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The application of electrocoagulation for the conversion of MSWI fly ash into nonhazardous materials

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ABSTRACT

This research investigated the electrocoagulation of municipal solid waste incineration (MSWI) fly ash at a liquid-to-solid ratio (L/S) of 20:1. The leachate that was obtained from this treatment was recovered for reutilization. Two different anodic electrodes were investigated, and two unit runs were conducted. In Unit I, the optimum anode was chosen, and in Unit II, the optimum anode and the recovered leachate were used to replace deionized water for repeating the same electrocoagulation experiments. The results indicate that the aluminum (Al) anode performed better than the iridium oxide (IrO_2) anode. The electrocoagulation technique includes washing with water, changing the composition of the fly ash, and stabilizing the heavy metals in the ash. Washing with water can remove the soluble salts from fly ash, and the fly ash can be converted into Friedel's salt ($3CaO \cdot Al_2O_3 \cdot CaCl_2 \cdot 10H_2O$) under an uniform electric field and the sacrificial release of Al^{+3} ions, which stabilizes the toxic heavy metals and brings the composition of the fly ash to within the regulatory limits of the toxicity characteristic leaching procedure (TCLP). Use of the Al anode to manage the MSWI fly ash and the leachate obtained from the electrocoagulation treatment is therefore feasible.

1. Introduction

Incineration has become the preferred option for the disposal of municipal solid waste (MSW) in Taiwan. The fly ash generated by MSW incineration (MSWI) is trapped in the air pollution control devices (APCDs). MSWI fly ash is classified as a hazardous waste and must be disposed of in landfills after the stabilization and solidification (S/S) of the heavy metals and dioxins. However, the increasingly limited landfill capacity and the restrictions of the new landfill policy developed by the Taiwanese Environmental Protection Administration (TEPA) in 2007 have caused the reuse of MSWI fly ash to become an essential issue in Taiwan.

The four methods for processing MSWI fly ash that have been described in research papers include physical, biological, thermal and chemical treatment techniques. Physical treatment techniques often use washing with water as a pretreatment to promote the formation of hydrate phases that convert heavy metals to less-reactive forms and remove significant amounts of soluble salts from the fly ash (Abbas et al., 2003; Arvelakis and Frandsen, 2005; Colangelo et al., 2012; Kirby and Rimstidt, 1994; Mangialardi, 2003). Wu and Ting (2006) used a biological technique to treat fly ash and found that fungal bioleaching was more beneficial than chemical leaching. However, bioleaching requires more time for processing than does chemical leaching. Thermal treatment techniques that are used to handle fly ash such as sintering, melting, fusion, and vitrification are usually operated at high temperatures. The products of these thermally treated fly ashes are highly crystalline on the surface and are highly durable chemically, which causes them to be considered suitable for reuse (De Casa et al., 2007; Károly et al., 2007; Karamanov et al., 2003; Lam et al., 2011). Nevertheless, these thermal treatment processes are economically undesirable on the commercial scale because of their high energy consumption and the high costs of building and maintaining the facilities used to operate the thermal treatment processes (Sakai and Hiraoka, 2000). Chemical extraction or fixation techniques have been studied much more often than the other three techniques. The chemical agents that are typically used include HCl, HNO_3, H_3PO_4, $Ca(OH)_2$, NaOH, ethylenediaminetetraacetic acid (EDTA) disodium salt, Na_2S, and thiourea (Astrup et al., 2006; Eighmy et al., 1997; Kida et al., 1996; Mizutani et al., 1996; Okada et al., 2007; Youcai et al., 2002). Many studies have

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addressed sequential extraction processes and confirmed the extractability of some heavy metals (Eighmy et al., 1995; Huang et al., 2007; Wan et al., 2006). These studies that have focused on the processing of MSWI fly ash appear to have resolved questions related to the toxicity of the fly ash; however, these methods are still defective in at least three aspects. The first defective area is the large amount of wastewater awaiting further disposal; the second defective area is the deficiency or absence of chlorine reduction in the fly ash; and the third defective area is the inability to keep up with the quantity of fly ash produced daily, which was the main reason that the chemical treatment methods of the fly ash were considered the most commercially optimal. With an increasing global focus on zero waste strategies and the continuous reuse of wastewater resources, this study investigated an innovative treatment of both MSWI fly ash and wastewater using direct electrolysis, wherein an electrocoagulation technique was used to resolve the above three problems, resulting in efficient handling of MSWI fly ash.

The electrocoagulation technique involves the generation of coagulants *in situ* by dissolving aluminum (Al) ions electronically from Al electrode, which is also known as a sacrificial electrode. Metal ions are generated at the anode. Hydrogen gas that is released from the cathode helps to float flocculated particles out of the water (Jiang et al., 2002). The reactions occurring in electrocoagulation are shown in Eqs. (1)–(4).

$$\text{Anode: } 2\ H_2O \rightarrow 4\ H^+ + O_2(g) + 4\ e^- \quad (1)$$

$$Al \rightarrow Al^{3+} + 3\ e^- \quad (2)$$

$$\text{Cathode: } 2\ H_2O + 2\ e^- \rightarrow 2\ OH^- + H_2(g) \quad (3)$$

$$Al^{3+} + 3\ OH^- \rightarrow Al(OH)_3 \quad (4)$$

The electrocoagulation technique is an effective method for removing toxic heavy metals from wastewater (Merzouk et al., 2009; Mouedhen et al., 2009; Zongo et al., 2009), and almost no research on electrocoagulation of MWSI fly ash is available in scientific literature. The main purposes of this research were to handle MSWI fly ash and retrieve the leachate (untreated wastewater) by means of the electrocoagulation technique while considering the following aspects: (a) the physicochemical features of the untreated fly ash, (b) a comparison of the untreated and the treated fly ash samples, as well as the wastewater that is generated within the regulatory limits after electrocoagulation, and (c) a mineralogical investigation to identify the composition of untreated and treated fly ash samples by scanning electron microscope/energy dispersive spectrometer (SEM-EDS) and X-ray diffraction (XRD).

2. Materials and methods

2.1. Sampling

The MSWI fly ash used in this study consisted of approximately 20 kg of material that was obtained from the Taichung City refuse incineration plant. This plant is a large-scale incinerator located in central Taiwan. The material was collected from a side tube of the duct into the fly ash hopper. The fly ash hopper is used to provide fly ash to carry out S/S treatments. The proportion of general (nonhazardous) waste to general industrial waste in the incineration source material was 78:22. The plant uses a set of three stoker grate-type incinerators. Fly ash is produced from a semi-dry scrubber system of APCDs, consisting of two spray dryer injections, one injecting slurry lime for the neutralization of acidic gases and one injecting powdered activated carbon for the removal of dioxins, followed by a fabric filter for the collection of particulate matter. The fly ash samples were dried at 105 ± 5 °C for 24 h and stored in PE bottles at 25 °C.

2.2. Experimental apparatus

A schematic diagram of the laboratory-scale setup for the electrocoagulation experiment comprising two similar reactors is shown in Fig. 1. The first reactor handles the fly ash mixed fluid consisting of untreated fly ash mixed with deionized water and conducts a solid/liquid separation of the treated fly ash mixed fluid. The second reactor handles the leachate from the separation. The two processes were operated under the same conditions: the electrical current was fixed at 10 A, and the pump flow rate was 600 mL/min. The main components of the reactor are a DC power supply device, an electrocoagulation reactor that consists of a reactor tank, Al or iridium oxide (IrO_2) stick anodes, a stainless steel cathode, an upper tank channel, and a quantitative pump. The upper tank channel uses triangular spill weirs to provide a uniform distribution of the inlet fluid. Prior to each batch experiment, an Al or IrO_2 anode was fixed in the middle of the reactor tank, and the tank was modified for electrical conduction while the stainless steel cathode board was enclosed inside the body of the reactor tank. The bottom three-way valve of the reactor tank was used for control purposes and was connected by a transparent PVC tube to a quantitative pump to control the mixed fluid reflux. The three-way valve was opened in the direction in which the pump was moving the fluid when the mixed fluid was being processed for electrocoagulation. The fluid temperature increased to approximately 50 °C after approximately 1 h of treatment time. The power was subsequently turned off, and the separation of solid and liquid proceeded while the treated fly ash mixed fluid rested for approximately 4 h to yield the treated fly ash and leachate. The clear supernatant liquor was carefully collected for the leachate and prepared for the next electrocoagulation experiment at the second reactor. The volume of the reactor device was approximately 3.5 L, which was sufficient for conducting electrocoagulation experiments in 3.0-L batches.

2.3. Experimental methodology

The MSWI fly ash experiments involved three aspects: a reduction in the toxicity of the solid fraction of the fly ash, the recovery and reuse of the leachate, and the analysis of the fly ash samples by SEM-EDS and XRD. This research was divided into two unit experiments to be conducted on hazardous MSWI fly ash to confirm the feasibility of reusing the treated leachate. In the processing of Unit I experiments, two electrocoagulation experiments were conducted on the different Al and IrO_2 anodes. The L/S ratio was maintained at 20:1, which had been determined to be the optimal ratio for the hydrolysis of the MSWI fly ash in our previous study (Yang et al., 2012). The samples were untreated water (A and B) and treated ash (A and B). These untreated water samples were also treated in the electrocoagulation experiment to yield treated water (A and B). The untreated water A was also electrocoagulated with the IrO_2 anode to develop a contrast. In Unit I experiments, the total three ash samples and five leachate samples were checked to ensure that they adhered to the limits established by the toxicity characteristic leaching procedure (TCLP) regulations and the effluent wastewater regulations, respectively (TEPA website, 2012).

Unit II experiments were conducted on the untreated fly ash for the purpose of reducing the toxicity of the fly ash with the leachate that was recovered from Unit I. The acceptable treated leachate was mixed with untreated fly ash for the electrocoagulation experiments under the same set of operating conditions that were