Chemical Engineering Journal 351 (2018) 418–427

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Low-carbon and low-alkalinity stabilization/solidification of high-Pb contaminated soil

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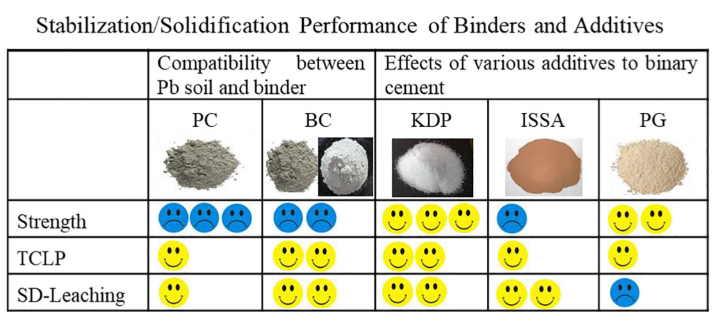
HIGHLIGHTS

- Binary cement showed superior compatibility with Pb contaminated soil.
- Pb evenly distributed on the mineral hydrates via precipitation and sorption.
- Phosphate-rich ISSA facilitated Pb stabilization and facilitated cement hydration.
- Sulphate-rich phosphogypsum promoted early strength development of S/S blocks.

GRAPHICAL ABSTRACT



PC: ordinary Portland cement; BC: binary cement; KDP: potassium dihydrogen phosphate; ISSA: incinerated sewage sludge ash; PG: phosphorus gypsum; TCLP: toxicity characteristic leaching procedure; SD: semi-dynamic.

ARTICLE INFO

Keywords:
Shooting range soil
Metal immobilization
Green remediation
Lead-phosphate precipitation
Waste recycling

ABSTRACT

Stabilization/solidification (S/S) is a low-cost and time-efficient method for soil remediation, however, delayed hydration reactions and high carbon footprint are major limitations for the treatment of high-Pb contaminated soil. This study develops a novel and low-carbon approach that combines ground granulated blast furnace slag (GGBS) and ordinary Portland cement (PC) with phosphate-/sulphate-rich byproducts to produce low-alkalinity, high-compatibility, high-strength binary cement (BC) for S/S process. Results show that contaminated soil with a large fraction of exchangeable and soluble Pb (e.g., shooting range sites) severely disturbed the formation of hydration products in conventional S/S treatment, whereas BC system could mitigate the Pb interference via precipitation and sorption evenly distributed on the BC hydrates as shown by elemental mapping. Thus, BC presented superior environmental performance in terms of toxicity characteristic leaching procedure (TCLP) and semi-dynamic leaching tests, which were substantiated by quantitative X-ray diffraction and thermogravimetric analyses. The addition of potassium dihydrogen phosphate reduced TCLP leachability of Pb by 86.9% and Pb

Abbreviations: BC, binary cement; CFL, cumulative fraction leached; CH, calcium hydroxide; C-S-H, calcium silicate hydrate; D_e, effective diffusion coefficients; GGBS, ground granulated blast furnace slag; ICP-AES, inductively coupled plasma-atomic emission spectrometry; ISSA, incinerated sewage sludge ash; KDP, potassium dihydrogen phosphate; PC, ordinary Portland cement; PG, phosphorus gypsum; QXRD, quantitative X-ray diffractometer; SD, semi-dynamic; SEM-EDX, scanning electron microscopy with energy dispersive X-ray spectroscopy; S/S, stabilization/solidification; TCLP, toxicity characteristic leaching procedure; TGA, thermogravimetric analysis; WPPF, whole powder pattern fitting; WR, water reducer; W/S, water-to-solid ratio

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https://doi.org/10.1016/j.cej.2018.06.118
Received 28 February 2018; Received in revised form 9 June 2018; Accepted 18 June 2018
Available online 20 June 2018
1385-8947/

diffusion coefficients by 69.4% due to the formation of insoluble $Pb_3(PO_4)_2$. Similarly, incinerated sewage sludge ash enhanced Pb stabilization, whereas waste phosphogypsum increased early strength via precipitation of $PbSO_4$, although the effectiveness of physical encapsulation was compromised due to reduction in hydration products. Therefore, the proposed binary binders with selected additives present a new and low-carbon S/S treatment for high-Pb shooting range soil remediation.

1. Introduction

Stabilization/solidification (S/S) is a chemical immobilization and physical encapsulation method for contaminated soil/sediment and hazardous waste [1–3], which offers many advantages such as relatively low cost, time-efficient and robust operation, and high compatibility with mixed contaminants [4,5]. However, inorganic contaminants have been recognized to interfere cement hydration reaction and compromise the hardening process and engineering properties of S/S products [6,7]. For example, Pb can react with calcium hydrates to form a dense membrane of complex mixtures (PbO, $Pb(OH)_2$, and $PbOPb(OH)_2$) and impede further cement hydration [8,9]. While most metals (e.g., Pb, Cu, Zn, Ni, and Cd) exhibit the lowest solubility at pH ~ 10, highly alkaline pore solution during cement hydration (pH > 13) leads to hydroxyl-promoted dissolution of metal precipitates [10,11]. As a result, the requirement for high content of ordinary Portland cement (PC) incurs high energy consumption and CO_2 emission [12]. Thus, low-carbon and low-alkalinity S/S approach should be developed for greener soil/sediment remediation.

Based on particle packing theory, supplementary cementitious materials and water reducer has been recently combined with PC to produce ultra-high performance concrete [13,14], which presents a cutting-edge material with high strength, low permeability, and high durability [15]. In view of these outstanding properties, this is the first study proposing an innovative and science-informed binder mixture for S/S treatment of field-contaminated, high-Pb shooting range soil. Ground granulated blast furnace slag (GGBS), an industrial by-product from steelworks, is associated with substantially low carbon footprint (60 kg CO_2 per tonne) compared to that of PC (660–820 kg CO_2 per tonne) [16–18]. As a green alternative cementitious material, the addition of GGBS can react with calcium hydroxide (CH) and generate insoluble (C-S-H) gel during cement hydration. This novel process effectively reduces the exorbitant pH increase while simultaneously enhancing metal stabilization and mechanical strength of S/S products [19]. Moreover, GGBS with small particle size can serve as filler for densifying pore structure, which facilitates physical encapsulation and low permeability [20,21]. To alleviate Pb interference with cement hydration, this study further proposes the incorporation of phosphate-rich and sulphate-rich materials, such as potassium dihydrogen phosphate (KDP), incinerated sewage sludge ash (ISSA), and phosphorus gypsum (PG), based on their proven effectiveness in recent studies [22–24].

In order to provide scientific insights for an innovative S/S technology, this study aims to: (i) assess the compatibility of PC-GGBS S/S for high-Pb shooting range soil in terms of Pb leachability and compressive strength at various dosages; and (ii) investigate the effectiveness of additives (KDP, ISSA, and PG) for enhancing Pb sequestration and long-term stability in semi-dynamic leaching. The corresponding stabilization mechanisms are elucidated through mineralogical and spectroscopic analysis.

2. Materials and methodology

2.1. Contaminated soil and S/S binary binders

The field-contaminated soil was collected from a shooting range (military site) in South Korea, where the concentration of Pb (43,198 ± 1941 mg kg^{-1}) exceeded legal regulation of Korean Military Site Soil Quality (700 mg kg^{-1}) by 61.7 times [25,26]. The exchangeable and soluble (Fraction 1) Pb contributed to 95.6% of the total Pb, whereas Fe-Mn oxyhydroxides (Fraction 2), organic matter and sulphides (Fraction 3), residual (Fraction 4) were only 1.5%, 2.5%, and 0.5%, respectively, using the fractionation analysis as described in

Table 1
Chemical compositions of soil and cementitious materials (wt%).

	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	K_2O	CaO
Soil	0.48	1.40	24.57	57.54	0.11	0.00	2.15	0.59
PC	0.17	1.38	4.79	18.99	0.08	4.52	0.79	65.72
GGBS	0.00	6.19	15.35	35.34	0.04	2.37	0.85	38.33
ISSA	6.52	3.52	13.72	27.33	9.23	2.07	2.07	10.96
PG	10.03	0.32	0.72	1.26	0.03	39.64	0.32	47.05

PC: ordinary Portland cement; GGBS: ground granulated blast furnace slag; ISSA: incinerated sewage sludge ash; PG: phosphogypsum.

Table 2
Mixture formulations (wt%) of S/S blocks with different soil to binder ratio.

	Soil	PC	GGBS	W/S	WR/S
PC	0	100	0	0.2	0.0075
S/PC = 1:1	50	50	0		
BC	0	50	50		
S/BC = 1:4	20	40	40		
S/BC = 1:3	25	37.5	37.5		
S/BC = 1:2	33.3	33.3	33.3		
S/BC = 1:1	50	25	25		
S/BC = 2:1	66.7	16.7	16.7		

PC: ordinary Portland cement; BC: binary cement; GGBS: ground granulated blast furnace slag; W/S: water to solid ratio; WR/S: water reducer to solid ratio.

Table 3
Mixture formulations (wt%) of S/BC = 1:1 blocks with different additives.

	Soil	PC	GGBS	KDP	ISSA	PG	W/S	WR/S
Pb/KDP = 4:1	50.00	24.85	24.85	0.31	0.00	0.00	0.2	0.0075
Pb/KDP = 2:1	50.00	24.70	24.70	0.61	0.00	0.00		
Pb/KDP = 1:1	50.00	24.39	24.39	1.22	0.00	0.00		
Pb/KDP = 1:2	50.00	23.78	23.78	2.44	0.00	0.00		
Pb/KDP = 1:4	50.00	22.56	22.56	4.88	0.00	0.00		
Pb/KDP = 1:8	50.00	20.12	20.12	9.76	0.00	0.00		
Pb/ISSA = 4:1	50.00	24.14	24.14	0.00	1.72	0.00		
Pb/ISSA = 2:1	50.00	23.28	23.28	0.00	3.45	0.00		
Pb/ISSA = 1:1	50.00	21.55	21.55	0.00	6.90	0.00		
Pb/ISSA = 1:2	50.00	18.10	18.10	0.00	13.80	0.00		
Pb/PG = 4:1	50.00	24.82	24.82	0.00	0.00	0.35		
Pb/PG = 2:1	50.00	24.65	24.65	0.00	0.00	0.70		
Pb/PG = 1:1	50.00	24.30	24.30	0.00	0.00	1.40		
Pb/PG = 1:2	50.00	23.60	23.60	0.00	0.00	2.80		
Pb/PG = 1:4	50.00	22.20	22.20	0.00	0.00	5.60		
Pb/PG = 1:8	50.00	19.40	19.40	0.00	0.00	11.21		

PC: ordinary Portland cement; GGBS: ground granulated blast furnace slag; KDP: potassium dihydrogen phosphate; ISSA: incinerated sewage sludge ash; PG: phosphorus gypsum. W/S: water to solid ratio; WR/S: water reducer to solid ratio.