



# Formation of lead ferrites for immobilizing hazardous lead into iron-rich ceramic matrix

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## HIGHLIGHTS

- Lead was incorporated to lead ferrites (i.e.  $\delta$ -,  $\gamma$ - and  $\beta$ -phase) by hematite.
- Crystal structure transformation between  $\delta$ -,  $\gamma$ - and  $\beta$ - phase was detected.
- $\delta$ -,  $\gamma$ - and  $\beta$ -phase dominantly formed at Pb/Fe of 1/(1–3), 1/(4–7) and 1/(7–12).
- Incongruent dissolution of  $\delta$ -,  $\gamma$ - and  $\beta$ -phase in leaching test was observed.

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## ABSTRACT

A strategy of immobilizing lead in the framework of ferrite-ceramic matrix, to reduce its environmental hazard was explored in this study. The mechanisms of incorporating lead into lead ferrites ( $\delta$ -phase ( $2\text{PbO} \cdot \text{Fe}_2\text{O}_3$ ),  $\gamma$ -phase ( $\text{PbO} \cdot (2-2.5)\text{Fe}_2\text{O}_3$ ) and  $\beta$ -phase ( $\text{PbO} \cdot (5-6)\text{Fe}_2\text{O}_3$ )) was revealed by observing the phase transformation in the products. The  $\delta$ -phase was dominantly formed at low temperature of 700–800 °C at Pb/Fe of 1/1–1/3. The significant growth of  $\gamma$ -phase was observed at 750–850 °C and Pb/Fe of 1/4–1/7. The  $\beta$ -phase substantially formed at 900–1000 °C with Pb/Fe of 1/7–1/12. The transformation of  $\delta$ -phase to  $\gamma$ -phase and/or  $\beta$ -phase indicated the destruction of  $\delta$ -phase unit and reconstruction of  $\gamma$ -phase and  $\beta$ -phase units during sintering process. However, the transformation of  $\gamma$ -phase into  $\beta$ -phase suggested a structure conversion process, local structural changes arose as a consequence of the addition of  $\text{Fe}_2\text{O}_3$ . When comparing the leaching ability of  $\delta$ -,  $\gamma$ - and  $\beta$ -phase, the results showed the superiority of  $\beta$ -phase for lead immobilization over the longer leaching period.

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## 1. Introduction

Lead (Pb) is a metal widely used for various technical purposes due to its versatile physical and chemical properties. Thus, lead-containing wastes increases constantly in the manufacturing of

lead batteries and oil-based paints, mining, plating, electronics and wood production (Conrad and Hansen, 2007; Venäläinen, 2011). The release of lead into the environment from its manufacturing industries is of serious environmental concern. Since lead is highly toxic and a non-biodegradable metal that tends to accumulate in the cells of living organisms, which causes severe damage to the kidneys, liver, and the nervous and reproductive systems of humans (Gupta et al., 2011). Therefore, remediation of the lead-containing wastes has become more and more important in recent years owing to the reinforcement in the environmental protection.

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Ceramic technologies are considered to be the versatile for rendering hazardous waste inert because they immobilize hazardous metals in a stable crystallization phase to reduce the hazardous metal leachability in waste materials (Xu et al., 2008, 2009). The immobilization mechanism is strongly dependent on phase transformation and evolution of heavy metals during the sintering process (Li et al., 2017; Li et al., 2011a,b; Hu et al., 2010). In addition, the leaching behaviors of heavy metals from the ceramic matrix are determined by the crystal structure of the metal-hosting phases (Hsieh et al., 2013; Lu et al., 2008). Besides aluminium and silica oxide, a commonly used material in ceramic processing is hematite ( $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>), which behaves as a refractory antileakage material in combination with alkalis. Owing to its relatively low cost and wide availability, hematite is known as a feasible matrix for the immobilization of various hazardous metals in waste, and shown effective stabilization ability (Mao et al., 2014; Li et al., 2011a,b; Shih et al., 2006). By sintering with hematite precursor, the hazardous Ni, Cu, Zn and Cd can be fairly fixed into ferrite spinal structures, and the hazardous metal leachability was significantly reduced (Shih et al., 2006; Su et al., 2017, 2018; Li et al., 2017). However, potential reactions between lead and hematite may strongly influence the leachability of hazardous metals from the matrix. To create a successful lead stabilization technology, a comprehensive understanding of the mechanisms for transforming lead into ferrite-ceramic matrix is crucial.

Because the composition of metals in waste streams can fluctuate and it would cause the varied stoichiometric values of Pb/Fe for ferrite synthesis. Furthermore, the relative incorporation of Pb and Fe was reported depends critically on the initial Pb/Fe molar ratio (Sahu et al., 2012; Diop et al., 2010). Previous studies have identified 11 ferrite complexes in PbO-Fe<sub>2</sub>O<sub>3</sub> system, i.e., 8PbO·Fe<sub>2</sub>O<sub>3</sub>, 3PbO·Fe<sub>2</sub>O<sub>3</sub>, 2PbO·Fe<sub>2</sub>O<sub>3</sub>, 3PbO·2Fe<sub>2</sub>O<sub>3</sub>, PbO·Fe<sub>2</sub>O<sub>3</sub>, PbO·2Fe<sub>2</sub>O<sub>3</sub>, 2PbO·5Fe<sub>2</sub>O<sub>3</sub>, PbO·3Fe<sub>2</sub>O<sub>3</sub>, PbO·4Fe<sub>2</sub>O<sub>3</sub>, PbO·5Fe<sub>2</sub>O<sub>3</sub> and PbO·6Fe<sub>2</sub>O<sub>3</sub> (Sahu et al., 2012; Diop et al., 2010; Rivolier et al., 1993; Raghavan, 1989). These ferrite complexes with different initial stoichiometric ratios were considered as the potential lead ferrites in sintered PbO/Fe<sub>2</sub>O<sub>3</sub> products (Sahu et al., 2012, 2013; Diop et al., 2010). Among these complexes,  $\delta$ -phase (2PbO·Fe<sub>2</sub>O<sub>3</sub>),  $\gamma$ -phase (PbO·(2–2.5)Fe<sub>2</sub>O<sub>3</sub>) and  $\beta$ -phase (PbO·(5–6)Fe<sub>2</sub>O<sub>3</sub>) were determined to be stable phases (Sahu et al., 2012, 2013; Diop et al., 2010). Although the formation of lead ferrites from a PbO-Fe<sub>2</sub>O<sub>3</sub> system is feasible, the reaction sequences involved in the incorporation of lead under different thermal conditions with various Pb/Fe molar ratios has remained unclear. To determine the potential mechanistic processes of lead incorporation, it is essential to identify the structures of crystalline phases in ceramic products. Furthermore, the robustness of the product(s) to acids should be examined with leaching tests to verify the metal stabilization effects. It has been demonstrated that constant-pH leaching test is a useful tool to assess the leachability of products, because CPLT could maintain the pH in a steady level (Al-Abed et al., 2007; Islam et al., 2004; Jackson et al., 1984). In TCLP leaching test, the decomposition of metal-containing crystal framework and cation exchange with solution protons may lead to the significant increase of leachate pH (Shih et al., 2006; Tang et al., 2011; Lu et al., 2013). However, the pH in constant-pH leaching experiments leachability can maintain stable. Thus, the leachability of different metal structures can be compared in the same condition.

The objective of this work is to in-depth study the lead stabilization mechanisms with hematite. The XRD technique was used to identify the structures of crystalline phases, and the metal incorporation mechanisms were further analyzed based on the XRD results. At the end of the study, the constant-pH leaching tests were carried out to evaluate the intrinsic phase durability and metal leachability in sintered products.

## 2. Materials and methods

### 2.1. Materials

PbO is common forms in the sewage sludge after thermal processes (Udayanga et al., 2018), in this study PbO (Sigma-Aldrich) was used as the lead source, representing its major form in the thermally treated lead-laden sludge. The PbO powder was identified by X-ray diffraction (XRD) assisted with the Powder Diffraction File (PDF) database published by International Centre for Diffraction Data (ICDD) as a mixture of the litharge ( $\alpha$ -PbO; ICDD PDF #77–1971) and massicot ( $\beta$ -PbO; ICDD PDF #05–0561) phases. Hematite powder was purchased from Sigma-Aldrich, and its XRD pattern matched that of standard hematite peaks (ICDD PDF #78–1996). The PbO and Fe<sub>2</sub>O<sub>3</sub> powders were calcined in air at 300 °C for 3 h to remove the moisture content before use.

### 2.2. Thermal reaction

To evaluate the feasibility of Pb incorporation, hematite precursor and PbO were mixed by ball milling in water slurry with Pb/Fe molar ratios of 1/1, 1/4 and 1/12 for 18 h. The ball milling was conducted in a planetary ball mill consisting of four tungsten carbide grinding jars (100 ml each) at a rotation speed of 500 rpm. Sequentially, the mixtures were dried at 105 °C for 48 h in a vacuum oven, and then further homogenized by extended mortar grinding for 10 min in an agate mortar. Before thermal treatment, the derived powder was pressed into 20-mm pellets at 650 MPa to ensure consistent compaction of the powder for the sintering process. For the incorporation reaction, a well-control thermal treatment scheme at the targeted temperature was applied. In most cases, the dwell time was fixed at 3 h for sintering temperatures ranging from 600 °C to 1050 °C.

### 2.3. X-ray diffraction

After sintering, the samples were ground using an agate mortar and pestle to a particle size of no more than 10  $\mu$ m for XRD analysis. The powder XRD patterns were recorded on a Bruker D8 Advance X-ray powder diffractometer equipped with Cu K $\alpha$  radiation and a LynxEye detector. The diffractometer was operated at 40 kV and 40 mA, and the  $2\theta$  scan range was from 10° to 80°, with a step size of 0.02° and a scan speed of 0.3 s per step. Phase identification was performed with the Eva XRD Pattern Processing software (Bruker, Germany) by matching the powder XRD patterns with those retrieved from the standard powder diffraction database published by the ICDD.

### 2.4. Leaching tests

The potential leaching ability of lead-bearing products were compared by constant-pH leaching experiments. The target product powder (<45  $\mu$ m) was subjected to batch leaching test in the plastic vessels at a liquid/solid ratio of 50 L/kg (4 g products powder and 200 ml pH 5.0 acetic acid solution) and a total equilibration time of 168 h. Acid (1 N HNO<sub>3</sub>) was added into plastic vessels to maintain the pH constant at 5.0 by a pH stat system. The continuously stirring was conducted to keep the mixture homogeneous and the leaching vessel was maintained at room temperature (23  $\pm$  0.5 °C). At different record times, the suspensions were allowed to settle for 5 min and small aliquots of 5 ml of the upper liquid were taken and centrifuged. Another 5 ml pH 5.0 acetic acid solution together with the filtered solids was added to the vessels. At the end of each agitation period, the leachates were filtrated using 0.2  $\mu$ m syringe filters. The total concentrations of Pb and Fe

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