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# Accelerated carbonation using municipal solid waste incinerator bottom ash and cold-rolling wastewater: Performance evaluation and reaction kinetics

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## ABSTRACT

Accelerated carbonation of alkaline wastes including municipal solid waste incinerator bottom ash (MSWI-BA) and the cold-rolling wastewater (CRW) was investigated for carbon dioxide (CO<sub>2</sub>) fixation under different operating conditions, i.e., reaction time, CO<sub>2</sub> concentration, liquid-to-solid ratio, particle size, and CO<sub>2</sub> flow rate. The MSWI-BA before and after carbonation process were analyzed by the thermogravimetry and differential scanning calorimetry, X-ray diffraction, and scanning electron microscopy equipped with energy dispersive X-ray spectroscopy. The MSWI-BA exhibits a high carbonation conversion of 90.7%, corresponding to a CO<sub>2</sub> fixation capacity of 102 g per kg of ash. Meanwhile, the carbonation kinetics was evaluated by the shrinking core model. In addition, the effect of different operating parameters on carbonation conversion of MSWI-BA was statistically evaluated by response surface methodology (RSM) using experimental data to predict the maximum carbonation conversion. Furthermore, the amount of CO<sub>2</sub> reduction and energy consumption for operating the proposed process in refuse incinerator were estimated.

**Capsule abstract:** CO<sub>2</sub> fixation process by alkaline wastes including bottom ash and cold-rolling wastewater was developed, which should be a viable method due to high conversion.

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## 1. Introduction

Accelerated carbonation is an effective method for stabilizing alkaline wastes; meanwhile, permanently storing CO<sub>2</sub> as solid carbonates (Bobicki et al., 2012; Lackner et al., 1995; Pan et al., 2013a). In many countries such as France and Canada, natural ageing and/or ambient air carbonation are the standard practice to stabilize alkaline solid wastes such as bottom ash (Assamoi and Lawryshyn, 2012; Rendek et al., 2006; Santos et al., 2013a). CO<sub>2</sub> may react with metal-oxide-bearing materials to form stable and insoluble carbonates, with calcium oxide (CaO) and magnesium oxide (MgO) being the most favorable metal oxides in reacting with CO<sub>2</sub>. Since mineral carbonation is an exothermic process, additional heat inputs and energy costs are minimal (Eloneva et al., 2008). The mineral carbonation process for CO<sub>2</sub> fixation was initially aimed at natural silicates (Lackner et al., 1995).

Recently, carbonation of alkaline wastes such as municipal solid waste (MSW) and steelmaking slag has been receiving more attention due to their availability, low cost, and high CaO and/or MgO contents (Bobicki et al., 2012; Pan et al., 2012). In addition, the carbonation of alkaline solids wastes can improve the chemical and physical characteristics of wastes and facilitate their reuse in a variety of applications, such as synthesis of construction materials (Fernandez Bertos et al., 2004a).

Incineration is one of the environmental friendly methods to dispose MSW, especially where recycling or reuse are not possible, because the mass and volume reductions of original waste can be up to 85% and 95%, respectively. Despite the substantial volume reduction of MSW by incineration, the MSW incinerator (MSWI) residues, i.e., bottom ashes (BA) and air pollution control (APC) ashes would be up to a mass fraction of 20% from the original MSW (Bobicki et al., 2012; Rendek et al., 2006). The MSWI-BA accounts for 80–90% of the total mass of the MSWI residues (Arickx et al., 2006). The elemental composition of MSWI-BA depends primarily on the composition of the waste input, which

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## Nomenclature

### Materials

CRW cold-rolling wastewater  
MSWI-BA municipal solid waste incinerator bottom ash

### Analytical methods and models

DTG differential thermo-gravimetric analysis  
DSC differential scanning calorimetric analysis

EDX energy dispersive X-ray spectroscopy  
PSD particle size distribution  
RSM response surface methodology  
SCM shrinking core model  
SEM scanning electron microscope  
TGA thermo-gravimetric analysis  
XRF X-ray fluorescence  
XRD X-ray diffraction

may vary with location, season and recycling schemes in operation (Fernandez Bertos et al., 2004a; Teir, 2008). Since MSWI-BA is typically classified as a non-hazardous waste according to the European Waste Catalogue (European Union, 2000), it is currently being utilized as an aggregate substitute in road bases and bituminous pavement in European countries (Astrup et al., 2006; Teir, 2008).

MSWI-BA is a heterogeneous mixture of slag including ferrous and non-ferrous metals, ceramics, and other non-combustible materials. The major elements in MSWI-BA are O, Cl, Ca, Si, Al, Fe, Na, K, Mg, and C, with trace elements including Cu, Zn, S, Pb, Cr, Ni, Sn, Mn, Sb, V, and Co (Teir, 2008; Todorovic and Ecke, 2006). Mineralogical studies indicate that the main crystalline phases of MSWI-BA are typically silicates (e.g., quartz [SiO<sub>2</sub>], gehlenite [Ca<sub>2</sub>Al<sub>2</sub>SiO<sub>7</sub>], olivine [(Mg, Fe)<sub>2</sub>SiO<sub>4</sub>], and augite [(Ca,Na)(Mg,Fe,Al,Ti)(Si,Al)<sub>2</sub>O<sub>6</sub>]), sulfates (e.g., anhydrite [CaSO<sub>4</sub>], ettringite [Ca<sub>6</sub>Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>(OH)], and gypsum [CaSO<sub>4</sub>·2H<sub>2</sub>O]), carbonates (e.g., calcite [CaCO<sub>3</sub>]), and hematite [Fe<sub>2</sub>O<sub>3</sub>] (Fernandez Bertos et al., 2004a; Teir, 2008). With the specific chemical characterizations of MSWI-BA, it exhibits its potential CO<sub>2</sub> fixation capability due to its calcium content and alkaline properties. In addition, the mineral carbonation of MSWI-BA can immobilize heavy metals and effectively prevent their leaching, especially for Cr, Cu, Pb, Zn, and Sb (Arickx et al., 2006; Fernandez Bertos et al., 2004a; Todorovic and Ecke, 2006).

Mineral carbonation processes conducted in the aqueous phase have proven to be more effective than those utilizing the dry gas–solid method (Bobicki et al., 2012; Fernandez Bertos et al., 2004g; Pan et al., 2012). In addition, aqueous carbonation of industrial solid wastes can be coupled with alkaline wastewater resulting in further lowering material and energy consumptions (Chang et al., 2013; Pan et al., 2013a). Therefore, the objectives of this study were (1) to evaluate the performance of aqueous carbonation using MSWI-BA coupled with cold-rolling wastewater (CRW) via a slurry reactor for CO<sub>2</sub> fixation, (2) to determine the reaction kinetics and the rate-limiting steps of accelerated carbonation based on the shrinking core model (SCM), (3) to establish a response surface model for visualizing the effect of different operating parameters on carbonation conversion, and (4) to estimate the energy consumption and operating cost of the developed process for CO<sub>2</sub> fixation.

## 2. Materials and methods

### 2.1. Materials

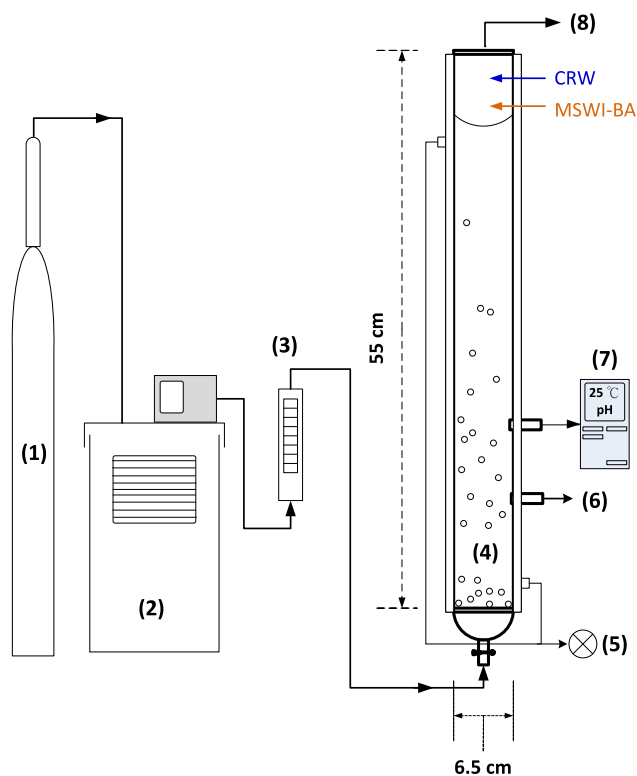
The ground MSWI-BA provided by Bali Refuse Incineration was sieved into three categories. i.e., <125 μm, 125–350 μm, and 350–500 μm, and dried at 105 °C for 8 h to eliminate moisture. After that, the MSWI-BA was stored at room temperature in small airtight containers and then placed in a larger capped container. In addition, high-pressure CO<sub>2</sub> gas with a volumetric concentration

of 99% was supplied by Ch'ing-Feng Gas Corporation (Taipei, Taiwan).

On the other hand, CRW produced from a steel manufacturer (Company B) was used as the liquid agent during the carbonation reaction in a slurry reactor. The CRW is the inevitable wastewater from cold rolling process in steelmaking industry, which typically needs additional neutralization and treatment before its discharge from the industry. Generally, the characteristics of CRW can be classified into three groups: alkaline (inorganic), acidic (inorganic), and oily (organic). In this study, the alkaline CRW was used (i.e., pH ~ 11.3), containing sodium of 800–1000 mg/L, potassium of 150–270 mg/L, chloride of 1400 mg/L, and sulfate of 150–230 mg/L.

### 2.2. Aqueous carbonation experiments

The effects of the operating conditions including reaction time, liquid-to-solid (L/S) ratio, gas flow rate, and particle size on the carbonation conversion of MSWI-BA were evaluated. Fig. 1 shows a



**Fig. 1.** Schematic diagram of the experimental set-up for carbonation of MSWI-BA in a slurry reactor. (1) CO<sub>2</sub> gas cylinder; (2) circulating bath; (3) rotameter; (4) slurry reactor; (5) heating jacket; (6) sampling; (7) thermo couple and pH analyzer; (8) vent to hood.

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