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Heavy metals leaching in bricks made from lead and zinc mine tailings with varied chemical components

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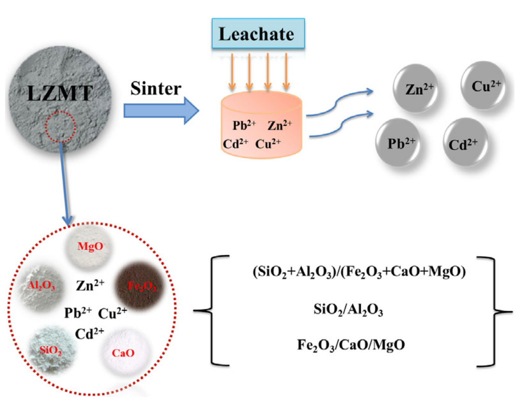
HIGHLIGHTS

- Impacts of SiO_2:Al_2O_3 and Fe_2O_3:CaO:MgO on metals leaching were examined.
- Leaching of Zn, Pb are highly depended on the chemical compositions in bricks.
- Presence of $PbAl_2Si_2O_8$ could facilitate the immobilization of Pb^{2+} in bricks.
- Higher C_{Zn} could be attributed to the formation of Zn_2SiO_4 in bricks.
- Concentrations of Cd, Cu, Pb, Zn in leachate are all below the standard.

GRAPHICAL ABSTRACT



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ABSTRACT

The stabilization/solidification approach has received considerable attentions for the disposal of hazardous metal-bearing mine tailings. A poor understanding of the immobilization mechanism of heavy metals in final products always restricts its practical applications. We reported that, the stabilization of toxic heavy metals (Zn, Pb, Cd and Cu) in bricks made from the lead and zinc mine tailings (LZMT) was highly dependent on the relative contents of SiO_2, Al_2O_3, Fe_2O_3, CaO and MgO in LZMT. As the $(SiO_2 + Al_2O_3)/(Fe_2O_3 + CaO + MgO)$ ratios (SA/FCM ratios) increased, the C_{Pb}, C_{Cd}, or C_{Cu} in leachates slightly decreased first and then increased. Much differently, C_{Zn} firstly increased and then decreased, having highest C_{Zn} (3.92 mg/L at 30th day) at the SA/FCM ratios of 0.80. Compared with C_{Zn} and C_{Pb}, a much small C_{Cd} and C_{Cu} can be found. Notably, the C_{Zn} and C_{Pb} are also impacted by the change of SiO_2:Al_2O_3 ratios (and Fe_2O_3:CaO:MgO ratios as well). The C_{Pb} decreased as the SiO_2:Al_2O_3 ratios increased from 1:0.046 to 1:1.354, while the C_{Zn} reached a highest value at SiO_2:Al_2O_3 ratio of 1:0.084. The XRD analysis suggests that, the Al and Si can interact with Pb to form $PbAl_2Si_2O_8$, which facilitates the immobilization of Pb^{2+}. Excessive amount of SiO_2 and Al_2O_3 bring the difficulties to forming stabile skeletons, resulting in the leaching of Pb readily. As for Zn, higher C_{Zn} is generally owing to the occurrence of acid-soluble Zn_2SiO_4 in bricks. The stabilization of Zn was achieved once Al^{3+} was introduced as host to be substituted by Zn^{2+} in the aluminate or aluminosilicate matrix. The findings could enrich the mechanistic understanding and pathway of heavy metals immobilization in eco-environmental products from LZMT.

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1. Introduction

The rapid development of industrialization has produced ever-growing amount of hazardous mine tailings. According to a statistics, more than 5.97 billion tons of mine tailings have been discarded in China till 2011 [1]. These solid-state mine tailings, such as lead and zinc mine tailing (LZMT), are frequently rich in toxic heavy metals, such as Pb, Zn, Cu, Cd, Mn, Ni, As, and so on [2–4]. Leaching of the toxic heavy metals inevitably will pose risks to the environment and public health worldwide [5–7].

Over the past years, many technologies have been proposed to minimize the environmental impact of these hazardous mine tailings [1,8–10]. The stabilization/solidification (S/S) approach has proved to be one of the most promising method [11] available for practical applications. In S/S method, sintering of hazardous wastes with solidified matrix not only helps to immobilize the heavy metals [12,13], but also might transfer the wastes into eco-friendly products, such as bricks, lightweight aggregates and ceramsites [14–18]. One of the keys to support the successful application of S/S method is to prevent the leaching of the heavy metals from the final products. To achieve this goal, efforts have been dedicated to searching the way by adding curing agents, or other materials (e.g. cement, silica fume, sodium silicate, potassium dihydrogen phosphate and ferric chloride hexahydrate [19–22]). Generally, the addition of additional curing agents or other materials are beneficial for heavy metals immobilization. Notwithstanding these advantages, the use of curing agents could increase the cost, which greatly restricts their practical applications. Another problem remains unsolved is that, a part of immobilized heavy metals (in mine tailings) might also be leached out, if the environmental conditions changed (e.g. the pH of leachates, and the types and amount of curing agents) [23]. Hence, a systematical study on immobilization mechanism of heavy metals in mine tailings is still highly desirable for finding a cost-effective way. Much recently, a few researches start to realize that, the leaching behavior of the heavy metals in the final products seems to have a close relationship with the chemical compositions of the products themselves [24,25]. It is possible that, the solidification characteristics of the heavy metals in sludge were strongly influenced by the mass ratios of acidic and basic oxides. However, it still lacks fundamental researches in this field. It brings great inspiration to the study of heavy metals immobilization in products made by mine-tailings as precursor.

In this work, the leaching behavior of heavy metals from a LZMT sample, which was collected from a mining company in a province of China, was investigated. This process is beyond the use of clay (the popular raw materials in bricks fabrication), the calcination of which usually much high temperature over 1100 °C [16,26,27]. In this work, to investigate the influence of the content of oxides on metals leaching, proportional amounts of SiO_2, Al_2O_3, Fe_2O_3, CaO, and MgO were added into the tailing to simulate tailings with different minerals ratios. The relationships between the leaching of heavy metals (particularly Zn and Pb) and chemical composition of the tailing were investigated. In briefly, the impact of the ratios of $(SiO_2 + Al_2O_3)/(Fe_2O_3 + CaO + MgO)$, and the ratios of SiO_2:Al_2O_3 and Fe_2O_3:CaO:MgO on metal leaching was investigated. A possible mechanism for the immobilization of Zn, Pb and other metals in mine tailings vitrified bricks was proposed. We anticipate the findings here can provide a general guide for managing the mine tailings with different compositions.

2. Experiment section

2.1. Materials

The LZMT used in this study was collected from a Cement Co. Ltd. in Fujian, China. The dewatering of LZMT was performed with a belt filter press, wherein the cationic polymeric flocculants were used. Subsequently, the LZMT was dried at 120 °C for 12 h in an electric oven thermostat [28]. After that, the dried LZMT was grounded into powder with uniform particle size for subsequent use.

As shown in Table 1, the major chemical composition of LZMT was determined by a Philips MagiX PRO XR spectrometer (X-ray fluorescence-XRF, Amsterdam, The Netherlands). The phases of minerals in LZMT were further identified by Powder X-ray diffraction (XRD) (Bruker D8 Advance) using Cu (Ka) radiation (40 kV, 40 mA).

As shown in Table 1, the heavy metals in LZMT mainly include the Zn, Cu, Pb, Mn, Cd, and so on. In addition, the relative content of SiO_2, Al_2O_3, Fe_2O_3, CaO and MgO in the LZMT is 32%, 3.38%, 18.1%, 21.3% and 1.47%, respectively. In the following experiments, small amount of analytical-grade oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO and/or MgO) were added into the LZMT. After that, the mixed samples were sintered in Muffle Furnace at 1000 °C by procedure showing in the following. All of the used oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO and MgO) are of sizes below 10 um.

2.2. Methods

As illustrated in Scheme 1, the LZMT and metal oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO and/or MgO) were mixed by stirring in a blender for 10 min. After that, H_2O (about 1–3 wt%) was added to the mixture under stirring for another 10 min to form the semi-dried molded brick samples. As-generated mixtures were pressed into cylinder (diameter of 40 mm and length of 40 mm) for producing brick specimen, under a pressure of 15 MPa using a hydraulic press (WE-30B). After that, the specimens were dried in an oven at 105 °C for 8 h, and subsequently, was sintered. The temperature was set at 20 °C at initial, and it rose from 20 to 1000 °C, with a duration time of 20 min at 850 °C for and 60 min at 1000 °C. Finally, the products were cooled to the room temperature.

According to the "Chinese standard solid waste extraction procedure for leaching toxicity – acetic acid buffer solution method" (HJ/T 300-2007), the leach ability of brick samples was determined. The acetic acid (CH_3COOH) solution ($pH = 2.64 \pm 0.05$) and as

Table 1
Components Analyses of the Dried LZMT (wt.%).

Components analyses								
SiO_2	CaO	Fe_2O_3	Al_2O_3	MgO	K_2O	ZnO	Others	Carbonaceous Matter
32.0	21.3	18.1	3.38	1.47	0.484	0.281	<6.185	<16.8
Elements analyses								
Zn	Fe	Mn	Si	Cu	Ca	Mg	O	Pb
0.23	12.7	0.2	15.0	0.02	15.30	0.88	45.6	0.17
P	Na	K	Al	Ti	C	S	Cd	Others
0.03	0.05	0.40	1.79	0.07	4.57	1.22	0.01	<1.76