Closed-loop multi-target optimization for discovery of new emulsion polymerization recipes

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Electronic Supplementary Information

1. MOAL algorithm
1.1 Definition of the input variables

Reaction conditions:
- reaction temperature
- feeding time 1
- feeding time 2
- post-processing time (time of the reaction to continue after feeding time has finished)
- P (ratio of initiator solution fed in the reactor during feeding time 1 and 2)

Starting material:
- amount of water
- amount of initiator solution (7% concentrated)
- amount of seed (30% solid content)

Feeding:
- amount of surfactant solution (15% concentrated)
- amount of CTA
- amount of styrene
- amount of butyl-acrylate
- amount of water for initiator solution which get fed into the reactor
- amount of initiator which needs to get solubilized in the water

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The highly dimensional decision space of fourteen variables was chosen to allow the discovery of new recipes for the target of high conversion and particle size of 100 nm. The fourteen variables were relaxed as much as possible to allow the discovery of new recipes with the MOAL algorithm. However, only physical constraints were made to obtain feasible recipes, as for the amount of surfactant and initiator the water solubility was taken into account or another example for the reaction temperature the activation temperature of the initiator and the boiling point of water were taken into account.

1.2 Definition of constraints for variables

The following are the reaction settings (input variables for the model considered):

- $M^1_i$, $M^2_i$, $I_i$, $E_i$, and $W_i$ are the initial amounts of monomers, initiator, emulsifier and water respectively. $T$ is the reaction temperature, $CTA$ is the amount of chain transfer agent and $P_0$ is the amount of polymer in seed.
- $M^1_F$, $M^2_F$, $E_F$, $I_F$ are the fed amounts of monomers, emulsifier and initiator respectively. $WE_F$ and $WI_F$ are emulsifier and initiator solutions in feed. $WI_F$ is split into two parts $pWI_F$ and $(1-p)WI_F$ (where $p \in (0, 1)$) and is added in two stages.
- $F^1_t$, $F^2_t$ are feeding times for adding $pWI_F$ and $(1-p)WI_F$ respectively and $P_t$ is the post feeding time. The total reaction duration is the sum of $F^1_t$, $F^2_t$ and $P_t$.

The ranges of the variables and constraints are:

1. $M^1_i = M^2_i = 1 \cdot 10^{-10}$ g and $E_i = E_F = 1 \times 10^{-3}$ g. These are kept constant for all recipes.
2. $W_i$, $WE_F$, $WI_F \in (0, 1500]$ g and $225 \leq WI + WE_F + WI_F \leq 1500$.
3. $I_i \in (0, 0.2WI)$ and $I_F \in (0, 0.2WI_F)$.
4. $F^1_t$, $F^2_t$, $P_t \in (0, 180\text{min})$ and $30 \leq F^1_t + F^2_t \leq 180$.
5. $T \in [65, 95]$ in degrees Celsius, $CTA \in (0, 20]$ g and $P_0 \in [5, 30]$ g.
6. $M^1_F$, $M^2_F \in (0, 1500]$ g and $150 \leq M^1_F + M^2_F \leq 1500$.
7. The final constraint is that the overall volume of the ingredients must not exceed the capacity of the reactor (3L).
2. *In silico* optimisation using a physical model of emulsion copolymerisation

*In silico* optimisation using a physical model of emulsion copolymerisation revealed 18 feasible recipes within 84 experiments.
Table S1: Given recipes from the MOAL algorithm to gain the target of full conversion and 100 nm particle sizes during the simulations.

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\(^2\) Amount of water used in the reactor as starting material
\(^3\) Amount of initiator used in the reactor as starting material
\(^4\) Amount of seed used in the reactor as starting material
\(^5\) Amount of CTA solution used in the reactor as starting material
\(^6\) Amount of SDBS solution used in the reactor as starting material
\(^7\) Proportion of initiator in feed
| Exp. No. | T (°C) | water^8 (g) | initiator^9 (g) | feeding time 1 (min) | feeding time 2 (min) | post-processing time (min) | seed^10 (g) | CTA^11 (g) | SDBS solution^12 (g) | water for initiator feed (g) | P^13 | amount of initiator in feed (g) | styrene (g) | butyl acrylate (g) |
|---------|--------|------------|----------------|--------------------|--------------------|-------------------------|------------|----------|---------------------|-----------------------------|      |-----------------------------|------------|-------------------|
| 18      | 83.87  | 655.0419   | 40.0829        | 147.45            | 10.56              | 111.44                  | 12.37      | 1.69     | 66.2844             | 56.82                       | 0.76 | 10.5528                      | 244.29     | 50.60             |
| 19      | 92.86  | 274.1051   | 80.3030        | 29.23             | 24.99              | 34.70                   | 12.17      | 6.05     | 372.4776            | 240.80                      | 0.93 | 41.5467                      | 513.92     | 24.82             |
| 20      | 73.76  | 142.7280   | 47.2565        | 104.35            | 49.27              | 169.91                  | 7.94       | 13.16   | 121.2492            | 8.03                        | 0.90 | 0.3706                      | 82.45      | 366.35            |
| 21      | 84.81  | 476.9154   | 187.5606       | 91.78             | 10.22              | 156.19                  | 24.58      | 2.94     | 48.9056             | 85.51                       | 0.52 | 19.2582                      | 280.87     | 76.59             |
| 22      | 66.63  | 816.0207   | 225.3879       | 76.78             | 1.55               | 171.41                  | 6.64       | 4.22     | 162.5487            | 38.81                       | 0.78 | 3.4212                      | 299.03     | 45.50             |
| 23      | 66.72  | 818.1133   | 226.1891       | 77.45             | 2.26               | 171.69                  | 6.84       | 4.14     | 162.3188            | 43.07                       | 0.78 | 2.5460                      | 301.00     | 42.37             |
| 24      | 94.35  | 1305.2877  | 267.1772       | 59.25             | 40.69              | 151.53                  | 8.05       | 5.02     | 12.2650             | 13.48                       | 0.69 | 3.0595                      | 197.34     | 208.11            |
| 25      | 76.29  | 257.2994   | 62.5430        | 101.75            | 29.97              | 164.92                  | 13.91      | 0.52     | 31.1544             | 461.69                      | 0.96 | 68.1298                      | 372.37     | 21.01             |
| 26      | 82.49  | 388.9864   | 50.8130        | 13.85             | 62.12              | 165.37                  | 13.56      | 6.45     | 242.8972            | 131.39                      | 0.63 | 24.3324                      | 263.23     | 119.17            |
| 27      | 92.79  | 678.2930   | 104.4728       | 20.42             | 9.27               | 117.23                  | 6.65       | 2.97     | 284.1747            | 49.87                       | 0.38 | 3.1411                      | 370.16     | 8.42              |
| 28      | 88.11  | 106.6388   | 29.5456        | 30.56             | 29.21              | 120.89                  | 16.94      | 12.33   | 31.1767             | 15.54                       | 0.83 | 3.7097                      | 158.43     | 227.35            |
| 29      | 90.88  | 442.8715   | 177.8230       | 133.62            | 20.48              | 149.92                  | 5.98       | 11.83   | 958.1551            | 3.42                        | 0.11 | 0.0920                      | 324.33     | 72.58             |
| 30      | 92.89  | 207.2628   | 14.3615        | 35.24             | 6.78               | 79.65                   | 7.15       | 12.21   | 4.1564              | 291.10                      | 0.80 | 14.7623                      | 435.79     | 23.82             |
| 31      | 73.21  | 570.1065   | 180.0786       | 42.00             | 0.72               | 174.46                  | 8.02       | 2.85     | 45.8876             | 6.64                        | 0.68 | 0.1352                      | 116.03     | 223.58            |
| 32      | 94.89  | 837.1270   | 244.6972       | 111.24            | 2.37               | 171.08                  | 20.99      | 7.62     | 30.5690             | 28.51                       | 0.02 | 0.9960                      | 425.02     | 11.71             |
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| 36      | 93.11  | 217.8358   | 41.6004        | 37.55             | 0.42               | 147.20                  | 13.59      | 3.38     | 66.3183             | 142.81                      | 0.46 | 25.5331                      | 296.71     | 78.18             |
| 37      | 94.28  | 854.0755   | 330.0775       | 0.70              | 0.65               | 175.71                  | 13.76      | 8.95     | 36.3154             | 13.35                       | 0.47 | 1.4960                      | 326.82     | 106.84            |

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^8 Amount of water used in the reactor as starting material  
^9 Amount of initiator used in the reactor as starting material  
^10 15% solid content, particle size 70 nm  
^11 Chain transfer agent  
^12 15% surfactant solution  
^13 Ratio of initiator solution fed in the reactor during feeding time 1 and 2
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$^{14}$ Amount of water used in the reactor as starting material  
$^{15}$ Amount of initiator used in the reactor as starting material  
$^{16}$ 15% solid content, particle size 70 nm  
$^{17}$ Chain transfer agent  
$^{18}$ 15% surfactant solution  
$^{19}$ Ratio of initiator solution fed in the reactor during feeding time 1 and 2
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<sup>22</sup> 15% solid content, particle size 70 nm
<sup>23</sup> Chain transfer agent
<sup>24</sup> 15% surfactant solution
<sup>25</sup> Ratio of initiator solution fed in the reactor during feeding time 1 and 2
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\(^{26}\) Amount of water used in the reactor as starting material
\(^{27}\) Amount of initiator used in the reactor as starting material
\(^{28}\) 15% solid content, particle size 70 nm
\(^{29}\) Chain transfer agent
\(^{30}\) 15% surfactant solution
\(^{31}\) Ratio of initiator solution fed in the reactor during feeding time 1 and 2
Figure S1. Results of the simulation of the emulsion polymerization model with the MOAL algorithm: the figures show a) monomer-liquid ratio to the particle size; b) the correlation of the amount of initiator feed to particle size; c) the behaviour of conversion and amount of monomer.

Figure S2. Results of the simulation of the emulsion polymerization model with the MOAL algorithm: shows the behaviour of the amount of surfactant on particle size and the amount of seed particles on the particle size.
Table S2. Results of conversion and particle size of *in silico* optimisation using a physical model of emulsion copolymerisation.

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Table S3. Results of the validation of the \textit{in silico} recipes.

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3. Experimental closed-loop optimization

3.2 Reaction setup

Figure S3. Automated semi-batch system incorporated feedback for discovery new recipes for emulsion polymerization reactions.
3.3 Emulsion polymerization recipe:

General composition of the system for copolymerization of styrene and butyl acrylate.[1]

Butyl acrylate: 77 g
Styrene: 7 g
K₂S₂O₈: 0.6 g
NaHCO₃: 0.03g
Aerosol MA80: 3.6 g
Aerosol 22N: 0.7
H₂O: variable to feed rate
Table S4. Given recipes from the MOAL algorithm to gain the target of full conversion and 100 nm particle size experimentally.

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<th>initiator(^{33}) (g)</th>
<th>feeding time 1 (min)</th>
<th>feeding time 2 (min)</th>
<th>post-processing time (min)</th>
<th>seed(^{34}) (g)</th>
<th>CTA(^{35}) (g)</th>
<th>SDBS(^{36}) (g)</th>
<th>water for initiator feed (g)</th>
<th>p(^{37})</th>
<th>amount of initiator in feed (g)</th>
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\(^{32}\) Amount of water used as starting material
\(^{33}\) Amount of initiator used as starting material
\(^{34}\) 15% solid content, particle size 70 nm
\(^{35}\) Chain transfer agent
\(^{36}\) 15% surfactant solution
\(^{37}\) Ratio of initiator solution fed in the reactor during feeding time 1 and 2
Table S5. Performance of the experimental optimization of the recipe with MOAL algorithm. In red the unfeasible experiments are marked.

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Figure S4. Results of the experiment of the emulsion polymerization model with the MOAL algorithm: the figures show the correlation of the amount of initiator feed to the amount of seed particles.
Figure S5. Each plot shows the correlation of one of the fourteen variables with conversion. In none of these plots a trend can be seen. Due to the low number of experiments and only three experiments which fulfilled the targets, it is not surprising that no trend can be obtained.
Figure S6. Pair-wise correlation of the fourteen variables regarding particle size. Although the correlation of particle size with monomer, surfactant and initiator is known, this trend cannot be obtained from the low number of experiments.

Reference: