Geotechnique

PC-based and MgO-based binders stabilised/solidified heavy metal contaminated model soil: Strength and heavy metal speciation in early stage

--Manuscript Draft--

Manuscript Number:	17-P-194R1
Full Title:	PC-based and MgO-based binders stabilised/solidified heavy metal contaminated model soil: Strength and heavy metal speciation in early stage
Article Type:	Technical Note
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Abstract:	The investigation of using PC-based and MgO-based binders in treating contaminated model soil was carried out to study the benefit of novel binders over conventional ones in Stabilisation/Solidification systems (S/S), as well as their involved binding mechanism. Binders used in this study include Portland cement (PC), ground granulated blastfurnace slag (GGBS), pulverised fly ash (PFA) and magnesia (MgO). The strength and the leaching properties of S/S treated samples via unconfined compressive strength (UCS) and sequential extraction tests are presented. The results showed that the early-age strength of these mixes is influenced by the reactivity of binders; heavy metals were principally distributed in the carbonate and the Fe-Mn oxide fractions in all mixes after 28-days curing time; the speciation distribution characteristics are not the same among Zn, Cu, Ni and Pb; and the stability of Zn, Cu, Ni and Pb benefits from a longer curing time and the usage of MgO.
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PC-based and MgO-based binders stabilised/solidified heavy metal contaminated model soil: Strength and heavy metal speciation in early stage

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A Paper Submitted for possible Publication as Technical Notes to Geotechnique.

ABSTRACT: The investigation of using PC-based and MgO-based binders in treating contaminated model soil was carried out to study the benefit of novel binders over conventional ones in Stabilisation/Solidification systems (S/S), as well as their involved binding mechanism. Binders used in this study include Portland cement (PC), ground granulated blastfurnace slag (GGBS), pulverised fly ash (PFA) and magnesia (MgO). The strength and the leaching properties of S/S treated samples via unconfined compressive strength (UCS) and sequential extraction tests are presented. The results showed that the early-age strength of these mixes is influenced by the reactivity of binders; heavy metals were principally distributed in the carbonate and the Fe-Mn oxide fractions in all mixes after 28-days curing time; the speciation distribution characteristics are not the same among Zn, Cu, Ni and Pb; and the stability of Zn, Cu, Ni and Pb benefits from a longer curing time and the usage of MgO.

Keywords: sequential extraction, MgO, GGBS, stabilization/solidification, strength

1. INTRODUCTION

Stabilisation/Solidification (S/S) has been widely used to remediate contaminated soils over the past a few decades especially in the US, UK and Netherlands. According to "2014-2020 China Soil Remediation Market (2014)", around 28% of contaminated sites in China has been remediated using S/S (Du *et al.*, 2014; Quina *et al.*, 2014; Wang *et al.*, 2014; Wang *et al.*, 2015a). Extensive studies have been focused on cement-based S/S, as cement produces high strength, low permeability, and has potential to neutralize acidic wastes and precipitate heavy metals into insoluble hydroxides (La Grega *et al.*, 2001). However, it was argued that magnesia-based binders have advantages over cement-based binders and provide additional unique benefits (Shand, 2006; Al-Tabbaa, 2013). MgO was considered as having superior advantages in immobilising a wide range of contaminants because of its pH neutralisation range (9-11), has good ion-exchange ability and strong complexation ability to metal ions (Jin *et al.*, 2015). The application of MgO in stabilising/solidifying contaminated soils is a recently development (Garcia *et al.*, 2004; Jin *et al.*, 2015). Iyengar (2008) used MgO-PFA blend to treat Zn contaminated soil and found its Zn immobilisation degree was significantly higher that of PC-PFA blend. Goodarzi and Movahedrad (2017) compared the performance of ground granulated blast-furnace slag (GGBS) alone and the slag activated with cement (C-GGBS) and MgO (M-GGBS) treated Zn contaminated clayey soil and found that MgO gives a better cementation structure-bonding and pH-buffering capacity to the slag-amended soil. Du *et al.* (2016) investigated the impact of drying-wetting cycle on the engineering properties of GGBS-MgO stabilised low plasticity clay and claimed that the GGBS-MgO stabilised clay display higher dry density and lower mass loss than the PC stabilised clay. Therefore, MgO-based binders are promising materials for S/S.

Sequential extraction chemical test provides detailed information about the origin, mode of occurrence, biological and physicochemical availability, mobilisation and transport of trace metals (Filgueiras, 2002). Using sequential extraction test to study heavy metals' speciation is able to enhance stakeholders' confidence in S/S. Zhang *et* *al.* (2011) investigated the impact of different agents in stabilising heavy metal contaminated clayish soil, and reported that metal speciation involved numerous mechanisms based on metal's properties, species and the interference between metals. Li *et al.* (2001) studied the binding mechanisms and chemical partitioning of Zn in cement-based stabilised wastes. Their results showed that the leaching of Zn from the cement treated waste took place at the second and the third extraction steps. Wang *et al.* (2014) investigated the metal speciation of 17-year-old cement-based binders stabilised field soil and claimed that the leaching of heavy metals occurred at the third extraction step. All of the above mentioned studies show the function of sequential extraction in exploring binding mechanisms of heavy metals. However, rare study can be found on the application of sequential extraction in MgO-based binder treated samples.

Accordingly, this study compares the strength and heavy metal speciation of PC-based and MgO-based binders treated contaminated model soil in their early age. The detailed objectives were: 1) strength; 2) estimate the mobility of Cu, Ni, Zn and Pb in these samples; 3) compare MgO-based binders with PC-based binders in stablised/solidified contaminated soil and understand their binding mechanisms of heavy metals.

2. MATERIAL AND METHODS

The model soil was made from sharp sand, silica flour and kaolin; the composition of which was 91%:4%:5%. The values were based on site soils investigated in previous studies (SMiRT project) (Wang *et al.*2015b; Wang *et al.*2016). The particle sizes of sharp sand are ranging from 0.07 to 4 mm with a median particle size (D50) of ~0.75 mm. The chemical composition of kaolin and silt is detailed in Table S1. The particle size distribution of the model soil is detailed in Abunada (2015). The moisture content of the model soil used in this study was 10% by weight. According to the maximum concentrations of Cu, Ni, Zn and Pb in site soils from previous studies (Wang *et al.* 2014; Wang *et al.*2015a), a relative high contamination level of these metals was determined at 1500,1200, 1600 and 2500 mg/kg soil, respectively.

PC(CEM I, 52.5N), PFA, GGBS and MgO were used as binders here. The detailed compositions of these binders are listed in Wang (2015). The mix design is based on Wang *et al.*(2015a) and is shown in Table 1.

Table 1.Soil-binder constituents in percentage weight of the laboratory treated model soil (wt%).

	No	Binder components ratio				Soil	Water
		PC	MgO	GGBS	PFA		
PC-based mixes	P	8.4	-	-	-	83.3	8.3
	PF	4.2	-	-	4.2	83.3	8.3
	PG	4.2	-	4.2	-	83.3	8.3
MgO-based mixes	M	-	8.4	-	-	83.3	8.3
	MF	-	4.2	-	4.2	83.3	8.3
	MG	-	4.2	4.2	-	83.3	8.3

The specimen preparation was carried out in laboratory and was closely followed the description in ASTM D1632-07 (2007). The detailed mixing work can be found in supporting information. After being prepared, the specimens were cured under the relative humidity of ~99% and temperature of 20^{o}C±2^{o}C. These samples were then de-moulded after 9 days and 28 days.

The unconfined compressive strength (UCS) of the stablilised/solidified soil was determined using the UCS test in accordance with the ASTM D4219-08. The details of UCS and sequential extraction tests are shown in supporting information and can be found in Wang *et al.* (2014).

3. RESULTS AND DISCUSSION

3.1 Bulk Density and UCS

The bulk densities of two group mixes are presented in Fig. 1 showing generally a consistent range of 2215kg/m^3 to 2285kg/m^3 at 28 days. Figure 2 shows the corresponding average UCS values of mixes at 28 days. The deviation of these values is in the range of 0.01-0.63 MPa. It can be seen that P produced the highest strength at 28 days (~1000kPa). This was followed by M, ~660kPa. The strength values of MF and MG are lower than M, at ~200kPa. Mixes PF and PG are the weakest, <70kPa.

Only P and M are able to produce enough strength as required in the Environment Canada WTC (440 kPa) for controlled utilisation (Stegemann & Cote, 1996).

Since the early-age strength of binders is known to be influenced by the reactivity of cement blends (Li & Zhao, 2003), the reactivity of different materials is used here to explain samples' strength development. The reactivity was determined by the chemical modulus ([CaO + MgO]/SiO_2) such that, the higher the value, the more reactive the binders are (Wainwright & Rey, 2000). The reactivity of PC, MgO, GGBS and PFA used here was calculated at 4.6, 40.9, 1.3 and 0.17 in sequence (Wang, 2015).

Because the reactivity of PFA and GGBS is very low, and the presence of both PFA and GGBS was reported as retarding the hydration of PC at the early-age (Li & Zhao, 2003; Zhang *et al.*, 1998; Hogan & Meusel, 1981), the early strengths of PF and PG were lower than the strength of P as shown in Fig. 2, equally the early strengths order is: M>MG>MF. Although the strength of $Mg(OH)_2$ (the main hydration product of MgO) was found lower than that of the hydration products of PC, both MF and MG blends produced higher strengths than PF and PG blends respectively at 28 days. This agrees well with a paste study reported by Yi *et al.*(2013) that the reactive MgO activated GGBS achieved higher 28-day compressive strength than that of the equivalent $Ca(OH)_2$-GGBS system due to the larger content of the voluminous hydrotalcite-like phases formed.

Although MgO was found more efficient in activating GGBS compared to PC (Jin, 2014), MgO was not fully consumed in MG, as strong peaks of MgO was identified in MgO-GGBS at a ratio of 1:1, by X-ray diffraction (XRD) test (Yi *et al.*,2013). The unhydrated MgO may cause cracks in the treated samples, due to delayed expansions, hence reducing the strength of this mix. This is also one of the reasons why the strength of MG is lower than M at both time points. In Wang *et al.* (2015a), it was found that the average strength of the GGBS and MgO mixes at a ratio of 9:1 at 28 days in the SMiRT project was ~3.2 Mpa, which is much higher than that of MG laboratory samples. This is due to the fact that 1) the high w/c ratio (the impact of w/c was studied in Wang (2015); 2) a ratio of 9:1 is identified as the best combination in MgO-GGBS blends. Yi *et al.* (2014) claimed that the strength of GGBS-MgO blends using 90% GGBS is three times higher than that of blends using 50% GGBS in stabilising uncontaminated soil. The reason of using 1:1 in the laboratory study was to keep consistent with the PC-based samples.

3.2 Sequential extraction

The recovery rate of sequential extraction was defined as a ratio of total concentrations from 5-step sequential extraction and the full acid digestion of these samples. The recovery rates of Cu, Ni, Zn and Pb are in the ranges of 72.7-99.2%, 76.5-90%, 78.6-92.1%and 73-94.5%, respectively.

After 28-days curing, the percentages of heavy metals obtained from the extractable, carbonate, Fe-Mn oxide, organic matter and residual phases from each sample were calculated and discussed in this study. In Fig.3, it is clear that heavy metals after 28-days curing time are principally distributed in the second and third fractions.

The percentages of Zn in the extractable, organic matter and residual phases are very low compared to their starting concentrations. Clearly, except ~60% Zn in M was found mainly bounded to the Fe/Mn oxide phase, it was principally dissolved in the carbonate phase in other mixes, at a range of ~78%-90% (Fig.3a). The high percentage of Zn in the carbonate phase is due to the dissolution of ZnO and other Zn-cement hydration products (Li *et al.*, 2001a). Since PFA and GGBS retard the hydration of PC (Zhang *et al*., 1998; Hogan and Meusel, 1981; Li and Zhao, 2003), PF and PG leached more Zn in the carbonate phase than P. In addition, the high percentage of Zn leached from PF and MF in the second extraction step was in agreement with Li *et al*. (2001a) that PFA has negative effects on immobilising Zn. Compared to other mixes, M was selected as the most effective mix in treating Zn, as ~60% of Zn in M leached from the third extraction step. MG was less effective than M in immobilising Zn, while Wang *et al*., (2015a) reported that MgO-GGBS blend works better than M. This is because the field condition is different with the laboratory one, and the ratios of MgO-GGBS used in two studies are different at 1:1 and 1:9, respectively. It was reported by Jin and Al-Tabbaa (2014) that unreacted MgO was identified from MgO-GGBS paste and suggested that the slag hydration did not fulfil. Hence, the residual unhydrated MgO together with these did not fully consumed slag hydration may be the cause of the higher percentage of Zn in the second extraction step in MG compared to that in M (Yi et al., 2014a).

In Fig. 3b, ~25% Cu in PG and ~30% Cu in MG were extracted from the fourth step instead of the third step. Although Cu was reported as more stable with organic matter, the presence of organics in the model soil is very low. The possible explanation is because of the existence of Cu-hydrotalcite ($Mg_6Al_2CO_3(OH)_{16}\cdot4H_2O$)-like phases (Ht) complex, one of the hydration product of PG and MG detected by XRD and scanning electron microscopy (SEM) (Yi et al., 2014b), is more stable than other hydration products.

In Fig.3c, the percentage of Ni in MF presents the highest value in the carbonate fraction indicates that more Ni can be easily released from MF. The effectiveness of M is not as significant as its function in immobilising Zn, Cu and Pb. In Fig.3d, 0.4-3.8% of Pb was found from extractable phase. This agrees well with Wang *et al.*, (2014)'s study that Pb was more mobile than Zn, Cu and Ni. In the alkali environment, Pb precipitates into lead hydroxide, which then was transformed into a more insoluble PbO phase (Li *et al.*, 2001b), together with its adsorption binding mechanism, it was mainly released in the secondary extraction step. The benefit of using M and MG in immobilsing Pb is significant (Fig. 3d).

The sequential extraction results of S/S samples under 9 and 28 days' curing time are displaced in Fig. 4 to study the effect of curing time. The raise of percentages leached in the third/forth fraction suggesting that the stability of these metals benefits from a longer curing time.

4. CONCLUSIONS

The present study investigated the strength and heavy metal speciation of stabilized/solidified heavy metals contaminated model soil using PC-based and MgO-based binders. This study found that:

- The early-age strength of samples is influenced by the reactivity of the binders, where P (PC only), M (MgO only) produced higher strength than other mixes;
- After 28-days curing time, heavy metals were principally distributed in the carbonate

and the Fe-Mn oxide fractions in all mixes;

- GGBS put a significant impact on the Cu distribution in treated soil.
- The speciation distribution characteristics of Zn, Cu, Ni and Pb are varied;
- MgO-based binders have advantages in treating heavy metals;
- The stability of Zn, Cu, Ni and Pb benefits from a longer curing time.

Acknowledgements

This research is financially supported by the National Natural Science Foundation of China, China (Grant No. 51608113). The second author would like to thank the Killam Trusts from Canada for kindly providing the Izaak Walton Killam Memorial Postdoctoral Fellowship.

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List of Figure Captions

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Dear Editors:

Enclosed is the revised version of the paper entitled "PC-based and MgO-based binders stabilised/solidified heavy metal contaminated model soil: Strength and heavy metal speciation in early stage". I appreciated the constructive and thorough reviews provide by the journal and the positive response of both two reviewers that found the research of this manuscript is suitable for the Geotechnique. I have followed your suggestion and re-written this article into a technical note. In the revised paper, the manuscript count is cut into 2000 excluding the abstract, acknowledgements, references, figure and table legends. These changes I have made in response to reviewers' comments had been marked in Red in the body of the revised manuscript. Detailed response to their comments are as follows:

Reviewer #1:

This paper presents an interesting study comparing a few PC and MgO mixtures for S/S of several metals. The paper has the potential to be a contribution to the profession.

RESPONSE: We appreciate the positive comments about our manuscript.

The paper contains far too many grammatical errors to permit reader to fully engage in the paper. Please carefully edit this paper.

RESPONSE: Thank you for your review comment. These changes had been marked in Red in the body of the revised manuscript.

Secondly, the paper presents UC and leach test results and proceeds to identify various chemical mechanisms for any given specific result without evidence supporting the claim. No doubt the authors have reasons for the speculations but no evidence is provided in terms of test results on the specific formulations reported in the paper.

RESPONSE: Thank you so much for your very useful review comment. "detected by XRD and scanning electron microscopy (SEM)" was added in section 3.2 and was marked in Red. The findings of Yi and Jin (my previous research group members) using the X-ray diffraction (XRD) test and the scanning electron microscopy (SEM) test were used in this paper to explain my results. Later, I will follow your suggestion and do these kinds of tests myself.

To this reviewer, that is a major technical flaw in the paper. Finally, the graphics are of low quality.

RESPONSE: Thank you for your review comment. This paper had been re-written into a technical note, and some efforts had been put on these graphics.

Reviewer #2:

Present submission compares the strength and heavy metal speciation of PC-based and MgO-based binders treated heavy metal contaminated model soil in their early stage. The study is restricted to a specific model soil and to different maximum concentrations of four heavy metals: Cu, Ni, Zn and Pb. Novelty in the manuscript is a small increment regarding previous publications of the authors. Background, mainly the use of Portland cement for S/S, is poor.

So, it is the reviewer opinion that the manuscript should be enhanced and re-written as a technical note.

RESPONSE: Thank you a lot for your review comments. This paper had been re-written into a technical note.

Yours sincerely,

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Manuscript reference number: 17-P-194

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Figure1

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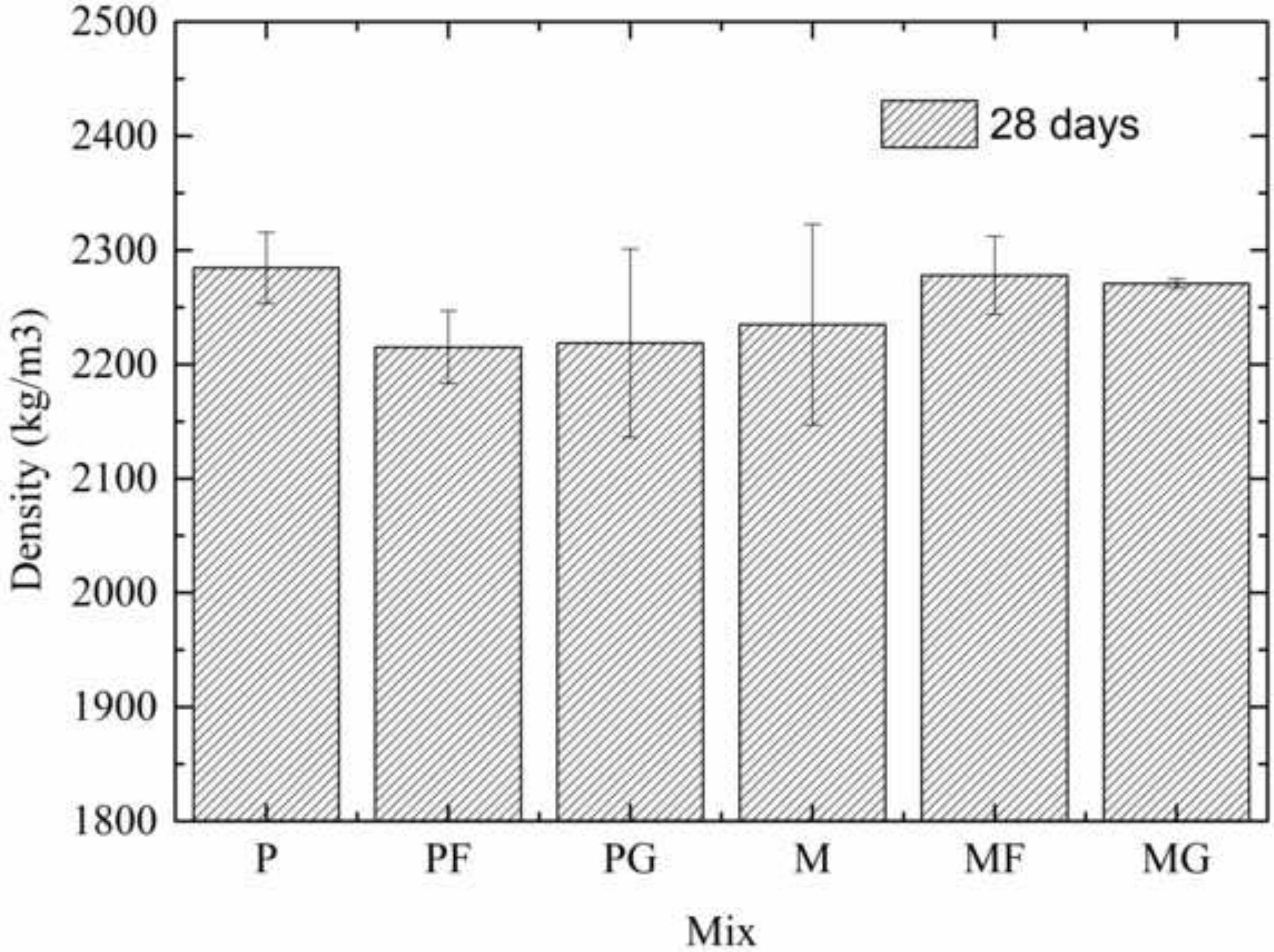

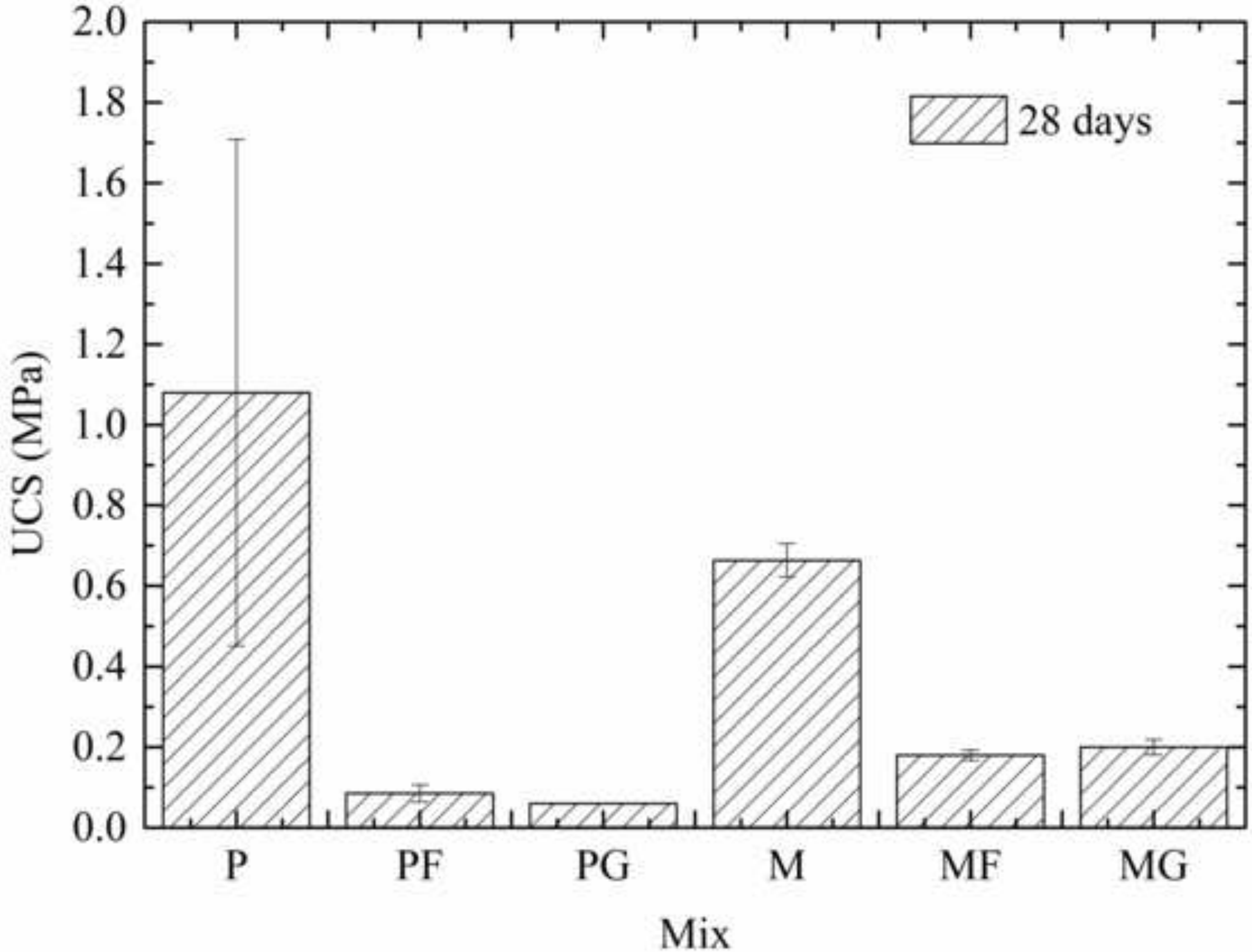

2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
UCS (MPa)
28 days
P
PF
PG
M
MF
MG
Mix

Figure 3a

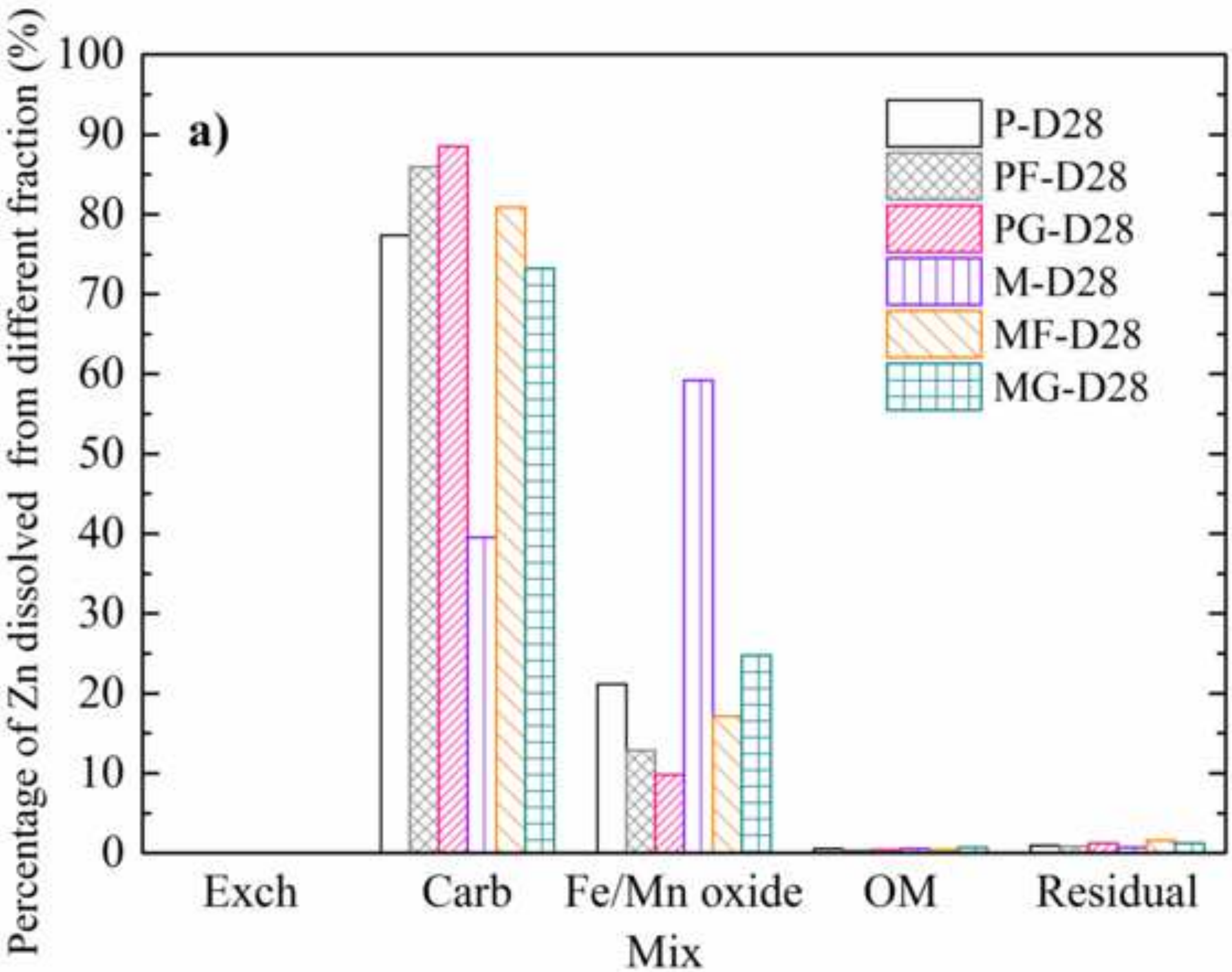
a)
Percentage of Zn dissolved from different fraction (%)
100
90
80
70
60
50
40
30
20
10
0
P-D28
PF-D28
PG-D28
M-D28
MF-D28
MG-D28
Exch
Carb
Fe/Mn oxide
OM
Residual
Mix

Figure 3b

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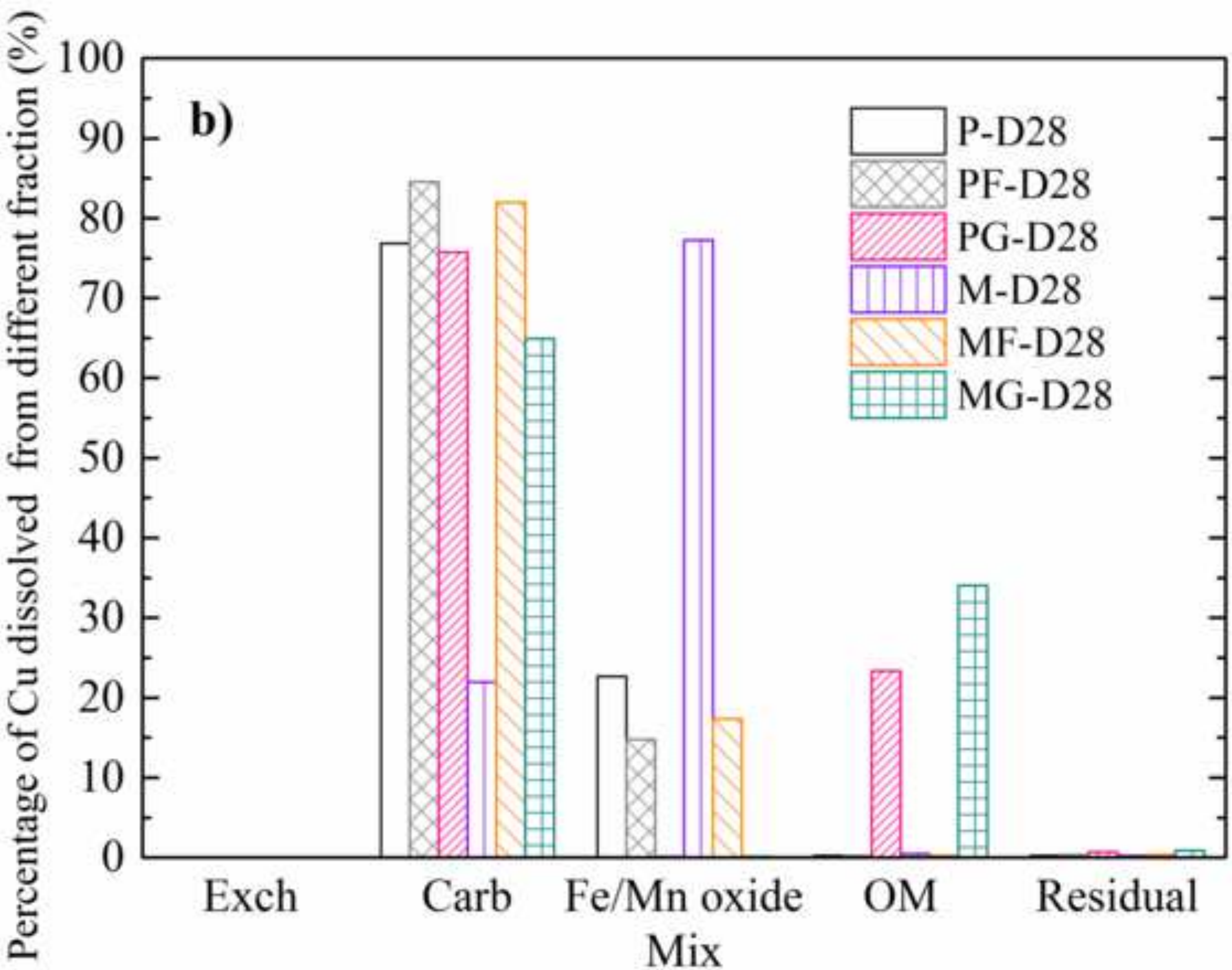

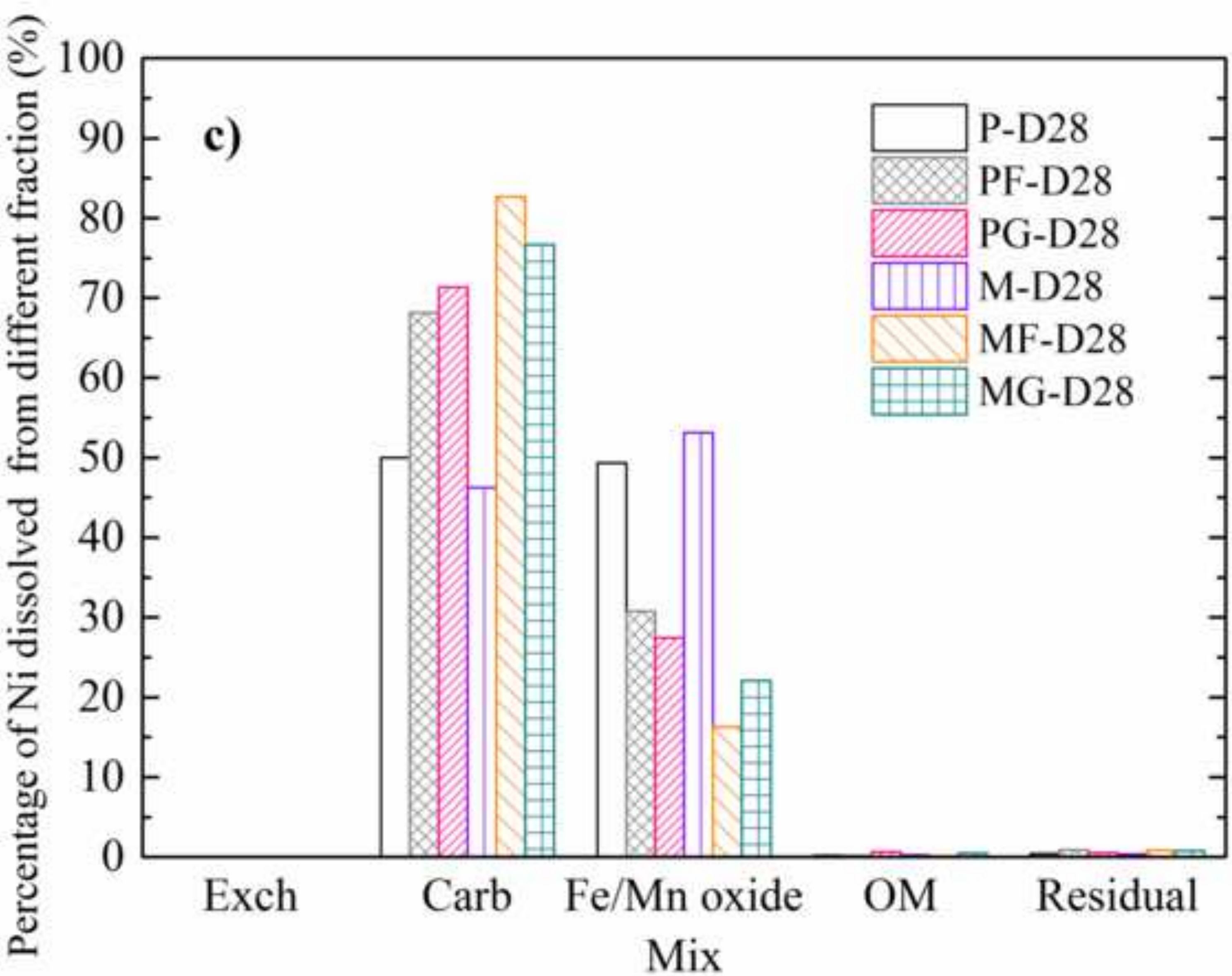
c)
Percentage of Ni dissolved from different fraction (%)
0
10
20
30
40
50
60
70
80
90
100
Exch
Carb
Fe/Mn oxide
OM
Residual
Mix
P-D28
PF-D28
PG-D28
M-D28
MF-D28
MG-D28

Figure 3d

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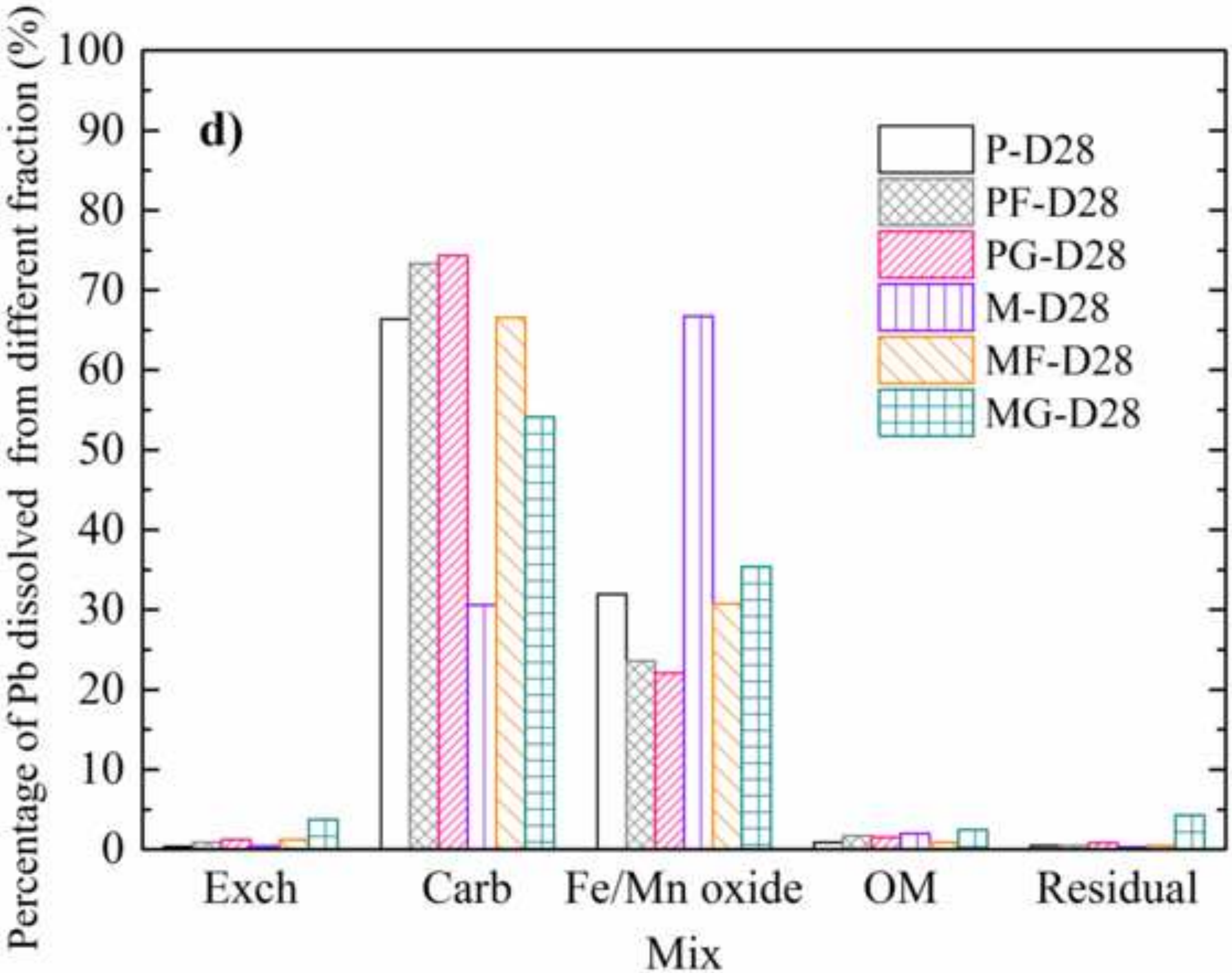

Figure 4a

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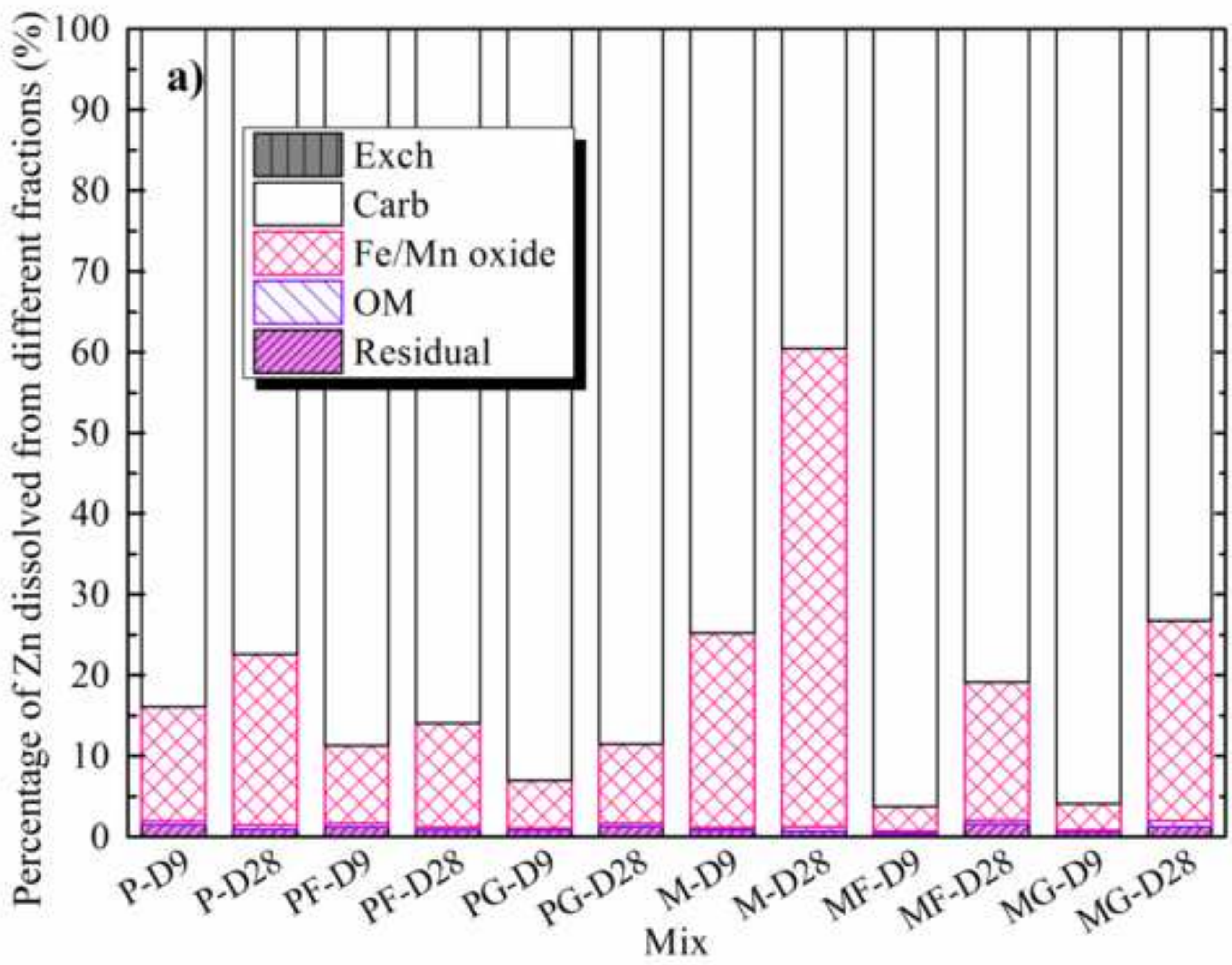

Figure 4b

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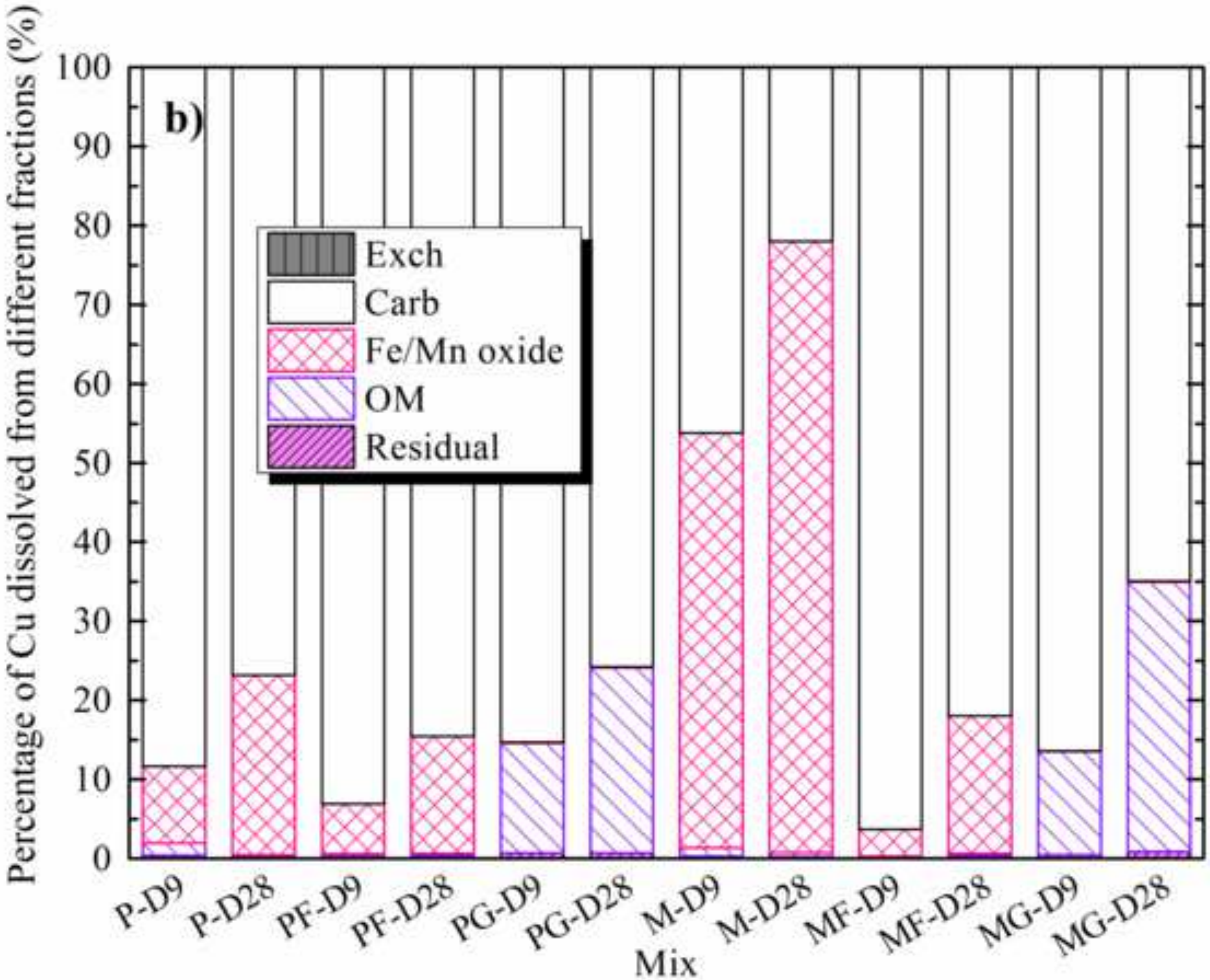

Figure 4c

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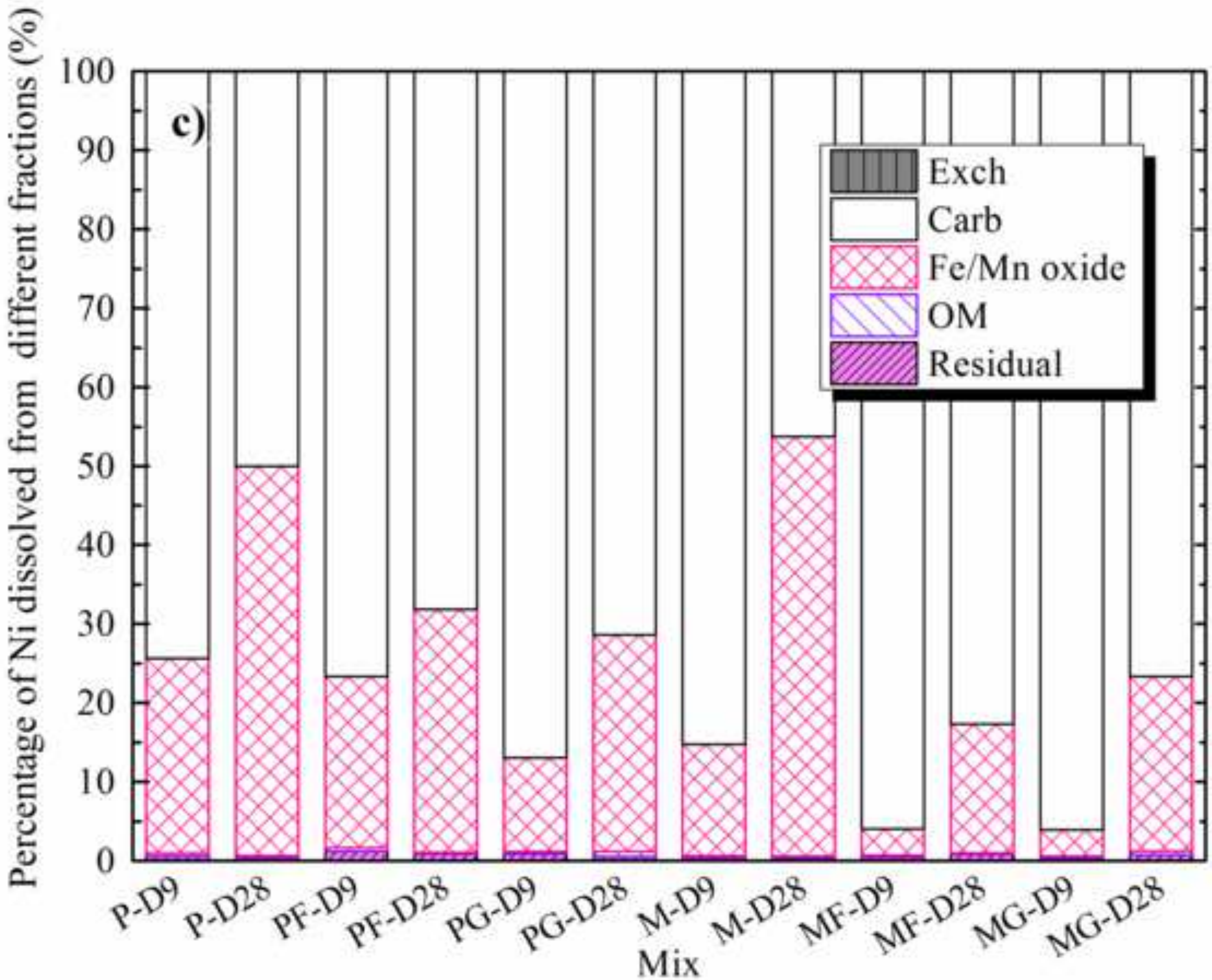

Figure 4d

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