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Transferring the Carbonation of Biomass Bottom Ash from Batch to Continuous Operation

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Abstract

Biomass incineration residues may be used for CO_2 capture and storage by carbonation of waste-borne metal hydroxides. Moist carbonation of these ashes can be performed under mild conditions (ambient pressure and temperature) and accelerated by using a CO_2-rich gas. The process requires the wetting of the typically dry-discharged ashes and highly depends on the moisture content. Handling of the moist ashes in continuously operated reactors is challenging, especially due to build-up of incrustations. In this study, the carbonation of biomass bottom ash was performed in fixed-bed and rotating drum reactors. Wetting was performed either manually prior to fixed-bed carbonation or with spray-nozzles. The moisture dependent CO_2 uptake was evaluated by a gas balance. Results show that the CO_2 uptake in the fixed-bed and rotating drum was consistently between 20 and 35 g/kg, although the CO_2 concentration in the rotating drum was significantly lower (10 vol% vs. 100 vol%). In the fixed bed, the uptake strongly depended on the moisture content, while in the rotating drum the effect was less pronounced. Application of such rotating drums allows for a continuous solids feed while combining wetting and carbonation.

Keywords: mineral carbonation, alkaline waste, carbon capture and storage, rotating drum

1. Introduction

Mitigating climate change requires a substantial reduction of greenhouse gas emissions from the energy supply sector. To meet the global warming target of 1.5 °C at maximum, the CO_2-neutral supply from biomass would need to significantly increase until 2050. A CO_2-negative energy supply could be implemented if biomass combustion were combined with carbon capture and storage. The main obstacle to this is the small scale of many biomass combustion plants resulting in disproportionally high costs for CO_2 capture units and the transport of captured CO_2 from decentralized sources to disposal sites (Gough and Upham 2011).

Worldwide, the combustion of solid biomass generates over 450 million tons of mostly alkaline ashes of which the majority is disposed of in landfills (Voshell et al. 2018). Considering that these ashes act as CO_2 absorbents (He et al. 2019), after disposal they may be regarded a final CO_2 sink. This sink is primarily driven by carbonation of readily available alkaline metal (hydr-)oxides. Other benefits of carbonation include an easier handling of the material due to granulation and hardening as well as potentially lower disposal costs due to reduced leachability of some toxic trace metals.

At ambient temperature and pressure, carbonation requires the presence of a free water phase were alkaline components and CO_2 dissolve, combine, and precipitate as carbonates. Available routes include the so-called wet carbonation with excess water (liquid to solid ration, L/S > 2) or the moist carbonation were the reaction takes place in a water film (L/S < 1). In the presence of water, hydration of metal oxides (i.e. calcium oxide, equation 1) precedes the actual carbonation reaction (equation 2). The carbonation rate has been shown to be sensitive towards the L/S (Schnabel et al. 2021b) due to the mass transfer of reactants through the ash or liquid layer and/or the liquid-solid interface.

$$\mathrm{CaO} + \mathrm{H_2O} \rightarrow \mathrm{Ca(OH)_2} \quad (1)$$

$$\mathrm{Ca(OH)_2} + \mathrm{CO_2} \xrightarrow{\mathrm{H_2O_{(l)}}} \mathrm{CaCO_3} + \mathrm{H_2O} \quad (2)$$

Depending on the incinerator design (e.g. inclusion of a quenching step) and the ash origin (fly ash, bottom ash etc.) the material is discharged in wet or dry state. The latter is the case for fly ash from combustion plants with a dry flue gas cleaning system (cyclones, filters). Dry discharge also holds for bottom ash from most of the smaller-scale biomass combustion plants (da Costa et al.

2019). Therefore, carbonation of such ashes needs humidification to allow for mineral carbonation within reasonable timeframes.

Rotating drum reactors have proven to overcome the limitations of fixed bed setups in the carbonation of alkaline waste materials. The continuous mixing of the solid in rotating drum reactors substantially increases the level and the reaction speed of alkaline waste carbonation (Brück et al. 2018; dos Reis et al. 2020; Łączny et al. 2015; Lombardi et al. 2016). Reactor hold-up, residence time and, thus, capacity can be controlled by rotation and mass flow rate using a proportionality between rotation-normalized flow rate and reactor hold-up (Schnabel et al. 2021a).

Here, we compared the carbonation of initially dry biomass ash under conditions of a fixed bed and a rotating drum. The rotating drum was operated as a batch reactor to evaluate the influence of moisture conditions. Additional operation in continuous-feed mode served to maximize reactor hold-up and residence time.

2. Materials and methods

2.1. Waste material

Carbonation experiments were performed with biomass ash originating from the incineration of waste wood chips and green waste. The incineration plant with a grate furnace and a dry-discharge system has a capacity of 2250 Mg/a. Samples were taken directly from the discharge container and screened with a mesh size of 5 mm to remove metal particles like nails, screws or brackets. Until experimental testing, the ash was stored in air-tight clamping ring drums.

2.2. Carbonation reactors

The carbonation reaction was performed in two reactors of different size representing lab- to pilot-scale.

A lab-scale fixed bed reactor was used for the initial characterization of the material and to determine the carbonation potential at optimum moisture conditions. The reactor was a gas-tight vessel (V≈1 L) equipped with a pressure sensor (WTW OxiTop) and a reactant gas supply (100 vol% CO_2) controlled by a two-point control (switching points at gauge pressure of ± 75 hPa). A sample rack with stainless steel frames covered with nylon grids served to expose the solid sample to the reactant gas.

The pilot-scale rotating drum reactor (V≈100 L) was used for batch and continuous carbonation experiments. The rotating drum was driven by variable-speed motor via a chain drive attached to a sprocket on the outer reactor mantle.

Reactant gas (10 vol% CO_2) was continuously supplied at 50 L/min by a gas mixing station (HiTec Zang GmbH Gmix) mixing CO_2 (N4.5) and compressed air. The exhaust was routed to sensors for temperature, relative humidity (AREXX TSN-TH70E) and CO_2 concentration (MRU VarioPlus Industrial).

For batch operation, the rotating drum was equipped with two spray nozzles for uniform humidification along the reactor axis. During continuous operation, the solid was fed with a screw-conveyor and humidified with a spray nozzle mounted at the reactor inlet (see Figure 1).

2.3. Experimental procedure

For the carbonation experiments the ash was humidified to yield desired liquid-to-solid ratios (L/S). In the fixed bed carbonation L/S of 0.1 to 0.5 were adjusted to test the complete range of moist carbonation. Humidification and mixing were performed outside the reactor before preparing the sample rack. Exceeding L/S 0.5 led to slurries that could not be carbonated as a fixed bed.

Considering the results of the fixed bed experiments the pilot-scale batch experiments were performed with L/S 0.1 to 0.3. Humidification and mixing were performed inside the rotating drum via the spray nozzles.

Continuous carbonation experiments were performed at L/S 0.1 while varying the feed rate to influence the reactor hold-up and residence time. An exit dam obstructing approx. 86% of the cross-sectional area was used to adjust a minimum hold-up of ~ 27 L.

The influence of the rotation-normalized flow on the reactor hold-up was evaluated from the carbonation tests (moist ash) and with dry ash to characterize the rotating drum reactor.

Specific CO_2 uptake was calculated from a gas balance using the measured pressures (fixed bed reactor, equation 3), the in- and output CO_2 concentrations (rotating drum reactor, equation 4).

$$\zeta_{\text{pressure}} = \frac{V}{\text{m}_{\text{BBA}} * R_S} * \int \frac{\Delta \text{p}}{T} \quad \textbf{(3)}$$

$$\zeta_{concentration} = \frac{\rho_{CO2}}{m_{BBA}} * \int \dot{V}_{in} * c_{in} - \dot{V}_{out} * c_{out} \quad \textbf{(4)}$$

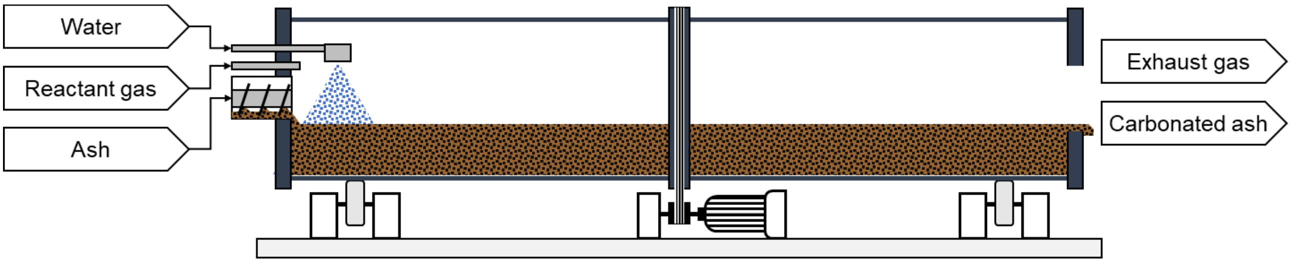

Figure 1. Configuration of the continuously operated rotating drum reactor.

3. Results and discussion

3.1. Carbonation performance

Carbonation results (Figure 2) show that the maximum CO_2 uptake within 2 hours was 25 and 35 g/kg in both reaction system (fixed bed / rotating drum), although the CO_2 concentration in the rotating drum was significantly lower (10 vol% vs. 100 vol%). In the fixed bed, the uptake strongly depended on the moisture content, while in the batch rotating drum the effect of the moisture was less pronounced. Rotating drums provide an intensive mixing and permanently renew the solid's surface (Brück et al. 2018). This promotes the distribution of water in the solid bed and avoids a locking effect where the diffusion of CO_2 is hampered by aggregation and formation of passivating layers.

Comparison of error bars of the CO_2 uptake shows a more pronounced variability of the results obtained under fixed bed conditions compared to the rotating drum. This may be attributed to the inherent small-scale heterogeneity of the ash. Unlike the fixed bed reactor (void volume 1 L) the pilot-scale rotating drum (100 L) integrates over the small-scale heterogeneities thereby yielding less variable results.

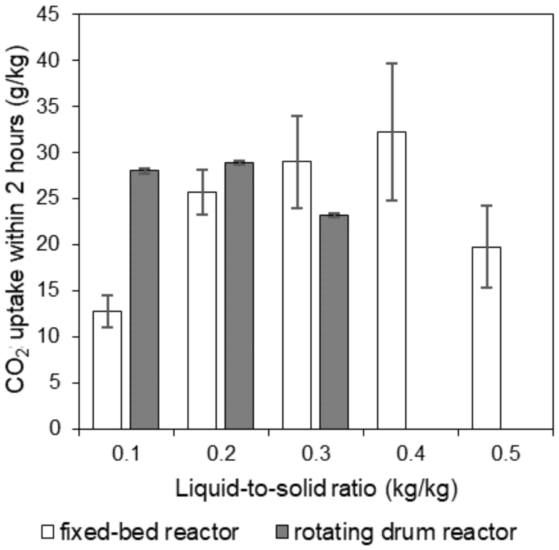

Figure 2. Influence of moisture conditions on the CO_2 uptake in the fixed bed (100 vol% CO_2) and rotating drum reactors (10 vol% CO_2). Error bars denote the standard deviation of replicated experiments.

3.2. Maximization of reactor hold-up

Proportionality of the reactor hold-up vs. the rotation-normalized flow rate is depicted for the dry and moist ash in Figure 3. The mass hold-up (in kg) was lower for the moist ash due its lower bulk density compared to the dry ash (~0.48 vs. ~0.60 kg/L). Considering the volumetric hold-up and flow rate, a linear relation for both materials can be derived. The characteristics can be beneficially applied to select appropriate operating points for the continuous carbonation.

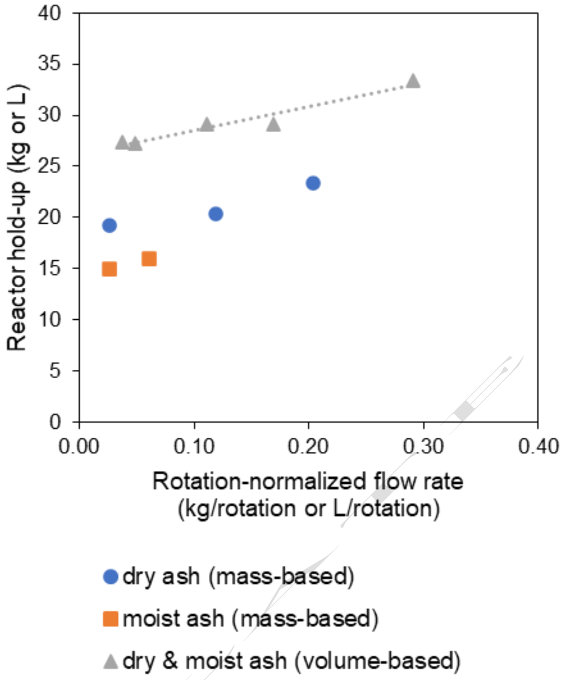

Figure 3. Influence of the rotation-normalized flow rate on the reactor hold-up. The representation includes mass-based as well as volume-based parameters. These were derived considering the material's bulk densities.

4. Conclusions

In this study the carbonation of dry biomass ash was successfully transferred from a batch fixed bed reactor to a continuously operated rotating drum reactor.

The carbonation in the rotating drum reactor has proven beneficial in terms of a lower water demand yet with a similar CO_2 uptake as in the fixed-bed reactor. The reactor hold-up and, thus, reactor capacity could be maximized by variation of flow rate and rotation frequency.

Ongoing work aims at determining the effect of process conditions on the BBA granulation and evaluating the CO_2 uptake at L/S below 0.1.

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