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Hydrogen gas generation from metal aluminum-water interaction in municipal solid waste incineration (MSWI) bottom ash

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ABSTRACT

In the present research, municipal solid waste incineration (MSWI) bottom ash (BA) residues from three incinerators (N, K, and R) in Japan were collected for hydrogen gas generation purpose. The samples were split into four particle size fractions: (1) $d \leq 0.6$, (2) $0.6 \leq d \leq 1.0$, (3) $1.0 \leq d \leq 2.0$, and (4) $2.0 \leq d \leq 4.75$ mm for the characterization of metal aluminum, the relationship between the present metal aluminum and hydrogen gas production, and the influence of external metal aluminum on the enhancement of hydrogen gas. The batch experiments were performed for each BA fraction under agitated (200 rpm) and non-agitated conditions at 40 °C for 20 days. The highest amount of hydrogen gas (cumulative) was collected under agitation condition that was 39.4, 10.0, and 8.4 L/kg of dry ash for N2, R2, and K2 (all fraction 2), respectively. To take the benefit of the BA high alkalinity (with initial pH over 12), 0.1 and 1 g of household aluminum foil were added to the fractions 2 and 3. A Significantly larger amount of hydrogen gas was collected from each test. For 0.1 g of aluminum foil, the cumulative amount of gas was in the range of 62 to 78 L/kg of dry ash and for 1 g of aluminum foil the cumulative amount of hydrogen was in the range of 119–126 L/kg of dry ash. This indicated that the hydrogen gas yield was significantly a function of supplementary aluminum and the intrinsic alkaline environment of the BA residues rather than ash source or particle size.

1. Introduction

1.1. Overview

Incineration is an effective technique for the treatment of various types of wastes, as it can approximately reduce waste mass by 70% and volume by up to 90% (Lam et al., 2010), and can recover energy from waste in the forms of heat and electricity. Incineration results in the generation of solid residues including BA and FA that are essentially inorganic substances. BA generally accounts for almost 80% of the total amount of solid residues generated in a MSWI plant, and is typically composed of glass, ceramics, metals, minerals, and unburned matter. In Japan, in excess of 75% of the total solid waste generated in 2013 was treated by incineration. The amount of BA produced in this year was approximately 3.33×10^6 t (MOE, 2013). Metallic components, particularly metal aluminum, are the minor constituents of BA. Metal aluminum might reach up to 2 vol% in the fine fraction of BA (Saffarzadeh et al., 2016a). Although metal aluminum may be seen as a drawback for BA reuse purpose, its potential to produce hydrogen gas as a clean source of energy might be considered an advantage.

1.2. Literature review

The mechanism of hydrogen gas generation from metallic aluminum-water interaction has been extensively discussed by several research groups around the globe (Armstrong and Braham, 1996; Belitskus, 1970; Dupiano et al., 2011; Hiraki et al., 2005; Macanás et al., 2011) as a source of hydrogen for fuel cell and other industrial applications.

Generation of hydrogen gas from MSWI residues has also been the subject of a number of investigations essentially focusing on the explosion risk issues in a few incinerators and landfills in the US, Japan, Germany, Sweden and Switzerland (Arm and Lindeberg, 2006; Mizutani et al., 2000; Musselman et al., 2000). Hydrogen gas evolution and its hindrance effects on BA utilization

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Abbreviations: BA, bottom ash; BSE, backscattered electron; ECS, Eddy Current Separation; EDX, energy-dispersive X-ray spectroscopy; FA, fly ash; L/S, liquid/solid; MSWI, municipal solid waste incineration; RL, reflected light; SEM, scanning electron microscopy; XRD, X-ray diffraction.

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in cement and concrete production, and road base construction have also been reported (Bertolini et al., 2004; Kim et al., 2015; Pera et al., 2000). Hydrogen gas leads to disruptive expansion, decreased strength, swelling, and increased permeability of cement that might compromise the performance of the resultant product and limit or prevent the ash reuse (Kim et al., 2015; Lynn et al., 2017; Saikia et al., 2015; Todorović, 2006). Alternatively, instead of pretreating BA to remove metallic aluminum, Song et al. (2016) proposed to use it as an aerating agent to replace costly aluminum powder in the production of aerated concrete.

The recovery of hydrogen gas has been the subject of few investigations as an alternative reuse method of BA (Biganzoli et al. 2013; Ilyas et al., 2010; Larsson, 2014; Nithiya et al., 2016; Saffarzadeh et al., 2016a, 2016b). The literature has reported the characteristic of gas generation from BA and investigated factors affecting the degree of this reaction. The rate of hydrogen production has extensively varied in each research ranging from 1 to 7 L/kg of dry ash in average within highly variable time frames. The results were incomparable in detail, as each research group has used different experimental parameters for hydrogen gas generation from BA. It was realized that the maximum hydrogen generated was obviously a function of ash particle size, pH, reaction time, type of BA sample (fresh vs. aged) and BA cooling method, L/S ratio, additional components, temperature, agitation, and the experimental setup configuration.

1.3. Problem statement and aims

The efficient recovery of *metal aluminum* from the ash residues has been the target of several researches in recent years (Biganzoli et al., 2012a; Grosso et al., 2011; Hu et al. 2011). Nevertheless, Biganzoli et al., 2012b suggested that the residual amount of metal aluminum concentrates in the fine fraction of BA (and FA) and its efficient recovery might be practically impossible using the standard Eddy Current Separation (ECS) technology. The recovery rate of the metallic aluminum in the 0–4 mm fraction of BA was only 10% when a standard ECS was used (Biganzoli et al., 2013). Therefore, the fine fraction of BA could be considered a potential source for the production of hydrogen due to the presence of non-recyclable metal aluminum; an energy resource which is currently lost in the landfills.

Japan is an incineration-oriented country that generates huge amount of ash residues annually, as mentioned earlier. The co-disposal of BA residues with treated FA in the sanitary landfills is currently the major technique for the management of incineration residues in Japan. In 2011, almost 72% of BA was directly landfilled, about 20% was treated through melting process, less than 7% was used as recycled aggregate for cement production, and around 1% was sintered (Sakata, 2013). Therefore, the idea of generating hydrogen from BA could be an alternative to other sources of hydrogen by developing a sustainable and environmentally sound technology that can partially offset the economic and environmental burden of waste to energy industry. In addition, producing hydrogen from BA might facilitate its reuse as a cement and concrete additive with less swelling and expansion problems (Biganzoli et al. 2013).

In Japan, hydrogen is either imported as liquid hydrogen or essentially generated through hydrocarbon reforming and water electrolysis. In recent years, efforts have been made to produce hydrogen from other sources such as reforming biogas from sludge fermentation (NILIM, 2015). As hydrogen is going to be "the energy of the future" for Japan (METI, 2014), particularly in the auto and transport industries, huge investment on alternate technologies will be highly demanded in order to allow the country to diversify energy sources, reduce energy costs and dependence on overseas resources, and to address the global warming problems.

In the present research, priority was given to identifying the characteristics of metal aluminum in MSWI BA, and to evaluating the relationship between the present metal aluminum and hydrogen gas production in the fine fraction of BA (less than 4.75 mm). Moreover, the influence of commercial aluminum foil as a proxy for scrap metal aluminum on the enhancement of hydrogen production rate from BA was investigated. The mechanism of metal aluminum corrosion in BA and hydrogen production was also discussed in detail.

2. Materials and methodology

Samples of *water-quenched* BA products were collected from three stoker-type incineration plants (K, N, and R) in Japan. Approximately, 60 kg of BA was collected from each plant at different periods in 2014. The collected samples were air-dried and then oven-dried at 65 °C for 24 h to remove the moisture. The samples were also freeze-dried for efficient drying. The dried ash samples were made into respective sub-samples using the cone and quartering technique. They were sieved and grouped into four size fractions including (1) $d \leq 0.6$ mm, (2) $0.6 \leq d \leq 1.0$ mm, (3) $1.0 \leq d \leq 2.0$ mm and (4) $2.0 \leq d \leq 4.75$ mm; hereinafter only referred to as fractions (1), (2), (3), and (4), respectively. This set of sieves were selected for splitting the less than 5 mm BA samples into four fractions based on the American Standard Sieves ASTM Designation: E 11-04.[2] The fraction larger than 4.75 mm was not considered in this research, as they contained a greater amount of unburned substances.

The detailed methodology for the recovery of hydrogen from the BA products was described in Nithiya et al. (2016) and Saffarzadeh et al. (2016a, 2016b). The techniques are succinctly described here as well. All four fractions from the three incineration plants were examined for hydrogen generation potential in 150 ml reactors in triplicate. A mixture of 15 g of BA and 75 ml of distilled water (L/S ratio of 5) was supplied into each reactor. Hydrogen evolution was initially examined under three different temperatures (20, 40 and 70 °C). Although hydrogen generation increased as a function of temperature, the 40 °C was selected as the optimum reaction temperature in order to consider the energy and cost efficiency, particularly for the possibility of future industrial applications. Therefore, all the reactors were incubated at 40 °C for 20 days under both agitated (200 rpm) and non-agitated conditions. Since the initial pH values in the samples from all sources were significantly high ($\text{pH} \geq 12$), no additional alkali was added to the experimental setup.

To enhance the recovery rate of hydrogen, standard household aluminum foil was used as the external source of metal aluminum. It was found that the aluminum foil is made up of at least 98 wt% of elemental Al and insignificant amounts of O, Si, and, Fe based on SEM-EDX analysis. For hydrogen enhancement, only fractions 2 and 3 of BA from all three sources (K, N, and R) were chosen, because the cumulative hydrogen produced from these two fractions was the highest. Pieces of aluminum foil were crumpled into small spheres (ca. 1 cm in diameter) and two scenarios were adopted by adding of 0.1 and 1 g of aluminum spheres into each reactor (Fig. 1). The hypothesis was to take advantage of the intrinsic alkaline property of BA in order to achieve a pH high enough to promote the aluminum corrosion under relatively low temperatures.

The fresh and reacted BA particles as well as the reacted aluminum foils from different scenarios were selected in order to prepare standard petrographic thin polished sections

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[2] ASTM E11-04 Standard Specification for Wire Cloth and Sieves for Testing Purposes, ASTM International, West Conshohocken, PA, 2004, www.astm.org.

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