



Solidification/stabilization of chromite ore processing residue using alkali-activated composite cementitious materials



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H I G H L I G H T S

- The optimum ratio of BFS and FA for preparing alkali-activated binder was obtained.
- Alkali-activated composite cementitious material was applied to immobilize COPR.
- The product as an available construction material has a high compressive strength.
- The immobilization effect of alkali-activated binder is much better than that of OPC.

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Chromite Ore Processing Residue (COPR) produced in chromium salt production process causes a great health and environmental risk with Cr(VI) leaching. The solidification/stabilization (S/S) of COPR using alkali-activated blast furnace slag (BFS) and fly ash (FA) based cementitious material was investigated in this study. The optimum percentage of BFS and FA for preparing the alkali-activated BFS-FA binder had been studied. COPR was used to replace the amount of BFS-FA or ordinary Portland cement (OPC) for the preparation of the cementitious materials, respectively. The immobilization effect of the alkali-activated BFS-FA binder on COPR was much better than that of OPC based cementitious material. The potential for reusing the final treatment product as a readily available construction material was evaluated. X-ray diffraction (XRD), Fourier transform infrared spectrometry (FTIR) and scanning electron microscope with energy dispersive spectrometer (SEM-EDS) analysis indicated that COPR had been effectively immobilized. The solidification mechanism is the combined effect of reduction, ion exchange, precipitation, adsorption and physical fixation in the alkali-activated composite cementitious material.

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1. Introduction

Chromium salt is widely used in the field of electroplating, leather tanning, wood preservative, catalyst, glass, ceramic, enamel, chemical reagent, organic synthesis, dyestuff, perfume, medicine and paint. In the process of chromium salt production, large amount of chromite ore processing residues (COPR) are generated (Zhang et al., 2009; Brose and James, 2013; Kameswari et al., 2015). COPR comprises of water and acid soluble hexavalent chromium ions, which is highly mobile and its toxicity is far greater than

relative stability of Cr³⁺ ions. So proper handling of COPR is necessary to save environment and human health risks (Wazne et al., 2008; Chrysochoou et al., 2009; Jagupilla et al., 2009; Wu et al., 2015). In recent years, chromium pollution increased due to ignorance of COPR. Therefore, COPR is considered as a hazardous waste, and needs much consideration regarding the treatment of its toxicity.

Stabilization/solidification (S/S) technology has been evaluated by the United States Environmental Protection Agency (USEPA) as most effective for treatment of hazardous waste (Moon et al., 2009; Yoon et al., 2010). This technology relies on reduction of mobility and toxicity of heavy metals. Ordinary Portland cement (OPC) is studied at domestic and international level for treatment of COPR but cannot be used at large scale due to certain limitations i.e. high

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leakage rate, poor durability, high capacity ratio, etc. But new type of cementitious material with blast furnace slag (BFS) or fly ash (FA) as main raw material has more advantages over OPC (Fu et al., 2011; Shen et al., 2011; Mejia et al., 2013; Lee and Lee, 2015). BFS is a waste slag from smelting iron in steel plants while FA is a powdery ash collected from boiler flue gas in thermal power plants. The main reaction product of alkali-activated BFS binder mainly involves the dissolution of calcium and the formation of C-(A)-S-H gel (CaO-Al₂O₃-SiO₂-H₂O). Whereas the main reaction product of alkali-activated FA binder is N-A-S-H gel (Na₂O-Al₂O₃-SiO₂-H₂O) with a three dimensional framework of [SiO₄] and [AlO₄] tetrahedrons. The study shows that by adding FA can significantly improve the performance of alkali-activated BFS binder, including the fluidity and setting time. Moreover, the cost of the product is also reduced (Ismail et al., 2014). On the other hand, the strength development of alkali-activated FA binder is slower due to its low activity, while by introducing BFS can improve the strength properties of alkali-activated FA binder. Certainly, the most important is that FA and BFS are two kinds of materials, which are abundant and environmental friendly (Kumar et al., 2010; Bagheri and Nazari, 2014). The key to the establishment of the cementitious material system is to check the synergistic effect of FA and BFS. Alkali-activated BFS-FA binder has low porosity and small pore size, which is favorable for reducing the leaching rate of Cr(VI), and has a high compressive strength, which is favorable for fixation of Cr(VI) in the matrix (Xu et al., 2014; Wang et al., 2015). Moreover, their hydration product has high adsorption capacity to fix metal ions, which is beneficial to the adsorption and solution of Cr(VI) (Deja, 2002; Srivastava et al., 2008; Nikolic et al., 2014). It is concluded that FA and BFS have significant advantages for the treatment of COPR, and reduces the risk of secondary pollution.

Duchesne and Laforest (2004) investigated the effect of several fixed binder on chromium ions, the results showed that the effect of BFS was better. Batchelor (2006) found that cement provides oxidation medium so reduction of Cr(VI) was not possible. Addition of BFS caused S²⁻ ions released and as a result reduction became possible. In another study, Bulut et al. (2009) used cement to solidify COPR, but the Cr(VI) leaching cannot be suppressed. Zhang et al. (2008) noted that alkali-activated FA-based geopolymer had good immobilization effect on Cd²⁺ and Pb²⁺ ions and had strong resistance to Cr(VI) leaching in presence of Na₂S as a reducing agent. These studies show that alkali-activated BFS-FA binder has inhibitory effect on the Cr(VI) leaching in the process of stabilization/solidification. It is feasible to use alkali-activated BFS-FA binder to solidify COPR. The real mechanism by which chromium ions is incorporated into the structure is not fully understood, so needs further study.

In this paper, alkali-activated BFS-FA based cementitious material was proposed to solidify COPR. This study discussed the properties of binders under different combinations of BFS-FA and

determined the optimum percentage of BFS and FA. The COPR was immobilized by the alkali-activated BFS-FA binder, and the stability of the solidified body was investigated. The OPC was used to solidify COPR, and the immobilization efficiency of OPC based cementitious material was compared with that of alkali-activated BFS-FA binder. X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy with energy dispersive spectrometry (SEM-EDS) were used to further analyse the characteristics of the binders and solidified bodies and to explore the solidification mechanism of COPR.

2. Materials and methods

2.1. Materials

The blast furnace slag (BFS) used in this work was supplied by a steel plant in Chongqing, China. The coefficients of quality was 1.53. Class F fly ash (FA) was collected from a power industrial plant in Chongqing, China. Chromite ore processing residue (COPR) was obtained from a chemical plant in Chongqing, China. Ordinary Portland cement (OPC) used in the experiments was from a cement plant in Chongqing, China. The chemical composition of raw materials is listed in Table 1. Alkali activators used in the experiments were NaOH (Analytical grade) and water glass (Industrial grade). The water glass composed of SiO₂ (56.03%) and Na₂O (19.30%) with density of 1.46 g cm⁻³ and modulus was 3.0.

2.2. Methods

2.2.1. Mechanical activation of BFS and FA

Mechanical activation of BFS and FA was carried out in a ball-grinding mill. Basically ball-grinding mill used steel balls for grinding of samples. The mass ratio of materials (BFS and FA) and steel balls was 1:6. The samples of BFS and FA were mechanically grinded for 12 h. While the rotation speed of the ball-grinding mill was 250 rpm. Mechanically activated BFS and FA passed through a 200-mesh sieve.

2.2.2. Sample preparation

The mixtures were prepared by mixing BFS-FA and alkali activator solution with “alkali activators/BFS-FA” ratio of 1:9. The alkali activator solution was obtained from NaOH and water glass dissolved in deionized water and cooled to room temperature, the mass ratio of water glass/NaOH was 7:3 whereas the liquid-to-solid ratio was 0.25. Then BFS/FA was added into the above alkali activator solution and was stirred for 10 min until homogeneous colloid was formed. The colloid was then placed into the stainless steel cubes (20 mm × 20 mm × 20 mm) and vibrated for 5 min to remove entrapped air bubbles and then cured in a curing box with a constant temperature of 25 °C and 90% humidity. After 24 h, the

Table 1

The chemical composition of raw materials (w%).

Material	CaO	SiO ₂	Al ₂ O ₃	MgO	TiO ₂	SO ₃	Fe ₂ O ₃	K ₂ O
BFS	38.32	32.06	12.62	7.35	5.58	1.80	0.65	0.51
FA	7.17	45.73	24.75	0.79	3.17	1.58	14.13	1.11
OPC	68.78	17.72	3.36	1.57	0.38	3.46	2.79	1.16
COPR	0.72	5.82	12.34	14.54	1.11	0.08	47.94	–
Material	MnO	Na ₂ O	Cr ₂ O ₃	ZrO ₂	P ₂ O ₅	CuO	ZnO	LOI ^a
BFS	0.49	0.30	–	0.04	0.02	–	–	0.26
FA	0.07	0.83	–	0.08	0.18	0.02	0.01	0.38
OPC	0.02	0.02	–	–	0.05	–	–	0.69
COPR	–	5.18	11.73	–	0.01	–	0.18	0.35

LOI^a: Loss on ignition at 1000 °C.

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