

HYDROGEN GAS EVOLUTION IN MUNICIPAL SOLID WASTE INCINERATION(NSWI) BOTTOM ASH RESIDUES

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論 文 名 : HYDROGEN GAS EVOLUTION IN MUNICIPAL SOLID
WASTE INCINERATION (MSWI) BOTTOM ASH RESIDUES
(一般廃棄物焼却灰における水素ガス生成に関する研究)

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論 文 内 容 の 要 旨

Approximately 3.6 million tons of aluminium (Al) is consumed in Japan annually in various industrial applications. About 29% of the Al used are able to be recycled while 7,000 tons of Al end up in landfills. In the presence of moisture (extreme pH) and oxygen limited condition, metal Al in the municipal solid waste incineration (MSWI) bottom ash could react to generate hydrogen (H_2) gas, which could lead to severe problems in recycling activities. Many studies have been conducted to increase metal Al recovery rate in MSWI bottom ash. However, the recovery rate for metal Al in particle size less than 4mm is nearly zero. Therefore, the main objective of this study is to determine the H_2 evolution capacity in MSWI bottom ash with optimal parameters and to enhance H_2 evolution with particle size refinement and addition of Al foil. Concurrently, the physical, chemical and mineralogical characteristics of MSWI bottom ash (bulk and fractionated air-cooled (AC-BA) and quenched (QBA)) were studied to understand its influence in H_2 evolution.

In Chapter 1, current status of MSW generation and management, MSWI bottom ash recycling activities and aluminium flow in MSW and MSWI were addressed. Besides, the obstacles faced due to the presence of metal Al and H_2 evolution from metal Al were also discussed. The objectives and structure of study were presented.

In Chapter 2, research materials and methodologies and instruments used in this study were comprehensively discussed. Two types of MSWI bottom ash (AC-BA) and (QBA) were collected from three stocker-type incineration facilities: N, K and R located in Osaka, Kurume and Fukuoka cities, respectively. The collected bottom ash residues were grouped as Bulk ($d \leq 4.75$ mm), Fraction 1 ($d \leq 0.6$ mm), Fraction 2 ($0.6 \leq d \leq 1$ mm), Fraction 3 ($1 \leq d \leq 2$ mm) and Fraction 4 ($2 \leq d \leq 4.75$ mm). Physical, chemical and mineralogical characteristics of bulk and fractionated AC-BA and QBA (both before and after hydrogen reaction) were analyzed. Hydrogen reaction set-up and volume calculation method was discussed in detailed. X-ray diffraction (XRD) and X-ray fluorescence (XRF) were used to analyze the chemical and mineralogical compositions of the solid bottom ash, while inductively coupled plasma atomic emission spectroscopy (ICP-OES) and Ion chromatography (IC) were used to analyze the concentrations of the inorganic elements and salts in bottom ash both before and after hydrogen evolution experiments.

In Chapter 3, the physical, chemical and mineralogical characteristics of both bulk and fractionated AC-BA and QBA were investigated. AC-BA consists of finer particles ($> 50\%$ of Fraction 1), while QBA contains coarser particles. Bottom ash from all three incineration facilities exhibited high alkalinity ($pH >$

11.5). Ca, Si, Al and Fe oxides were the most common elements in bottom ash which represented approximately 80% of the total chemical components. Quartz, calcite, pseudowollastonite and gehlenite are the common crystal phases observed in bottom ash. Phases such as hydrocalumite / Friedel's salt and portlandite were not observed in AC-BA. Existence of primary hydroxide layer was observed in QBA particles indicates H₂ evolution started during quenching process. AC-BA was hypothesized to yield higher H₂ due to the presence of individual Al particles and absence of quench phase.

In Chapter 4, the H₂ evolution in MSWI bottom ash residues were analyzed with various controlling factors. Both AC-BA and QBA observed generating H₂. However cumulative H₂ in QBA (bulk) was 3-8 folds lower than AC-BA of N (56 L/kg -dry ash) and K (66 L/kg dry ash) Facilities which indicates that quenching process and quench phase have great influence on cumulative H₂. Cumulative H₂ in Fraction 1 ($d \leq 0.6\text{mm}$) of AC-BA of N (80.6 L/kg dry ash) and K (67 L/kg dry ash) was 22-30% and 22-55% higher than other particle fractions for 20 days of reaction. Fraction 2 ($0.6 \leq d \leq 1\text{mm}$) of QBA (N) showed 28-44% higher cumulative H₂ than other fractions. Similarly all F2 of Facility K and R showed higher cumulative H₂. Hydrogen evolution under different experimental condition revealed that metal Al distribution in bottom ash (as single particle) and quenching process has direct influence.

In Chapter 5, H₂ evolution enhancement in bulk AC-BA and QBA of Facility N was carried out. Particle size refinement leads to H₂ evolution enhancement where cumulative H₂ in AC-BA and QBA increased 23% and 69% respectively after grinding. Addition of Al foil at 1 and 3 wt.% into bulk AC-BA and QBA showed an increase of 64% and 87% respectively. However, in this study addition of Al foil has no influence in enhancing H₂ evolution in bottom ash. Instead, the intrinsic alkalinity of bottom ash products with $\text{pH} > 11.5$ enhanced H₂ evolution in Al foil.

Chapter 6 consists of general discussion and conclusions. Hydrogen generation can be observed in both bulk and fractionated AC-BA and QBA. However, cumulative hydrogen and its generation rate vary among bottom ash samples depending on their physical, chemical and mineralogical characteristics. Metal Al distribution and concentration has great impact on the cumulative hydrogen produced from the bottom ash residues. However, the former showed more impact on hydrogen evolution. Quenching process has great effect on bottom ash particle size distribution, elements leachability, metal distribution and hydrogen generation rate.

A considerable amount of H₂ gas was released to the atmosphere during quenching process. Therefore, in-situ hydrogen recovery during quenching process could be a reasonable option as air-cooling of bottom ash is not a practical method to be used in the incineration facilities as a substitute for water-quenching process. Approximately 560 l H₂ could be recovered for 1 tons of bottom ash (AC-BA), if effective in-situ H₂ recovery technique is applied. Recovered hydrogen gas could provide an additional economic and environmental values if directed as clean fuel for electricity generation.