



Metal stabilization mechanism of incorporating lead-bearing sludge in kaolinite-based ceramics

Xingwen Lu, Kaimin Shih*

Department of Civil Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong, Hong Kong SAR, China

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ABSTRACT

The feasibility and mechanism of incorporating simulated lead-laden sludge into low-cost ceramic products was investigated by observing the reaction of lead with two kaolinite-based precursors under sintering conditions. To investigate the phase transformation process of lead, lead oxide (PbO) mixed with a kaolinite or mullite precursor were fired at 500–950 °C for 3 h. Detailed X-ray diffraction analysis of sintered products revealed that both precursors had crystallochemically incorporated lead into the lead feldspar ($\text{PbAl}_2\text{Si}_2\text{O}_8$) crystalline structure. By mixing lead oxide with kaolinite, lead feldspar begins to crystallize at 700 °C; maximum incorporation of lead into this structure occurred at 950 °C. However, two intermediate phases, $\text{Pb}_4\text{Al}_4\text{Si}_3\text{O}_{16}$ and a polymorph of lead feldspar, were detected at temperatures between 700 and 900 °C. By sintering lead oxide with the mullite precursor, lead feldspar was detected at temperatures above 750 °C, and an intermediate phase of $\text{Pb}_4\text{Al}_4\text{Si}_3\text{O}_{16}$ was observed in the temperature range of 750–900 °C. This study compared the lead leachabilities of PbO and lead feldspar using a prolonged leaching test (at pH 2.9 for 23 d) modified from the toxicity characteristic leaching procedure. The results indicate the superiority of lead feldspar in stabilizing lead and suggest a promising and reliable strategy to stabilize lead in ceramic products.

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1. Introduction

A variety of treatments based on high-temperature phase transformation have been used to stabilize radioactive waste in vitrified glass or ceramic materials (Lewis et al., 1993; Wronkiewicz et al., 1997). However, the end products of these processes are radioactive and must be stored in geologic repositories. For non-radioactive hazardous metals, nickel and copper can be stabilized with well-controlled thermal treatment schemes via the formation of spinel structures (Shih et al., 2006a,b; Tang et al., 2010). The use of these products as raw materials for marketable ceramic products has also been proposed. In previous studies, hazardous metals were incorporated into aluminum-based precursors by forming spinel-type crystalline products during the sintering process; the metal leachability of these sintered products was significantly reduced.

Most common ceramic products are made from natural clays and silicates. Kaolin, which consists chiefly of kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$), is a common clay type, and most ceramics contain high percentages (40–65%) of kaolinite (Velde, 1992; Reed, 1995). Because it is widely available, kaolinite is one of the most commonly used starting materials for aluminosilicate-based ceramics (Sainz et al., 2000). Calcined kaolin is a primary byproduct generated by heating spray-dried kaolin particles to render them anhydrous through

their transformation into mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) and silica (SiO_2) (MacKenzie et al., 1996; Chen and Tuan, 2002). Mullite, the only stable binary crystalline phase in the SiO_2 – Al_2O_3 system, can be produced by thermal decomposition of kaolinite (Lee et al., 2008). Recent publication of the PbO – Al_2O_3 – SiO_2 phase diagram has provided an opportunity to form lead feldspar at equilibrium (Chen et al., 2001). Understanding whether this incorporation mechanism can be initiated during the industrial sintering of ceramic products, which may be subject to limited processing time and the use of clay-based precursors, is crucial in determining the stability of hazardous lead in the products. To investigate the metal incorporation mechanism(s) of lead-laden sludge under ceramic sintering conditions, kaolinite-based materials were used as inexpensive ceramic precursors in this study.

Using acetic acid (pH 2.9) as a leaching fluid to simulate the presence of organic materials in municipal landfill leachate (US EPA, 1998; MacKenzie et al., 2000; Halim et al., 2004), the toxicity characteristic leaching procedure (TCLP) simulates the worst-case scenario for co-disposing of waste materials in landfill. However, the TCLP produces a single-point, short-term leaching result and its use in predicting leaching may result in misclassification of materials, which consequently leads to the underestimation of long-term leachability or to unnecessary treatment cost (Kosson et al., 2002). Extending the duration of this standard leaching procedure may be more useful to determine the long-term leaching behavior of metal-bearing products in the environment.

* Corresponding author. Tel.: +852 2859 1973; fax: +852 2559 5337.

E-mail address: kshih@hku.hk (K. Shih).

In this study kaolinite and mullite precursors were assessed for their potential to incorporate lead into aluminosilicate structures through ceramic sintering. The primary goals of the sintering experiments were to observe lead phase transformation under varying thermal conditions and verify the potential formation of lead feldspar in the sintered products. Finally, a prolonged leaching procedure modified from the TCLP was carried out to evaluate the long-term stabilization of lead in the product phases.

2. Materials and methods

As most metals in sludge oxidize into respective metal oxides under high temperature, PbO (Sigma–Aldrich) was used to simulate lead-laden sludge. The phase composition of 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 (α -PbO; ICDD PDF #77-1971) and massicot (β -PbO; ICDD PDF #05-0561) phases. Kaolinite powder (Sigma–Aldrich) was used as one precursor and its XRD pattern matched that of standard kaolinite peaks (ICDD PDF #78-1996). The mullite precursor was prepared by calcining the kaolinite powder at 1350 °C for 24 h to yield mullite (ICDD PDF #79-1455) and cristobalite (SiO₂; ICDD PDF #76-0938) confirmed by XRD pattern.

Lead incorporation experiments were conducted using PbO to simulate the high-temperature phase of lead in sludge under sintering conditions. The kaolinite precursors and PbO were mixed by ball milling in water slurry (Pb:Al molar ratio of 1:2) for 18 h. The slurry samples were then dried and homogenized by mortar grinding. The derived powder was pressed into 20-mm pellets at 650 MPa to ensure consistent compaction of the powder for the sintering process. The pellets were sintered at the targeted temperatures for 3 h (Sun et al., 2001) and then quenched in air to room temperature. The total mass loss after sintering was less than 1 wt.%. After sintering, the samples were ground by agate mortar and pestle to a particle size of no more than 10 μ m for XRD analysis and leaching tests.

Phase transformation during sintering was determined by powder XRD, and the data were collected with a Bruker D8 Advance X-ray powder diffractometer equipped with Cu K α radiation and a LynxEye detector. The diffractometer was operated at 40 kV and 40 mA, and the 2θ scan range was from 10° to 80°, with a step size of 0.02° and a scan speed of 0.3 s per step. Qualitative phase identification was performed with Eva XRD Pattern Processing software (Bruker) by matching powder XRD patterns with those retrieved from the standard powder diffraction database of the ICDD PDF-2 Release 2008.

Before leaching test, the surface areas of single-phase powders were measured by a Beckman Coulter SA3100 Surface Area and Pore Size Analyzer using the BET method after degassing by heating at 300 °C with He-gas purging for 3 h. The leachabilities of single-phase samples were evaluated with a procedure modified from the US EPA SW-846 Method 1311-TCLP with a pH 2.9 acetic acid solution (extraction fluid #2) as the leaching fluid. Each leaching vial was filled with 10 mL TCLP extraction fluid and 0.5 g powder. The leaching vials were rotated end-over-end at 60 rpm for 0.75–23 d. At the end of each agitation period, the leachates were passed through 0.2- μ m syringe filters, pH was measured, and Pb concentrations were determined with a flame-type Perkin Elmer model 3300 atomic absorption spectrometer (Perkin Elmer).

3. Results and discussion

3.1. Formation of lead feldspar from kaolinite precursor

Nickel incorporation with a kaolinite precursor was reported to be strongly dependent on sintering temperature (Shih et al.,

2006a,b). To investigate the effect of temperature on initiating the lead reaction with the kaolinite precursor during ceramic sintering processes, we carried out a short (3-h) sintering scheme at temperatures ranging from 500 to 950 °C. The XRD patterns revealed that the sample sintered at 700 °C started to develop distinguishable lead feldspar (ICDD PDF #89-6435) phase as shown in Fig. 1. The chemically bound water of kaolinite is generally removed at temperatures above 550 °C and amorphous metakaolin (Al₂Si₂O₇) forms (MacKenzie et al., 1996; Chen and Tuan, 2002). XRD patterns of products sintered at 700 °C showed that lead incorporation may be activated by a recrystallization process when sintering PbO with kaolinite:



The intensity of the lead feldspar phase increased at higher temperatures, with a substantially higher lead feldspar signal at 800 °C. By high-temperature XRD technique (temperature increase rate of 5 °C min⁻¹), Sorrell (1962) reported the formation of lead feldspar started at this temperature and therefore the formation of lead feldspar is strongly influenced by the diffusion process. Lead feldspar was the only lead-containing phase in the system when the sintering temperature reached 950 °C. However, two intermediate phases, namely lead aluminium silicate (Pb₄Al₄Si₃O₁₆; ICDD PDF #32-0505) and a polymorph of lead feldspar (ICDD PDF #14-0326) were also observed at the temperature range of 700–900 °C. The appearance of these two intermediate phases suggests that the reaction between PbO and metakaolin was in a nonequilibrium system at low temperatures (700–900 °C) during the short 3-h sintering time. As the homogeneous solid-state reaction is usually influenced by the encounter rate between reactant molecules (Kukukova et al., 2009), the short sintering time and low temperature did not allow sufficient contact, resulting in incomplete lead feldspar formation and the two intermediate compounds. However, higher temperatures enabled more intensive interaction between reactants, and the two intermediate silicates were not observed in the sintered samples at these high temperatures.

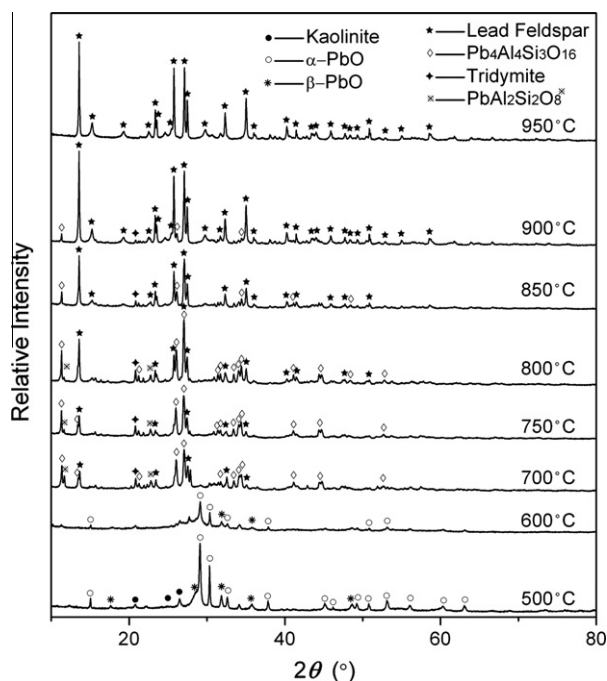


Fig. 1. XRD patterns of the PbO + kaolinite system (Pb:Al molar ratio of 1:2) show the formation of lead feldspar (PbAl₂Si₂O₈, ICDD PDF #89-6435) sintered at temperatures between 500 °C and 950 °C for 3 h.

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