



Performance Evaluation of Stabilised/Solidified Contaminated Model Soil Using PC-Based and MgO-Based Binders

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Abstract. The physical and chemical performances of stabilised/solidified contaminated model soil were investigated to reveal the benefit of stabilisation/solidification treatment using novel binders over conventional binders. Different combinations of binders selected from materials including Portland cement (PC), ground granulated blastfurnace slag (GGBS), pulverised fly ash (PFA) and magnesia (MgO) were mixed with contaminated soil, the water/cement (w/c) ratio at 0.5:1 was used in this study. The strength and the leaching properties of these mixes via the unconfined compressive strength (UCS) test and the batch leaching test are presented. The immobilisation degree under different mixes and strength difference under two w/c ratios are discussed. The results show that although less binder dosage was applied in mixes with a w/c ratio at 0.5:1, all these mixes produced higher UCS values than mixes with a w/c ratio at 1:1 (the ratio used in the field taken from previous studies). Moreover, the leachate concentrations of Ni, Cu and Zn in all mixes were far below their drinking water standards at 0.02 mg/l, 2 mg/l and 3 mg/l, respectively. Although most mixes cannot meet the regulative requirement of immobilising Pb, the Pb immobilisation degrees of MgO-based mixes (>99.95%) were found higher than PC-based mixes (98.8%).

Keywords: Heavy metals · Strength · Stabilisation/solidification
MgO · Soil remediation

1 Introduction

Soil contamination is one of the most important environmental issues, as it is closely related to people's daily life. Among a large amount of soil remediation techniques, stabilisation/solidification (S/S) is a well-established and widely used waste management technique (Zampetakis et al. 2014). It involves the introduction and mixing of a binder with contaminated soil and hence relies on the reaction between the binder and

soil to stop, prevent or reduce the mobilisation and hazardous nature of contaminants into the environment (Conner 1990).

In the past a few years, although many binders have been investigated, cement-based binders are the mostly used materials (Poon et al. 2001). Due to the disadvantages of cement, such as the large amount of greenhouse gases emission during production and the limited effectiveness in treating high contents of soluble sulfates etc., industrial by-products (e.g., ground granulated blastfurnace slag (GGBS) and pulverised fly ash (PFA)) were considered to replace cement for higher sustainability (Goodarzi and Movahedrad 2017). In addition, a less commonly used material, magnesia (MgO), was used alone or blended with other materials in S/S system recently (Wang et al. 2015a). It was claimed that MgO-based binders have a high priority over cement-based binders due to its high surface area, adequate pH neutralisation range and strong ion-exchange ability (Al-Tabbaa 2013; Jin et al. 2015).

Based on previous studies, it was found that the water/cement ratio (w/c) of 1:1 that used in field studies may not be optimal for laboratory studies, and a smaller water/cement ratio may result in a better stabilisation performance (Wang et al. 2018). Therefore, to study the parameter differences between laboratory study and field study, this paper presents the physical and chemical performances of stabilised/solidified heavy metal contaminated model soils using PC-based and MgO-based mixes under a w/c at 0.5:1. Unconfined compressive (UCS) strength was used to reveal the physical properties of soil. Batch leaching test was used to investigate the chemical characteristics of PC-based and MgO-based binders stabilised/solidified contaminated model soil.

2 Materials and Methods

2.1 Contaminated Model Soil and Soil Characterization

The model soil used in the present study was prepared in laboratory. In line with Wang et al. (2015a, b) and (2016), the composition of the model soil was consisted of 91% sharp sand (from Ridgeons, UK), 4% silica flour (silt, from David Ball Group, UK) and 5% kaolin (from Richard Baker Harrison, UK). The particle sizes of sharp sand range from 0.07 to 4 mm with a median particle size (D50) of ~ 0.75 mm. The detailed particle size distribution of the model soil can be found in Abunada (2015). The moisture content of the model soil is designed as 10% by weight.

The contaminants used in this study are Cu, Ni, Zn and Pb. A relative high contamination level of these metals was determined at 1500, 1200, 1600 and 2500 mg/kg soil, respectively, based on the maximum concentrations of metals reported in contaminated site soils from previous studies (Wang et al. 2014, 2015a). A mixture of analytical grade copper nitrate $\text{Cu}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, nickel nitrate $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, zinc nitrate $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and lead nitrate $\text{Pb}(\text{NO}_3)_2$, all obtained from Fisher Scientific, UK, was used to prepare the contaminated model soil. The contaminant compound and the quantities used in this study can be found in Table 1.

Table 1. The details of the metal salt compounds used in this study to spike the model soil with.

Unit		Compound			
		$\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$	$\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	$\text{Pb}(\text{NO}_3)_2$
Molecular mass	g/mol	241.6	136.9	290.79	331.2
Element mass	g/mol	63.5	65.4	58.69	207.2
Quantity spiked in the model soils	mg/kg soil	1500	1600	1200	2500

2.2 Materials and Specimen Preparation

The PC-based and MgO-based materials studied in this paper include PC (CEM I, 52.5N), PFA, GGBS and MgO. The detailed compositions of these binders are presented in Wang et al. (2018). Different binder compositions consisting of PC (P), PFA (F), GGBS (G) and MgO (M) were grouped into PC-based mixes and MgO-based mixes according to our previous work (Wang et al. 2015a). Since the w/c ratio (1:1) adopted from the field of SMiRT project did not perform well in the laboratory study, the w/c ratio used here is at 0.5:1. Table 2 presents the compositions of these mixes.

Table 2. Details of the laboratory model soil-binder mixes tested.

No.	Binder components (%)				w/c	Slurry content (%)
	PC	MgO	GGBS	PFA		
L-P2	100	0	0	0		
L-PF2	33	0	0	67		
L-PG2	50	0	50	0	0.5	9.3
L-M2	0	100	0	0		
L-MG2	0	10	90	0		

Following the description in ASTM D1632-07 (2007), the mix preparation was carried out in laboratory. One portion of the calculated water was used to mix the dry soil components and binders in a mixer, while the rest of the mixing water was used to dissolve heavy metals before adding into the model soil. The dry grout components, water and the contaminated model soil were then mixed thoroughly. The proportions of each part are shown in Table 2.

The size of split PVC moulds is at 100 mm high and 50 mm in diameter. The placement process was performed in three layers with light tamping, in order to achieve a uniform filling and compaction with minimum air entrapment. The top surface of each sample was flattened by a hand trowel and then covered with a plastic sheet to preserve moisture. These mixes were then cured in their moulds, sealed in plastic boxes (at ~99% RH) to avoid moisture loss and incubated in laboratory at $20\text{ }^\circ\text{C} \pm 2\text{ }^\circ\text{C}$. These samples were de-moulded after 28 days curing and were then subjected to different tests.

2.3 Testing Procedure

The UCS test was performed by applying a constant axial strain of 0.5 to 2% per minute based on ASTM D4219-08 using a Controls Testing Uniframe 70-T0108/E loading frame. After UCS test, the crushed samples were sieved into particle sizes between 1 and 4 mm before BSEN 12457-2 batch leaching test (BSEN 12457-2 2002). Briefly, 20 g sieved samples was mixed with 200 ml carbonated deionised water (by pumping CO₂, pH = 5.6). After 24 h of agitation at 30 ± 2 rpm, the leachate solutions were filtered through 0.45 µm filters and tested for pH values and metal concentrations with a PerkinElmer 7000 inductively coupled plasma optical emission spectroscopy (ICP-OES). Both tests were performed in triplicates.

3 Results and Discussion

3.1 UCS

Figure 1 presents the average strength results of mixes at 28 days, with a deviation range at 0.01–0.63 MPa. One set of data from previous paper was used for comparison (Wang et al. 2018). The columns represent the average strengths of mixes using a w/c ratio at 0.5:1, whereas the strengths of mixes using a w/c ratio at 1:1 were drawn as triangles. The value of L-P2 is the highest at ~1500 kPa, followed by L-M2 at ~1100 kPa and L-MG2 at ~600 kPa. L-PG2 and L-PF2 showed the lowest strengths, which could not meet the design values of 350 kPa used in the UK (Wheeler 1995). This trend agrees well with the mixes using a w/c ratio of 1:1. Less binder dosage (w/c of 5:1) resulted in higher UCS strengths of the samples compared with those at w/c of 1:1. The results highlight the importance of using appropriate parameters according to required applications.

3.2 BSEN Batch Leaching Test

The leachate pH values and metal concentrations at the end of the BSEN batch leaching test on mixes (w/c of 0.5:1) at 28 days are displayed in Fig. 2. Since the leachate concentrations of Ni in all these mixes were lower than its detection limit at 0.003 mg/L, they were not shown in Fig. 2. It can be seen that the leachate pH values of the three PC-based binders (L-P2, L-PF2 and L-PG2) were all higher than 12, while those of the two MgO-based binders (L-M2 and L-MG2) were at ~10.5 and 11.5, respectively. This is related to the pH of PC, slag and MgO/brucite alone.

Although the leachate concentrations of Pb in most mixes were higher than its drinking water standard at 0.01 mg/L, those of Cu and Zn were far below their drinking water standards at 2 mg/L and 3 mg/L, respectively (HMSO 2009). This is due to (1) the high concentration of Pb-salt at 2500 mg/kg used in the model soil; (2) the high solubility of Pb hydroxide at pH > 11 in its single metal system and a five metallic compounds system (Wang and Al-Tabbaa 2014); and (3) its adsorption mechanism (Dermatas and Meng 2003). The lower pH of MgO/brucite compared with cement lowered the Pb solubility. The high adsorption ability of brucite may also aid the immobilisation of metals (Jin 2014; Harrison 2003; Tresintsi et al. 2014). Therefore,

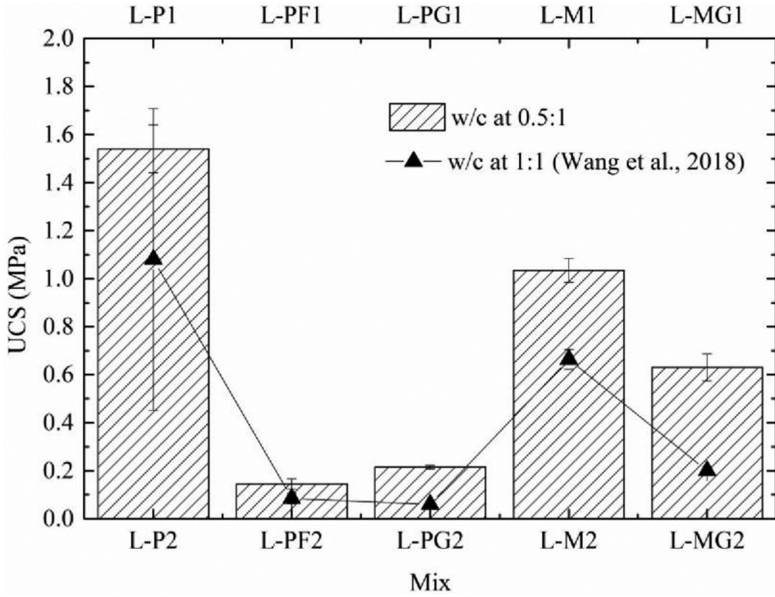


Fig. 1. The average UCS values of all samples at 28 days.

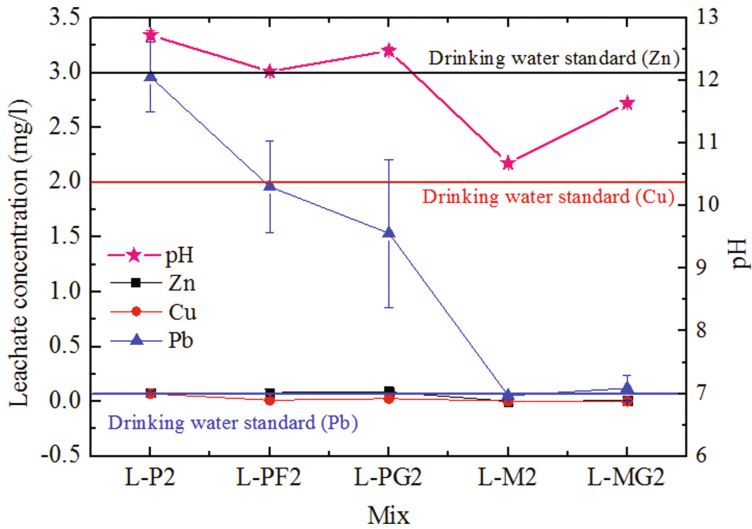


Fig. 2. The BSEN batch test leachate pH and concentrations of Zn, Cu and Pb at 28 days.

MgO-based mixes leached less Pb compared to PC-based mixes. The concentrations of Ni, Cu and Zn used in the model soil were at 1200, 1500 and 1600 mg/kg respectively. Since their solubilities were dominated by pH-controlled precipitation mechanisms, the leachate concentrations of Ni, Cu and Zn were below drinking water standards due to the suitable pH range for their immobilisation, caused by the addition of MgO-based binders. This indicates that all the mixes used in the laboratory study can immobilise these three metals effectively at 28 days. Among them, MgO-based binders were found more efficient in immobilising Pb, Ni, Cu and Zn compared to PC-based binders.

3.3 Immobilisation Degree

The immobilisation degree was calculated by dividing the immobilised fraction by its initial concentration in each mix. The immobilisation degrees of Zn, Cu and Pb obtained from BSEN test in mixes with a w/c ratio of 0.5:1 are shown in Fig. 3. For Cu and Ni, all mixes achieved >99.95% immobilisation degree, followed by Zn at >99.94%. In the PC-based mixes, the immobilisation degrees of these three metals are in the order of L-PG2 > L-PF2 > L-P2. This agrees well with the finding of Giergiczny and Krol (2008) that mineral additions in PFA and GGBS favour the formation of specific microstructure promoting the immobilisation of hazardous elements. In addition, MgO-based mixes further immobilised these metals to >99.99%, which is more efficient compared to PC-based mixes. This is in agreement with the findings of Jin and Al-Tabbaa (2014) that reactive MgO activated slag was significantly more efficient compared to lime activated slag in immobilising metals. For Pb contamination, although relatively high concentrations of Pb were leached from PC-based mixes using BSEN test, only a slightly lower immobilisation degree at >98.8% was found when comparing with other three metals. The replacement of PC with either PFA or GGBS was found to improve the efficiency of immobilising Pb. MgO-based mixes improved the immobilisation degree of Pb significantly to >99.95%, suggesting the great benefit of using MgO in treating high level Pb contaminated soil.

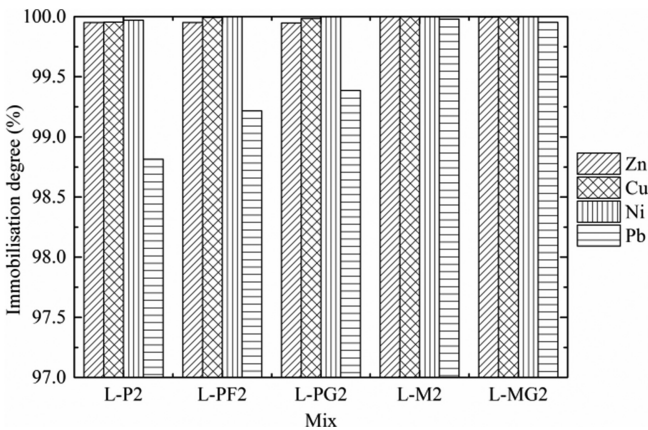


Fig. 3. The heavy metal immobilisation degree of mixes at 28 days.

4 Conclusions

The strength and leaching performance of stabilised/solidified heavy metals contaminated model soil treated by PC-based and MgO-based mixes were investigated. The main findings are as follows:

- The strengths of L-P2, L-M2 and L-MG2 at 28 days curing time met the strength requirement of the Environment Canada WTC (440 kPa) for controlled utilisation.
- Mixes with less binder dosage (w/c of 0.5:1) showed higher strengths than mixes at a w/c ratio of 1:1 (the ratio used in the field). The difference of which can be a guidance for real application.
- The BSEN leachate concentrations of Ni, Cu and Zn in all mixes were far below their drinking water standards. However, PC-based mixes cannot meet the drinking water requirement of Pb at its early stage.
- The replacement of PC with either PFA or GGBS was found effective to improve the efficiency in treating metals.
- MgO-based mixes improved the immobilisation degree of Pb to >99.95%, compared to PC-based mixes at <99.3%.

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