



Towards zero-waste mineral carbon sequestration via two-way valorization of ironmaking slag



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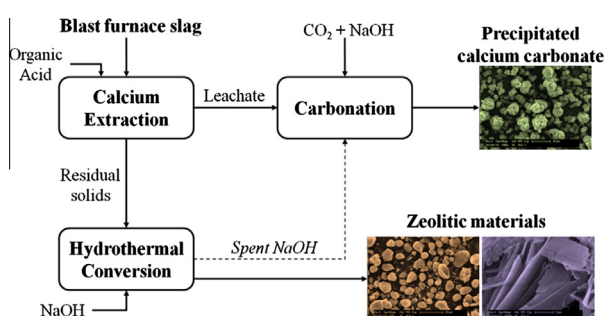
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HIGHLIGHTS

- A symbiotic process is developed to valorize blast furnace slag into two products.
- Carbonation of extracted acetic acid leachate results in the production of PCC.
- Hydrothermal conversion of the extraction residues forms various zeolitic materials.
- Leaching selectivity is a key factor in the valorization potential of both products.

GRAPHICAL ABSTRACT



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ABSTRACT

A three-stage process was developed to transform blast furnace slag (BFS) into two valuable products: precipitated calcium carbonate (PCC) and zeolitic materials. The conceptualized process aims to simultaneously achieve sustainable CO₂ sequestration and solid waste elimination. Calcium is first selectively extracted by leaching with an organic acid, followed by carbonation of the leachate to precipitate CaCO₃. In parallel, the hydrothermal conversion of the extracted solid residues in alkali solution induces the dissolution/precipitation mechanism that leads to the formation of micro- and meso-porous zeolitic materials. Leaching selectivity was identified as a key factor in the valorization potential of both products. Acetic acid satisfactorily limited the leaching of aluminum, required for the subsequent synthesis of zeolites, and carbonation of the acetic acid leachate resulted in the production of PCC of varied mineralogy and morphology, depending on processing conditions. In the hydrothermal conversion stage, the formation of zeolitic phases was observed, and their characteristics were found to vary depending on the calcium extraction efficiency in the previous stage, and the alkali (NaOH) concentration. The zeolitic phases produced, in order of increasing valorization potential, were: tobermorite, sodalite, lazurite, and analcime.

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1. Introduction

The utilization of waste materials such as steelmaking slags for CO₂ sequestration could be more appealing to the industry if the benefits of mineral carbonation and waste stabilization/valorization

could be combined symbiotically. Indirect carbonation is especially interesting due to the production of high-purity products, particularly precipitated calcium carbonate (PCC) [1–6]; however, the generation of a large amount of destabilized solid residue (e.g. 0.73 t per tonne steel converter slag [5]), wherein hazardous heavy metal and metalloid components can become concentrated and mobilized [7], may hinder this approach. In order to meet the goal of sustainable CO₂ sequestration, a mineral carbonation processing scheme that results in zero solid waste is desirable. This study investigates the potential of two-way valorization of blast furnace slag (BFS), derived from carbon steel production, via: (i) production of PCC from the initial-stage extraction step; and (ii) utilization of the solid residue for the production of zeolitic materials via hydrothermal conversion.

Fig. 1 presents a schematic flowsheet of the conceptualized process. Its principal advantage is the generation of two marketable products, which can potentially off-set the cost of carbon sequestration. In addition, the approach presents opportunities for process integration, such as that highlighted in Fig. 1 (dashed line) whereby the residual alkalinity of the hydrothermal stage can be re-routed to the carbonation stage to induce carbonate precipitation. This integrated flowsheet can potentially contribute to the improvement of economic and environmental sustainability of mineral carbonation, thus advancing this route for carbon sequestration closer to industrial implementation.

Recent works on the production of PCC from steelmaking slags [3–6] have primarily focused on converter slag (also known as Basic Oxygen Furnace (BOF) slag), as this residue is in pressing need of new valorization routes [8]. However, this slag presents several inefficiencies as a carbon sink, which include: its relatively small production output compared to CO₂ emissions of steelworks; its monolithic morphology, requiring energy intensive milling to impart high reactivity; and its high intrinsic content of regulated heavy metals and metalloids [8]. In the present study, blast furnace slag was chosen as the feedstock to the proposed process due to three factors: (i) its large and global production (0.25–0.30 t BFS per tonne crude iron, resulting in approximately 300 Mt BFS produced globally in 2011 [9]), allowing for meaningful carbon storage capacity; (ii) its rather consistent chemical and mineralogical composition compared to other residual materials, which have allowed it to be commercialized in stringent applications such as slag cements [10]; and (iii) its relatively high aluminum content (10–14 wt% Al₂O₃ [10]), an essential component of aluminosilicate-based zeolites [11]. BFS is currently valorized to a large extent in

the cement industry for the production of blended cements where it partially replaces traditional Portland cement [10]. As the latter material has a large CO₂ footprint, due to calcination of limestone during production, this replacement results in reduction of CO₂ emissions. The present study aims to investigate an alternative and innovative valorization route that may result in higher value generation (PCC and zeolite) coupled to concomitant CO₂ abatement.

The production of PCC from steelmaking slags has been a major focus of research due to the numerous commercial applications for this material such as filler, pigment and color stabilizer, paint extender, pH buffer and neutralizer, fertilizer and animal feed [12]. Of particular note are the works of Teir et al. [1] and Eloneva et al. [2], who used acetic acid (CH₃COOH) for the extraction of calcium from BFS, and later carbonated the leachate with the aid of sodium hydroxide (NaOH) as an acid neutralization agent, to precipitate calcium carbonate crystals. Based on these investigations, acetic was utilized as the leaching agent in the present study for the first-stage calcium leaching from BFS. The question pursued in the present study was if the use of acetic acid in the extraction stage allows the symbiotic balancing of PCC quality and zeolitic material utility, by delivering the necessary leaching extent and selectivity.

The hydrothermal conversion of silica- and alumina-rich solids into zeolitic minerals via heat treatment in highly alkaline solutions has been sought for the valorization of fly ashes and bottom ashes from coal combustion and waste incineration [11,13]. The synthesis of zeolitic minerals from industrial and synthetic slags utilizing direct and indirect hydrothermal conversion methods has been reported by a limited number of researchers [14–16]. Sugano et al. [14] reported that when the CaO content of the raw material is higher than 20 wt% large quantities of undesirable phases form, due to their low microporosity; these include tobermorite (Ca₅Si₆O₁₆(OH)₂·4(H₂O)) and hydrogarnet (3CaO·Al₂O₃·(3–x)SiO₂·2xH₂O, 0 ≤ x ≤ 3). This indicates that extensive removal of calcium is required in the extraction stage, which is in line with maximizing the output of PCC in the carbonation stage of the present conceptual process flowsheet.

Prior researches on the hydrothermal conversion of BFS did not, however, consider using extraction agents amiable to PCC production. Rather, Kuwahara et al. [15] applied concentrated hydrochloric acid (HCl) as the leaching agent, which is not only impractical for mineral carbonation due to its high acid strength and energy intensive regeneration [16], but also undesirably leached the

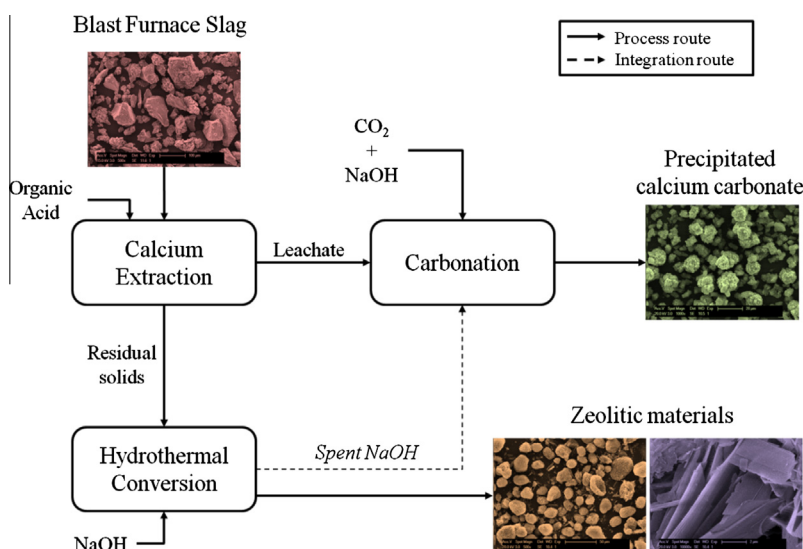


Fig. 1. Conceptualized flowsheet of two-way valorization of blast furnace slag.

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