

# Incorporation Mechanism of Stabilizing Simulated Lead-Laden Sludge in Aluminum-Rich Ceramics

Xingwen Lu, Kaimin Shih

**Abstract**—This study investigated a strategy of blending lead-laden sludge and Al-rich precursors to reduce the release of metals from the stabilized products. Using PbO as the simulated lead-laden sludge to sinter with  $\gamma$ - $\text{Al}_2\text{O}_3$  by Pb:Al molar ratios of 1:2 and 1:12,  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$  were formed as final products during the sintering process, respectively. By firing the  $\text{PbO} + \gamma$ - $\text{Al}_2\text{O}_3$  mixtures with different Pb/Al molar ratios at 600 to 1000 °C, the lead transformation was determined through X-ray diffraction (XRD) data. In Pb/Al molar ratio of 1/2 system, the formation of  $\text{PbAl}_2\text{O}_4$  is initiated at 700 °C, but an effective formation was observed above 750 °C. An intermediate phase,  $\text{Pb}_9\text{Al}_8\text{O}_{21}$ , was detected in the temperature range of 800-900 °C. However, different incorporation behavior for sintering PbO with Al-rich precursors at a Pb/Al molar ratio of 1/12 was observed during the formation of  $\text{PbAl}_{12}\text{O}_{19}$  in this system. In the sintering process, both temperature and time effect on the formation of  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$  phases were estimated. Finally, a prolonged leaching test modified from the U.S. Environmental Protection Agency's toxicity characteristic leaching procedure (TCLP) was used to evaluate the durability of PbO,  $\text{Pb}_9\text{Al}_8\text{O}_{21}$ ,  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$  phases. Comparison for the leaching results of the four phases demonstrated the higher intrinsic resistance of  $\text{PbAl}_{12}\text{O}_{19}$  against acid attack.

**Keywords**—Sludge; Lead; Stabilization; Leaching behavior

## I. INTRODUCTION

THE contamination of wastewater and soil with lead ions from the discharge of industrial wastewater is a world wide environmental problem. Lead is widely used in lead batteries, oil-based paints, mining, plating, hot dip galvanizing, petroleum refining, and pigment industries, paper and pulp, electrodes in electrochemistry and chemical industries [1]-[3]. Available data from International Lead and Zinc Study Group estimated that a significant amount of lead consumption was reaching more than 9 million tons in 2010. Lead is nonbiodegradable and highly toxic to human being, which causes severe damage to the kidneys, liver, and the nervous and reproductive systems of humans [4]. Therefore, hazardous metals must be removed from wastewater before they can be discharged into environment. Physicochemical treatment for lead removal from wastewater and soil commonly included precipitation, coagulation, reduction, ion exchange, and membrane processes [5]-[9].

However, these treatments produce large amounts of hazardous metal-laden sludge, which requires additional treatment.

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In many regions of the world, traditional disposal methods (e.g., landfills) no longer accept hazardous metal waste [10]. Consequently, metal stabilization strategies have been sought to replace traditional disposal methods and safely reuse the sludge. Previous studies have reported that nickel and copper sludge can be incorporated into aluminate and ferrite structures in ceramics through a well-controlled sintering process, significantly reducing product leachability through the intrinsic properties of the product phases [11]-[14]. Incorporating lead-laden sludge may be a promising strategy to stabilize the hazardous metal and largely reduce the risk of their environmental hazard.

Studies have shown that the calcining of lead oxide (PbO) on  $\gamma$ - $\text{Al}_2\text{O}_3$  produces the various phases of nonlead octaaluminum oxide ( $\text{Pb}_9\text{Al}_8\text{O}_{21}$ ), lead aluminate ( $\text{PbAl}_2\text{O}_4$ ) and lead dodecaaluminate ( $\text{PbAl}_{12}\text{O}_{19}$ ) [15], [16]. However, the incorporation mechanism and phase transformation pathway have not been discussed in detail. The recent published  $\text{PbO-Al}_2\text{O}_3\text{-SiO}_2$  equilibrium phase diagram also confirmed the presence of  $\text{Pb}_9\text{Al}_8\text{O}_{21}$ ,  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$  [17]. These equilibrium studies have provided the opportunity to observe the potential interaction between PbO and aluminum-rich precursors under industrial sintering processes. Therefore, incorporating lead-laden sludge into ceramic sintering may be able to initiate a beneficial phase-transformation process to further stabilize lead-containing waste.

In this study,  $\gamma$ - $\text{Al}_2\text{O}_3$  was evaluated as possible ceramic precursors to react with lead oxide, which is often the lead-containing phase produced by heating lead-laden sludge. The incorporation experiment was firstly conducted by heating powder mixtures of PbO and precursors at 600 to 1000 °C in a 3-h short-sintering scheme. Secondly, the influence of time on phase transformation mechanism was analyzed on certain temperatures. Finally, three potential lead phases ( $\text{Pb}_9\text{Al}_8\text{O}_{21}$ ,  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$ ) in the products were prepared separately and leached by an acetic acid solution (pH 2.9), which is similar to the leaching fluid used in toxicity characteristic leaching procedures (TCLP), for up to 23 days to characterize the intrinsic stabilization effect of lead in these three product phases.

## II. EXPERIMENTAL METHODS

### A. Materials

PbO powder was purchased from Sigma Aldrich. The phase composition of the PbO powder was identified by X-ray diffraction (XRD) method as a mixture of  $\alpha$ -PbO (litharge) and  $\beta$ -PbO (massicot) phases. HiQ-7223 alumina powder was obtained from PURAL SB powder fabricated by Sasol with an average particle size  $\sim 45$   $\mu\text{m}$  and identified as boehmite ( $\text{AlOOH}$ ) by XRD, which was then converted to the  $\gamma$ - $\text{Al}_2\text{O}_3$  precursor upon thermal treatment at 650 °C for 3 h [18], [19].

### B. Methods

Lead incorporation efficiency was determined by firing lead oxide mixed with  $\gamma$ -alumina. Raw materials were prepared by mixing PbO and Al-rich precursors with molar ratios of Pb:Al = 1:2 and 1:12 to a total dry weight of 60 g. The mixing process was carried out by ball milling the powder in water slurry for 18 h. The slurry samples were then dried, homogenized again by mortar grinding, and pressed into 20-mm pellets at 650 MPa to ensure consistent compaction of the powder sample for the sintering process. The pellets were firstly sintered at targeted temperatures from 600 to 1000 °C and then quenched in air to room temperature.

### C. XRD Analysis

To identify the potential phases responsible for metal incorporation during sintering, pelletized samples were fired in a high-temperature furnace under air atmosphere in the heating conditions. Phase transformation during sintering was monitored by a Bruker D8 Advance X-ray powder diffractometer equipped with Cu K $\alpha$ 1,2 X-ray radiation and a LynxEye detector. The  $2\theta$  scanning range was 10° to 80°, the step size was 0.02°, and the scan speed was 0.3 s step<sup>-1</sup>. Qualitative phase identification was executed by matching the powder XRD patterns with those retrieved from the standard powder diffraction database of the International Centre for Diffraction Data (ICDD PDF-2 Release 2008). Five lead-containing crystalline phases were found in the products:  $\alpha$ -PbO (ICDD PDF #77-1971),  $\beta$ -PbO (ICDD PDF #05-0561), Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> (ICDD PDF #73-1875), PbAl<sub>2</sub>O<sub>4</sub> (ICDD PDF #83-1016), Pb<sub>3</sub>(CO<sub>3</sub>)<sub>2</sub>(OH)<sub>2</sub> (ICDD PDF # 73-4362) and PbAl<sub>12</sub>O<sub>19</sub> (ICDD PDF #80-1174).

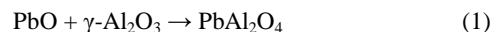
### D. Prolonged TCLP Test

As an important goal of this study was to distinguish the leaching characteristics of different Pb-bearing phases, single-phase samples were considered preferable in the leaching experiment. Single-phase Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub>, PbAl<sub>2</sub>O<sub>4</sub> and PbAl<sub>12</sub>O<sub>19</sub> were fabricated from samples of PbO +  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> with Pb/Al molar ratios of 9/8, 1/2 and 1/12, respectively. The leaching experiments for PbO, Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub>, PbAl<sub>2</sub>O<sub>4</sub> and PbAl<sub>12</sub>O<sub>19</sub> were conducted by a method modified from the U.S. Environmental Protection Agency's SW-846 Method 1311: Toxicity Characteristic Leaching Procedure (TCLP), with an acetic acid solution (extraction fluid #2, pH 2.9) used as the leaching fluid. Each leaching vial was filled with 10 mL of leaching fluid and 0.5 g of the powder sample and rotated end-over-end at 60 rpm for 0.75 to 23 days. At the end of each agitation period, the leachates were filtered using 0.2- $\mu$ m syringe filters, the pH was determined, and the concentrations of lead were derived by inductively coupled plasma atomic emission (Perkin-Elmer Optima 3300 DV).

## III. RESULTS AND DISCUSSION

### A. Mechanisms of Incorporating Lead into PbAl<sub>2</sub>O<sub>4</sub> Structure

Lead can be incorporated into the PbAl<sub>2</sub>O<sub>4</sub> structure by sintering PbO with Al-rich precursors (e.g.,  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>). This result is qualitatively demonstrated by the XRD diffraction pattern of the product sintered at 600-1000 °C for 3 h (Fig. 1). The result showed that when sintered at 700 °C, the formation of crystalline PbAl<sub>2</sub>O<sub>4</sub> phase was first observed. The thermal reactions between PbO and Al<sub>2</sub>O<sub>3</sub> can be expressed by the following equations according to the observed XRD pattern.



The reported formation temperature of PbAl<sub>2</sub>O<sub>4</sub> was at 600 °C with 1-2 months of dwelling time [20]. Therefore, this difference may suggest that the formation of PbAl<sub>2</sub>O<sub>4</sub> at temperatures below 700 °C is largely limited by the prevailing slow diffusion, although it is thermodynamically feasible at temperatures above 600 °C. The solid-state reaction is affected by both thermodynamic constraint and kinetic process. Below 700 °C, the PbAl<sub>2</sub>O<sub>4</sub> phase formed by the short sintering scheme might only be limited at the grain boundary of reactants, and this small quantity in the sample was not reflected in the XRD results. The intensities of PbAl<sub>2</sub>O<sub>4</sub> peaks were weak in the 700 °C sintered samples, but the substantial formation of PbAl<sub>2</sub>O<sub>4</sub> in the system was detected at temperature of 750 °C. The findings may indicate the energy needed to overcome the major diffusion barrier in the system. As the intensity of the PbAl<sub>2</sub>O<sub>4</sub> phase increased when the sintering temperature increases, a significantly higher intensity of PbAl<sub>2</sub>O<sub>4</sub> signal was achieved at 750 °C, and no Bragg reflection of  $\alpha$ -PbO or  $\beta$ -PbO phase was observed when the temperature was above 800 °C. An intermediate product of Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> was found when the sintering temperature was around 750 °C, but its Bragg diffraction peaks disappeared in the sample sintered at 900 °C. Increasing the sintering temperature to 950 °C results the completing lead transformation to PbAl<sub>2</sub>O<sub>4</sub> in the Pb/Al of 1/2 system.

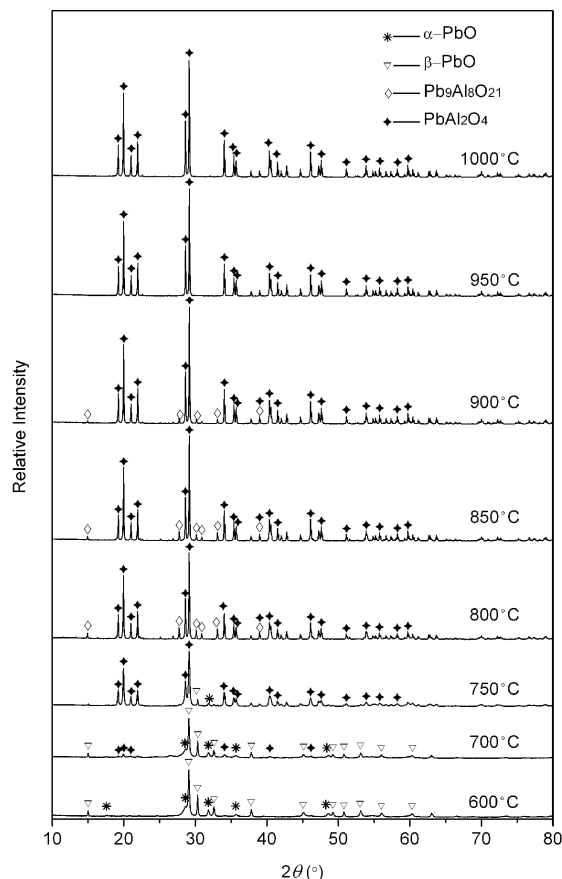


Fig. 1 The XRD patterns of the PbO +  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> system with a Pb:Al molar ratio of 1:2 when sintering at temperatures between 600 and 1000 °C for 3 hours. The result shows the formation of PbAl<sub>2</sub>O<sub>4</sub> and Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> in the system

The effect of the reaction time on the lead stabilization by the formation of PbAl<sub>2</sub>O<sub>4</sub> was investigated at temperatures of 700 °C (Fig. 2a) and 800 °C (Fig. 2b) for 0.25 to 10 hours.

The X-ray diffraction patterns of 700 °C sintering indicated that no reaction between PbO and  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> was found when the treatment time is less than 3 h. With the increase of thermal treatment time, PbAl<sub>2</sub>O<sub>4</sub> and Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> co-existed in the products, but only the peak intensities of Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> were significantly increased during the sintering process. The formation of Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> may probably initiate by the reaction between PbO and  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> in a non-equilibrium system due to the lower temperature.

Due to the efficiency of a homogeneous reaction generally depends on the encountering rate between reactant molecules [21], the low temperature was not able to provide sufficient contact, and hence the formation of Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> appeared and increased in the products. At higher temperature (800 °C), more intensive interaction between reactants was achieved.

Even with the short sintering time (0.25 hour), PbAl<sub>2</sub>O<sub>4</sub> was observed as the dominate crystal phase in the sintered samples. But the prolonged thermal treatment time cannot achieve the complete transformation of lead into PbAl<sub>2</sub>O<sub>4</sub>.

Therefore, to more effectively achieve the formation of PbAl<sub>2</sub>O<sub>4</sub>, the temperature of a short sintering scheme should be higher than 800 °C.

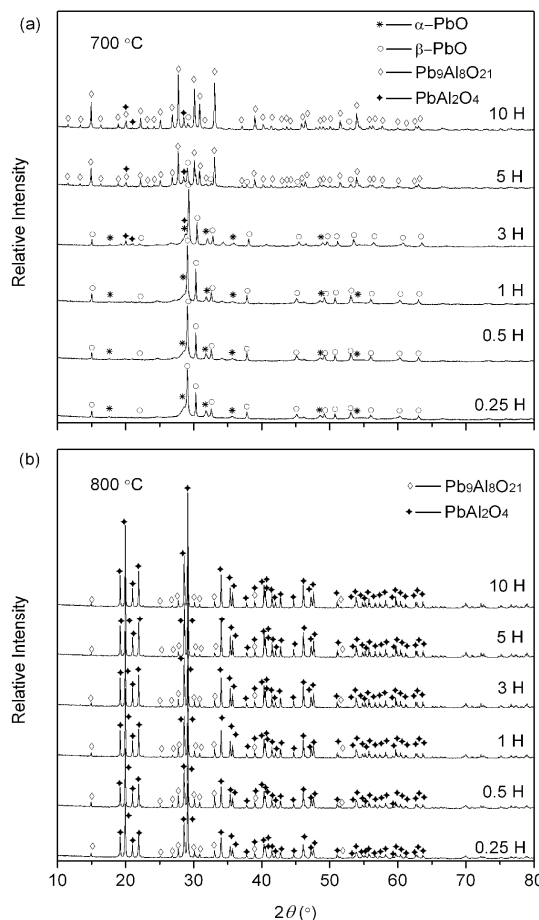


Fig. 2 The XRD patterns of the PbO +  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> system with a Pb:Al molar ratio of 1:2 when sintering at temperatures of 700 and 800 °C for different hours

#### B. Mechanisms of Incorporating Lead into PbAl<sub>12</sub>O<sub>19</sub> Structure

When the PbO molar content in the PbO-Al<sub>2</sub>O<sub>3</sub> system was lower than 50%, PbAl<sub>12</sub>O<sub>19</sub> was the only product phase reported in equilibrium experiments [17], [22]. Therefore, a potential thermal reaction of incorporating lead by  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> precursor at lower lead level is described by Eq. (2):



Fig. 3 presents the XRD patterns of the 600-1000 °C sintered PbO +  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> mixtures with a Pb/Al molar ratio of 1:12, and shows that the peaks of the PbAl<sub>12</sub>O<sub>19</sub> phase appeared when the sintering temperature exceeded 950 °C. The lowest temperature at which PbAl<sub>12</sub>O<sub>19</sub> was observed after a short sintering scheme was about 200 °C higher than that derived from the equilibrium experiment carried out by [22]. Similarly, this discrepancy can be explained by the shorter sintering time and the potential diffusion barrier created by the newly formed PbAl<sub>12</sub>O<sub>19</sub> layer between the PbO and  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> grains.

At 1000 °C, substantial growth of  $\text{PbAl}_{12}\text{O}_{19}$  phase was found and  $\text{PbAl}_{12}\text{O}_{19}$  was the only lead-containing phase in the product. However, an intermediate phase, hydrocerussite ( $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$ ), formed in the products sintered at temperatures between 700 °C and 950 °C.  $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$  was identified as a common lead corrosion product by other studies [23], [24].

Thus, the formation of poor crystalline  $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$  phase was probably due to the instability of product phase(s) sintered at 700-950 °C, which might be vulnerable to the attack of  $\text{CO}_2$  and moisture in the air during sample quenching.

Nevertheless, when sintered at 1000 °C, the significant conversion to the  $\text{PbAl}_{12}\text{O}_{19}$  phase completely eliminated the formation of  $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$  in the product and  $\text{PbAl}_{12}\text{O}_{19}$  was found to be the only lead-containing phase in the sample. This result confirms the potential of forming  $\text{PbAl}_{12}\text{O}_{19}$  to incorporate lead into the aluminum-rich ceramics for systems with lower Pb/Al ratios.

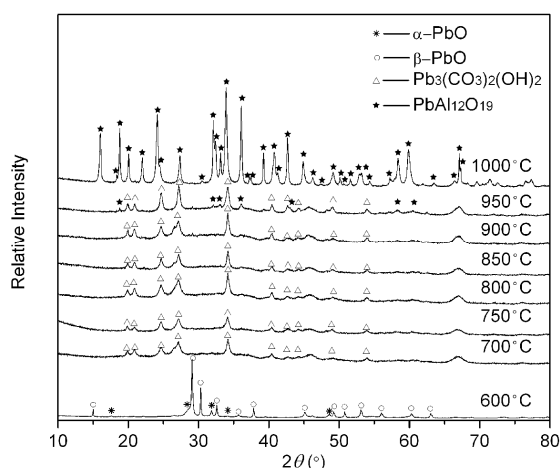


Fig. 3 The XRD results of products sintered from the  $\text{PbO} + \gamma\text{-Al}_2\text{O}_3$  samples with a Pb:Al molar ratio of 1:12 at temperatures between 600 and 1000 °C for 3 hours

Time influence to the crystal growth of  $\text{PbAl}_{12}\text{O}_{19}$  was examined at temperatures of 950 and 1000 °C by observing the reaction of  $\text{PbO}$  and  $\gamma\text{-Al}_2\text{O}_3$  with Pb/Al molar ratio of 1/12 (Fig. 4). Starting from 950 °C, the unstable thermal treatment products can be easily attack by  $\text{CO}_2$  and moisture for the formation of hydrocerussite under short sintering time (0.25-3 H).

The initiation of poor crystalline  $\text{PbAl}_{12}\text{O}_{19}$  phase from sintering the  $\text{PbO} + \gamma\text{-Al}_2\text{O}_3$  mixture could be observed under 3 h sintering, and the significant increase of  $\text{PbAl}_{12}\text{O}_{19}$  was found with the increasing treatment time.

At 1000 °C, the formation of hydrocerussite started to appear with the short sintering time, but significant growth of the  $\text{PbAl}_{12}\text{O}_{19}$  phase clearly occurred with the increase of reaction time from 50 to 180 min. Therefore, longer sintering time and higher temperature enabled more intensive interaction between reactants to the significant increase of  $\text{PbAl}_{12}\text{O}_{19}$ .

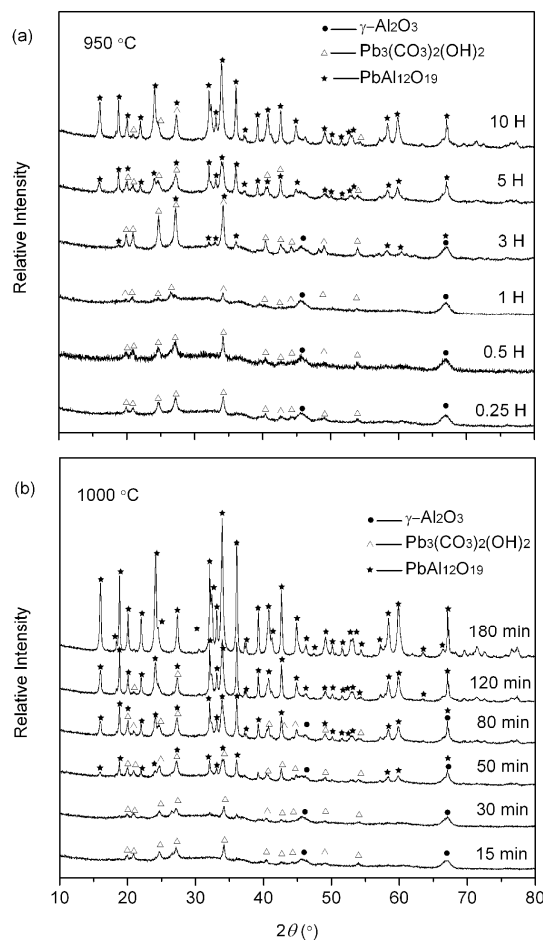


Fig. 4 The XRD patterns of the  $\text{PbO} + \gamma\text{-Al}_2\text{O}_3$  system with a Pb:Al molar ratio of 1:12 when sintering at temperatures of 950 and 1000 °C for different times.

### C. Lead Leachabilities of Product Phases

Because  $\text{Pb}_9\text{Al}_8\text{O}_{21}$ ,  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$  are the potential lead-containing phases in the sintered products, their differences in lead leachability must be taken into account to optimize the lead stabilization strategy. This study thus prepared single-phase samples to determine their intrinsic leachability and leaching behavior in a prolonged (23-d) leaching experiment. The single-phase  $\text{Pb}_9\text{Al}_8\text{O}_{21}$  sample was obtained by sintering the  $\text{PbO} + \gamma\text{-Al}_2\text{O}_3$  mixture with Pb/Al molar ratio of 9/8 at 800 °C for 24 h, pure phase  $\text{PbAl}_2\text{O}_4$  was prepared from the sintered the  $\text{PbO} + \gamma\text{-Al}_2\text{O}_3$  mixture with Pb/Al molar ratio of 1/2 and the single-phase  $\text{PbAl}_{12}\text{O}_{19}$  sample was obtained from the  $\text{PbO} + \gamma\text{-Al}_2\text{O}_3$  mixture with Pb:Al molar ratio = 1:12 sintered at 1000 °C for 3 h. In addition, the property of  $\text{PbO}$  powder used in sintering the raw material was also examined by the leaching experiment to compare it with those of aluminate phases.

The pH values of  $\text{PbO}$ ,  $\text{Pb}_9\text{Al}_8\text{O}_{21}$ ,  $\text{PbAl}_2\text{O}_4$  and  $\text{PbAl}_{12}\text{O}_{19}$  leachates during the 23 d leaching are shown in Fig. 5a, revealing significant pH increases of the  $\text{PbO}$ ,  $\text{Pb}_9\text{Al}_8\text{O}_{21}$  and  $\text{PbAl}_2\text{O}_4$  leachates.

Within the first few days, the pH of the PbO and Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> leachates increased from 2.9 to 9.4 and 8.1, respectively. Similarly, the pH of the PbAl<sub>2</sub>O<sub>4</sub> leachate increased in the early part of the leaching period from 2.9 to 6.3, and the pH was then maintained at approximately 6.5 throughout the rest of the leaching period. In contrast, the pH of the PbAl<sub>12</sub>O<sub>19</sub> leachate remained relatively stable throughout the entire leaching period. The leachate pH increase may be due to the dissolution of cations through ion exchange with protons in the solution, which may be accompanied by the destruction of crystals initiated by the acidic leaching fluid. Therefore, these results suggest a substantially higher intrinsic resistance of PbAl<sub>12</sub>O<sub>19</sub> toward acidic attack, compared with PbO, Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> and PbAl<sub>2</sub>O<sub>4</sub>. The leaching of solids is likely dominated by surface reactions and is therefore expected to be proportional to the sample surface area. The BET surface areas of the powder samples were 0.51 m<sup>2</sup>/g for PbO, 0.48 m<sup>2</sup>/g for Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub>, 0.63 m<sup>2</sup>/g for PbAl<sub>2</sub>O<sub>4</sub> and 3.76 m<sup>2</sup>/g for PbAl<sub>12</sub>O<sub>19</sub>. As the same weight was used for each sample in this experiment (0.5 g), the total lead content in the sample, subject to the different lead phases, should also be normalized to compare leached lead from the solid. Fig. 5b summarizes the amounts of leached lead normalized to the surface area and total lead content of the tested samples. The normalized lead concentrations in PbO leachates were about 1.5 times greater than that of Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub> leachate, 3 times larger than that of PbAl<sub>2</sub>O<sub>4</sub> and four orders of magnitude higher than that from PbAl<sub>12</sub>O<sub>19</sub> leachate at the end of the experiment, indicating that the formation of PbAl<sub>12</sub>O<sub>19</sub> is the preferred stabilization mechanism for incorporating lead in ceramic products.

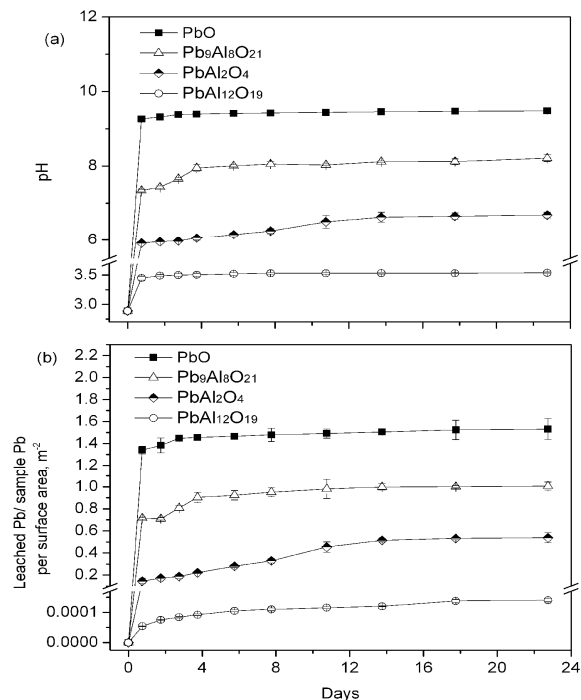


Fig. 5 The pH values (a) and the normalized lead concentrations (b) of the leachate samples of PbO, Pb<sub>9</sub>Al<sub>8</sub>O<sub>21</sub>, PbAl<sub>2</sub>O<sub>4</sub> and PbAl<sub>12</sub>O<sub>19</sub>

#### IV. CONCLUSION

The sintering of lead-laden waste with the Al-rich precursors widely used for ceramic products may result in significant lead hosted by the PbAl<sub>2</sub>O<sub>4</sub> and PbAl<sub>12</sub>O<sub>19</sub> structure with different Pb/Al molar ratios during the sintering process in this study. Although intermediate phases is sometimes achieved in ceramics at low sintering temperatures (e.g. < 950 °C), it is worthwhile to identify the crystal structure hosting the targeted metal(s) and promote a superior metal stabilization mechanism to enhance product safety. A lower Pb/Al molar ratio (e.g. 1/12) and attainable sintering temperature (1000 °C) is suggested to facilitate the preferred lead speciation into the PbAl<sub>12</sub>O<sub>19</sub> structure when incorporating lead-laden sludge into ceramics.

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#### REFERENCES

- [1] R. Jalali, H. Ghafourian, Y. Asef, S. J. Davarpanah, S. Sepehr, "Removal and recovery of lead using nonliving biomass of marine algae." *Journal of Hazardous Materials*, vol. 92(3), pp. 253-262, 2002.
- [2] V. K. Gupta, M. Gupta, S. Sharma, "Process development for the removal of lead and chromium from aqueous solutions using red mud - An aluminium industry waste." *Water Research*, vol. 35(5), pp. 1125-1134, 2001.
- [3] K. Conrad, H. C. Bruun Hansen, "Sorption of zinc and lead on coir." *Bioresource Technology*, vol. 98(1), pp. 89-97, 2007.
- [4] V. K. Gupta, S. Agarwal, T. A. Saleh, "Synthesis and characterization of alumina-coated carbon nanotubes and their application for lead removal." *Journal of Hazardous Materials*, vol. 185(1), pp. 17-23, 2011.
- [5] S. W. Lin, R. M. F. Navarro, "An innovative method for removing Hg<sup>2+</sup> and Pb<sup>2+</sup> in ppm concentrations from aqueous media." *Chemosphere*, vol. 39(11), pp. 1809-1817, 1999.
- [6] D. Petruzzelli, M. Pagano, G. Tiravanti, R. Passino, "Lead removal and recovery from battery wastewaters by natural zeolite clinoptilolite." *Solvent Extraction and Ion Exchange*, vol. 17(3), pp. 677-694, 1999.
- [7] A. Saeed, M. Iqbal, M. W. Akhtar, "Removal and recovery of lead(II) from single and multimetal (Cd, Cu, Ni, Zn) solutions by crop milling waste (black gram husk)." *Journal of Hazardous Materials*, vol. 117(1), pp. 65-73, 2005.
- [8] B. Dubey, T. Townsend, "Arsenic and lead leaching from the waste derived fertilizer ironite." *Environmental Science and Technology*, vol. 38(20), pp. 5400-5404, 2004.
- [9] I. Ali, V. K. Gupta, "Advances in water treatment by adsorption technology." *Nature Protocols*, vol. 1(6), pp. 2661-2667, 2007.
- [10] D. Laner, J. Fellner, P. H. Brunner, "Flooding of municipal solid waste landfills-An environmental hazard?" *Science of the Total Environment*, vol. 407(12), pp. 3674-3680, 2009.
- [11] K. Shih, T. White, J. O. Leckie, "Spinel formation for stabilizing simulated nickel-laden sludge with aluminum-rich ceramic precursors." *Environmental Science and Technology*, vol. 40(16), pp. 5077-5083, 2006.
- [12] K. Shih, T. White, J. O. Leckie, "Nickel stabilization efficiency of aluminate and ferrite spinels and their leaching behavior." *Environmental Science and Technology*, vol. 40(17), pp. 5520-5526, 2006.
- [13] Y. Tang, K. Shih, K. Chan, "Copper aluminate spinel in the stabilization and detoxification of simulated copper-laden sludge." *Chemosphere*, vol. 80(4), pp. 375-380, 2010.
- [14] C. Y. Hu, K. Shih, J. O. Leckie, "Formation of copper aluminate spinel and cuprous aluminate delafossite to thermally stabilize simulated copper-laden sludge." *Journal of Hazardous Materials*, vol. 181(1-3), pp. 399-404, 2010.

- [15] G. Wendt, C. D. Meinecke, W. Schmitz, "Oxidative dimerization of methane on lead oxide-alumina catalysts." *Applied Catalysis*, vol. 45(2), pp. 209-220, 1988.
- [16] S. E. Park, J. S. Chang, "Oxidative coupling of methane over PbO/PbAl<sub>2</sub>O<sub>4</sub> catalysts." *Studies in Surface Science and Catalysis*, vol. 75, pp. 2233-2236, 1993.
- [17] S. Chen, B. Zhao, P. C. Hayes, E. Jak, "Experimental study of phase equilibria in the PbO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system." *Metallurgical and Materials Transactions B: Process Metallurgy and Materials Processing Science*, vol. 32(6), pp. 997-1005, 2001.
- [18] R. S. Zhou, R. L. Snyder, "Structures and transformation mechanisms of the eta, gamma and theta transition aluminas." *Acta Crystallographica Section B*, vol. 47(5), pp. 617-630, 1991.
- [19] Y. Wang, C. Suryanarayana, L. An, "Phase transformation in nanometer-sized  $\gamma$ -alumina by mechanical milling." *Journal of the American Ceramic Society*, vol. 88(3), pp. 780-783, 2005.
- [20] R. F. Geller, E. N. Bunting, "Report on the systems lead oxide-alumina and lead oxide-alumina-silica." *Journal of research of the National Bureau of Standards*, vol. 31(5), pp. 255-270, 1943.
- [21] A. Kukukova, J. Aubin, S. M. Kresta, "A new definition of mixing and segregation: Three dimensions of a key process variable." *Chemical Engineering Research and Design*, vol. 87(4), pp. 633-647, 2009.
- [22] U. Kuxmann, P. Fischer, "Lead monoxide-aluminum oxide, lead monoxide-calcium oxide, and lead monoxide-silicon dioxide phase diagrams." *Erzmetall*, vol. 27(11), pp. 533-537, 1974.
- [23] E. J. Kim, J. E. Herrera, D. Huggins, J. Braam, S. Koshowski, "Effect of pH on the concentrations of lead and trace contaminants in drinking water: A combined batch, pipe loop and sentinel home study." *Water Research*, vol. 45(9), pp. 2763-2774, 2011.
- [24] E. J. Kim, J. E. Herrera, "Characteristics of lead corrosion scales formed during drinking water distribution and their potential influence on the release of lead and other contaminants." *Environmental Science and Technology*, vol. 44(16), pp. 6054-6061, 2010.



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