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Lead glass-ceramics produced from the beneficial use of waterworks sludge

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ABSTRACT

This study quantified the effects of lead stabilization by blending lead-containing sludge into glass-ceramics through sintering with aluminum- and silica-rich precursors. Using lead oxide (PbO) to simulate lead-laden sludge under thermal conditions, the predominant $PbAl_2O_4$ phase was found at temperatures of 900–1000 °C while sintering with γ-Al_2O_3 for 3 h. To analyze the influence of silica, amorphous SiO_2 and quartz were blended with γ-Al_2O_3 for lead stabilization. The results revealed both silica precursors could crystallochemically incorporate lead into the $PbAl_2Si_2O_8$ structure, and the weight percentage of the $PbAl_2Si_2O_8$ phase ranged from 30% to 40% in the product after 3 h of sintering at 1000 °C. Furthermore, the calcined waterworks sludge was applied as a sintering precursor and found to result in 46 wt.% of $PbAl_2Si_2O_8$ in the sintered product, suggesting an effective and feasible waste-to-resource strategy for the beneficial use of waterworks sludge. Finally, a prolonged acid leaching experiment (lasting 23 d) evaluated the stability of lead in the $PbAl_2O_4$ and $PbAl_2Si_2O_8$ phases. The concentration of lead in the $PbAl_2O_4$ leachate was 30-times higher than that in the $PbAl_2Si_2O_8$ leachate at the end of the experiment, suggesting a preferred lead stabilization strategy of forming the $PbAl_2Si_2O_8$ phase.

1. Introduction

Global production and consumption of lead have substantially increased in recent decades. Available data from the International Lead and Zinc Study Group estimates that lead consumption reached more than 9 million tons in 2010. Wastewaters generated from the industries of lead batteries, oil-based paints, mining, plating, electronics, and wood production are the main lead pollution sources (Jalali et al., 2002; Gupta et al., 2001; Conrad and Bruun Hansen, 2007). Lead can easily accumulate in the vital organs of humans and animals, inducing lead poisoning (Gupta et al., 2011). Physicochemical techniques of removing lead from wastewater, employed to treat lead-containing effluents, usually include precipitation, coagulation, reduction, ion exchange, and membrane processes (Husein et al., 1998; Lin and Navarro, 1999; Petruzzelli et al., 1999; Saeed et al., 2005; Doyurum and Çelik, 2006; Ali and Gupta, 2007). These treatments often result in large amounts of sludge laden with hazardous metals, which is generally solidified in cementitious materials and disposed in landfills. Considering of alternative solution to better immobilize hazardous metals, thermal treatment offers the advantage of producing a stable and usable product (Basegio et al., 2002). Previous studies have

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reported mechanisms for stabilizing nickel and copper into aluminum-rich ceramic matrices and reducing metal leachability through the use of spinel properties (Shih et al., 2006a, 2006b; Tang et al., 2010; Hu et al., 2010).

Water treatment systems generate approximately 10,000 ton/day of waterworks sludge on a worldwide scale (Dharmappa et al., 1997). The costs of handling the enormous amount of water treatment sludge are likely to multiply due to increasingly stringent regulations (Babatunde and Zhao, 2007). Recent studies evaluated the feasibility of using water treatment sludge as a raw material for ceramic production, and suggested the success of incorporating harmful metals in crystal structures to largely reduce their risks to the environment (Lin and Weng, 2001; Vicenzi et al., 2005; Xu et al., 2008). Recycling water treatment sludge for ceramic production can encourage a more sustainable use of natural resources, and provide economic incentives. Moreover, if the application of waste sludge in ceramic systems can further benefit the stabilization of harmful materials, such as hazardous metals, it can simultaneously reduce both the burdens of waste management and environmental hazard.

The recently published PbO–Al_2O_3–SiO_2 equilibrium phase diagram (Chen et al., 2001) suggests that the mixture of PbO, Al_2O_3 and SiO_2 may thermally result in glass-ceramic materials with crystallizations of lead aluminate ($PbAl_2O_4$) and lead feldspar ($PbAl_2Si_2O_8$) phases. To create a successful metal stabilization technology, a quantitative understanding of the partition of lead into the different hosting phases and of lead existing as amorphous content under different thermal conditions is crucial, and the stabilities of product phases need to be distinguished. Our study first quantitatively compared the reaction efficiencies among lead oxide, Al_2O_3 and/or different forms of SiO_2 through ceramic sintering to delineate the potential mechanistic processes of lead incorporation. Then, the feasibility of using water treatment sludge as a source of material used for stabilizing hazardous metals during the sintering process was evaluated. Finally, a prolonged leaching experiment examined the stability of the lead and leaching behavior of the sintered product phases.

2. Materials and methods

Lead incorporation experiments were conducted using PbO, together with alumina and silica, in order to simulate the phase transformation of lead in sludge under high temperature sintering conditions (Yao and Naruse, 2009). X-ray diffraction (XRD) used in conjunction with the Powder Diffraction File (PDF) database published by International Centre for Diffraction Data (ICDD) identified the phase composition of the received PbO (Sigma–Aldrich) powder as a mixture of the litharge (α-PbO; ICDD PDF #77-1971) and massicot (β-PbO; ICDD PDF #05-0561) phases. Commercial PURAL SB powder (Sasol Ltd., ~45 μm) was used to prepare γ-Al_2O_3. The phase of PURAL SB powder was identified by XRD as boehmite (AlOOH; ICDD PDF #74-1875), and it was converted successfully to the γ-Al_2O_3 phase after heat treatment at 650 °C for 3 h (Zhou and Snyder, 1991), as shown in Fig. S1 of Supplementary Information. Amorphous silica (Sigma–Aldrich, 0.2–0.3 μm) and crystalline quartz (Sigma–Aldrich, 14–19 μm; ICDD PDF#79-1910) were used as Si-rich precursors. The water treatment sludge employed in this study was collected from the waterworks in Hong Kong, which employs a typical water treatment process using an aluminum-based coagulant. Before the sintering experiments to facilitate the observation of metal transformation processes among different phases were undertaken, the dried sludge was heated at 900 °C for 30 min to simulate the sludge incineration ash before sintering experiments (Turovskiy and Mathai, 2006). The crystalline phases of the heat-treated sludge were analyzed by a Bruker D8 Advance X-ray powder diffractometer (Cu Kα radiation) as shown in Fig. S2 of Supplementary Information. Moreover, the major chemical compositions (expressed in their oxide forms) of the heated waterworks sludge were identified by XRF (JSX-3201z, JEOL), listed as Al_2O_3 (56.53), SiO_2 (30.16), Fe_2O_3 (6.17), P_2O_5 (3.82), K_2O (1.28), CaO (1.07) and others (0.97) in decreasing order of weight %.

The alumina and silica precursors were homogeneously mixed with PbO by ball milling with Pb/Al or Pb/Si molar ratios of 1:2. Mixed powders pressed into 20-mm pellets at 650 MPa ensured adequate compaction for sintering. The samples were heated at a rate of 5 °C/min to targeted temperatures, varying from 500 to 1000 °C; the samples were quenched in air to room temperature after 3 h at each firing temperature. Thermogravimetric analysis for the four samples (PbO/γ-Al_2O_3, PbO/γ-Al_2O_3/Amor.-SiO_2, PbO/γ-Al_2O_3/Quartz and PbO/waterworks sludge) were conducted by a TGA92 (SETARAM Inc., Newark, CA) thermoanalyser with an initial sample weight of 6–8 mg. Using a heating rate of 5 °C/min, the TG analysis of the four samples was tested from room temperature to 1000 °C under air purging. The TG curves demonstrate that the weight loss was less than 1% for all four samples at temperatures below 1000 °C. After sintering, an agate mortar and pestle was used to grind the samples to a particle size less than 10 μm for the XRD analysis and leaching test. The X-ray 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/step. Phase identification was done using Eva XRD Pattern Processing software (Bruker Co. Ltd.) by matching powder XRD patterns with those retrieved from the standard powder diffraction database of the International Centre for Diffraction Data (ICDD PDF-2 Release, 2008).

The TOPAS (version 4.0) crystallographic program processed the Rietveld refinements for quantitative analysis of the phase compositions. To monitor the refinement quality of this analytical scheme, its application was compared to samples with known compositions; the derived reliability values are provided in the Supplementary Information of Tables S1, S2, S3, S4 and S5. For systems potentially containing amorphous or poorly crystalline phases (i.e., γ-Al_2O_3, amorphous SiO_2 and waterworks sludge), a refinement method using CaF_2 as the internal standard (De La Torre et al., 2001; Magallanes-Perdomo et al., 2009; Rendtorff et al., 2010) was used to quantify the amorphous content in the samples. Scans were collected from 10° to 120° 2θ-angle, with a step width of $2\theta = 0.02°$ and a sampling time of 0.5 s per step.

To determine the role and characteristics of each Pb stabilization mechanism, the potential Pb-bearing product phases were compared in a leaching experiment. The leachabilities of the single-phase samples were tested using a leaching