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## A fast and simple method to monitor carbonation of MSWI bottom ash under static and dynamic conditions



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#### ABSTRACT

Accelerated carbonation may be employed to improve the leaching behaviour and the geotechnical properties of MSWI bottom ash (BA). Here we report on a novel method to monitor and evaluate the progress of carbonation in both static and dynamic reactor systems. The method is based on following the pressure drop in the gas phase induced by the  $CO_2$ -uptake of BA and was benchmarked against carbonate contents as measured by thermogravimetry. Laboratory results demonstrated the serviceability and reproducibility of the method. Complementary logging of relative humidity and temperature showed constant moisture conditions and self-heating induced by the exothermal carbonation reaction, respectively. Under dynamic conditions BA carbonation was higher than in the static reactor. Consistently, the self-heating was more pronounced. After a reaction time of 120 min the pressure records indicated a  $CO_2$ -uptake of 1.5 g  $CO_2/100$  g BA (static tests) and of 2.6 g  $CO_2/100$  g BA (dynamic tests). The proposed method is suited to study carbonation processes at minimum analytical expense and integrates over the small-scale heterogeneity of BA.

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#### 1. Introduction

Bottom ash (BA) is the dominant solid residue generated in municipal solid waste incinerators and waste-to-energy plants. Worldwide, the installation of incineration capacities has increased (Santos et al., 2013) causing increasing quantities of BA. Disposal options for BA (landfilling or geotechnical reuse) depend on country-specific guidelines especially regarding the leachability of trace elements (Liu et al., 2015; Verbinnen et al., 2016).

Fresh BA contains highly reactive constituents and their transformation under ambient conditions has been referred to as the ageing of BA. One of the most important ageing processes is carbonation by atmospheric carbon dioxide (CO<sub>2</sub>). Due to the presence of alkaline mineral phases (mainly Ca(OH)<sub>2</sub>; Rendek et al. 2006a, 2006b) the pore water of BA acts as a CO<sub>2</sub>-trap (Fernández Bertos et al., 2004b). The simplified carbonation reaction (1) shows that by CO<sub>2</sub>-uptake Ca(OH)<sub>2</sub> is converted to CaCO<sub>3</sub> and alkalinity is consumed.

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O, rH = -113kJ/mol$$
 (1)

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For a more detailed description of the reaction routes refer to Costa et al. (2007) and Pan (2012). Consumption of BA alkalinity by carbonation is beneficial since it reduces the leachability of amphoteric trace metals (Saffarzadeh et al., 2011) and thereby lowers disposal costs. For Germany, cost savings between  $10~\epsilon$  and  $45~\epsilon$  per metric tonne of BA have been estimated after improving the leacheate characteristics by carbonation (our own unpublished data). Carbonation may also improve the geotechnical properties of BA, e.g. by increasing the compressive strength and modulus of elasticity (Chimenos et al., 2005; Fernández Bertos et al., 2004b).

Under ambient conditions, carbonation is slow (Costa et al., 2007) and limited by the access of  $CO_2$  requiring storage periods of up to several months (Astrup, 2007; Santos et al., 2013). Therefore, considerable efforts have been made to accelerate  $CO_2$ -uptake by increasing the  $CO_2$  partial pressure under static and dynamic conditions.

In static laboratory-scale reactors a thin layer of BA is contacted with the CO<sub>2</sub> enriched gas phase without agitation of the solid. Such systems with flow-through supply of CO<sub>2</sub> have frequently been described (Arickx et al.,2006; Lin et al.,2015; Sakita and Nishimura,2015; Nam et al.,2012; Um et al.,2013; van Gerven et al.,2005). As an alternative approach, pressurized stainless-steel

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vessels with discontinuous gas supply operated under  $CO_2$  partial pressures of  $2 \cdot 10^2$  kPa to  $1.7 \cdot 10^3$  kPa have been used by Baciocchi et al. (2010), Fernández Bertos et al. (2004a), Johnson (2000), and Rendek et al. (2006a).

In view of industrial scale applications dynamic reactor systems, e.g. rotating drums, have been suggested (Fernández Bertos et al., 2004b; Zingaretti et al., 2013). These simultaneously provide agitation, exposure to the reaction gas, and mixing. Rotating drum reactors have been tested under flow-through conditions and showed superior CO<sub>2</sub>-uptake as compared with static reactor systems (Lombardi et al., 2016).

In practice, the management of BA ageing is solely based on whether or not leachate properties comply with legal values. A comprehensive and systematic development of appropriate methods for the accelerated carbonation of BA including quality control for  $\rm CO_2$ -uptake is lacking. As a prerequisite these require a time-resolved monitoring of the process capable of integrating over the heterogeneity of BA.

In this study we tested the hypothesis that CO<sub>2</sub>-uptake of BA can adequately be deduced from the pressure drop observed in closed reactor systems. This would spare classical analyses and provide a better understanding of processes. In addition, pressure monitoring integrates over the whole reactor loading which circumvents problems related to small-scale variability of BA. Quantification of CO2-uptake from pressure data was first suggested by Sun et al. (2008) for air pollution control residues. However, their application was restricted to pressures >200 kPa. This is beyond practical conditions where exhaust gas from thermal processes is an available CO<sub>2</sub> source. Here, we expanded the approach to atmospheric pressure conditions to monitor and evaluate the progress of carbonation. The method is based on following the pressure drop in the gas phase induced by the CO2-uptake of BA. Suitability was tested both in a static reactor system with manual CO2-dosage and a rotating drum reactor with automated  $CO_2$  – supply.

#### 2. Materials and methods

#### 2.1. Bottom ash sampling and characterisation

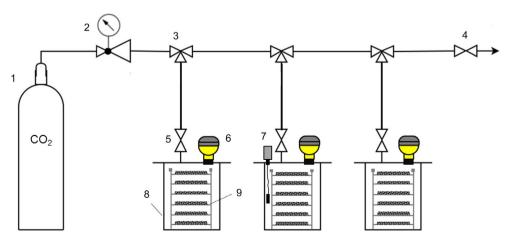
Freshly quenched BA was obtained from a grate-type refuse derived fuel incinerator located in central Germany. Annually, the incinerator handles 25,000 metric tons of sorted commercial waste residues providing 75,000 MWh to a district heating system. Total BA production amounts to 5000 metric tons per year, approximately. The BA was sampled at the end of an apron conveyor which transports the material from the quenching tank to the collection containers. The sample was screened to remove the 11 mm oversize fraction (about 40%-wt. of the raw sample). This considered findings by Lombardi et al. (2016) according to which the coarse material contributes little to the overall carbonation. Moisture content was determined gravimetrically after drying subsamples at 105 °C for 24 h (DIN EN 12880). The particle sizedistribution was determined by sieving with stainless-steel screens (mesh size 0.125, 1, 2, and 4 mm) in conformity with DIN 18123. Substance density was obtained by pycnometry (EN 11508). The loose bulk density of the fresh BA was determined by weighing a graduated cylinder holding 100 mL of the material. Table 1 summarizes the results. These are consistent with values reported elsewhere (Chimenos et al., 2005; Costa et al., 2007).

#### 2.2. Reactor configuration

Fig. 1 shows the experimental set-up for the accelerated carbonation under static conditions. Experiments were conducted in glass reaction vessels with a void volume of 1 L. Six vessels connected in series were equipped with piezoresistive pressure sensors (OxiTop™, WTW, Weilheim, Germany) and sealed air-tightly. The OxiTop respirometry system is commonly used to monitor biological aerobic activity in water, soil, or waste (Ilyas et al., 2014; Rendek et al., 2006b). In the classical set-up, CO₂ evolved by

**Table 1**Moisture content, loose bulk density, particle density and particle size distribution of fresh bottom ash.

Water content	Loose bulk density	Particle density	<0.125 mm	0.125-1.00 mm	1.00-2.00 mm	2.00-4.00 mm	4.00–11.0 mm
% wt.	kg/L	kg/L	% wt.				
15.2	1.14	2.78	18.7	35.8	17.9	16.8	10.8



**Fig. 1.** Set-up of the laboratory-scale carbonation experiments under static conditions. (1) CO<sub>2</sub> supply; (2) ball valve with flow rate display; (3) 3-way valve; (4) exhaust gas; (5) 2-way valve; (6) OxiTop pressure sensor; (7) temperature and relative humidity sensor; (8) reaction vessel; (9) sample rack.

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