

Mineral carbonation from metal wastes: Effect of solid to liquid ratio on the efficiency and characterization of carbonated products[☆]



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HIGHLIGHTS

- Steel, phosphorus and blast furnace slags are suitable feedstock for mineralization.
- Efficiency of carbonation increases for all 3 materials when S/L ratio decreases.
- Silica from the raw material did not dissolve and formed the core of final particles.
- CaSO₄ left from incomplete carbonation also formed the core of final particles.
- CaCO₃ was deposited on the surface of the carbonated final particles.

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ABSTRACT

Mineral carbonation technologies aim at permanently storing CO₂ into materials rich in metal oxides. A multi-step mineralization process employing Ca-rich waste streams to precipitate calcium carbonate is investigated in this paper. Ground granulated blast furnace slag (GGBS), phosphorus slag (PS) and steel slag (SS) were employed as feeding materials for the process. Solid to liquid ratio (S/L) is an important factor which affects mineral carbonation and this study examines its effect on the carbonation efficiencies. The main phases present in the carbonated residues were identified using XRF, XRD and SEM-EDS analytical techniques. For the three materials investigated, the carbonation efficiency increased when the S/L ratio decreased (from 50 g/L to 25 g/L and then 15 g/L) because of the dilution effect. In a previous study, where an analog process was employed, efficiency using serpentine was found lower than that calculated here for GGBS and SS, and slightly above PS. This confirms that, in general, waste materials require less energy-intensive carbonation conditions, in comparison to mineral rocks. Finally, the structure of the carbonated particles is also discussed.

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1. Introduction

Nowadays fossil fuels account for 80–85% of the total of world energy use and they cause the release of about 30 Gt/year of CO₂ into the atmosphere [1]. Considering that CO₂ is a greenhouse gas, such large and increasing atmospheric CO₂ levels are causing climatic consequences [2]. In fact, it has been shown that the global average air and ocean temperatures have been increasing, global sea levels raising [3] and the number of severe weather events intensifying [4].

Carbon dioxide capture and storage (CCS) is a portfolio of technologies aiming to capture the CO₂ produced from the combustion of fossil fuels and other energy intensive process and then store it underground for a very long time [5]. Recently, interest for an

alternative route to geological storage, called mineral carbonation or mineralization, has increased because of its advantages of vast and permanent storage capacity [6]. The mineralization process consists of reacting CO₂ with a divalent metal oxide (MO) to produce a metal carbonate (MCO₃). However, despite the intrinsic advantages of mineralization, including being an ex-situ and permanent process, its costs are currently considered not competitive compared to those of geological storage [7].

At ambient conditions, the gas–solid mineralization reaction takes place on geological time-scales and, therefore, research has focused on developing different technologies to speed up the process [2]. A closed-loop, multi-step process using ammonium salts has recently been developed to extract magnesium from serpentine and produce separated streams of pure by-products at ambient pressure [6,8]. Furthermore, this process allows recovery of the chemicals used, reducing the overall environmental and economical impact, but thus far it has only been applied to serpentine minerals. It is interesting to highlight that there is increasing interest in using alkaline industrial waste residues as feedstock material

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for mineral carbonation [9–13]. This is because these materials require a lower degree of pre-treatment and less energy-intensive carbonation conditions, in comparison to mineral rocks [14,15]. Moreover, waste materials have the potential of storing 1 Mt/year CO_2 in the UK alone [16] and, furthermore, their use would avoid disposal requirements and could transform them into a potential revenue resource instead of a costly waste stream. However, their storage potential is limited if compared to the annual CO_2 emissions in the UK (470 Mt) [17]. This technology could be attractive for cases where the producer of suitable waste materials and the CO_2 emitter are located in close proximity [16].

This paper focuses on using a process similar to that previously developed for magnesium-rich serpentine minerals by Wang and Maroto-Valer [6,8], but employing calcium-rich waste streams, and therefore resulting in the precipitation of calcium carbonate instead of magnesium carbonate. The proposed mineralization process includes four main steps (Fig. 1): (1) mineral dissolution, (2) pH adjustment and precipitation of impurities, (3) carbonation reaction, and (4) regeneration of additives.

The overall process utilizes ammonium bisulfate (NH_4HSO_4) solution for the mineral dissolution phase (step 1), extracting calcium from the waste feeding materials and producing solid calcium sulfate (CaSO_4). This is an intrinsic difference with the process developed for serpentine, where, following the mineral dissolution step, MgSO_4 is formed in solution, while here CaSO_4 is precipitated. In fact, the two sulfates have different values of solubility in water, CaSO_4 has been reported of being slightly soluble [18] while MgSO_4 has a solubility of 26 g/100 mL (20 °C) [19]. Afterwards, ammonium carbonate ($(\text{NH}_4)_2\text{CO}_3$), from the CO_2 capture step, is added in the carbonation step (step 3), allowing the precipitation of calcium carbonate (CaCO_3). Raising the pH (step 2) is an important step between dissolution and carbonation because it allows the following precipitation of CaCO_3 . While the pH is raised, the impurities (Mg, Fe, Al) precipitate as hydroxides. This carbonation process could also re-circulate and regenerate the chemicals involved (step 4), i.e. NH_4HSO_4 and NH_3 . Therefore, this paper investigates the process described above for three differ-

ent metal wastes: ground granulated blast furnace slag (GGBS), phosphorus slag (PS) and steel slag (SS). For each waste material, the dissolution, adjustment of pH and carbonation steps of the overall close-loop mineralization process were studied.

Furthermore, solid-to-liquid (S/L) ratio is an important parameter which has been shown to affect significantly the efficiency of mineralization of serpentines [9,20,21,22,23]. Therefore, this work also investigates the effect of three different S/L ratios, starting from 50 g/L, the same employed by Wang and Maroto-Valer [6], and then reducing to 25 and 15 g/L in an attempt to improve the efficiency of mineralization, as previously reported for coal fly ash and steel slag [22] [24].

2. Experimental methods

2.1. Characterization of the parent samples

The three feedstock materials, namely, SS, GGBS and PS employed for this study were obtained from production plants of steel, iron and phosphorus respectively where a representative amount of 5 kg of each sample was collected from the residue of the manufacturing process and safely stored indoor for the experiments. SS is the by-product of the manufacture of steel from pig iron (blast furnace) and metal scrap and it was obtained from an electric arc furnace. GGBS is a by-product from the production of iron, resulting from the fusion of fluxing stone (fluorspar) with coke, ash and the siliceous and aluminous residues remaining after the reduction and separation of iron from the ore. PS is the by-product from yellow phosphorus production obtained from electric furnaces.

SS, GGBS and PS were characterized employing different techniques to assess the parameters important for mineral carbonation. The loss on ignition (LOI) was determined gravimetrically by taking ~1 g of a crushed representative sample from each material and drying for 1 h at 950 °C. Oxide composition was characterized using a PANalytical Axios Advanced X-ray Fluorescence (XRF) spectrometer. For XRF analysis, a fused bead sample was prepared using ~1 g of a crushed and finely ground representative sample.

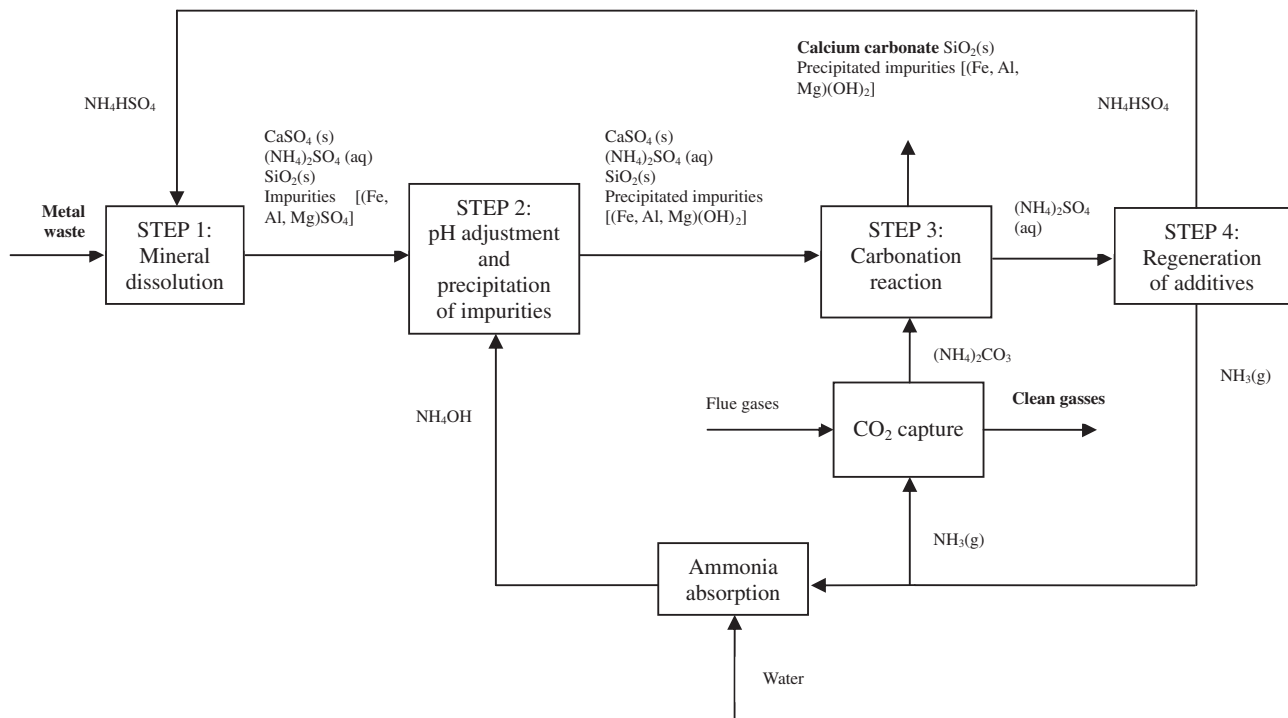


Fig. 1. Multi-step close loop mineral carbonation process.

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