this kind.

The structure of this chapter is as follows. Section 4.1 provides an introduction to the problem and a review of the publications related to this topic. Section 4.2 details the solution procedure proposed to solve this problem. In Section 4.3, case studies of this problem are formulated as per the proposed solution procedure. Section 4.4 presents the implementation details and Section 4.5 discusses the results obtained. Section 4.6 contains a summary and conclusions of the chapter, and further discussions involving the proposed methodology.

4.1 Introduction and literature review

In the preceding chapters, the problems examined did not consider any uncertainty in the values of the kinetic parameters present in the underlying process models. That is, the kinetic parameters, such as the deactivation rate constant in the rate equation for catalyst decay, and the Arrhenius pre-exponential factor and the activation energy in the rate equation for the product formation reaction, were assumed to be known and fixed in value. However, as these parameters are experimentally obtained, their values are susceptible to a degree of uncertainty because of inevitable inaccuracies arising from measurement errors, estimation errors, interpolation and extrapolation errors etc.

With regard to the problems involving optimisation of maintenance scheduling and production in industrial processes using decaying catalysts, such uncertainties in kinetic parameters can have a considerable impact. Since the outputs of the kinetic models form the basis for making optimal decisions, there can be many implications to not considering uncertainties in kinetic parameters. For instance, uncertainty in the rate of product formation or catalyst decay can result in variable and unpredictable processing times and production yields. This can create difficulties in identifying the optimal maintenance schedules, operating conditions, and the appropriate production amounts and inventory levels to effectively meet time-varying product demand. In some cases, an uncertainty in the rate of catalyst decay could affect the durability of the reactor and threaten the safe operation of the process, because of the lack of accuracy in the rate of decay causing possibilities of the catalyst being used beyond its recommended lifespan. Hence, it is critical to take uncertainty in kinetic parameters into account while optimising process operations.

A literature survey revealed a very limited set of studies that considered problems related to that involving a consideration of uncertainty in kinetic parameters while optimising maintenance scheduling and production planning in processes using decaying catalysts. Only two such studies have been found and these have been based on online or data-driven methods of optimisation. However, it is highlighted that even in these studies, the product demand to be met was constant and not seasonal, and therefore these studies cannot be considered as addressing the complete problem of interest here.

One of these is a work by Lim et al. (2009), which proposed a proactive scheduling strategy to handle uncertainties in coke thickness and growth rates in the scheduling of decoking operations in a naptha cracking furnace system. As per this strategy, when the gap between model predictions and the actual measurements becomes larger than a certain threshold value, the model is updated using the measurements and this updated model is used to obtain a new schedule. This technique has shown advantages in terms of productivity and risk management, in comparison to reactive scheduling and heuristic decoking strategies.

The other one is a work by Jahandideh et al. (2019), which considered production planning and scheduling of decaying catalysts in a process, while including uncertainty in the rate of catalyst decay, by formulating this problem as a semi-Markov decision process. They use a a two-level heuristic to make the problem tractable and report attaining low percentage gaps with the lower bounds on the optimal costs.

The online methods, used in the aforementioned literature, involve making decisions to handle the effect of uncertainties on the basis of data continuously obtained from the process. However, as the data obtained encompasses the variations in all parameters present, the impact of uncertainty in a particular parameter cannot be quantified. That is, the use of these methods does not enable a sensitivity analysis regarding the effect of each uncertain parameter to be performed. In addition, for large scale complex processes operating over long time horizons, it would be desirable to understand the effect of uncertainties before the process begins execution. Obtaining prior predictions of the optimal maintenance schedules and operating conditions, as well as forecasts of expected profits, while considering uncertainties can greatly improve investment and operating decisions. To fulfil such purposes, preventive methods for handling uncertainties need to be used.

The popular preventive methods of handling uncertainty in problems involving scheduling optimisation include stochastic programming, fuzzy programming, robust optimisation and parametric programming (Li and Ierapetritou, 2008a). The use of one of these techniques in combination with a mixed-integer formulation represent the conventional methodology of solving scheduling problems involving parametric uncertainties. There have been several publications that have used one of these conventional methodologies in order to optimise scheduling of varied processes while considering uncertainties in parameters such as processing times, demand and prices, to name a few.

A few examples of works using stochastic programming to handle uncertainty in optimisation problems involving scheduling include those by Balasubramanian and Grossmann (2002) for flowshop plants involving uncertain processing times, by Bonfill et al. (2005) for batch processes with uncertain operation times, by Restrepo et al. (2017) for multi-activity tours under demand uncertainty, and by Palacín et al. (2018) for evaporator networks while considering uncertainty in the outdoor weather and production plan.

Some examples of works using fuzzy programming to handle uncertainty in scheduling optimisation problems are those by Balasubramanian and Grossmann (2003) for flowshop plants and new product development processes with uncertain processing times, by Wang (2004) for product development projects with uncertain temporal parameters, by Felizari and Lüders (2006) in oil refineries facing uncertain time delays, and by Kilic (2007) for parallel machines with uncertain processing times and flexible due dates.

Some works using robust optimisation to consider uncertainty in scheduling optimisation include those by Li and Ierapetritou (2008c) for processes facing uncertainty in processing times, demand and prices, by Yan and Tang (2009) for problems involving inter-city buses under variable market share and uncertain market demand, by Li et al. (2012) for crude oil operations under demand uncertainty, and by Ye et al. (2014) for continuous casting processes in steel-making involving demand uncertainty.

Examples of publications using parametric programming to consider uncertainty in scheduling optimisation problems are those by Ryu et al. (2007) for multiproduct batch scheduling under uncertainty in processing time and equipment availability, by Li and Ierapetritou (2007) for processes facing uncertainties processing times, demand and prices, by Li and Ierapetritou (2008b) for processes facing uncertain disruptions of rush orders and machine breakdowns, and by Umeozor and Trifkovic (2016) for operation of microgrids containing uncertainty in power outputs, among others.

However, there has been no work that has used one of the conventional methodologies to consider parametric uncertainty in the optimisation of maintenance scheduling and production in processes using decaying catalysts. Further, none of the existing publications have explicitly considered uncertainties in the values of the kinetic parameters of the underlying process model. In reality, due to the involvement of mixed-integer techniques, these conventional methodologies would face difficulties in obtaining good quality solutions to the complex large-scale problem under consideration here: that of optimising maintenance scheduling and production planning in a process using decaying catalysts while incorporating uncertainty in kinetic parameters.

As mentioned in Section 1.4 of Chapter 1, mixed-integer optimisation techniques are combinatorial in nature and these methods approximate the differential equations present in the problem as a collection of steady state equations, which are imposed as additional constraints in the problem. In that section, these features were highlighted as the drawbacks involved in the use of mixed-integer techniques, in that such features could cause these techniques to face difficulties in convergence to optimal and accurate solutions when used in large scale problems. It was also detailed in that section how the drawbacks of these techniques apply in a problem related to the one under consideration in this chapter, which was deterministic in that there were no uncertainty considerations present.

If, as in this chapter, uncertainty considerations are present, and one of the aforementioned conventional methodologies are used, a problem of similar or larger size in comparison to the deterministic problem will have to be solved using mixed-integer optimisation techniques. Therefore, the drawbacks of these techniques will once again be manifested or possibly even be further aggravated. This is elaborated upon next.

In stochastic programming, an overview of which is given by Birge and Louveaux (2011), the uncertain parameters are considered as stochastic variables, which can take a known set of values. The instance of occurrence of each value is called a scenario and the probability of occurrence of each scenario is assumed known. However, as mentioned in the work of Li and Ierapetritou (2008c), in the techniques of this category, the number of scenarios increases exponentially with the number of uncertain parameters. And as the number of scenarios increases, the number of decision variables, differential equations and constraints increase proportionately. Therefore, if stochastic programming techniques are applied to the large scale problem under consideration here and solved using mixed-integer optimisation methods, even for a small number of uncertain parameters involving a small number of scenarios, the number of variables and constraints involved would be enormous. Hence, the final problem size would be intractable and so, obtaining good quality solutions will be difficult.

In fuzzy programming, an uncertain parameter is considered as a fuzzy number, which is essentially an interval of values. The membership function of a fuzzy number describes the degree of membership of any numerical value to the interval. This membership function takes values within the range, $[0,1]$, with a larger value of the membership function indicating a greater degree of membership to the fuzzy number (Zadeh, 1965). If uncertainty in a parameter is to be appreciably taken into account in an optimisation problem, it is essential to use numerical values of this parameter corresponding to a large number of values of the membership function, across the range of $[0,1]$. And when a larger number of membership function values, and so, a larger number of values of the uncertain parameter are considered, the number of variables and constraints involving that parameter in the problem increase proportionately. For large scale problems, this could lead to intractable problem sizes.

As such, in scheduling optimisation problems using fuzzy programming to consider parametric uncertainty, the mixed-integer optimisation techniques, due to the underlying drawbacks, are able to solve only relatively small scale problems but are unable to solve large scale problems due to the intractable problem sizes encountered (Balasubramanian and Grossmann, 2003). For such large scale problems, a number of publications, examples of which include some of the previously mentioned papers of scheduling optimisation under uncertainty using fuzzy programming, resort to using meta-heuristic techniques over mixed-integer optimisation methods. However the use of meta-heuristic techniques faces the drawback of not being able to provide a theoretical guarantee of convergence to optimality. Therefore, for the large scale problem under consideration here, if such methodologies are followed, similar difficulties will be encountered. That is, using a mixed-integer optimisation technique with a fuzzy programming method to consider parametric uncertainty, would likely be unable to obtain solutions as the problem size would become intractable. Alternatively, if a meta-heuristic technique is used in place of the mixed-integer optimisation method, the optimality of the obtained solutions cannot be guaranteed.

In robust optimisation, the objective is to obtain a solution that is feasible for any realisation of uncertainty in a given set. The techniques of this category function by constructing a robust counterpart of the deterministic problem, the solution of which enables attaining the aforementioned objective (Gorissen et al., 2015). An attractive feature of this category of techniques is that the robust counterpart problem formulated is comparable in size to that of the deterministic problem. However, if for the problem under consideration here, a robust counterpart is formulated and mixed-integer optimisation methods are used for solution, the drawbacks of these methods will be encountered once again. That is, since mixed-integer methods can face difficulties in converging to optimal and accurate solutions even for the deterministic version of the problem under consideration here, similar difficulties will be experienced if these methods are used to solve the corresponding robust counterpart problem.

Parametric programming methods seek to obtain mappings between uncertain parameters and optimal solution alternatives, and thereby provide an analytical tool for the identification of solutions under uncertainty. For problems involving integer and continuous variables, this is done by solving a series of parametric and mixed-integer programming problems (Dua and Pistikopoulos, 2000; Faísca et al., 2009; Wittmann-Hohlbein and Pistikopoulos, 2012), the latter which are larger in size compared to the deterministic problem. However, if such procedures are applied to the problem under consideration here, with mixed-integer optimisation techniques being used to obtain solutions, once again, the drawbacks of these techniques will come into play. That is, since these techniques can cause difficulties in converging to optimal and accurate solutions even for the deterministic version of the problem under consideration here, using these techniques for the solution in the series of problems will also face similar drawbacks.

From the preceding discussion, it is concluded that conventional methods would not be well suited to solve the complex large scale problem being considered here: that of optimising maintenance scheduling and production planning in a process using decaying catalysts while incorporating uncertainty in kinetic parameters. A methodology is needed that can effectively solve this problem. In this chapter, such a methodology is developed using optimal control theory. This optimal control methodology is in fact an extension of the forms of optimal control methodologies used to solve deterministic problems in Chapters 2 and 3.

As in Chapters 2 and 3, in this optimal control methodology, the basic MSMIOCP of the form of equation (2.4) is to be used for the initial formulation of the problem under consideration in this chapter. The elements of this MSMIOCP are to represent the various elements of the process in an analogous manner to that in those chapters. That is, the set of binary controls in this formulation is to indicate when to schedule catalyst changeovers, the set of continuous controls is to decide the reactor operating conditions and the sales of the process, the DAEs are to represent the process model, the constraints are to represent the operating limits of the process and the binary restrictions on the catalyst changeover controls, and the objective function is to represent the net costs of the process. However, unlike in those chapters, the DAEs in this MSMIOCP formulation contain uncertain parameters, with the uncertain parameters being the kinetic parameters, the values of which are regarded uncertain.

In the next section, the solution procedure to be used in this optimal control methodology is presented. As the basic MSMIOCP formulation of the form of equation (2.4) is to be used for the initial formulation of the problem under consideration here, the solution procedure is demonstrated using this formulation. It is also highlighted what advantages the methodology using this formulation in combination with this solution procedure would potentially offer over conventional methods of solving problems of this kind. In the sections following this, this optimal control methodology is applied to solve case studies of an industrial process and the results obtained are discussed.

4.2 Problem solution procedure

The solution procedure to be used in this optimal control methodology seeks to solve an MSMIOCP of the form of equation (2.4) which contains uncertainties in the parameters present in the DAEs, as a standard nonlinear optimisation problem using a feasible path approach, while ensuring that the model-inherent parametric uncertainties are accounted for. In Chapter 2, the solution procedure of Implementation II, detailed in Section 2.4.3, was used to solve a deterministic MSMIOCP of the form of equation (2.4) as a standard nonlinear optimisation problem using the feasible path approach. The solution procedure presented in this section is a modified form of the procedure presented in Section 2.4.3, with the modification involved being the inclusion of an additional step in order to incorporate model-inherent parametric uncertainties.

First, as done in Section 2.4.3, the binary restriction on the integer controls in the basic MSMIOCP formulation of the form of equation (2.4) is relaxed in order to form an MSMIOCP with relaxed binary controls of the form of equation (2.61).

Next, in order to consider parametric uncertainty, a multiple scenario approach is used. That is, multiple scenarios are generated, where in each scenario, each uncertain parameter takes a particular value within a pre-specified range. These scenarios can be generated through any random sampling method.

As mentioned previously, the uncertainties in this problem occur in the parameters present in the DAEs of the MSMIOCP formulation. Thus, for each scenario generated, a new DAE system, comprised of the DAEs, initial and junction conditions, is formed, the parameter values of which correspond to the scenario generated. And the state variables in each scenario can be determined by the solution of the DAE system corresponding to that scenario. The uncertainty in the problem is represented by the different scenarios: by all the different parameters and state variables values attainable. The aim is to perform the optimisation while ensuring that the uncertainty represented by the different scenarios are accounted for.

A feature of the solution procedure of any optimal control problem is that the integration phase is independent of the optimisation phase. That is, any DAE system present has to be integrated completely to obtain values of the state variables, which in turn are used to formulate the objective function and constraints based on which the optimisation can occur. This feature will be exploited here to perform the optimisation under uncertainty.

A new MSMIOCP is formulated wherein the DAE systems corresponding to all scenarios are stacked together. However, while the state variables obtained from the DAE system for each scenario lead to a unique set of constraints and objective function corresponding to that scenario, only the averages of these over all scenarios are used in this MSMIOCP. That is, the decision (or control) variables have to be chosen to optimise only the average of the objective functions over all scenarios while fulfilling only the average of each set of constraints over all scenarios. By considering the averages of the objective function and constraints over all scenarios, this new MSMIOCP ensures that the uncertainty represented by all the different scenarios are accounted for. This new MSMIOCP is given by equation (4.1) and can be considered a stochastic version of the formulation presented in equation (2.61). In fact, a formulation of the form of equation (4.1) will be referred to as a "stochastic MSMIOCP with relaxed binary controls" in this thesis.

In equation (4.1), W_{st} is the performance index of the stochastic MSMIOCP with relaxed binary controls and SN is the number of scenarios considered. A separate term $\overline{C}^{(p)}$ is introduced to represent the average of the set of constraints over all scenarios in stage p, as shown in equation (4.1f). The remainder of the terminology used are similar to the MSMIOCP formulation in equation (2.61) with the only difference being that where subscript s occurs, it represents the variable in scenario s.

$$\begin{aligned}\min_{u,v} W_{st} \quad = \quad & \frac{1}{SN}\left[\sum_{s=1}^{SN}\left[\sum_{p=1}^{NP}\left\{\phi_s^{(p)}\left(x_s^{(p)}(t_p),\, z_s^{(p)}(t_p),\, u^{(p)},\, v^{(p)},\, t_p\right)\right.\right.\right. \\ & \left.\left.\left. + \int_{t_{p-1}}^{t_p} L_s^{(p)}\left(x_s^{(p)}(t),\, z_s^{(p)}(t),\, u^{(p)},\, v^{(p)},\, t\right)\,\mathrm{d}t\right\}\right]\right]\end{aligned} \tag{4.1a}$$

subject to

$$\begin{aligned}\dot{x}_s^{(p)}(t) = f_s^{(p)}\left(x_s^{(p)}(t),\, z_s^{(p)}(t),\, u^{(p)},\, v^{(p)},\, t\right) \\ t_{p-1} \le t \le t_p \\ p = 1,2,\ldots,NP \\ s = 1,2,\ldots,SN\end{aligned} \tag{4.1b}$$

$$\begin{aligned}x_s^{(1)}(t_0) = h_s^{(1)}\left(u^{(1)},\, v^{(1)}\right) \\ s = 1,2,\ldots,SN\end{aligned} \tag{4.1c}$$

$$x_s^{(p)}(t_{p-1}) = h_s^{(p)}\left(x_s^{(p-1)}(t_{p-1}), z_s^{(p-1)}(t_{p-1}), u^{(p)}, v^{(p)}\right)$$
$$p = 2, 3, \ldots, NP \tag{4.1d}$$
$$s = 1, 2, \ldots, SN$$

$$0 = g_s^{(p)}\left(x_s^{(p)}(t), z_s^{(p)}(t), u^{(p)}, v^{(p)}, t\right)$$
$$t_{p-1} \leq t \leq t_p$$
$$p = 1, 2, \ldots, NP \tag{4.1e}$$
$$s = 1, 2, \ldots, SN$$
$$\overline{C}^{(p)} \leq 0$$

where

$$\overline{C}^{(p)} = \frac{1}{SN}\left[\sum_{s=1}^{SN} c_s^{(p)}\left(x_s^{(p)}(t), z_s^{(p)}(t), u^{(p)}, v^{(p)}, t\right)\right] \tag{4.1f}$$
$$t_{p-1} \leq t \leq t_p$$
$$p = 1, 2, \ldots, NP$$

$$u^{(p)} \in [0, 1]$$
$$p = 1, 2, \ldots, NP \tag{4.1g}$$

$$v^{(p)} \in \mathscr{V}$$
$$p = 1, 2, \ldots, NP \tag{4.1h}$$

In this stochastic MSMIOCP formulation, it is seen how the feature of the integration and optimisation phases being independent of each other is used to advantage: the use of multiple scenarios leads to the size of the DAE system in the integration phase to increase proportionately compared to a single scenario case, but the number of control variables and constraints involved in the optimisation is the same as in a single scenario case. Hence, only a single optimisation problem has to be solved, which has the same number of decision variables and constraints compared to the deterministic problem, but with a larger DAE system compared to the latter.

A schematic describing the underlying principle of this formulation is shown in Figure 4.1. A similar principle has been used for optimisation under uncertainty in works by Al Ismaili et al. (2019) for scheduling of heat exchanger network cleaning operations and by Kanavalau et al. (2019) in model predictive control for batch process intensification.

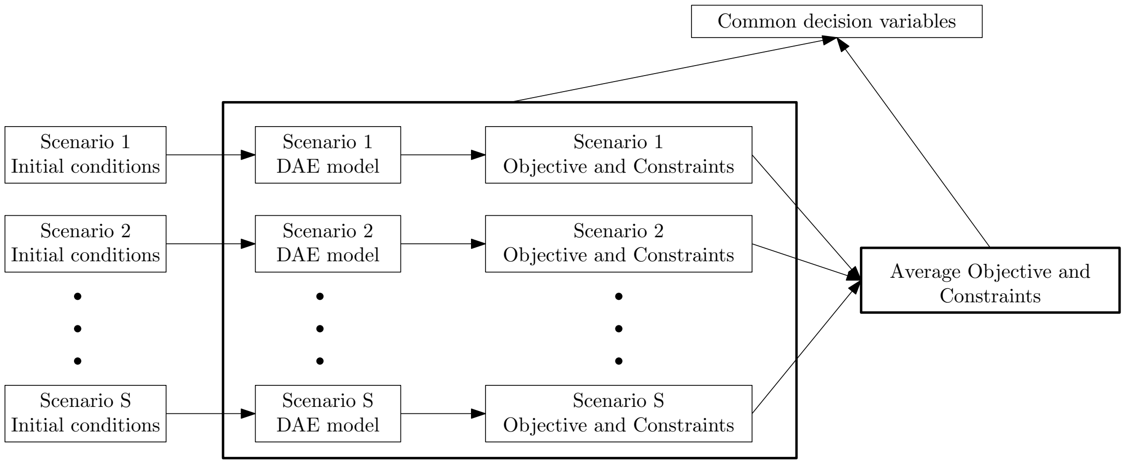


Fig. 4.1 The schematic of the principle of a stochastic optimal control formulation.

Problems of the form of equation (4.1) are essentially standard multistage optimal control problems and hence, standard nonlinear optimisation problems, as no discrete controls are involved and only an optimal set of continuous controls have to be identified, while using the feasible path approach to solve the differential equations. However, in order to obtain solutions equivalent to that of the original problem of the form of equation (2.4), wherein the controls, $u^{(p)}$, for each stage $p = 1, 2, \ldots NP$, take values of only 0 or 1, a penalty term homotopy technique, similar to that done in Section 2.4.3, is used. That is, a monotonically increasing penalty term is added to the objective function in equation (4.1a) and a series of standard nonlinear optimisation problems of the following generic form are solved:

$$Fs_k : \min \left\{ W_{st} + Ms_k \sum_{p=1}^{NP} u^{(p)} \left[1 - u^{(p)} \right] \right\} \tag{4.2}$$

subject to equations (4.1b) – (4.1h), for

$$k = 1, 2, 3 \ldots$$

$$Ms_1 = 0$$

As in Section 2.4.3, the problem, Fs_1, in the first major iteration ($k = 1$) of the series, is designated a weight $Ms_1 = 0$. An iterative procedure is followed wherein problem Fs_{k+1} is solved using the solution of problem Fs_k as initial guesses and with weight $Ms_{k+1} > Ms_k$. This iterative procedure is continued until iteration K ($K \geq 1$) such that all controls in u, in the solution of problem Fs_K, are forced by weight Ms_K to take values of either 0 or 1. Once again, the progression for the increase of weights, Ms_k, have to be chosen by trial and error, while ensuring that the weight is not increased too slowly or too fast, in order to avoid large computational times or an indefinite continuation of iterations, respectively.

Thus, the proposed solution procedure is a modified form of the solution procedure of Implementation II in Section 2.4.3. While the the penalty term homotopy technique in Implementation II was used to solve a deterministic MSMIOCP formulation, here, a similar penalty term homotopy technique is applied to solve a stochastic MSMIOCP formulation. Therefore, this optimal control methodology proposed to solve MSMIOCP formulations involving model-inherent parametric uncertainties will be referred to as a "stochastic MSMIOCP formulation in combination with a solution procedure of the principle of Implementation II". As will be seen, the implementation details of the proposed solution procedure are also similar to those used for Implementation II, which was shown to produce optimal solutions in Chapter 2. Therefore, the results obtained using such a solution procedure and implementation in this optimal control methodology can be expected to be optimal as well. The advantages that this optimal control methodology can potentially offer over the conventional methods of solving the type of problem under consideration in this chapter follow next.

4.2.1 Potential advantages over conventional methodologies

The preceding text presented the details of the optimal control methodology proposed to consider uncertainties in the kinetic parameters involved while optimising maintenance scheduling and production in a process containing a reactor using decaying catalysts. As mentioned in Section 4.1, the conventional methods of solving problems of this kind involve using one of the popular preventive methods of handling uncertainty in combination with mixed-integer optimisation methods. The proposed optimal control methodology's features, of using a stochastic MSMIOCP formulation and a solution procedure as a standard nonlinear optimisation problem using the feasible path approach, can provide potential advantages over the conventional methodologies of solving this problem by potentially overcoming the drawbacks introduced by the use of mixed-integer optimisation techniques in those methodologies. These potential advantages are similar to those mentioned in Section 2.5.1 in the conclusions of Chapter 2. However, these potential advantages are stated here, in the context of the mixed-integer techniques being used in combination with the popular preventive methods of handling uncertainty:

1. The practice in mixed-integer techniques of approximating differential equations as a collection of steady state equations, can cause the problem to end up containing a very large number of variables and constraints. This can lead to difficulties in convergence to optimal solutions, even for a deterministic problem. In addition, if a large number

catalyst loads are available to be used, the problem size becomes large and this can lead to further difficulties in convergence. Hence, when uncertainties are involved, and if conventional methodologies are used for solution, these difficulties can be further aggravated, especially if stochastic or fuzzy programming approaches are used.

However, in this formulation, by virtue of a feasible path approach being used in combination with a multiple scenario approach to consider uncertainties, although a large DAE system will be obtained, the number control actions to be applied and the number of constraints to be fulfilled will be the same as in a single scenario or a deterministic problem case. In addition, the use of the feasible path approach ensures that the differential equations are solved by an integrator without creating additional variables or constraints to be considered in the optimisation phase.
Further, by virtue of the penalty term homotopy technique, the 0 or 1 values for the controls to schedule catalyst changeover will be obtained inherently when solving this problem as a standard nonlinear optimisation problem, without the use of mixed-integer optimisation methods. Hence, even if an infinite number of catalyst loads are available, the problem size will not increase as the decisions on how many catalyst loads to use and when to schedule catalyst changeovers will be taken inherently during the optimisation.

Therefore, due to these features of the methodology, the number of variables and constraints will be considerably smaller than when existing preventive methods of handling uncertainty are applied to mixed-integer formulations, and obtaining solutions will be well within the scope of existing solvers, regardless of the initial guesses used or the number of scenarios considered. Hence, the methodology is expected to be more robust in obtaining solutions in comparison to the conventional methodologies.

2. In the feasible path approach, state-of-the-art integrators are used to solve nonlinear DAEs to a high accuracy. On the other hand, the mixed-integer formulations, when used with any of the existing preventive methods of handling uncertainty, approximate such DAEs as a collection of steady state equations which are solved as equality constraints in the optimisation, and this is likely to cause errors in the solution. Thus, the solutions obtained by this formulation are expected to be more reliable than those obtained by the conventional methodologies.

As was mentioned in Chapter 2, by enabling potential advantages of robustness and reliability, the use of the feasible path approach also provides a further potential advantage of avoiding making the difficult compromise between accuracy and ease of convergence, which would be faced when using the mixed-integer techniques in the conventional methodologies.

3. As mentioned previously, by virtue of the penalty term homotopy technique, regardless of the number of catalyst loads involved, the 0 or 1 values for the controls corresponding to the catalyst changeover actions in the problem under consideration here, will be decided inherently during the optimisation, without using mixed-integer techniques.

   Hence, no additional computational effort will be spent in deciding when to schedule catalyst changeovers, thereby underlining the potential efficiency of this methodology. In the event of a large number of catalyst loads being involved, this feature will enable saving the substantial amount of computational effort which would be required in the huge number of combinations to be considered when mixed-integer formulations are used with the any of the existing preventive methods of handling uncertainty.

However, in this formulation, since the number of DAEs scales with the number of scenarios involved, if a large number of scenarios are considered, as would be needed to sufficiently take uncertainty into account, the number of DAEs in the formulation would become very large. The use of the feasible path approach implies considerable computational effort will be spent in solving each such equation to a high accuracy in each iteration of the optimisation, even in those iterations away from the optimal solution. Hence, the computational time is expected to be large when using this methodology, considerably larger than the methodologies used to solve the deterministic problems in Chapters 2 and 3.

While this can be perceived as a drawback of this methodology, it is highlighted that the scalability of the formulation can be exploited to reduce computation time. Using high performance computing facilities, each DAE set can be simulated entirely on a separate computer. Further, using parallel computing facilities, any required gradient evaluations can be parallelised within the computer on which each simulation occurs.

Another issue is that, since the problem is non-convex, only local optima can be obtained by this methodology. Thus, several runs using different start points have to be performed to identify the global optimum. The high performance and parallel computing facilities would make such a task feasible as well.

The preceding discussion suggests that the proposed methodology has the potential to effectively solve the problem of optimisation of maintenance scheduling and production in an industrial process using decaying catalysts while considering uncertainties in the kinetic parameters of the underlying process model, and can potentially overcome the major drawbacks that would be introduced by mixed-integer optimisation methods in the conventional methodologies if these methodologies are used to solve this problem.

In essence, the optimal control methodology, termed stochastic MSMIOCP formulation in combination with a solution procedure of the principle of Implementation II, that is proposed to solve this problem can be summarised as follows. First, the industrial process under consideration is formulated as a basic MSMIOCP of the form of equation (2.4) which is then converted into an MSMIOCP with relaxed binary controls of the form of equation (2.61). Using a multiple scenario approach to consider uncertainties in the kinetic parameters, this formulation as an MSMIOCP with relaxed binary controls is converted to a stochastic MSMIOCP with relaxed binary controls of the form of equation (4.1), to which a penalty term homotopy technique, as per an implementation similar to in Section 2.4.3, is applied to attain binary values for the catalyst changeover controls. In the next set of sections, the proposed methodology is applied to solve different case studies of such a problem.

4.3 Case studies: Problem formulation

The industrial process under consideration in this chapter is similar to the single reactor process considered in Chapter 2. The form of kinetics for the catalyst deactivation and the product formation reaction in this process correspond to that of Case Study A in Chapter 2, with the only difference here being that there is uncertainty regarding the values of the kinetic parameters present in the rate equations for these reactions. Different case studies are examined in this chapter, in order to analyse the effects on the optimal operation of each uncertain parameter individually as well as an analysis that considers the effect of uncertainty in all parameters simultaneously alongside the effect of the number of scenarios generated.

This section focuses on the problem formulation for these case studies. As per the proposed methodology, first, the industrial process is formulated as an MSMIOCP of the form of equation (2.4) which is then converted into an MSMIOCP with relaxed binary controls of the form of equation (2.61). As something similar was done for the industrial process considered in Chapter 2, and since the process considered in this chapter is similar to the process considered in Chapter 2, the formulation as an MSMIOCP with relaxed binary controls for Case Study A in Section 2.2 of Chapter 2 will be used for the construction of the similar formulation here. The terminology to used for this formulation is also similar to the the terminology used in that section, with the exception that a subscript, *un*, is added to each symbol to indicate that this is a formulation belonging to the "uncertainty" study.

The assumptions involved in constructing this formulation are similar to those mentioned in Section 2.2, with the exception that there is uncertainty here regarding the values of the kinetic parameters involved. Rather than be regarded as fixed constants, each of the kinetic parameters of the catalyst deactivation rate constant, Kd_{un}, and the pre-exponential factor, Ar_{un}, and the activation energy, Ea_{un}, for the main reaction is considered as taking values over a certain range. However, the mean of the range of values for each of these uncertain parameters is assumed to be known, and the known mean values for the parameters of Kd_{un}, Ar_{un} and Ea_{un} are designated symbols $\overline{Kd_{un}}$, $\overline{Ar_{un}}$ and $\overline{Ea_{un}}$, respectively.

As in Section 2.2, the controls of this formulation include the catalyst changeover decision variables, $y_{un}(i)$, for each month, $i \in \{1,2,...,NM_{un}\}$ and the feed flow rate, $ffr_{un}(i,j)$, the temperature, $T_{un}(i,j)$, and the sales, $sales_{un}(i,j)$, for each week $j \in \{1,2,3,4\}$ of each month $i \in \{1,2,...,NM_{un}\}$ of the process time horizon.

However, unlike in Section 2.2, the state variables in the formulation here do not include the catalyst age and the only state variables considered are the catalyst activity, cat_act_{un}, the reactant exit concentration, cR_{un}, the inventory level, inl_{un}, and the cumulatve inventory cost, cum_inc_{un}. As the differential equation (2.19) for the catalyst age in Section 2.2 does not directly or indirectly involve any parameters considered uncertain in this chapter, that state variable is omitted in this study. Therefore, there will be no differential equation, initial and junction conditions pertaining to the catalyst age in the formulation here. However, the differential equations, initial and junction conditions for the other state variables here are formulated similar to those of the corresponding variables in Section 2.2.

The ODEs for the state variables in this formulation, for each week $j \in \{1,2,3,4\}$ of each month $i \in \{1,2,...,NM_{un}\}$ of the process time horizon, are given by equations (4.3) – (4.7) and these are formulated according to a similar logic used to develop equations (2.20), (2.24), (2.28), (2.29) and (2.30), respectively. The kinetic rate equations for the catalyst deactivation and the product formation reaction corresponding to Case Study A, of the form of equations (2.21) and (2.25), respectively, have been directly inserted into the ODEs, as has an Arrhenius expression, of the form of equation (2.27), for the rate constant of the product formation reaction.

$$\frac{\mathrm{d}\left(cat\_act_{un}\right)}{\mathrm{d}t} = y_{un}(i) \times \left[-Kd_{un} \times cat\_act_{un}\right] \tag{4.3}$$

$$\begin{aligned}\frac{\mathrm{d}\left(V_{un} \times cR_{un}\right)}{\mathrm{d}t} &= ffr_{un}(i,j) \times \left(CR0_{un} - cR_{un}\right)\\ &\quad - y_{un}(i) \times \left[V_{un} \times Ar_{un} \times \exp\left(-\frac{Ea_{un}}{R_g \times T_{un}(i,j)}\right) \times cat\_act_{un} \times cR_{un}\right]\end{aligned} \tag{4.4}$$

$$\frac{\mathrm{d}\left(inl_{un}\right)}{\mathrm{d}t} = y_{un}(i) \times \left[V_{un} \times Ar_{un} \times \exp\left(-\frac{Ea_{un}}{R_g \times T_{un}(i,j)}\right) \times cat\_act_{un} \times cR_{un}\right] \tag{4.5}$$

$$\frac{\mathrm{d}\left(cum\_inc_{un}\right)}{\mathrm{d}t} = inl_{un} \times icf_{un} \tag{4.6}$$

$$icf_{un} = base\_icf_{un} \times \left(1 + inflation_{un}\right)^{\lfloor i/12 \rfloor} \tag{4.7}$$

The initial conditions for the solutions of the ODEs for these state variables, in week 1 of month 1 of the time horizon, are given by equations (4.8) – (4.11), and these have been derived as per reasonings similar to the derivation of the initial conditions (2.32) – (2.35), respectively in Section 2.2.

$$init\_cat\_act_{un}(1,1) = start\_cat\_act_{un} \tag{4.8}$$

$$init\_cR_{un}(1,1) = CR0_{un} \tag{4.9}$$

$$init\_inl_{un}(1,1) = 0 \tag{4.10}$$

$$init\_cum\_inc_{un}(1,1) = 0 \tag{4.11}$$

The junction conditions that link the state variables between any two consecutive weeks are given by equations (4.12) – (4.15) and these have been formulated along the lines of junction conditions (2.37) – (2.40), respectively in Section 2.2.

$$\begin{gathered} init\_cat\_act_{un}(i, j+1) = end\_cat\_act_{un}(i, j) \\ j = 1,2,3 \qquad i = 1,2,\ldots,NM_{un} \end{gathered} \tag{4.12a}$$

$$init\_cat\_act_{un}(i,\,1) \;=\; [y_{un}(i) \times end\_cat\_act_{un}(i-1,\,4)] + [(1-y_{un}(i)) \times start\_cat\_act_{un}]$$
$$i = 2,3,\ldots,NM_{un} \tag{4.12b}$$

$$\begin{aligned} init\_cR_{un}(i,\,j+1) \;&=\; end\_cR_{un}(i,\,j) \\ j = 1,2,3 \qquad & i = 1,2,\ldots,NM_{un} \end{aligned} \tag{4.13a}$$

$$\begin{gathered} init\_cR_{un}(i,\,1) = [y_{un}(i) \times end\_cR_{un}(i-1,\,4)] + [(1-y_{un}(i)) \times CR0_{un}] \\ i = 2,3,\ldots,NM_{un} \end{gathered} \tag{4.13b}$$

$$\begin{aligned} init\_inl_{un}(i,\,j+1) \;&=\; end\_inl_{un}(i,\,j) - sales_{un}(i,\,j) \\ j = 1,2,3 \qquad & i = 1,2,\ldots,NM_{un} \end{aligned} \tag{4.14a}$$

$$\begin{gathered} init\_inl_{un}(i,\,1) \;=\; end\_inl_{un}(i-1,\,4) - sales_{un}(i-1,\,4) \\ i = 2,3,\ldots,NM_{un} \end{gathered} \tag{4.14b}$$

$$\begin{aligned} init\_cum\_inc_{un}(i,\,j+1) \;&=\; end\_cum\_inc_{un}(i,\,j) \\ j = 1,2,3 \qquad & i = 1,2,\ldots,NM_{un} \end{aligned} \tag{4.15a}$$

$$\begin{gathered} init\_cum\_inc_{un}(i,\,1) \;=\; end\_cum\_inc_{un}(i-1,\,4) \\ i = 2,3,\ldots,NM_{un} \end{gathered} \tag{4.15b}$$

The constraints that apply in this formulation, for each week $j \in \{1,2,3,4\}$ of each month $i \in \{1,2,...,NM_{un}\}$ of the process time horizon, are given in equations (4.16) – (4.24). Constraints (4.16) – (4.23) have been formulated as per reasonings similar to formulation of the constraints (2.41) – (2.48), respectively in Section 2.2. However, constraint (4.24) has been exclusively derived for this formulation and the explanation for its derivation is as follows. In order to prevent the productivity of the process from becoming too low, any of the decaying catalysts in this process is not recommended to be used beyond when its age crosses a certain maximum value or when its activity falls below a certain minimum value. In Section 2.2, as a decision was required to be made on whether to replace a catalyst or not on a monthly basis, constraint (2.49) was used to enforce that any catalyst's age does not cross a stipulated maximum value (max_cat_age) at the end of each month. However, the catalyst age is not considered as a state variable in the formulation here. As this formulation also requires making decisions on a monthly basis as to whether to replace a catalyst or not, constraint (4.24), which enforces that the catalyst activity does not fall below a specified minimum value ($min\_cat\_act_{un}$) at the end of each month, i, is used in place of constraint (2.49) in the formulation here.

$$0 \leq y_{un}(i) \leq 1 \tag{4.16}$$

$$0 \leq ffr_{un}(i,j) \leq FU_{un} \tag{4.17}$$

$$0 \leq sales_{un}(i,j) \leq demand_{un}(i,j) \tag{4.18}$$

$$TL_{un} \leq T_{un}(i,j) \leq TU_{un} \tag{4.19}$$

$$ffr_{un}(i,j) - [FU_{un} \times y_{un}(i)] \leq 0 \tag{4.20}$$

$$TL_{un} \leq T_{un}(i,j) \leq [(TU_{un} - TL_{un}) \times y_{un}(i)] + TL_{un} \tag{4.21}$$

$$\sum_{i=1}^{NM_{un}} y_{un}(i) \geq NM_{un} - n_{un} \tag{4.22}$$

$$end\_inl_{un}(i,\, j) - sales_{un}(i,j) \geq 0 \tag{4.23}$$

$$end\_cat\_act_{un}(i,4) \geq min\_cat\_act_{un} \tag{4.24}$$

And finally the elements of the objective function and the objective function itself in this formulation are given in equations (4.25) – (4.35), wherein $j \in \{1,2,3,4\}$ and $i \in \{1,2,...,NM_{un}\}$, and these have been derived as per a logic similar to the derivation of equations (2.50) – (2.60) in Section 2.2.

$$GRS_{un} = \sum_{i=1}^{NM_{un}} \sum_{j=1}^{4} psp_{un}(i,j) \times sales_{un}(i,j) \tag{4.25}$$

$$psp_{un}(i,j) = base\_psp_{un} \times (1 + inflation_{un})^{\lfloor i/12 \rfloor} \tag{4.26}$$

$$TIC_{un} = end\_cum\_inc_{un}(NM_{un},4) \tag{4.27}$$

$$TCCC_{un} = \sum_{i=1}^{NM_{un}} crc_{un}(i) \times (1 - y_{un}(i)) \tag{4.28}$$

$$crc_{un}(i) = base\_crc_{un} \times (1 + inflation_{un})^{\lfloor i/12 \rfloor} \tag{4.29}$$

$$unmet\_demand_{un}(i,j) = demand_{un}(i,j) - sales_{un}(i,j) \tag{4.30}$$

$$NPUD_{un} = \sum_{i=1}^{NM_{un}} \sum_{j=1}^{4} pen_{un}(i,j) \times unmet\_demand_{un}(i,j) \tag{4.31}$$

$$pen_{un}(i,j) = base\_pen_{un} \times (1 + inflation_{un})^{\lfloor i/12 \rfloor} \tag{4.32}$$

$$TFC_{un} = \sum_{i=1}^{NM_{un}} \sum_{j=1}^{4} cof_{un}(i,j) \times ffr_{un}(i,j) \tag{4.33}$$

$$cof_{un}(i,j) \quad = \quad base\_cof_{un} \times (1 + inflation_{un})^{\lfloor i/12 \rfloor} \tag{4.34}$$

$$\min NC_{un} \quad = \quad -GRS_{un} + TIC_{un} + TCCC_{un} + NPUD_{un} + TFC_{un} \tag{4.35}$$

This concludes the formulation of the industrial process as an MSMIOCP with relaxed binary controls. This formulation applies to all of the following case studies of the industrial process that will be examined in this chapter:

Case Study E: Effect of uncertainty in the catalyst deactivation rate constant, Kd_{un} on the optimal maintenance schedule and production operations

Case Study F: Effect of uncertainty in the pre-exponential factor, Ar_{un}, of the product formation reaction, on the optimal maintenance schedule and production operations

Case Study G: Effect of uncertainty in the activation energy, Ea_{un}, of the product formation reaction, on the optimal maintenance schedule and production operations

Case Study H: Parametric uncertainty study that considers the effect of simultaneous uncertainty in all kinetic parameters and the number of scenarios generated on the optimal maintenance schedule and production operations

The formulation as an MSMIOCP with relaxed binary controls, that applied for all of the above case studies, presented in the preceding text, represented the first step of the use of the proposed optimal control methodology to solve each of these case studies. As per the next step of this methodology, using this preceding formulation, a formulation as a stochastic MSMIOCP with relaxed binary controls, of the form of equation (4.1), is developed for each of these case studies. In the following text, such a formulation is developed for Case Study H, wherein all kinetic parameters are considered uncertain. Similar formulations for Case Studies E, F and G are derived from minor modifications of this formulation, and these modifications will be detailed later.

As mentioned in Section 4.2, this formulation incorporates parametric uncertainty by generating multiple scenarios, with each uncertain parameter taking a particular value within a pre-specified range about its mean value in each scenario. Here, the total number of scenarios generated is given by NS. The kinetic parameters of the catalyst deactivation rate constant, the pre-exponential factor and the activation energy of the product formation reaction generated in scenario $s \in \{1,2,\ldots,NS\}$, of this stochastic MSMIOCP formulation are represented by $\widetilde{Kd^s_{un}}$, $\widetilde{Ar^s_{un}}$ and $\widetilde{Ea^s_{un}}$, respectively and these are the counterparts of Kd_{un}, Ar_{un} and Ea_{un}, respectively, in the formulation as an MSMIOCP with relaxed binary controls.

Following from Section 4.2, the decision variables in this stochastic formulation remain the same as in the formulation as an MSMIOCP with relaxed binary controls and so the same terminology as in the latter formulation, applies here for the decision variables. The catalyst changeover decision variables, $y_{un}(i)$, for each month $i \in \{1, 2, \ldots, NM_{un}\}$, corresponds to the binary control u in equation (4.1g). The decisions of the amount of reactor feed flow rate, $ffr_{un}(i, j)$, temperature of operation, $T_{un}(i, j)$ and quantity of sales, $sales_{un}(i, j)$ for each week, $j \in \{1, 2, 3, 4\}$, of each month $i \in \{1, 2, \ldots, NM_{un}\}$, correspond to the continuous control v in equation (4.1h).

However, as per Section 4.2, the unique kinetic parameter values in each scenario lead to a corresponding unique set of state variables and their deriving ODE systems, comprised of ODEs, initial conditions and junction conditions. The state variables of catalyst activity, reactant exit concentration, inventory level and cumulative inventory costs attained in scenario $s \in \{1, 2, \ldots, NS\}$, are represented by the symbols $\widetilde{cat\_act^s_{un}}$, $\widetilde{cR^s_{un}}$, $\widetilde{inl^s_{un}}$ and $\widetilde{cum\_inc^s_{un}}$, respectively, and these are the counterparts of cat_act_{un}, cR_{un}, inl_{un} and cum_inc_{un}, respectively, in the formulation as an MSMIOCP with relaxed binary controls. The ODE systems comprising the ODEs, initial conditions and junctions conditions, for each of these state variables, in each scenario $s \in \{1, 2, \ldots, NS\}$ are presented next. The explanations for their formulation are similar to those of their MSMIOCP counterparts.

The ODEs for each state variable in each scenario $s \in \{1, 2, \ldots, NS\}$, of the form of equation (4.1b), are given by equations (4.36) – (4.39) and these are the counterparts of ODEs (4.3), – (4.6), respectively, in the formulation as an MSMIOCP with relaxed binary controls. These ODEs are to be solved repeated over a weekly time span, over each week $j \in \{1, 2, 3, 4\}$, of month $i \in \{1, 2, ..., NM_{un}\}$ of the time horizon.

$$\frac{\mathrm{d}\left(\widetilde{cat\_act^s_{un}}\right)}{\mathrm{d}t} = y_{un}(i) \times \left[-\widetilde{Kd^s_{un}} \times \widetilde{cat\_act^s_{un}}\right] \tag{4.36}$$

$$\begin{aligned}\frac{\mathrm{d}\left(V_{un}\times\widetilde{cR^s_{un}}\right)}{\mathrm{d}t} &= ffr_{un}(i,j)\times\left(CR0_{un}-\widetilde{cR^s_{un}}\right)\\ &-y_{un}(i)\times\left[V_{un}\times\widetilde{Ar^s_{un}}\times\exp\left(-\frac{\widetilde{Ea^s_{un}}}{R_g\times T_{un}(i,j)}\right)\times\widetilde{cat\_act^s_{un}}\times\widetilde{cR^s_{un}}\right]\end{aligned} \tag{4.37}$$

$$\frac{\mathrm{d}\left(\widetilde{inl^s_{un}}\right)}{\mathrm{d}t} = y_{un}(i)\times\left[V_{un}\times\widetilde{Ar^s_{un}}\times\exp\left(-\frac{\widetilde{Ea^s_{un}}}{R_g\times T_{un}(i,j)}\right)\times\widetilde{cat\_act^s_{un}}\times\widetilde{cR^s_{un}}\right] \tag{4.38}$$

$$\frac{\mathrm{d}\left(\widetilde{cum\_inc^s_{un}}\right)}{\mathrm{d}t} = \widetilde{inl^s_{un}}\times icf_{un} \tag{4.39}$$

where icf_{un} is given by equation (4.7).

The initial conditions for week 1 of month 1 of the time horizon, for each of the state variables in each scenario $s\in\{1,2,\ldots,NS\}$, of the form of equation (4.1c), are given by equations (4.40) – (4.43) and are the counterparts of initial conditions (4.8) – (4.11), respectively, in the formulation as an MSMIOCP with relaxed binary controls:

$$init\_\widetilde{cat\_act^s_{un}}(1,1) \;=\; start\_cat\_act_{un} \tag{4.40}$$

$$init\_\widetilde{cR^s_{un}}(1,1) \;=\; CR0_{un} \tag{4.41}$$

$$init\_\widetilde{inl^s_{un}}(1,1) \;=\; 0 \tag{4.42}$$

$$init\_\widetilde{cum\_inc^s_{un}}(1,1) \;=\; 0 \tag{4.43}$$

The junction conditions that link the state variable values between any two consecutive weeks in each scenario $s\in\{1,2,\ldots,NS\}$, of the form of equation (4.1d), are given by equations (4.44) – (4.47) and these are the counterparts of the junction conditions (4.12) – (4.15), respectively, in the formulation as an MSMIOCP with relaxed binary controls:

$$\begin{gathered}init\_\widetilde{cat\_act^s_{un}}(i,\,j+1) \;=\; end\_\widetilde{cat\_act^s_{un}}(i,\,j)\\ j=1,2,3 \qquad i=1,2,\ldots,NM_{un}\end{gathered} \tag{4.44a}$$

$$\begin{aligned}init\_\widetilde{cat\_act^s_{un}}(i,\,1) \;&=\; \left[y_{un}(i)\times end\_\widetilde{cat\_act^s_{un}}(i-1,\,4)\right]\\ &+\left[(1-y_{un}(i))\times start\_cat\_act_{un}\right]\end{aligned} \tag{4.44b}$$

$$i = 2,3,\ldots,NM_{un}$$

$$\begin{array}{c} init\_\widetilde{cR^s_{un}}(i,\, j+1) \;=\; end\_\widetilde{cR^s_{un}}(i,\, j) \\ j = 1,2,3 \qquad i = 1,2,\ldots,NM_{un} \end{array} \tag{4.45a}$$

$$\begin{array}{c} init\_\widetilde{cR^s_{un}}(i,\, 1) = \left[y_{un}(i) \times end\_\widetilde{cR^s_{un}}(i-1,\, 4)\right] + \left[(1-y_{un}(i)) \times CR0_{un}\right] \\ i = 2,3,\ldots,NM_{un} \end{array} \tag{4.45b}$$

$$\begin{array}{c} init\_\widetilde{inl^s_{un}}(i,\, j+1) \;=\; end\_\widetilde{inl^s_{un}}(i,\, j) - sales_{un}(i,\, j) \\ j = 1,2,3 \qquad i = 1,2,\ldots,NM_{un} \end{array} \tag{4.46a}$$

$$\begin{array}{c} init\_\widetilde{inl^s_{un}}(i,\, 1) \;=\; end\_\widetilde{inl^s_{un}}(i-1,\, 4) - sales_{un}(i-1,\, 4) \\ i = 2,3,\ldots,NM_{un} \end{array} \tag{4.46b}$$

$$\begin{array}{c} init\_\widetilde{cum\_inc^s_{un}}(i,\, j+1) \;=\; end\_\widetilde{cum\_inc^s_{un}}(i,\, j) \\ j = 1,2,3 \qquad i = 1,2,\ldots,NM_{un} \end{array} \tag{4.47a}$$

$$\begin{array}{c} init\_\widetilde{cum\_inc^s_{un}}(i,\, 1) \;=\; end\_\widetilde{cum\_inc^s_{un}}(i-1,\, 4) \\ i = 2,3,\ldots,NM_{un} \end{array} \tag{4.47b}$$

The constraints in this stochastic formulation, of the form of equation (4.1f), are formulated on the basis that only the averages of the sets of constraints generated over all scenarios are required to be fulfilled. Among the constraints in the formulation as an MSMIOCP with relaxed binary controls, given by (4.16) – (4.24), only constraints (4.23) and (4.24) are influenced by the generation of multiple scenarios. Hence, in this stochastic formulation, constraints (4.16) – (4.22) apply as in the formulation as an MSMIOCP with relaxed binary controls, while constraints (4.23) and (4.24) are modified into their stochastic counterparts, given by equations (4.48) and (4.49), respectively:

$$\frac{1}{NS}\left[\sum_{s=1}^{NS} end\_\widetilde{inl^s_{un}}(i,\, j)\right] - sales_{un}(i,j) \geq \;\; 0 \tag{4.48}$$

$$\frac{1}{NS}\left[\sum_{s=1}^{NS} end\_\widetilde{cat\_act^s_{un}}(i,\, 4)\right] \geq \;\; min\_cat\_act_{un} \tag{4.49}$$

And finally, the objective function of this formulation, of the form of equation (4.1a), is formulated. It is highlighted that among the components of the objective function in the formulation as an MSMIOCP with relaxed binary controls (equations (4.25) – (4.34)), only the net inventory cost, given by equation (4.27), is influenced by the generation of multiple scenarios and its counterpart in the stochastic formulation is given by equation (4.50):

$$\begin{aligned} \widetilde{TIC^s_{un}} &= \widetilde{end\_cum\_inc^s_{un}}(NM, 4) \\ s &= 1, 2, \ldots, NS \end{aligned} \tag{4.50}$$

As per the proposed solution methodology, only the average of the net inventory costs over all scenarios will be included in the objective function of the stochastic formulation. The remaining components of this objective function are the same as those given by (4.25), (4.26) and (4.28) – (4.34) in the formulation as an MSMIOCP with relaxed binary controls, as these are not influenced by the generation of multiple scenarios. Thus, the objective function in this stochastic formulation, which is the counterpart of (4.35), is given by equation (4.51):

$$\min NC_{st} = -GRS_{un} + \left[\frac{1}{NS} \left(\sum_{s=1}^{NS} \widetilde{TIC^s_{un}} \right) \right] + TCCC_{un} + NPUD_{un} + TFC_{un} \tag{4.51}$$

where NC_{st} is the net costs in this stochastic formulation.

This concludes the formulation of the stochastic MSMIOCP with relaxed binary controls for Case Study H, wherein all kinetic parameters are considered uncertain. The formulation as a stochastic MSMIOCP with relaxed binary controls to be used for Case Study E is largely similar to that of Case Study H with the exception that because Kd_{un} is the only uncertain parameter involved, the pre-exponential factor and activation energy values in the formulation are fixed to their mean values of $\overline{Ar_{un}}$ and $\overline{Ea_{un}}$, respectively, which are assumed to be known. That is, the formulation as a stochastic MSMIOCP with relaxed binary controls for Case Study E is a special case of that for Case Study H, wherein $\widetilde{Ar^s_{un}} = \overline{Ar_{un}}, \forall s \in \{1, 2, \ldots, NS\}$ and $\widetilde{Ea^s_{un}} = \overline{Ea_{un}}, \forall s \in \{1, 2, \ldots, NS\}$.

Similarly, in Case study F where Ar_{un} is the only uncertain parameter, the formulation as a stochastic MSMIOCP with relaxed binary controls is a special case of that for Case Study H, wherein $\widetilde{Kd^s_{un}} = \overline{Kd_{un}}, \forall s \in \{1, 2, \ldots, NS\}$ and $\widetilde{Ea^s_{un}} = \overline{Ea_{un}}, \forall s \in \{1, 2, \ldots, NS\}$. And, in Case Study G where Ea_{un} is the only uncertain parameter, the formulation as a stochastic MSMIOCP with relaxed binary controls is a special case of that for Case Study H, wherein $\widetilde{Kd^s_{un}} = \overline{Kd_{un}}, \forall s \in \{1, 2, \ldots, NS\}$ and $\widetilde{Ar^s_{un}} = \overline{Ar_{un}}, \forall s \in \{1, 2, \ldots, NS\}$.

The set of parameters that apply to all case studies is given in Table 4.1. These are mostly similar to the parameters used in Chapter 2, wherein the deterministic version of this problem was considered. The values used for the pre-exponential factor and the activation energy of the product formation reaction, and the catalyst deactivation rate constant corresponding to Case Study A in the deterministic study in Chapter 2 are used as the mean values of those parameters which are considered uncertain in this chapter. The minimum allowable catalyst activity ($min\_cat\_act_{un}$) is set to a value that is attained after a duration of 1.5 years (which was the maximum catalyst age value used in Chapter 2) for the form of catalyst deactivation kinetics considered in this chapter. Details of the implementation to obtain solutions for each of these case studies are presented in the next section.

Table 4.1 List of parameters.

Parameter Symbol	**Value**
$\overline{Ar_{un}}$	885 (1/day)
$base\_cof_{un}$	\$ 210 /week
$base\_crc_{un}$	\$ 10^7
$base\_icf_{un}$	\$ 0.01 /(kmol day)
$base\_pen_{un}$	\$ 1250 /kmol
$base\_psp_{un}$	\$ 1000 /kmol
$CR0_{un}$	1 kmol/m^3
$demand_{un}$	1st quarter of year: 8000 kmol/week
	2nd quarter of year: 7200 kmol/week
	3rd quarter of year: 3300 kmol/week
	4th quarter of year: 4500 kmol/week
$\overline{Ea_{un}}$	30,000 J/gmol
FU_{un}	9600 m^3/day
$inflation_{un}$	5%
$\overline{Kd_{un}}$	0.0024 (1/day)
$min\_cat\_act_{un}$	0.2983
n_{un}	5
NM_{un}	36 months (= 3 years)

Table 4.1 List of parameters.

Parameter Symbol	**Value**
R_g	8.314 J/(gmol.K)
$start\_cat\_act_{un}$	1.0
TL_{un}	400 K
TU_{un}	1000 K
V_{un}	50 m^3

4.4 Implementation details

The impact of parametric uncertainty in all case studies is analysed by solving sub-problems within each case study. That is, the results of all sub-problems of the case study together represent the impact of the considered parametric uncertainty in that case study. Depending on the case study, the sub-problems differed either in the range of values considered for the uncertain parameter or the number of scenarios. The details of the sub-problems investigated in each case study are as follows:

Case Study E: To analyse the effect of uncertainty in Kd_{un}, three sub-problems are examined, that consider values for this parameter in the ranges of 10%, 20% and 30% relative standard deviations (RSDs) around the mean, $\overline{Kd_{un}}$. In each of these sub-problems, $NS = 20$ scenarios are sampled uniformly from the respective ranges.

Case Study F: To analyse the effect of uncertainty in Ar_{un}, three sub-problems are examined, that consider values for this parameter in the ranges of 10%, 20% and 30% RSDs around the mean, $\overline{Ar_{un}}$. In each of these sub-problems, $NS = 20$ scenarios are sampled uniformly from the respective ranges.

Case Study G: To analyse the effect of uncertainty in Ea_{un}, three sub-problems are examined, that consider values for this parameter in the ranges of 5%, 7.5% and 10% RSDs around the mean, $\overline{Ea_{un}}$. As Ea_{un} appears within an exponential function in the model, smaller ranges are considered in the sub-problems here compared to the parameters in Case Studies E and F. For each of these sub-problems, $NS = 20$ scenarios are sampled uniformly from the respective ranges.

Case Study H: In this parametric uncertainty study the effects of simultaneous uncertainty in all kinetic parameters and the impact of the number of scenarios generated are investigated. Five sub-problems are examined that consider NS values of 5, 10, 15, 20 and 25. For each of these sub-problems, the respective number of scenarios are sampled uniformly from only one range of values for each uncertain parameter: 10% RSD around the mean, $\overline{Kd_{un}}$, for Kd_{un}, 10% RSD around the mean, $\overline{Ar_{un}}$, for Ar_{un} and 5% RSD around the mean, $\overline{Ea_{un}}$, for Ea_{un}.

In addition, a deterministic (single scenario) run is carried out using values of $\overline{Kd_{un}}$, $\overline{Ar_{un}}$ and $\overline{Ea_{un}}$ for Kd_{un}, Ar_{un} and Ea_{un}, respectively. This deterministic run is very similar to what was done in Chapter 2 for Case Study A using Implementation II, with the exception that as there is no state variable of catalyst age present, there is no ODE, initial or junction condition involving that state variable and the limit on the maximum duration of catalyst use is imposed using a constraint on the catalyst activity, of the form of equation (4.24), in place of a constraint on the catalyst age, of the form of equation (2.49). It is intended to analyse the effects of uncertainty in the kinetic parameters by comparing the results of the sub-problems of all case studies with the solution of the deterministic run.

For the chosen time horizon of 3 years, Table 4.2 provides details of the number of decisions variables and constraints, which as mentioned previously, remain the same size as in the deterministic (single scenario) case, for all sub-problems of all case studies considered here. It is only the size of the ODE system that varies between the deterministic run and the case studies, and details regarding this, for the time horizon of 3 years, are given in Table 4.3.

For each sub-problem in each case study, the implementation was performed in Python™ 3.7.1 under PyCharm 2019.3.3 (Community Edition). The multiple scenario values for each uncertain parameter in each sub-problem were generated by constructing Sobol sequences (Sobol, 1976), of the length of the number of scenarios, within the specified range for that uncertain parameter. Using quasi-random low-discrepancy Sobol sequences provided the advantage of ensuring that the range of uncertainty is covered evenly. The Sobol sequences were generated using the *i4_sobol_generate* method in the *sobol_seq* module (version 0.1.2) in Python. The notion that a uniform sampling of values from the specified ranges of uncertainty would aid a more reliable consideration of uncertainty was the motivation behind using Sobol sequences to generate samples, but it noted that any other uniform sampling

Table 4.2 Size specifications for the decision variables and constraints, applicable for the deterministic problem as well as all sub-problems of all case studies.

Property		**Size**
Decision variables	Catalyst changeover actions (y_{un})	36
	Feed flow rate (ffr_{un})	144
	Sales ($sales_{un}$)	144
	Temperature (T_{un})	144
	Total	*468*
Constraints	Constraints (4.16)	72
	Constraints (4.17)	288
	Constraints (4.18)	288
	Constraints (4.19)	288
	Constraints (4.20)	144
	Constraints (4.21)	288
	Constraint (4.22)	1
	Constraints (4.48)	36
	Constraints (4.49)	144
	Total	*1549*

method can also fulfil this purpose.

The stochastic MSMIOCP formulated in each sub-problem of each case study, was solved as standard nonlinear optimisation problem using the feasible path approach, by applying the penalty term homotopy technique, of the form of equation (4.2), and solving the following series of standard multistage optimal control problems:

$$Gs_k : \min \left\{ NC_{st} + Ms_k \sum_{i=1}^{NM_{un}} y_{un}(i)\left[1 - y_{un}(i)\right] \right\} \tag{4.52}$$

subject to the ODEs, initial conditions, junction conditions and constraints of the respective case study, which were presented in Section 4.3, for

$$k = 1, 2, 3 \ldots$$
$$Ms_1 = 0$$

Table 4.3 Details of the size of the ODE systems present in the deterministic run and the sub-problems of all case studies.

Case Study	**Sub-problem**	**Number of ODEs**
Deterministic run	N/A	576
Case Study E	All sub-problems	11520
Case Study F	All sub-problems	8784
Case Study G	All sub-problems	8784
Case Study H	5 scenario sub-problem	2880
	10 scenario sub-problem	5760
	15 scenario sub-problem	8640
	20 scenario sub-problem	11520
	25 scenario sub-problem	14400

In accordance with this procedure, if in the solution of problem Gs_k, the condition, $y_{un}(i) \in \{0,1\}$ for $i = 1,2,\ldots NM_{un}$, does not apply, then problem Gs_{k+1} is solved using the solution of Gs_k as initial guesses, with weight $Ms_{k+1} > Ms_k$. The weight term, Ms_k, is increased as per the arithmetic progression given by equation (4.53). This progression is similar to the progression of increase of weights in equation (2.64) in Chapter 2, and is chosen as such because the parameters used here are similar to those used in Chapter 2.

$$\begin{aligned} Ms_{k+1} &= (2 \times Ms_k) + \left(5 \times 10^7\right) \\ Ms_1 &= 0 \\ k &= 1,2,3\ldots \end{aligned} \tag{4.53}$$

For the deterministic run, as well as for all sub-problems of all case studies, the initial guesses for the decision variables were set to their respective upper bounds. The same initial guesses were used for all these runs in order to facilitate a comparison between the results of these runs.

The implementation of the solution procedure for each sub-problem was chosen similar to that of Implementation II in Section 2.4.3, which produced optimal solutions. That is, CasADi in Python was once again used to write the code, with the elements of the series of problems given by equation (4.52), for each sub-problem of each case study, defined as symbolic expressions, this time using CasADi v3.5.1. CasADi plug-ins to the *IDAS* solver of the open source SUNDIALS suite and IPOPT by COIN-OR were once again used for the integration

Table 4.4 Criteria for termination of optimisation by IPOPT

Property	Termination tolerance	Acceptable termination tolerance
Optimality error	10^{-4}	10^{-4}
Dual infeasibility	1	10^{6}
Constraint violation	10^{-4}	10^{-2}
Complementarity	10^{-4}	10^{-2}

of ODEs and optimisation, respectively, with similar termination criteria to those used in the previous chapters. That is, the integration by *IDAS* had the following termination criteria: an absolute tolerance of 10^{-6} and a relative tolerance of 10^{-6}. For the optimisation by IPOPT, Table 4.4 presents the termination and ‘acceptable’ termination criteria, wherein the ’acceptable’ number of iterations concerning the latter was set at 15, as was done in Chapter 2.

The implementations were performed on a 2.80 GHz Intel(R) Core(TM) i5-8400 CPU, 16 GB RAM, Windows machine running on Microsoft Windows 10 Enterprise. Next, the results obtained for all case studies are discussed.

4.5 Results and discussions

It was found that the proposed methodology faced no difficulties in obtaining solutions for any sub-problem in any of the case studies, regardless of the parameter considered uncertain, the ranges of values examined or number of scenarios involved. In each sub-problem, the ODEs were solved to the specified integration tolerances and the optimisation converged to within the stipulated tolerances, thereby underlining the high quality of solutions obtained.

The major impacts of the uncertainties in kinetic parameters were seen in the values of objective functions and the time and frequency of catalyst replacements. These properties showed variations between the results of the different sub-problem within the same case study as well as in comparison to the result of the deterministic (single scenario) run, all of which were performed using the same initial guesses.

In the solution of each sub-problem, the trends of the variations of the decision and state variables over the time horizon were found to be very similar to those obtained in the

Table 4.5 Deterministic run solution details.

Property	Value
Profit (Million \$)	447.139
Number of catalyst replacements	4
Months of catalyst replacements	7, 14, 19, 26
Number of major iterations	2
Solution time (seconds)	9183

deterministic study involving Case Study A in Section 2.3.3 and Section 2.4.5 of Chapter 2. Therefore, these are not discussed in detail here. However, in order to provide insights into the kind of variations induced in the decision variables due to the consideration of uncertainty, the trends of decision variables over the time horizon for the deterministic run and all sub-problems of Case Study G are plotted and compared in Section 4.5.3 wherein the results of that case study are examined. This is done only for Case Study G, that considered the effect of uncertainty in Ea_{un}, because, as will be seen, it was in this case study that the maximum impact of uncertainty on the objective function was observed. Similar presentations for all other case studies are not informative enough to justify the large volume of the thesis that would be necessary to do so.

Details of the solution obtained from the deterministic run are given in Table 4.5. The properties of the results of the sub-problems of the four case studies, analogous to the properties of the solution of the deterministic run in Table 4.5, are given in Tables 4.6 – 4.9. In the following text, the effects induced by the consideration of the different aspects of uncertainty within a case study are analysed by making a comparison between the results of the sub-problems in each case study, as well as comparing the results of these sub-problems with the result of the deterministic run. Further insights are also drawn by making a comparison between the results of different case studies.

4.5.1 Case Study E

Details regarding the solutions obtained from the sub-problems investigated in this case study are given in Table 4.6, Figure 4.2 and Figure 4.3. As can be seen in Table 4.6, the inclusion of uncertainty in Kd_{un} introduces significant differences in comparison to the solution of the deterministic run presented in Table 4.5.

Table 4.6 Case Study E solution details.

Property	**%RSD about $\overline{Kd_{un}}$**		
	10	**20**	**30**
Mean profit over all scenarios (Million $)	418.832	419.640	420.685
Maximum profit over all scenarios (Million $)	418.905	419.785	420.901
Minimum profit over all scenarios (Million $)	418.760	419.496	420.427
Profit RSD (%)	0.0107	0.0214	0.0321
Number of catalyst replacements	3	3	3
Months of catalyst replacements	7, 19, 26	7, 19, 26	7, 19, 26
Number of major iterations	4	4	4
Solution time (seconds)	703187	811814	1049004

Table 4.7 Case Study F solution details.

Property	**%RSD about $\overline{Ar_{un}}$**		
	10	**20**	**30**
Mean profit over all scenarios (Million $)	447.451	441.436	439.689
Maximum profit over all scenarios (Million $)	447.772	442.081	440.667
Minimum profit over all scenarios (Million $)	447.123	440.785	438.715
Profit RSD (%)	0.0452	0.09145	0.1383
Number of catalyst replacements	4	4	4
Months of catalyst replacements	7, 13, 19, 26	7, 12, 19, 26	7, 13, 19, 26
Number of major iterations	2	2	2
Solution time (seconds)	825814	754913	488749

Table 4.8 Case Study G solution details.

Property	**%RSD about $\overline{Ea_{un}}$**		
	5	**7.5**	**10**
Mean profit over all scenarios (Million $)	426.323	465.965	443.435
Maximum profit over all scenarios (Million $)	426.884	466.816	444.523
Minimum profit over all scenarios (Million $)	425.740	465.044	442.213
Profit RSD (%)	0.0837	0.1187	0.1628
Number of catalyst replacements	3	4	3
Months of catalyst replacements	7, 19, 26	7, 13, 19, 26	7, 19, 26
Number of major iterations	4	2	4
Solution time (seconds)	934872	747505	802522

Table 4.9 Case Study H solution details.

Property	**Number of scenarios**				
	5	**10**	**15**	**20**	**25**
Mean profit over all scenarios (Million $)	458.362	435.290	423.175	425.849	459.550
Maximum profit over all scenarios (Million $)	459.001	435.774	423.901	426.606	460.317
Minimum profit over all scenarios (Million $)	457.731	434.471	422.448	425.102	458.796
Profit RSD (%)	0.1172	0.0990	0.1046	0.1010	0.1015
Number of catalyst replacements	3	4	3	3	4
Months of catalyst replacements	7, 19, 26	7, 13, 19, 26	7, 19, 26	7, 15, 22	7, 13, 19, 26
Number of major iterations	4	2	4	6	2
Solution time (seconds)	95150	308784	1060280	1367039	858033

The expected (mean) profits in the sub-problems of 10%, 20% and 30% RSDs about $\overline{Kd_{un}}$ show a considerable decrease of about 29, 28 and 27 million dollars, respectively, from the profit obtained in the deterministic run. Further, a comparison between the expected profits of the sub-problems gives an indication of the sensitivity to this parameter. While the expected profits increase with an expansion in the range of values considered, the increase between sub-problems is relatively small, showing an increase in about 2 million dollars for an expansion in range from 10% RSD to 30% RSD. This suggests that while consideration of uncertainty in Kd_{un} does have an impact on the profits of the process, the sensitivity of profits to further change in values of Kd_{un} is relatively less.

The low sensitivity to change in Kd_{un} is also indicated in the small values of the RSDs of the profits generated over all scenarios for all sub-problems. These RSD values increase as the range of values considered in the sub-problems increases, which is natural. This is also reflected in the increasing difference between the maximum and minimum profits generated over all scenarios as the range of values considered in the sub-problems increases.

Insights into the distribution of the profits over the 20 scenarios for each of the 3 sub-problems are provided in Figure 4.2. In the histograms presented in this figure, the height of each bin represents the number of scenarios out of 20 that result in profit values within the range specified by the horizontal edges of that bin.

It is highlighted that the RSDs of the profits generated over all scenarios are of small magnitudes in all case studies. This can be attributed to the fact that it is only the inventory costs that are impacted by the generation of multiple scenarios and as observed in the results of Chapter 2, these costs form the smallest proportion of the total expenses. However, different magnitudes are observed in different case studies, which suggests different levels of sensitivity of the profits to the change in values of different parameters.

Figure 4.3 provides an insight into the correlation between the profit and the value of Kd_{un} over all scenarios, for each sub-problem. Within all sub-problems, the profits seem to increase in a linear manner with the increase in value of Kd_{un}. This does not mean a generalisation that a higher value of Kd_{un} always leads to higher profit. It only implies that for a set of optimal decisions obtained in a particular sub-problem considering a set of scenarios, the nature of equations in the process model is such that the scenario containing a higher value of Kd_{un} attains a larger profit.

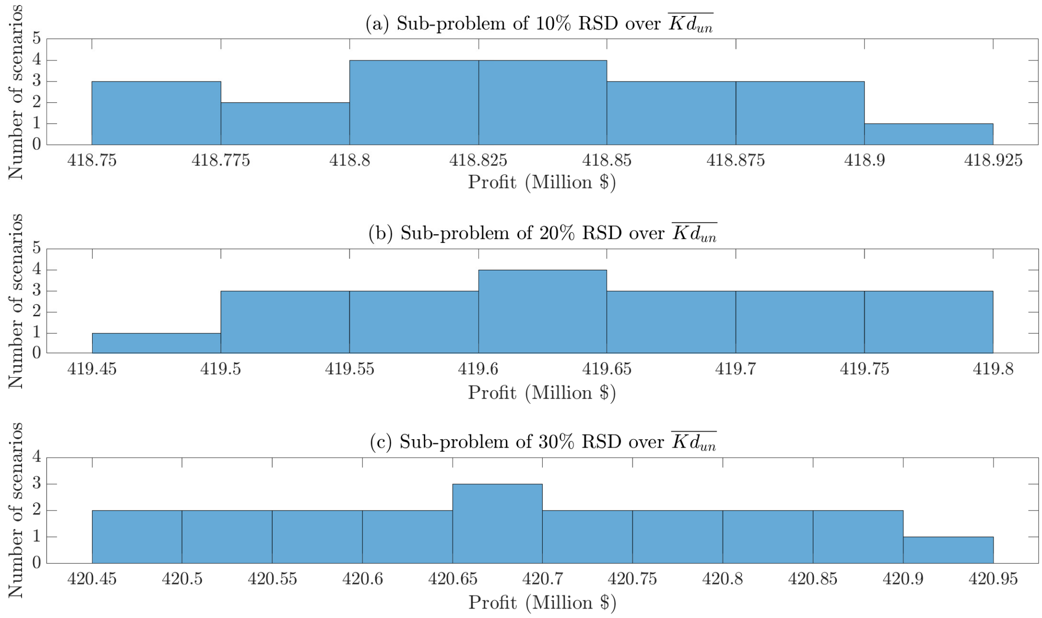


Fig. 4.2 The distribution of the profits over all 20 scenarios for the sub-problem of (a) 10% RSD over $\overline{Kd_{un}}$ (b) 20% RSD over $\overline{Kd_{un}}$ (c) 30% RSD over $\overline{Kd_{un}}$

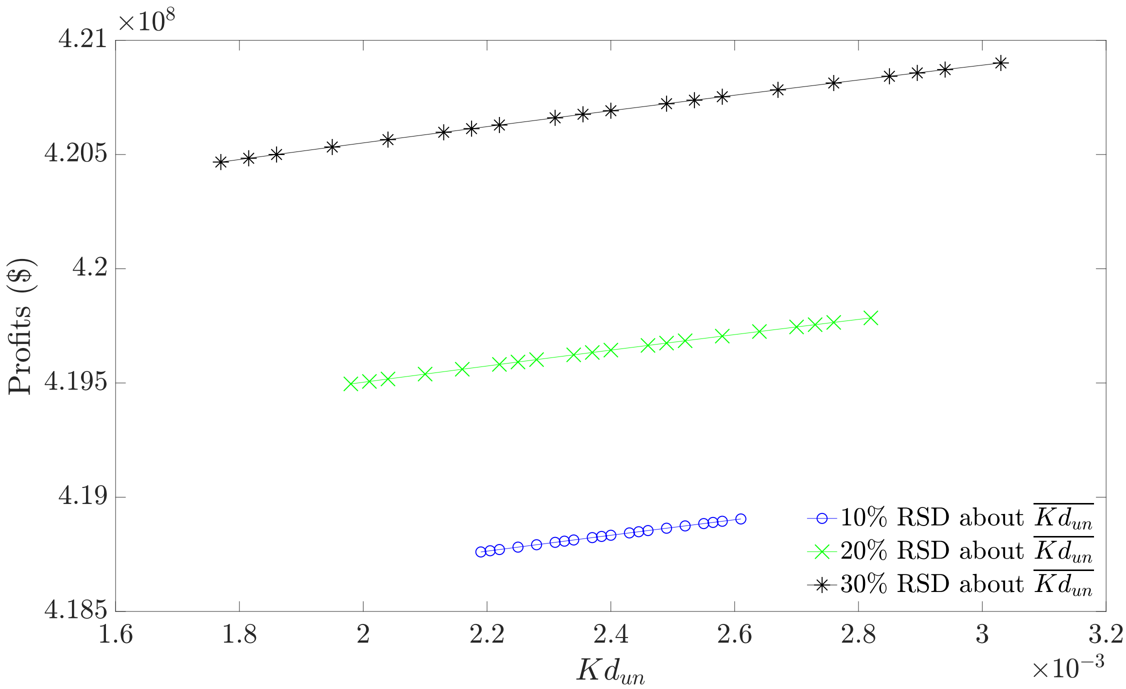


Fig. 4.3 The correlation between value of Kd_{un} and profit over all scenarios for the three sub-problems.

The number of catalyst replacements and the months in which they occur are the same for all sub-problems in this case study, indicating that these properties are not sensitive to a change in the range of uncertainty for Kd_{un}. But the number of catalyst replacements occurring in all of these sub-problems is 3, which is one less than in the deterministic case. However, the months in which these 3 catalyst replacements occur, are the same as the months of 3 of the 4 catalyst replacements in the solution of the deterministic run. Thus, in comparison to the deterministic run, the inclusion of uncertainty in Kd_{un} causes a change in the number of catalyst replacements, but does not cause a variation in the preferred months of catalyst replacement.

The introduction of uncertainty in Kd_{un} also causes the penalty term homotopy technique to operate differently compared to in the deterministic study. Each of the sub-problems require 4 major iterations, or 4 problems of the series given by equation (4.52) to be solved, to ensure binary values for the catalyst changeover controls. This is different from the deterministic study that required only 2 such major iterations.

The use of several different scenarios to consider parametric uncertainty causes the size of the ODE system to be solved in each of these sub-problems to become several times larger than that of the deterministic study (Table 4.3). This causes the solution time for each of these sub-problems to be much larger than that of the deterministic study.

4.5.2 Case Study F

Details of the solutions obtained from the sub-problems investigated in Case Study F are given in Table 4.7, Figure 4.4 and Figure 4.5. It is seen that the inclusion of uncertainty in Ar_{un} also introduces differences in comparison to the solution of the deterministic run.

Compared to the profit obtained in the deterministic run, the expected profit for the sub-problem of 10% RSD about $\overline{Ar_{un}}$ showed a relatively small difference of about 0.3 million dollars, but larger differences of about 6 and 8 million dollars are seen for the sub-problems of 20% and 30% RSD about $\overline{Ar_{un}}$, respectively. This suggests that uncertainty in Ar_{un} can result in expected profits significantly different from the deterministic profit only when larger ranges of uncertainty are considered. Further, for a given range of uncertainty in Ar_{un}, the difference of the expected profit from the deterministic profit is less than that seen for a similar range of uncertainty in Kd_{un} in Case Study E. This indicates that uncertainty in Ar_{un} has a smaller impact on the profits of the process in comparison to uncertainty in Kd_{un}. Another difference from Case Study E is that the expected profits decrease as the range of uncertainty in Ar_{un} increases, whereas an opposite trend was seen for uncertainty in Kd_{un} in Case Study E.

On the other hand, the change in the value of expected profits as the range of uncertainty in Ar_{un} is changed, is larger than that seen for a similar change of range in Kd_{un}. This suggests that the profits are more sensitive to a change in value of Ar_{un} compared to a change in value of Kd_{un}. This is also reflected in larger values of the RSDs of the profits generated over all scenarios as well as the larger difference between maximum and minimum profits in each range of uncertainty in this case study compared to the similar range in Case Study E. As in Case Study E, a natural increase occurs in the RSDs of the profits generated over all scenarios and the difference between maximum and minimum profits, as the range of uncertainty is increased in this case study.

Figure 4.4 provides insights into the distribution of the profits over the 20 scenarios for each of the 3 sub-problems. As in Figure 4.2, in the histograms presented in this figure, the height of each bin represents the number of scenarios out of 20 that result in profit values within the range specified by the horizontal edges of that bin.

An insight into the correlation between the profit and the value of Ar_{un} over all scenarios for each sub-problem is provided in Figure 4.5. Within all sub-problems, the profits seem to increase with an increase in Ar_{un} value, but unlike in Case Study E, this increase is not linear. Once again, it is stressed that a higher Ar_{un} value in a scenario leading to a higher profit in that scenario in each sub-problem, arises from the nature of the process model equations for a fixed set of optimal decisions and is not a trend that can be generalised.

The number of catalyst replacements for all sub-problems in this case study is the same and is equal to that in the deterministic run, thereby suggesting that this property is not affected by the inclusion of uncertainty in Ar_{un}.

However, on observing the months in which these replacements occur, the timing of the second catalyst replacement appears to vary, by occurring in the 13^{th} month in the sub-problems of 10% and 30% RSD about $\overline{Ar_{un}}$ and in the 12^{th} month in the sub-problem of 20% RSD about $\overline{Ar_{un}}$, in comparison to the 14^{th} month in the deterministic study. The months of all other catalyst replacements (7, 19 and 26) in all sub-problems are the same as in the deterministic run.

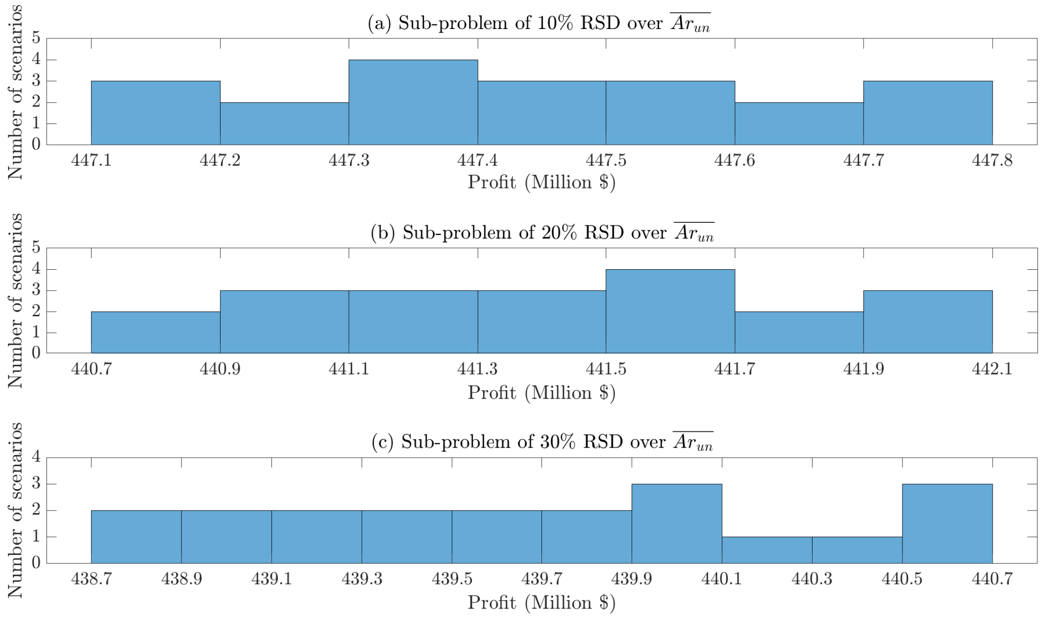


Fig. 4.4 The distribution of the profits over all 20 scenarios for the sub-problem of (a) 10% RSD over $\overline{Ar_{un}}$ (b) 20% RSD over $\overline{Ar_{un}}$ (c) 30% RSD over $\overline{Ar_{un}}$

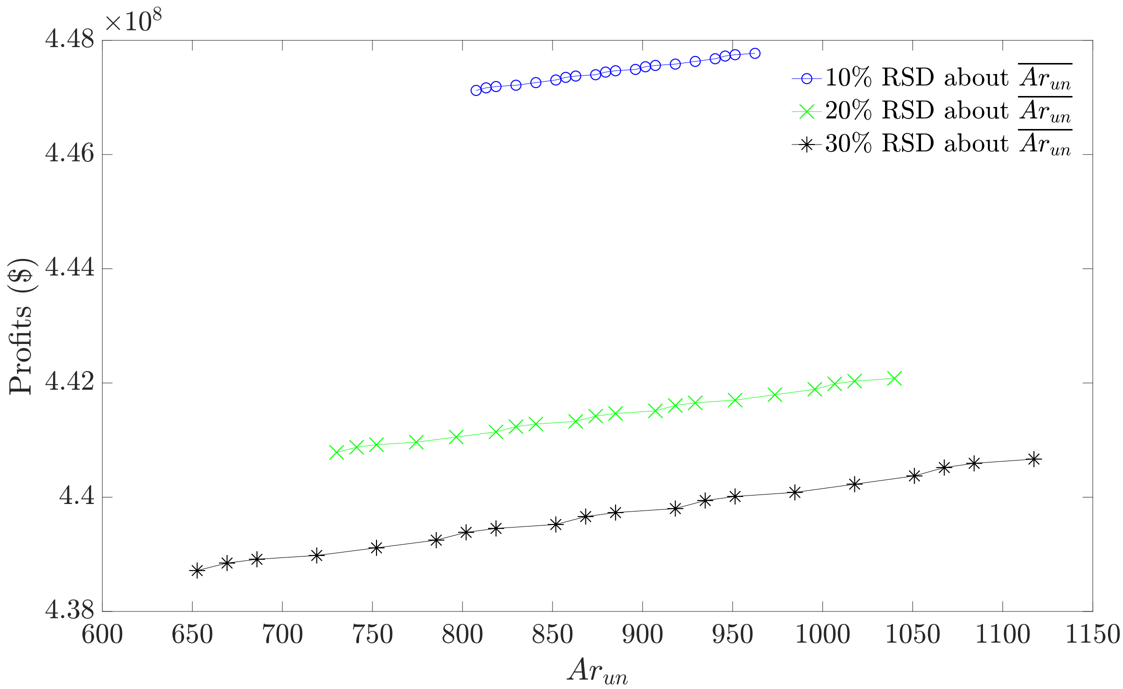


Fig. 4.5 The correlation between value of Ar_{un} and profit over all scenarios for the three sub-problems.

Thus, the observation is that even in the presence of uncertainty in Ar_{un}, majority of the catalyst replacements occur at the same time as in the deterministic run, and in the minority of situations when the timing is different, it is within one or two months of the time suggested in the deterministic run. This suggests that while the timing of catalyst replacements is impacted by the uncertainty in Ar_{un}, the effects are not drastic in the sense that the timing is only less sensitive to the uncertainty. It is also worth noting that majority of the months of catalyst replacements here overlap with the times suggested in the sub-problems of Case Study E.

As in the deterministic run, all sub-problems in this case study require two major iterations of the penalty term homotopy technique for the catalyst changeover controls to attain binary values. However, due to the large ODE system (Table 4.3), the solution times for all these sub-problems are several times larger than that of the deterministic run.

4.5.3 Case Study G

Details of the solutions obtained from the sub-problems investigated in Case Study G are given in Table 4.8, Figure 4.6 and Figure 4.7. The occurrence of the uncertain parameter, Ea_{un}, in an exponential term, causes the solutions of the sub-problems in this case study to be significantly different from the deterministic study, even while considering smaller ranges of uncertainty in comparison to the previous case studies.

As seen in Table 4.8, the expected profits for the sub-problems of 5%, 7.5% and 10% RSD about $\overline{Ea_{un}}$ were lower by approximately 21 million dollars, higher by approximately 18 million dollars and lower by approximately 4 million dollars respectively, in comparison to the profit of the deterministic run. Thus, it can be concluded that uncertainty in Ea_{un} has a significant effect on the expected profit. However, the wide variation in profit values between sub-problems and the lack of clearly increasing or decreasing trends in the expected profits with increasing ranges of uncertainty makes it difficult to compare the effect on the expected profit of uncertainty in Ea_{un} with that of the other kinetic parameters in the previous case studies.

Figure 4.6 provides insights into the distribution of the profits over the 20 scenarios for each of the 3 sub-problems. As in Figures 4.2 and 4.4, in the histograms presented in this figure, the height of each bin represents the number of scenarios out of 20 that result in profit values within the range specified by the horizontal edges of that bin.

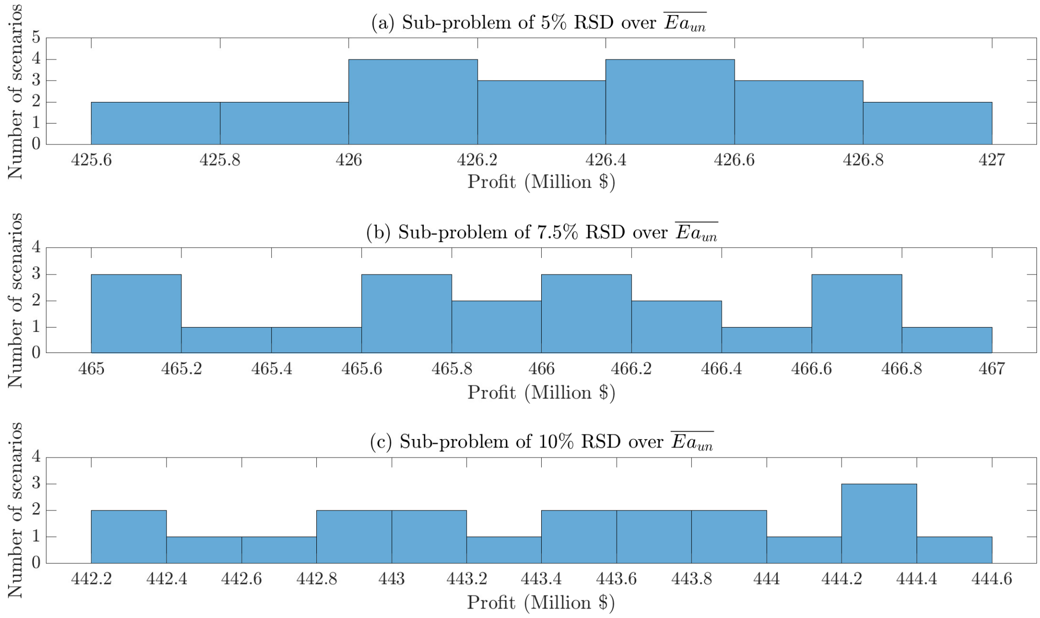


Fig. 4.6 The distribution of the profits over all 20 scenarios for the sub-problem of (a) 5% RSD over $\overline{Ea_{un}}$ (b) 7.5% RSD over $\overline{Ea_{un}}$ (c) 10% RSD over $\overline{Ea_{un}}$

As in the previous case studies, the RSDs of the profits generated over all scenarios as well as the difference between maximum and minimum profits in a sub-problem increases as the range of uncertainty for Ea_{un} increases. However, the profit RSD values are comparable to or even larger than those of the parameters in the previous case studies even though ranges of uncertainty used for Ea_{un} are smaller than those of the parameters in those case studies. This suggests that the profit values are more sensitive to a change in value of Ea_{un} compared to a change in value of Ar_{un} or Kd_{un}. This can be attributed to the occurrence of Ea_{un} within an exponential term in the process model against the linear occurrences of Ar_{un} and Kd_{un}.

The correlation between the profit and the value of Ea_{un} over all scenarios for each sub-problem is given in Figure 4.7. Within all sub-problems, the profits seem to increase with an increase in Ea_{un} value in a linear manner. Once again, this is not a trend that can be generalised due to reasons similar to those mentioned in the explanations of the analogous figures (Figures 4.3 and 4.5) in the previous case studies.

The number of catalyst replacements in the sub-problem of 7.5% RSD about $\overline{Ea_{un}}$ is 4, which is the same as in the deterministic run. But this number drops to 3 in the sub-problems of 5% and 10% RSD about $\overline{Ea_{un}}$. This suggests that this property is susceptible to change

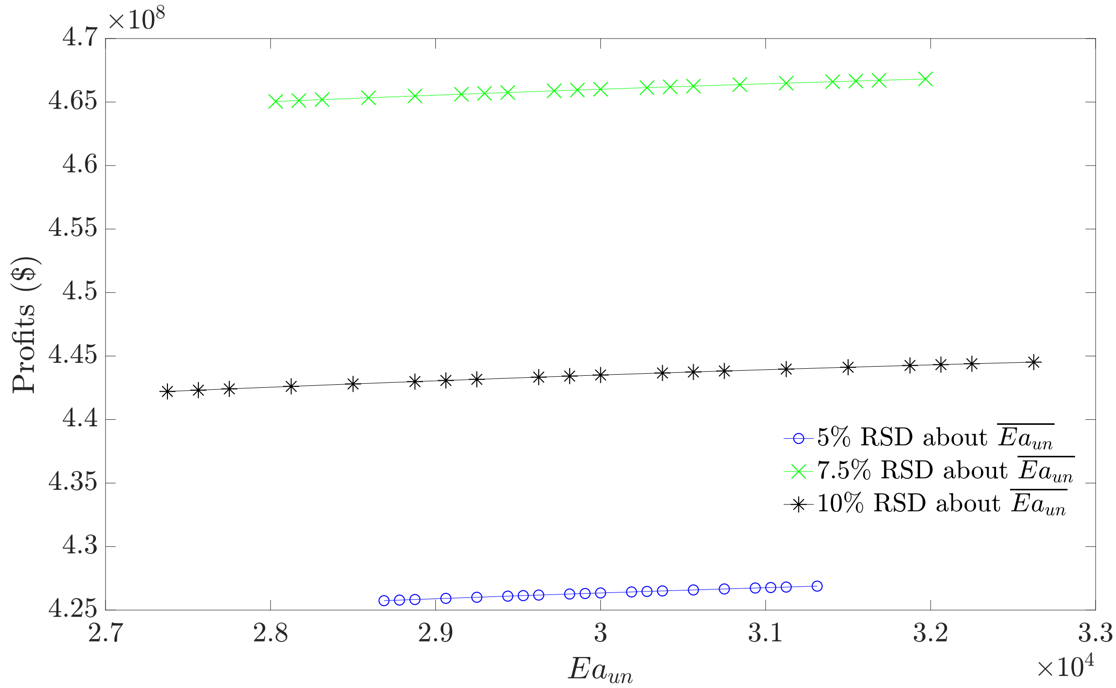


Fig. 4.7 The correlation between value of Ea_{un} and profit over all scenarios for the three sub-problems.

with varying ranges of uncertainty in Ea_{un}.

While the number of catalyst replacements in the sub-problem of 7.5% RSD about $\overline{Ea_{un}}$ is the same as in the deterministic run, the second catalyst replacement in this sub-problem occurs in the 13$^{\text{th}}$ month, in comparison to the 14$^{\text{th}}$ month in the deterministic run. The months of the other 3 catalyst replacements (7, 19 and 26) in the sub-problem are the same as in the deterministic run and in fact, these overlap with the months of the 3 catalyst replacements in the sub-problems of 5% and 10% RSD about $\overline{Ea_{un}}$.

Thus, even in presence of uncertainty in Ea_{un}, majority of the catalyst replacements occur at the same time as in the deterministic run, and in the minority of situations when the timing is different, it is within one month of the time suggested in the deterministic run. This indicates that while the timing of catalyst replacements is impacted by the uncertainty in Ea_{un}, the sensitivity to uncertainty in Ea_{un} is small. The reporting of similar observations in other case studies suggests that the timing of catalyst replacement is less sensitive to the presence of uncertainty in kinetic parameters.

While the number of major iterations in the penalty term homotopy technique, to guarantee binary values for the catalyst changeover controls, is 2 in the deterministic run and the sub-problem of 7.5% RSD about $\overline{Ea_{un}}$, it is 4 in the sub-problems of 5% and 10% RSD about $\overline{Ea_{un}}$. As in the previous case studies, due to the large ODE system (Table 4.3), the solution times for all these sub-problems are several times larger than that of the deterministic run.

As was mentioned at the beginning of Section 4.5, since uncertainty in Ea_{un} causes the maximum variation in the objective function, the trends over the time horizon of the decision variables of the sub-problems of Case Study G are discussed in order to provide insights into the kind of variations induced in the decision variables due to the consideration of uncertainty. The months of catalyst replacements and hence, the values of catalyst changeover controls, for all sub-problems have already been examined when discussing Table 4.8. Therefore, only the profiles of ffr_{un}, T_{un} and $sales_{un}$ over the time horizon, for all sub-problems of Case Study G alongside that of the deterministic run, are examined and these are shown in Figures 4.8, 4.9 and 4.10, respectively.

As seen in Figure 4.8, during times of catalyst operation, the profile of ffr_{un} for each sub-problem is very similar to that of the deterministic run: while in the deterministic run, the flow rate is decreased at a rate that maintains a constant exit concentration, in the other sub-problems the flow rate is decreased at a rate such that average of the exit concentrations over all scenarios considered is constant. The trends of the flow rates can be explained using reasoning similar to that offered for the explanation of the trend of the flow rate in Figure 2.8 in Section 2.3.3. It is noticed, however, that during times of catalyst operation, excluding the time when the sharp drop in value occurs at the end of the time horizon, the magnitudes of the flow rates become higher as the range of uncertainty for Ea_{un} is increased. But an opposite trend is observed during the time when the sharp drop in value occurs at the end of the time horizon. It is noted though that, at all of these times, the difference in magnitudes between the runs is quite small.

It can be seen in Figure 4.9 that the profiles of the temperatures over the time horizon for the deterministic run and all sub-problems are similar in that the temperatures are maintained at the upper bound during times of catalyst operation and drop to the lower bound during times of catalyst replacement. Such trends can be explained using reasoning similar to that provided for the explanation of the profile of the temperature in Figure 2.8 in Section 2.3.3.

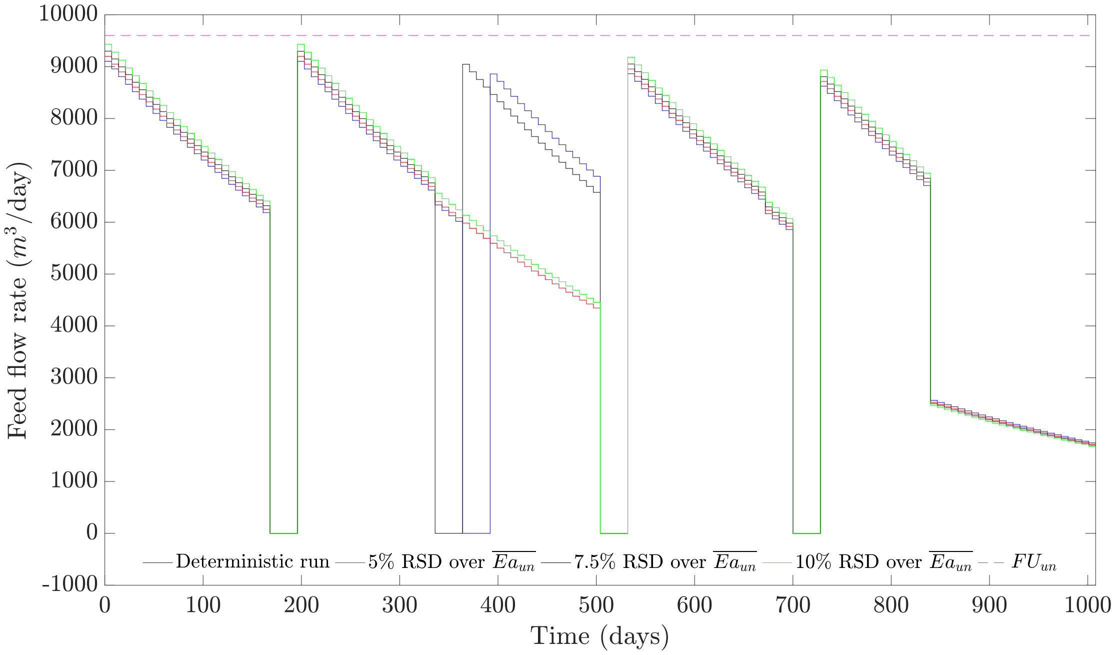


Fig. 4.8 A comparison between the profiles of the feed flow rates of the deterministic run and all sub-problems of Case Study G

And from Figure 4.10, it is seen that the profiles of the sales over the time horizon for the deterministic run and all sub-problems are largely similar in that in all these runs, there is some unmet demand at the beginning of the process and there is considerable unmet demand at the end of the first year, but the sales completely meet the demand during the second and third years. Such observations can be explained using reasoning similar to that given for the explanation of the profiles of the sales in Figure 2.9 in Section 2.3.3. However, there are some minor differences between the sub-figures in that the amount of sales and hence, the unmet demand, in the first year, differs between these runs. If a sub-figure has a greater amount of sales and hence, lesser unmet demand during the first year, it is because the run corresponding to that sub-figure has a more efficient catalyst replacement schedule that enables a greater amount of product to be produced and thus, a greater amount of sales to occur.

It is mentioned that in the sub-problems of the other case studies, namely Case Study E, F and H, the trends of the feed flow rate, temperature and sales were also similar to that of the deterministic run, with minor variations seen in the magnitudes of only the feed flow rate and the sales.

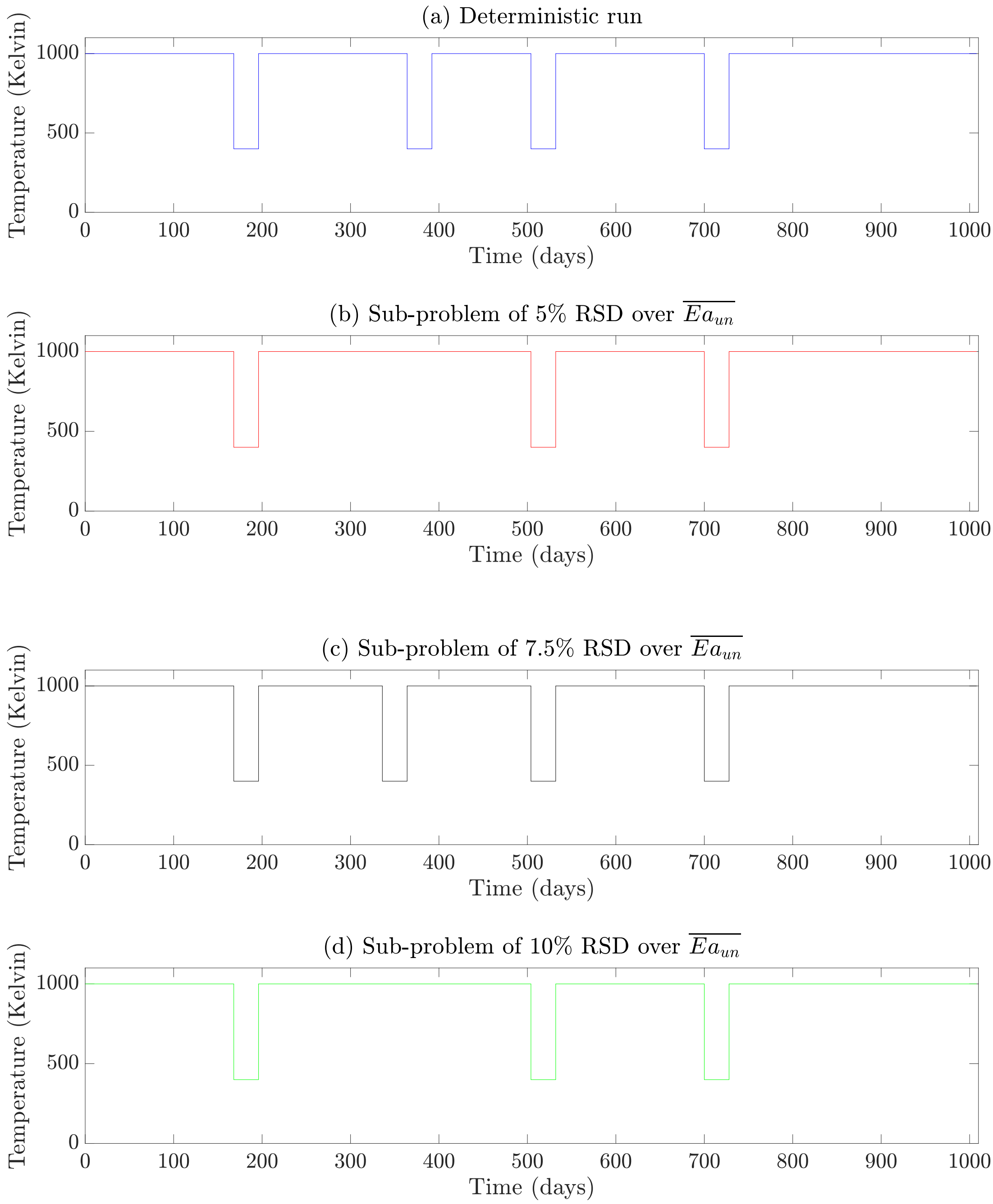


Fig. 4.9 The profile of the temperatures over the time horizon in the (a) Deterministic run (b) Sub-problem of 5% RSD over $\overline{Ea_{un}}$ (c) Sub-problem of 7.5% RSD over $\overline{Ea_{un}}$ (d) Sub-problem of 10% RSD over $\overline{Ea_{un}}$. This figure facilitates a comparison between the profiles of the temperatures of the deterministic run and all sub-problems of Case Study G

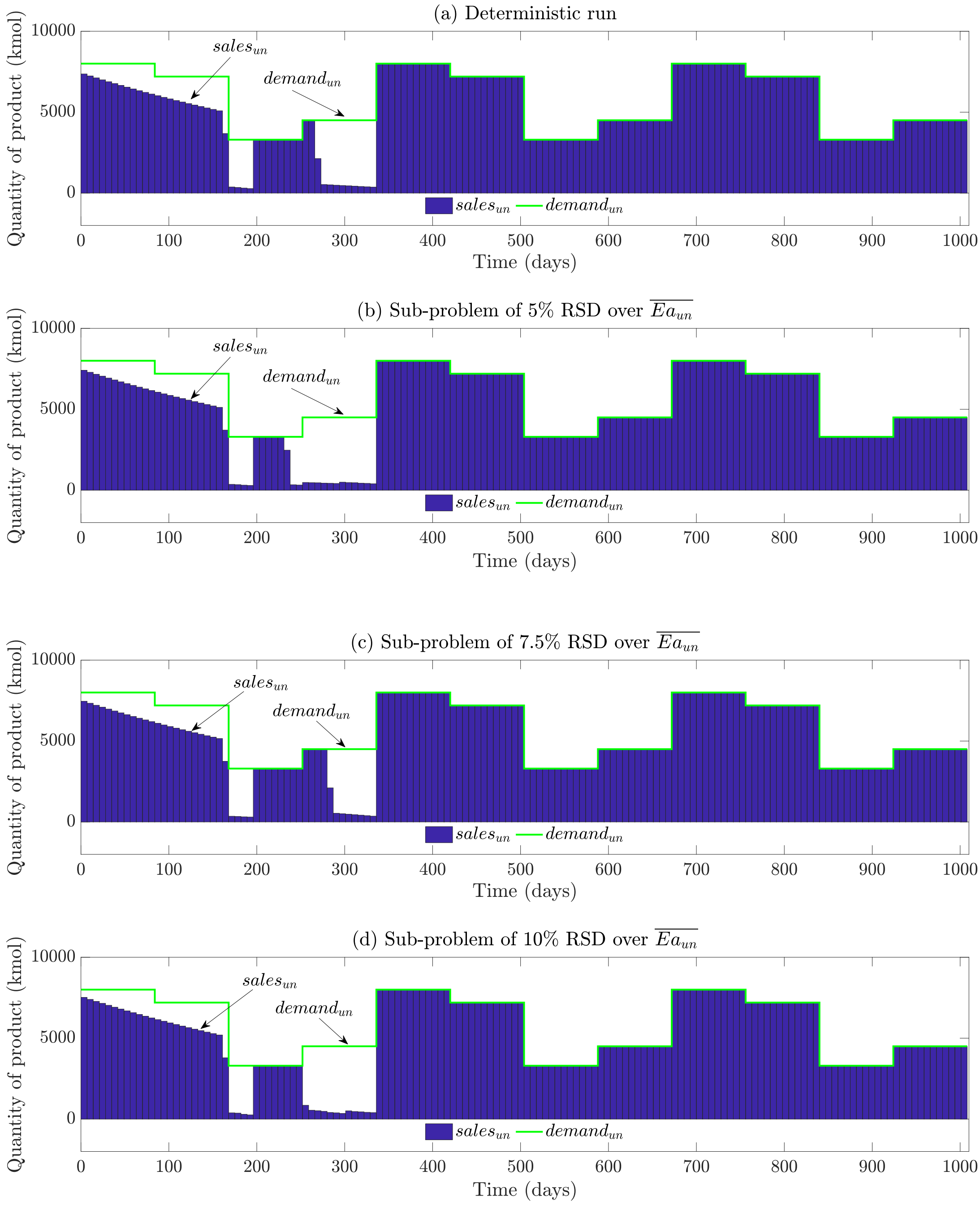


Fig. 4.10 The profile of the sales over the time horizon in the (a) Deterministic run (b) Sub-problem of 5% RSD over $\overline{Ea_{un}}$ (c) Sub-problem of 7.5% RSD over $\overline{Ea_{un}}$ (d) Sub-problem of 10% RSD over $\overline{Ea_{un}}$. This figure facilitates a comparison between the profiles of the sales of the deterministic run and all sub-problems of Case Study G

4.5.4 Case Study H

Table 4.9 provides details of the solutions of the sub-problems investigated in Case Study H, that considered the effect of simultaneous uncertainty in parameters Kd_{un}, Ar_{un} and Ea_{un} while generating different number of scenarios. It is noted that the scenarios for each parameter in each sub-problem were generated independently of each other. That is, for example, in the 10 scenario sub-problem, the 10 samples for each parameter were generated exclusively for this sub-problem and not by adding 5 samples to the samples of the 5 scenario sub-problem.

As is seen in Table 4.9, the expected profits show significant variations between sub-problems that considered different the number of scenarios, as well as in comparison to the profit in the deterministic run. The expected profits for the 5, 10, 15, 20 and 25 scenario sub-problems show an increase of about 11 million dollars, a decrease of about 12 million dollars, a decrease of about 24 million dollars, a decrease of about 22 million dollars and an increase of about 12 million dollars, respectively, from the profit in the deterministic run.

The difference between the maximum and minimum profit over all scenarios in each sub-problem shows a gradual increase as the number of scenarios considered in the sub-problem increases. The RSDs of the profit values over all scenarios show slight variations between sub-problems but are mostly around the value of about 0.1%.

Figure 4.11 provides insights into the distribution of the profits over the respective number of scenarios for each sub-problem. In the histogram presented for each sub-problem in this figure, the height of each bin represents the number of scenarios out of the total number of scenarios involved in that sub-problem, that result in profit values within the range specified by the horizontal edges of that bin. This presentation is merely to visualise the distribution of the profits in these sub-problems and there are no comments that can be drawn regarding the impact of the number of scenarios on these distributions.

The number of catalyst replacements also shows variations between sub-problems: while 4 replacements, the same as in the deterministic run, occur in the sub-problems of 10 and 25 scenarios, only 3 replacements occur in the sub-problems of 5, 15 and 20 scenarios.

While the number of catalyst replacements in the 10 and 25 scenario sub-problems are the same as in the deterministic run, the second catalyst replacement in these sub-problems occurs in the 13th month, in comparison to the 14th month in the deterministic run. The

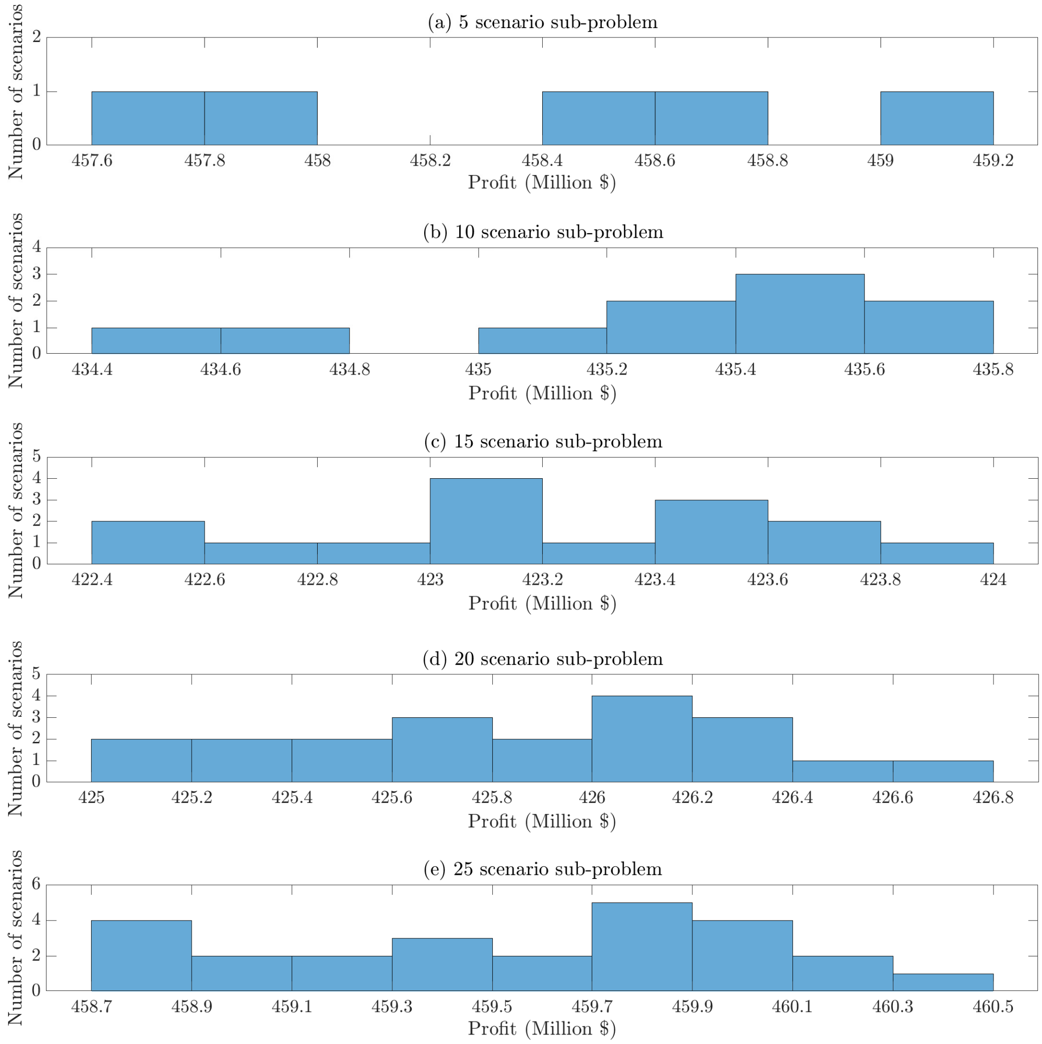


Fig. 4.11 The distribution of the profits over the respective number of scenarios runs for (a) 5 scenario sub-problem (b) 10 scenario sub-problem (c) 15 scenario sub-problem (d) 20 scenario sub-problem (e) 25 scenario sub-problem

months of the other 3 catalyst replacements (7, 19 and 26) in these sub-problems are the same as in the deterministic run and in fact, these overlap with the months of the 3 catalyst replacements in the sub-problems of 5 and 15 scenarios. In the 20 scenario sub-problem, while the timing of the first catalyst replacement (7^{th} month) is consistent with that of the deterministic run and all other sub-problems, the timings of the second (15^{th} month) and third (22^{nd} month) replacements are different from those in the deterministic run and the other sub-problems.

However, apart from a few anomalous cases like in the 20 scenario sub-problem, the observation is that the timings of the catalyst replacements in all sub-problems in this case study are mostly the same as in the deterministic run, or within a month of the timing in the deterministic run in the minority of situations. This suggests that the sensitivity of the timing of catalyst replacements to the simultaneous inclusion of uncertainty in all kinetic parameters as well as different number of scenarios is small, but not nil. This observation reinforces the suggestions made in the previous sub-sections that the timing of catalyst replacements is less sensitive to the presence of uncertainty in kinetic parameters.

The number of major iterations in the penalty term homotopy technique to ensure binary values for the catalyst changeover controls also varies between sub-problems. The sub-problems of 10 and 25 scenarios require 2 such iterations, as in the deterministic run, but the sub-problems of 5 and 15 scenarios require 4 such iterations and the 20 scenario sub-problem requires 6 such iterations.

The solution time for all sub-problems is several times larger than in the deterministic run, owing to their larger ODE systems compared to the latter (Table 4.3). However, the 5 scenario sub-problem has the smallest solution time among all sub-problems, followed by the 10 scenario sub-problem, and this follows from the size of their respective ODE systems. It may seem counter-intuitive that the solution time for the 25 scenario sub-problem is smaller than those of the 15 and 20 scenario sub-problems, despite the larger size of the ODE system in the former. This is because a greater number of major iterations are needed for the 15 and 20 scenario sub-problems (4 and 6, respectively) compared to the 25 scenario sub-problem (which required only 2 major iterations).

Thus, it is seen that the solutions of all sub-problems show variations from the deterministic run and this is an indication of the significant effects that the inclusion of uncertainty in all kinetic parameters simultaneously can produce. But there are also variations between the solutions of the sub-problems, each which considered a different number of scenarios, although the same range of uncertainty was considered for each uncertain parameter in all these sub-problems.

The number of major iterations and the solution times for the different sub-problems can be expected to be different, due to the different sizes of the ODE systems involved in each sub-problem. However, the variations in the RSDs of the profits over all scenarios, the number of catalyst replacements, the months of catalyst replacements and especially the large differences in the expected profit values between sub-problems suggest that an insufficient number of scenarios have been considered to solve this problem. That is, a much larger number of scenarios have to be considered in order to identify the optimal schedule of catalyst replacements and production plan under the given range of uncertainty for each parameter.

4.5.5 General discussion of results

In this sub-section, a comparative discussion of the results obtained in all case studies is carried out and some conclusions are drawn.

With regard to the expected profit of the process, a comparison of the results of the sub-problems of Case Studies E and F indicated that uncertainty in Ar_{un} seems to have a smaller impact compared to uncertainty in Kd_{un}. Another notable trend is that the expected profits decrease as the range of uncertainty for Ar_{un} increases, whereas an opposite trend was seen for an increase in range of uncertainty for Kd_{un}.

The results of the sub-problems of Case Study G indicate that uncertainty in Ea_{un} has a significant impact on the expected profit of the process. However, with increasing range of uncertainty in Ea_{un}, there is no pattern in the obtained values of the expected profit, in terms of a variation from the deterministic profit or an increasing or decreasing trend. Hence, no clear comparisons could be drawn between the impact on the expected profits of uncertainty in Ea_{un} and uncertainty in Kd_{un} or Ar_{un}.

On the other hand, a comparison of the RSDs of the profits over all scenarios indicates that the profits of the process are most sensitive to a change in value of Ea_{un}, the next highest sensitivity is to a change in value of Ar_{un} and the lowest sensitivity is to a change in value of Kd_{un}. The highest sensitivity to a change in Ea_{un} could be attributed to the occurrence of Ea_{un} in an exponential term in the process model, against the linear occurrences of Ar_{un} and Kd_{un}.

However, in the results of the sub-problems of Case Study H, which considered different number of scenarios, although the RSDs of the profits over all scenarios are all around the value of about 0.1%, the expected profits vary widely between the sub-problems, even though all considered the same range of uncertainty for each uncertain parameter. This indicates that an insufficient number of scenarios were being considered to solve this problem.

To obtain the actual solution of this problem, a much larger number of scenarios need to be considered, such that a further increase beyond this number would cause negligible variations in the solutions. However, the computational power required to consider such a large number of scenarios is currently unavailable and so, this has not been carried out in this thesis.

The conclusion from Case Study H, that a much larger number of scenarios is needed to effectively solve this problem, throws a question on the inferences drawn from the solutions of Case Studies E, F and G, as these case studies considered only 20 scenarios, which is certainly not a large number. Therefore, the apparent observations such as the profits being least sensitive to change in Kd_{un}, or uncertainty in Ar_{un} having a smaller impact on expected profit compared to uncertainty in Kd_{un}, cannot be concluded to be true unless the same are observed while considering the required larger number of scenarios.

However, with regard to the number and timing of catalyst replacements, the results obtained suggest that parametric uncertainty has a small impact. In the results of the sub-problems of all case studies, the optimal number of replacements is either 3 or 4, thus showing small or no variation from the deterministic solution that has 4 replacements. Further, the months of replacements are mostly similar or within one or two months of the times in the deterministic solution, apart from in the 20 scenario sub-problem of Case Study H which can be considered an anomalous result.

This observation suggests that, just by knowing the deterministic solution, the operator of the process can obtain good estimates of the optimal number and timing of catalyst replacements, in the presence of parametric uncertainty. However, the fact remains that variations, though small, are seen in these properties between the results of the different number of scenarios considered in Case Study H. The consideration of the required larger number of scenarios can enable obtaining exact values, rather than estimates, of the optimal values of these properties in the presence of parametric uncertainty.

In Table 4.10, the solution times of all sub-problems of all case studies, as well as that of the deterministic run are summarised. It is noted that no patterns can be drawn regarding these solution times, with respect to the size of the sub-problem, the range of values for the uncertain parameter or the number of scenarios considered. However, Table 4.10 enables an appreciation of the impact of the consideration of uncertainty on the solution times: the solution time of each sub-problem of each case study is several times larger than that of the deterministic run due to the size of the ODE system in each of those being

Table 4.10 A summary of the solution times of the sub-problems of all case studies

Case Study	Sub-problem	Solution time (seconds)
Deterministic run	N/A	9183
Case Study E	10% RSD about $\overline{Kd_{un}}$	703187
	20% RSD about $\overline{Kd_{un}}$	811814
	30% RSD about $\overline{Kd_{un}}$	1049004
Case Study F	10% RSD about $\overline{Ar_{un}}$	825814
	20% RSD about $\overline{Ar_{un}}$	754913
	30% RSD about $\overline{Ar_{un}}$	488749
Case Study G	5% RSD about $\overline{Ea_{un}}$	934872
	7.5% RSD about $\overline{Ea_{un}}$	747505
	10% RSD about $\overline{Ea_{un}}$	802522
Case Study H	5 scenario sub-problem	95150
	10 scenario sub-problem	308784
	15 scenario sub-problem	1060280
	20 scenario sub-problem	1367039
	25 scenario sub-problem	858033

significantly larger than that of the deterministic run. It is also noted that these solution times are specific to the set of initial guesses and the type of computer used. That is, these solution times may well be different if different initial guesses and a different type of computer is used.

And finally, it is highlighted that only one set of initial guesses was used for the optimisation in all sub-problems of all case studies. As the problem here is non-convex, the solution obtained for each sub-problem is only a local optimum. In order to find the global optimum in each of these problems, the best solution out of several optimisation runs, each carried out using different initial guesses, needs to be identified. This would require a huge amount of computational effort and hence, has not been carried out in this thesis.

Thus, the results of the different case studies obtained in this section can be said to provide useful insights into the effects of parametric uncertainty in this problem while demonstrating the application of the proposed solution methodology. But these results cannot be concluded as the actual solutions of these case studies. In order to fully identify the effect of each uncertain parameter in Case Studies E, F and G, the sub-problems in these case studies need to be solved while considering a large number of scenarios and by identifying the global optimum from a set of solutions obtained from several different initial guesses. Similarly, the global optimum of the parametric uncertainty problem in Case Study H can be identified from a set of solutions obtained from several different initial guesses, while considering a large number of scenarios for each uncertain parameter.

The advantage of the stochastic formulation in the proposed solution methodology is that only the size of the ODE system increases as the number of scenarios considered increases and the number of decision variables and constraints remain the same as in the deterministic case, regardless of the number of scenarios involved. This property has prevented an explosion in problem size and facilitated convergence to high quality solutions, within the stipulated tolerances, for all case studies considered. Thus, the proposed solution methodology would face no difficulties in obtaining the global optimum of the aforementioned case studies by considering the required number of large scenarios and the several different initial guesses, provided the required computational power is available. The use of high performance computing and parallel computing facilities would make such a task feasible.

4.6 Summary, conclusions and further discussions

In this chapter, an optimal control methodology has been developed for considering uncertainties in kinetic parameters in the optimisation of maintenance scheduling and production in a process containing a reactor using decaying catalysts. This methodology is an extension of the forms of optimal control methodologies used in the previous chapters to solve deterministic problems.

As in the previous chapters, this methodology involves an initial problem formulation as an MSMIOCP. The elements of the MSMIOCP represent elements of the process in an analogous manner to that done in the previous chapters, with the exception that here there is uncertainty regarding the values of the kinetic parameter present in the process model represented by the DAEs of the MSMIOCP.

The integer restrictions on the binary controls in this MSMIOCP formulation are first relaxed and then, using a multiple scenario approach to consider parametric uncertainty, a stochastic version of this MSMIOCP formulation with relaxed binary controls is developed. A unique feature of this formulation as a stochastic MSMIOCP with relaxed binary controls is that the size of the DAE system increases proportionately with the number of scenarios considered compared to a deterministic/single scenario case, but the number of control variables and constraints involved in the optimisation is the same as in the latter case. This is by virtue of the property that a set of controls analogous to that in deterministic/single scenario case is to be used in this formulation in order to optimise only the average of the set of objective functions over all scenarios while fulfilling only the average of each set of constraints over all scenarios.

This stochastic MSMIOCP with relaxed binary controls is solved as a standard nonlinear optimisation problem in a manner similar to that done in the previous chapters. That is, a penalty term homotopy technique is applied to this formulation wherein a series of standard multistage optimal control problems are solved using a feasible path approach, with a monotonically increasing penalty term added to the objective function to enforce values of 0 or 1 for the binary controls, which were assumed continuous in this formulation. As was mentioned in the previous chapters, this procedure possesses multiple favourable characteristics: the penalty term homotopy technique obviates the need for mixed-integer optimisation methods to schedule catalyst changeovers and the feasible path approach guarantees accuracy, and both these features enable a smaller problem size which facilitates an easier convergence to solutions. As this procedure is similar to that of Implementation II in Section 2.4.3 of Chapter 2, only here is is applied to a stochastic MSMIOCP formulation with relaxed binary controls, this methodology is referred to as a "stochastic MSMIOCP formulation in combination with a solution procedure of the principle of Implementation II".

This methodology was applied to an industrial process similar to a case study of the industrial process considered in Chapter 2, with the exception that here there was uncertainty regarding the values of the kinetic parameters of the catalyst deactivation rate constant, the pre-exponential factor and the activation energy of the product formation reaction. Four case studies were considered, three (Case Studies E, F and G) of which examined the effect of uncertainty of each parameter individually and the fourth (Case Study H) which studied the effect of all these parameters being uncertain simultaneously alongside the effect of the number of scenarios generated.

The methodology faced no difficulties in obtaining solutions for any case study. The obtained results were of high quality in the sense that all ODEs were solved to a high accuracy, to the specified integration tolerances, and the optimisation converged to within the stipulated optimality tolerances. The trends of the decision and state variables over the time horizon for all case studies were similar to that of the related case study in Chapter 2. The main effects of uncertainties were seen in the values of the objective functions, and the frequency and timings of catalyst replacements.

A comparison between the results of Case Studies E, F and G as well a comparison of these results with that of a deterministic run performed using the same initial points was done in order to gain insights into the relative effects of uncertainties in different parameters. However, the results of Case Study H, which showed wide variations in solutions when considering different numbers of scenarios, suggested that the number of scenarios being considered (20 / 25) in all of the case studies examined was an insufficient number. In addition, it was highlighted that the results of all case studies were obtained using only one set of initial points and the true effects of uncertainties can only be identified only when several different initial points are used.

To obtain the true solutions of all these case studies, several runs using different initial points and while considering a large number of scenarios have to be performed. But the computational power required for such a task was not available. However, in the proposed methodology, as only the number of DAEs increase with the number of scenarios considered, while the number of decision variables and constraints remain the same as in a deterministic case, it is reasonable to expect that the methodology can obtain high quality solutions when a large number of scenarios and several initial points are used as well, provided the computational power required for such a task is available.

In conclusion, in this chapter, a novel optimal control methodology has been developed that is capable of effectively optimising maintenance scheduling and production in an industrial process using decaying catalysts while considering uncertainties in the kinetic parameters. This optimal control methodology's features can provide potential advantages over the conventional methodologies of solving problems of this kind, which use mixed-integer optimisation techniques in combination with one of the popular preventive methods of handling uncertainty, by potentially overcoming the drawbacks introduced by the mixed-integer techniques in those methodologies. These potential advantages are stated as follows:

1. It is robust because the smaller problem size enabled facilitates convergence to optimal solutions, regardless of the number of scenarios considered for the uncertain parameters.

2. It is reliable because solutions can be obtained to a high degree of accuracy using state-of-the-art integrators, without the use of any approximation techniques.

3. It is efficient because a maintenance schedule for catalyst replacements can be obtained inherently during the optimisation, without using any additional computational effort in deciding when to schedule catalyst changeovers.

Thus, in this chapter, in line with the third objective of the thesis, a methodology has been developed that can effectively optimise the maintenance scheduling and production in a process containing a reactor using decaying catalysts while considering uncertainties in kinetic parameters, and which can overcome the disadvantages that the use of mixed-integer methods would introduce in the conventional methodologies, if such methodologies are used to solve problems of this kind.

Further discussion can be drawn regarding the methodology developed in this chapter. In this methodology, the constraints were "softened" in that only the average of each set of constraints over all scenarios was required to be satisfied. However, this may not be sufficient in the handling of critical operational constraints. For example, for constraints involving personnel and plant integrity and safety, using only a softened version of the constraints may not be acceptable. In such cases, it may be necessary to use a "hard" form of the constraints wherein the constraint arising in each scenario is required to be fulfilled.

In addition, it is highlighted that the use of a multi-scenario approach and the formulation as a stochastic MSMIOCP is just one way of including parametric uncertainty in this problem formulated as an MSMIOCP. Alternatively, parametric uncertainty can be considered in the initial MSMIOCP formulation of the problem by applying one of the other previously mentioned techniques of handling uncertainty on this formulation. For example, one of the robust optimisation techniques can be applied and a robust counterpart formulation of the MSMIOCP can be derived. Following techniques of parametric programming, an MSMIOCP formulation can be developed to obtain mappings between parametric uncertainties and optimal solution alternatives. By employing, as done throughout this thesis, a solution procedure as a standard nonlinear optimisation problem using a feasible path approach, such formulations can be solved effectively, especially in comparison to the conventional methodologies of using mixed-integer optimisation methods in combination with such techniques of handing uncertainty, in which case the drawbacks of the mixed-integer methods could cause difficulties in obtaining solutions. These can be regarded as directions of future research work.

Chapter 5

Conclusions and future work

As the title suggests, the focus of this chapter is on the conclusions of the thesis and suggestions for future research work. First, an overview of the research problems and objectives of the thesis is given in Section 5.1. This is done in order to facilitate the presentation of the conclusions of the thesis in Section 5.2. The recommendations for future research work are mentioned in Section 5.3.

5.1 Overview of research problems and objectives

The problems under consideration in this thesis involve the optimisation of large scale industrial processes using decaying catalysts. In order to optimise the performance of such processes, it is necessary to obtain an optimal maintenance schedule for the replacements of the decaying catalysts in tandem with an optimal production plan that specifies the best operating conditions of the process and the periodic sales to meet time-varying demand effectively.

Two publications, by Houze et al. (2003) and Bizet et al. (2005), exist that attempt to solve such a problem using mixed-integer optimisation methodologies. The problems considered in the two publications were similar in that the process contained only a single reactor using decaying catalysts that processed a single feed stream to produce a single product, there were no uncertainty considerations present and the scheduling of catalyst replacements had to be optimised in tandem with the optimisation of reactor operating conditions and the periodic product sales in order to meet time-varying demand. But the two publications differed in that different time horizons of the process were considered and different mixed-integer methodologies were utilised to obtain solutions.

However, there are major drawbacks involved in the use of mixed-integer optimisation techniques. When such techniques are used, differential equations present in the problem have to be approximated as a collection of steady state equations, which in turn are imposed as equality constraints in the problem. Not only does the steady state approximation reduce the accuracy of the results obtained, the fact that these steady state equations are imposed as equality constrains increases the number of variables and constraints present in the problem, and this could lead to difficulties in convergence to optimal solutions when a large number of differential equations are involved. In addition, if greater accuracy is desired, a finer steady state approximation of the differential equations is needed which, however, further increases the number of variables and constraints in the problem. Therefore, there is a difficult compromise between accuracy and ease of convergence involved in the use of mixed-integer methods.

Another major drawback is that, since mixed-integer methods are combinatorial in nature, the computational effort to solve a problem increases exponentially with the number of integer decisions involved. This adds to the drawback that if a large number of integer decisions are involved, the problem size also increases significantly. Therefore, in such cases, convergence to a solution may be difficult and even if solutions can be obtained, an enormous amount of computational power would be required.

While the two publications, by Houze et al. (2003) and Bizet et al. (2005), did not report any difficulties in convergence to optimal solutions, both did not reveal the underlying model or any of the parameters used, citing confidentiality reasons. Therefore, it was not possible to reproduce the results of those publications or even check for the accuracy of the solutions obtained. Further, these publications considered only a maximum of 3 or 4 catalyst loads to be used. If a larger number of catalysts was involved, the size of the problem would have increased greatly, and obtaining good quality solutions using methodologies similar to those employed by those publications would have faced considerable difficulties.

Further, more realistic problems compared to that considered by Houze et al. (2003) and Bizet et al. (2005) exist in this domain, which have not been considered by existing literature. For instance, industries commonly have parallel lines of reactors operating simultaneously. In order to maximise the profits of such a set up, it is necessary to optimise scheduling of catalyst replacements and operating conditions in all reactors, in addition to the inventory management and sales to meet seasonal demand, while also fulfilling a condition that no two reactors undergo catalyst replacement at the same time. While traditionally such a problem is solved using mixed-integer methodologies as well, it is highlighted that this problem is of larger size compared to the one considered by the works of Houze et al. (2003) and Bizet et al. (2005). Therefore, if mixed-integer optimisation techniques are used to solve this problem as well, the drawbacks of these techniques will be aggravated in comparison to the drawbacks faced by such techniques in solving the problem considered in those publications, and so, it would be difficult to obtain good quality solutions.

Further, the works of Houze et al. (2003) and Bizet et al. (2005) assumed that all kinetic parameters involved were known exactly, which is an unrealistic assumption. It is important to consider uncertainties in kinetic parameters in the optimisation of maintenance scheduling and production in processes using decaying catalysts, as such uncertainties can have a significant impact on the optimal process operations. The conventional methodologies of solving such a problem involve using one of the popular preventive methods of handling uncertainty in combination with a mixed-integer optimisation technique. However, this would involve using mixed-integer techniques to solve a problem of similar or larger size in comparison to when no uncertainty considerations are present, and so the drawbacks of these techniques will once again be encountered or possibly even be further aggravated. Therefore, obtaining good quality solutions for this problem using the conventional methodologies will be difficult.

The drawbacks involved in the use of mixed-integer optimisation techniques highlighted the need for an alternative set of methodologies to solve problems involving maintenance scheduling and production in processes using decaying catalysts, of the types mentioned above. This led to the objectives of the thesis, which were enumerated as follows:

1. To develop a methodology that can effectively optimise the maintenance scheduling and production in a process containing a reactor using decaying catalysts, and which can overcome the drawbacks faced by mixed-integer optimisation techniques in solving this problem

2. To develop a methodology that can effectively optimise maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts, and which can overcome the drawbacks that mixed-integer methods would face in solving problems of this kind

3. To develop a methodology that can consider uncertainties in kinetic parameters while effectively optimising the maintenance scheduling and production in a process containing a reactor using decaying catalysts, and which can overcome the disadvantages that the use of mixed-integer methods would introduce in the conventional methodologies, if such methodologies are used to solve problems of this kind

As was established, methodologies based on optimal control theory were developed to fulfil these objectives. Next, the conclusions of thesis are stated.

5.2 Conclusions

Firstly, the first objective was aimed to be fulfilled. An optimal control methodology was developed to solve the problem of interest: that of optimising maintenance scheduling and production in a process containing a reactor using decaying catalysts. The structure of the problem considered was similar to that in the works of Houze et al. (2003) and Bizet et al. (2005). However, since those works did not reveal the underlying model or any of the parameters used, citing confidentiality reasons, for the problem to be optimised by this methodology, the process model was constructed and an invented set of parameters were used. Four case studies of the problem were considered that differed based on either the kinetics of the catalyst deactivation or the product formation reaction in the process model.

The problem was formulated as a multistage mixed-integer optimal control problem (MSMIOCP) with the binary controls that decide the scheduling of catalyst changeovers occurring linearly in the formulation. A theoretical analysis suggested that due to the linear occurrence, these binary controls would exhibit a bang-bang behaviour in the optimal solution. Therefore, these binary controls were relaxed to be continuous controls in the range $[0,1]$, and for each of the four case studies, an MSMIOCP formulation with relaxed binary controls was used. In what was termed as Implementation I, the MSMIOCP formulation with relaxed binary controls in each of the four case studies was attempted to be solved as a standard nonlinear optimisation problem using the feasible path approach. And for each of the four case studies, in order to account for the non-convex nature of the problem, 50 runs were performed, each which used a different set of initial guesses.

It was found that Implementation I could obtain solutions only for two of the four case studies examined, which contained relatively less nonlinear process models. And even in the results of these case studies, out of the 50 runs carried out, only a very limited set of runs produced good quality solutions, with the vast majority of runs either converging prematurely or crashing due to problems in integration. And for the other two case studies which contained process models of relatively high nonlinearity, solutions could not be obtained because every one of the 50 runs crashed due to problems in integration. These integration problems could probably be attributed to the inadequacies of the ODE integrator suite of MATLAB, which was used for this implementation. However, despite these drawbacks, Implementation I was favourable from a theoretical point of view as in the limited set of successful runs, the catalyst changeover controls exhibited a bang-bang behaviour, consistent with the predictions of the theoretical analysis.

As an alternative to Implementation I, another implementation, termed Implementation II, was attempted on the formulation as an MSMIOCP with relaxed binary controls, which, however, did not exhibit the bang-bang behaviour for the catalyst changeover controls and instead required a penalty term homotopy technique to enforce binary values for those controls. The procedure of this technique was to solve a series of problems, of the form of the MSMIOCP with relaxed binary controls, using the feasible path approach, with a monotonically increasing weight term in the objective function to enforce values of 0 or 1 for the catalyst changeover controls. The lack of bang-bang behaviour for the catalyst changeover controls, despite the linear occurrence of those controls in the formulation, was probably due to a shortcoming of the IPOPT tool in the package of CasADi, which was used in this implementation.

Implementation II produced high quality solutions for all case studies in that it faced no difficulties in converging to optimal solutions, within the stipulated tolerances, for each of the 50 runs for each of the case studies, regardless of the set of initial guesses used and regardless of the nonlinearity of the process model involved. It was found that, for the two case studies for which solutions were possible to be obtained using Implementation I, the range of profit values obtained in the successful runs of those case studies when using Implementation I was comparable to the range of profit values obtained from the runs for the corresponding case studies when using Implementation II. And since the solutions obtained by Implementation I exhibited the optimal bang-bang behaviour for the catalyst changeover controls, consistent with the theoretical analysis, the aforementioned finding indicated that the solutions obtained by Implementation II were optimal as well. Therefore, the methodology proposed to be used to solve the problem under consideration was an MSMIOCP formulation in combination with the solution procedure of Implementation II.

In the optimal solutions obtained for all case studies, the profiles of the decision and state variables over the time horizon provided an indication of how to operate the process in order to maximise profits. In each case study, the notable results were the profiles of the temperature of the reactor and the concentration of the reactant exiting the reactor, during the times of catalyst operation.

Among the four case studies considered, the kinetics of catalyst deactivation in one case study was considered independent of reactant or product concentration while in the other case studies, the deactivation kinetics was dependent on the concentration of either the reactant or the product, and in all case studies, the rate constant of the catalyst deactivation was considered independent of temperature. In the optimal solutions of the case study involving composition independent deactivation kinetics, the policy for the temperature and the reactant exit concentration was to operate at the upper bound and maintain a constant value and respectively, and this was consistent with the work at the reactor level by Szépe and Levenspiel (1968), which predicted such optimal policies when the conditions on the catalyst deactivation kinetics were similar to that considered in this case study. However, while a work at the reactor level by Crowe (1976) predicted that, when the time scale for the catalyst deactivation is much larger than that of the main reaction and flow rate, the optimal policy was to operate all variables to maintain a constant exit concentration even when concentration dependent catalyst deactivation kinetics was involved, such trends were not obtained under similar conditions in the optimal solutions of the other case studies. This suggested that when concentration dependent deactivation kinetics is involved, policies similar to that predicted by Crowe (1976) at the reactor level do not hold when inventory, sales and demand considerations come into play.

Though the structure of the problem considered was similar to that considered in the works of Houze et al. (2003) and Bizet et al. (2005), a comparison of the trends of the variables over the time horizon or the profits obtained with that of those works was not possible, as those works did not reveal the underlying model or any of the parameters used, citing confidentiality reasons. However, those works revealed the time horizons of the process considered, the number of catalyst loads involved and the problem sizes that resulted when the respective mixed-integer methodologies were applied. The problem size for each of the time horizons in those works was compared with the problems size obtained for the corresponding time horizon when the MSMIOCP formulation of the proposed methodology was applied.

It was found that, for a given time horizon, the number of integer decisions, continuous decisions and especially the constraints were considerably smaller when using the MSMIOCP formulation of the proposed methodology in comparison that obtained by the mixed-integer methodologies used by those works. This was despite the fact that in the MSMIOCP formulation used, the continuous decisions such as the feed flow rate, the temperature and the sales were taken on a weekly basis, whereas in those works, such decisions were taken on a monthly basis. Further, the problem sizes in those works were obtained when considering only 2, 3 or 4 catalyst loads and due to the nature of the mixed-integer methodologies used, the problem size would increase if the number of available catalyst loads increased. However, in the proposed methodology, for a given time horizon, the problem size obtained using the MSMIOCP formulation applied regardless of the number of catalyst loads available to be used. In comparison to those works which use mixed-integer methodologies, the proposed methodology enables a smaller problem size due to the features of the feasible path approach, which solves differential equations without creating additional constraints, and the penalty term homotopy technique which enables scheduling of catalyst changeovers inherently during the optimisation without mixed-integer programming methods. The property of enabling a significantly smaller problem size in comparison to mixed-integer techniques provides the proposed methodology the advantage over those techniques of being more robust in convergence to optimal solutions in large scale problems.

Further, in the works of Houze et al. (2003) and Bizet et al. (2005), the differential equations present were approximated as a collection of weekly steady state equations, because of which the solutions obtained cannot be considered accurate. However, in the proposed methodology, all differential equations were solved to a high accuracy by the state-of-the-art integrators employed in the feasible path approach. This indicated the advantage of the proposed methodology over mixed-integer techniques that the solutions obtained by the methodology were more reliable than those obtained by such techniques in those works.

In addition, the works of Houze et al. (2003) and Bizet et al. (2005) considered only a maximum of 4 and 3 catalyst loads respectively, and these works would face a large increase in problem size and computation times if a larger number of catalyst loads was considered. However, in the proposed methodology, by virtue of the penalty term homotopy technique, the number and timings of catalyst replacements were decided inherently during the optimisation, without mixed-integer methods. Therefore, the proposed methodology was able to obtain optimal schedules for catalyst replacements in reasonable solution times even though there were six catalyst loads available to be used and this would be the case even if an infinite number of catalyst loads was available to be used. This indicated the efficiency of the proposed methodology over mixed-integer optimisation techniques.

Thus, the optimal control methodology developed was able to effectively solve the problem of optimising maintenance scheduling and production in a process containing a reactor using decaying catalysts, and by providing advantages of robustness, reliability and efficiency over the mixed-integer optimisation techniques, the methodology was able to overcome the drawbacks faced by those techniques in solving this problem. Hence, the first objective of the thesis was fulfilled.

Next, the second objective of the thesis was attempted to be fulfilled. Given the success of the optimal control methodology of using an MSMIOCP formulation in combination with a solution procedure of Implementation II in fulfilling the first objective, an optimal control methodology was developed using similar principles to solve the problem of interest: that of optimising maintenance scheduling and production in a process containing parallel lines of reactors using decaying catalysts. As no publication had worked on such a problem previously, the process considered while fulfilling the first objective was modified into a process containing a single feed split over 4 reactors using decaying catalysts, all of which produced the same product, and the optimal control methodology was applied to optimise maintenance scheduling and production in such a set up. In all reactors, the catalyst deactivation rate constant was independent of temperature and the catalyst deactivation kinetics were independent of reactant or product composition. Once again, due to the non-convex nature of the problem, 50 runs were performed, each which used a different set of initial guesses.

It was found that the methodology produced high quality solutions in that it faced no difficulty in obtaining solutions, while solving to within the stipulated optimality and integration tolerances, for each of the 50 runs. And among the profiles of the optimal decision and state variables over the time horizon, the notable profiles were that of the temperature of and the reactant exit concentration from each reactor, which were at their upper bounds and at a constant value, respectively, during the times when the catalyst was in operation in the reactor. This was once again, consistent with the work at the reactor level by Szépe and Levenspiel (1968) which predicted such policies for such variables when conditions on deactivation kinetics similar to that considered in this problem were involved.

The fact that the methodology faced no difficulties in converging to optimal solutions, in all of the 50 runs, regardless of the initial guesses used, indicated the robustness of the methodology. By means of the feasible path approach, all differential equations were solved to a high accuracy by state-of-the-art integrators and this suggested the reliability of the obtained solutions. By virtue of the penalty term homotopy technique, the optimal schedules for catalyst replacements for all reactors, which also fulfilled the condition that no two reactors undergo catalyst replacement at the same time, were obtained inherently during the optimisation, without mixed-integer methods, and this indicated the efficiency of the methodology. Thus, the methodology was able to solve this problem effectively while also offering potential advantages of robustness, reliability and efficiency over mixed-integer optimisation techniques if such techniques were to be used to solve the problem. Hence, the second objective of the thesis was fulfilled.

And finally, the third objective was attempted to be fulfilled. Using a modified version of the forms of the methodologies used to fulfil the first two objectives, an optimal control methodology was developed to solve the problem of interest: that of considering uncertainties in kinetic parameters in the optimisation of maintenance scheduling and production in an industrial process containing a reactor using decaying catalysts. The modification was that, using a multiple scenario approach to consider uncertainties, an initial formulation as an MSMICOP with relaxed binary controls was converted into its stochastic form, which was then solved in a manner similar to that in the previous methodologies, as per a solution procedure of the principle of Implementation II. The attractive feature of this stochastic formulation was that only the size of the DAE system increased proportionately with the number of scenarios considered compared to a deterministic/single scenario case, but the number of decision variables and constraints remained the same as in the latter case.

The problem considered involved a process which was a modification of a case study of the process considered while fulfilling the first objective, in order to include uncertainties in the values of the kinetic parameters of the catalyst deactivation rate constant, and the Arrhenius parameters of the pre-exponential factor and the activation energy of the product formation reaction. Four case studies were considered in this problem. For each of the three uncertain parameters, one case study was used to identify the individual effect of uncertainty in that parameter and this was done by attempting to solve three sub-problems within the case study, each which considered a different range of uncertainty for the uncertain parameter, while sampling 20 scenarios from within these range of values. The fourth case study was used to analyse the effect of all these parameters being uncertain simultaneously, alongside the effect of the number of scenarios generated. This was done by attempting to solve five sub-problems that considered 5, 10, 15, 20 and 25 scenarios for each of the three uncertain parameters simultaneously, while sampling from a specified range of uncertainty that, for a given parameter, remained the same across all sub-problems. And for all sub-problems of all case studies, the same set of initial points were used.

The methodology was successful in obtaining high quality solutions for all sub-problems of all case studies, in the sense that all ODEs were solved to the specified integration tolerances, and the optimisation converged to within the stipulated optimality tolerances. The trends of the decision and state variables over the time horizon for all case studies were similar to that of the related case involving no uncertainty considerations which was examined when attempting to fulfil the first objective. The main effects of uncertainties were seen in the values of the objective functions, and the frequency and timings of catalyst replacements.

A comparison between the results of the three case studies that identified the individual effects of uncertainty in each parameter with that of a deterministic run performed using the same initial points provided apparent insights into the relative effects of uncertainties in different parameters. However, the results of the fourth case study, which showed wide variations in solutions between sub-problems that considered different numbers of scenarios, suggested that an insufficient number of scenarios were being considered in all of the case studies examined. In addition, it was highlighted that the results of all case studies were obtained using only one set of initial points and that the true effects of uncertainties can only be identified only when several different initial points are used.

It was concluded that to obtain the true solutions of all these case studies, several runs had to be performed while using different initial points and while considering a large number of scenarios. But the computational power required for such a task was not available. However, the property of the proposed methodology, that only the number of DAEs increase with the number of scenarios considered, while the number of decision variables and constraints remain the same as in a deterministic case, suggested that the methodology can obtain high quality solutions when a large number of scenarios and several initial points are used as well, provided the computational power required for such a task was available.

Thus, a methodology was developed that is capable of solving the problem of optimisation of maintenance scheduling and production in a process containing a reactor using decaying catalysts, while considering uncertainties in kinetic parameters. Further, in this methodology, by virtue of the feasible path approach, a multiple scenario approach could be used to obtain a stochastic problem formulation that contained the same number of decision variables and constraints compared to a deterministic case, and this property, in addition to the relatively small problem size enabled by the feasible path approach and the penalty term homotopy technique, made the methodology robust in solving the problem, regardless of the number of scenarios considered. The features of the feasible path approach and the penalty term homotopy technique also enable the reliability and efficiency of the methodology, respectively, in a manner similar to that enabled for the methodologies used to fulfil the first two objectives. Thus, the methodology developed also offered potential advantages of robustness, reliability and efficiency over the use of mixed-integer optimisation techniques in the conventional methodologies, if such methodologies were to be used to solve this problem. Hence, the third objective of the thesis was fulfilled.

To sum up, the optimal control methodologies developed were able to effectively solve problems involving maintenance scheduling and production in processes using decaying catalysts. The features common to all methodologies developed include the feasible path approach to solve differential equations and a penalty term homotopy technique to enforce binary values for the controls that schedule catalyst changeovers, which are considered continuous in the problem formulation. These features, by providing advantages over the practices followed in mixed-integer optimisation techniques, enable the optimal control methodologies developed to overcome the drawbacks that are faced or would be faced by methodologies using mixed-integer optimisation techniques to solve these problems. These advantages are enumerated as follows:

1. Robustness: This is enabled by the relatively small problem size obtained which facilitates convergence to optimal solutions. The relatively small problem size is obtained by virtue of the following features:

   (i) The feasible path approach, that solves differential equations without creating additional constraints in the optimisation phase, as is done by mixed-integer methods

   (ii) The penalty term homotopy technique to schedule catalyst changeovers without mixed-integer methods, which allows to maintain the same problem size, regardless of the number of catalyst loads available to be used

   (iii) In the case where model-inherent uncertainties are present, the feasible path approach, when used with a multiple scenario approach to consider uncertainties, enables a stochastic problem formulation that has the same number of decision variables and constraints as in a deterministic case, regardless of the number of scenarios considered

2. Reliability: This is enabled by the feasible path approach that solves the differential equations present in the problem to a high accuracy using state-of-the-art integrators, unlike mixed-integer methods which approximate such differential equations as a collection of steady state constraints.

As a further comment, the feasible path approach, by enabling robustness and reliability, also enables the methodology to avoid making the difficult compromise between accuracy and ease of convergence, which is faced by mixed-integer methods.

3. Efficiency: This is enabled by the penalty term homotopy technique that can decide how many catalyst loads to use and when to replace each catalyst load, inherently during the optimisation. No additional computational effort would be required to identify optimal catalyst replacement schedules even if an infinite number of catalyst loads were available. This is unlike mixed-integer methods which require increasing computational effort with increasing number of catalyst loads available.

5.3 Future work

In this thesis, optimal control methodologies have been developed to effectively solve a few problems within the domain of problems involving optimisation of maintenance scheduling and production in processes using decaying catalysts. There are numerous directions of future research that can be drawn from the work presented in this thesis.

In all of the problems considered in this thesis, it has been assumed that the rate constant of the catalyst deactivation is independent of temperature. However, in applications of biochemical engineering, for example, enzymatic catalysts are used, and the deactivation rate constants of these enzymatic catalysts commonly have an even greater dependence on temperature compared to the main reaction. It is of interest to optimise maintenance scheduling and production in such processes, for cases which are deterministic as well as which involve uncertainty, by applying optimal control methodologies similar to those developed in this thesis, and to analyse the results obtained, especially how the optimal policy for the operating temperature comes out to be.

The problems considered in this thesis involve only a single reactant feed to either a single reactor or parallel lines of reactors, in order to produce a single product. However, in ethylene plants, for example, multiple reactors operate in parallel which process multiple feeds to produce multiple products. It is sought to derive an optimal control methodology, of the principle of those developed in this thesis, in order to optimise maintenance scheduling and production in such a set up.

Further, in this thesis, the problem of optimisation of maintenance scheduling and production while considering uncertainties in kinetic parameters has been examined only for a process containing a single reactor using decaying catalysts. It is of interest to analyse similar aspects in processes containing parallel lines of reactors using decaying catalysts, while considering a single feed producing a single product as well as multiple feeds producing multiple products.

As mentioned in the concluding section of Chapter 4, it is sought to derive optimal control methodologies alternative to that developed in this thesis, by drawing from concepts of the other popular preventive methods of handling uncertainty, to perform the optimisation under uncertainty of the maintenance scheduling and production in processes using decaying catalysts. That is, it is intended to apply techniques of methods such as fuzzy programming, robust optimisation and parametric programming to an optimal control formulation of such a problem and to obtain solutions as a standard nonlinear optimisation problem in a manner similar to that done throughout this thesis. It would be interesting to compare the results obtained using such methodologies with the results obtained using the stochastic optimal control formulation in this thesis. And when using such methodologies, it is of interest to identify the effect on the optimal operations of not only the uncertainty in kinetic parameters, as has been done in this thesis, but also the effect of uncertainty in parameters such as prices and demand.

It is highlighted that optimal control methodologies of the principle of those developed in this thesis can be applied to optimise maintenance scheduling and production planning in other processes experiencing decaying performances, whose problem structures are similar to the problems considered in this thesis. In the literary review section of Chapter 3, a number of publications were mentioned which use mixed-integer methodologies to optimise maintenance scheduling and operating conditions in varied processes that operate parallel lines of different types of units, such as gas engines, evaporators and compressors, to name a few examples, all of which experience decaying performances. Such processes are of similar structure to the problems considered in this thesis which involve processes using decaying catalysts. It is of interest to apply optimal control methodologies of the principle of those developed in this thesis in order to optimise maintenance scheduling and operating conditions in such processes and attempt to overcome the drawbacks that the use of mixed-integer optimisation techniques introduce in the solutions obtained in those publications.

There are limitations to the research presented in this thesis. While robust, reliable and efficient solutions for all problems considered in this thesis, that could overcome the drawbacks of mixed-integer optimisation techniques, could be obtained using solution procedures of the principle of Implementation II, this solution procedure faced the disadvantage of being unable to obtain a bang-bang behaviour for the catalyst changeover controls. It was possible to state that Implementation II could obtain optimal solutions only by the fact that the range of objective function values obtained by Implementation II for two case studies of the single reactor problem compared well with those obtained by Implementation I, which was able to obtain a bang-bang behaviour for the catalyst changeover controls. But Implementation I could obtain a limited set of solutions only for the case studies involving relatively less nonlinear models and failed completely when more nonlinear models were involved. The fact that two solution procedures are required is unfavourable, and it would be convenient if a single solution procedure could be obtained that combines the advantageous features of Implementations I and II.

Developing such a solution procedure could be a direction for future research. For example, research could be carried out on why the MATLAB ODE integrator suite is unable to integrate highly nonlinear process models and why the *fmincon* optimiser on MATLAB has a tendency to converge prematurely depending on the initial guesses used for the decision variables. Resolving such issues could result in a superior version of Implementation I and thereby, the development of the desired solution procedure. Alternatively, research could be carried out on why the IPOPT optimiser within the CasADi module in Python is unable to obtain bang-bang behaviour for the catalyst changeover controls. Resolving that issue could enable obtaining a superior version of Implementation II and thereby, result in the desired solution procedure.

And finally, the development of a dedicated software for optimisation of maintenance scheduling, which incorporates algorithms of the types presented in this thesis, can help in bridging the gap between the development of these methodologies and their wider application in the industrial context. The software can contain features such as requiring the specification of the initial guesses, parameters and the nature of the control parametrisation as inputs, with the outputs being the optimal solution values and graphs. A software capable of incorporating algorithms of the types presented in this thesis is not available currently: for example, gPROMS^{TM} (Process Systems Enterprise, 2020), which is one of the most advanced commercial simulators, cannot obtain multistage optimal control problem solutions as it does not allow for junction conditions. This is an avenue for future research that holds great commercial potential.

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