



## Leaching modelling of slurry-phase carbonated steel slag



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### HIGHLIGHTS

- Slurry-phase accelerated carbonation of BOF steelmaking slag were investigated.
- Acid neutralisation and leaching of untreated and carbonated slag were studied.
- Carbonation affected the leaching of both major elements and metal contaminants.
- Geochemical modelling of the leaching solutions was adopted to interpret leaching.

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### ABSTRACT

In the present work the influence of accelerated mineral carbonation on the leaching behaviour of basic oxygen furnace steel slag was investigated. The environmental behaviour of the material as evaluated through the release of major elements and toxic metals under varying pH conditions was the main focus of the study. Geochemical modelling of the eluates was used to derive a theoretical description of the underlying leaching phenomena for the carbonated material as compared to the original slag. Among the investigated elements, Ca and Si were most appreciably affected by carbonation. A very clear effect of carbonation on leaching was observed for silicate phases, and lower-Ca/Si-ratio minerals were found to control leaching in carbonated slag eluates as compared to the corresponding untreated slag sample as a result of Ca depletion from the residual slag particles. Clear evidence was also gained of solubility control for Ca, Mg and Mn by a number of carbonate minerals, indicating a significant involvement of the original slag constituents in the carbonation process. The release of toxic metals (Zn, V, Cr, Mo) was found to be variously affected by carbonation, owing to different mechanisms including pH changes, dissolution/precipitation of carbonates as well as sorption onto reactive mineral surfaces. The leaching test results were used to derive further considerations on the expected metal release levels on the basis of specific assumptions on the relevant pH domains for the untreated and carbonated slag.

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### 1. Introduction

The steel manufacturing industry generates considerable amounts of solid residues. The 2013 worldwide crude steel production has been reported to amount to  $1.65 \times 10^3$  Mt, with a major contribution of Asia ( $1.12 \times 10^3$  Mt) followed by Europe ( $2.05 \times 10^2$  Mt) [1]. The main technologies adopted are the basic oxygen furnace (BOF) and the electric arc furnace (EAF) processes, which account for 72.0 and 27.5% of the total amount of crude steel produced [1] and generate 125.8 and 168.6 kg of slag per tonne of steel, respectively [2]. The steel industry is also a significant contributor to the anthropogenic CO<sub>2</sub> emissions, with an average emission

factor of  $\sim 2.2$  t CO<sub>2</sub>/t crude steel [3], which however may vary considerably depending on the production route used, the product mix, the energy efficiency of the process, the fuel mix and its carbon intensity, as well as the electricity carbon intensity.

Over the last decades several studies have been conducted on the use of alkaline industrial residues, including steelmaking slag [4–14], for CO<sub>2</sub> sequestration from stationary sources.

Although the leaching of inorganic contaminants from steel slag generally complies with the established limits (see e.g. [15]), the release of toxic elements such as Ba, Cr, V and Mo may still be critical in some instances [16–23]. The mechanisms governing steel slag leaching still need to be further elucidated through systematic and comprehensive studies, particularly for carbonated slags. The available studies in this regard are relatively limited [5,24–33], and only a few specifically focus on the mechanisms governing the release of major elements and toxic metals from the material. Studies on wet

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carbonation of different types of steel and stainless steel slag (EAF, AOD, BOF and mixtures of these [5,24,25,28,31,33]) showed that major elements including Ca and Si were strongly affected by the process, and (especially at the native pH of the material) a notable decrease in Ca leaching due to Ca carbonate precipitation and an increased Si release associated to the precipitation of more soluble secondary Si minerals were reported. Fe leaching was shown to be slightly influenced by carbonation, with some reduction in the extent of release at the material's own pH [24,27,32], while the opposite trend was reported for Mg [5,24,27,32]. The reported effects of carbonation on Cr leaching differ depending on the type of slag and the leaching conditions [5,24,25,27,28,30,32], however, the observed changes are more likely associated to changes in pH upon carbonation rather than to variations in the nature of the solubility-controlling phases. Among the other elements of potential environmental concern in steel slag, the leaching of Cu, Ni, Mo and Zn was generally observed to decrease upon carbonation [25,33], while the opposite was found for V [25,27,30–33], Sr and Ba [30], Se [32] as well as fluoride [25].

The description and prediction of the potential environmental impacts associated to waste materials such as steelmaking slags are deemed to be very important to estimate the potential risks associated to their reuse or final disposal, and this explains the need for an improved prediction of their leaching behaviour. To this aim, the present study investigates the mechanisms governing the leaching of major elements and trace contaminants from carbonated BOF slags, as well as the changes induced by accelerated carbonation on leaching. The study was intended to contribute to improving the understanding of the environmental behaviour of the material and providing information about the release of metal contaminants from carbonated steel slag, to be used e.g. in environmental and risk assessment evaluations. While a previous paper focused on the leaching of thin-film (liquid-to-solid ratio, L/S = 0.4 l/kg) carbonated steel slag [33], the present work investigates the behaviour of slurry-phase (L/S = 5 l/kg) carbonated slag, which was found to display a better CO<sub>2</sub> sequestration performance with a more intensive involvement of the slag constituents in the process [34].

## 2. Materials and methods

### 2.1. Slag sampling and characterisation

Slag samples were collected from an Italian steel manufacturing plant employing the integrated steelmaking process, both at the outlet of the basic oxygen furnace (BOF1 slag) and from the storage site after metals recovery (BOF2 slag). Both samples were ground to  $d < 2$  mm at the industrial site before sampling.

Slag characterisation involved the determination of elemental composition, calcite content, mineralogy, acid neutralisation capacity (ANC) and leaching behaviour. The elemental composition was determined by alkaline digestion with Li<sub>2</sub>B<sub>4</sub>O<sub>7</sub> at 1050 °C, followed by dissolution in 10% HNO<sub>3</sub> and measurement of element concentrations using atomic absorption spectrometry (AAS). The carbonate content was measured with an inorganic carbon (IC) analyser. The mineralogical composition was evaluated by powder XRD analysis with Cu K $\alpha$  radiation using a diffractometer equipped with a copper tube operated at 40 kV and 40 mA. The leaching behaviour was investigated through the CEN/TS 14429 leaching pH dependence test. Eluate concentrations were determined by AAS analysis in either the air-acetylene flame or the graphite furnace configurations, allowing a detection limit in the order of ppb. All chemical analyses were performed in triplicate, while the pH dependence test was not replicated, since it consists of a series of individual batch tests each carried out on a separate test portion of the material and hence the resulting leachate concentration profiles

allow to implicitly take into account the intrinsic heterogeneity of the material.

### 2.2. Slag carbonation

Batch carbonation experiments were performed on ball-milled (<150  $\mu$ m) slag samples in a 300-ml heated and magnetically stirred pressurised stainless steel reactor. A suspension of slag and deionised water at a liquid to solid (L/S) ratio of 5 l/kg (adopted on the basis of previous results [5,24,25,34,35]) was contacted in batch mode by 100% CO<sub>2</sub> for 4 h at a total pressure of 10 bar and a temperature of 100 °C. Such operating conditions were selected on the basis of an optimisation study carried out using previous results [34].

After carbonation the slag samples were centrifuged at 4000 rpm and solid/liquid separation was accomplished through 0.45  $\mu$ m membrane filtration. The solid material was oven-dried at 105 °C and subjected to the ANC leaching test as described above for the untreated slag.

### 2.3. Leaching modelling

Geochemical modelling of the ANC eluates was conducted using Visual Minteq [36], extending the standard thermodynamic database with additional solid phases of potential interest for steel slag, for which the stability constants were retrieved from numerous literature studies (see Ref. [37]). The code was applied following a three-step procedure: (1) a first run used the measured eluate concentrations and pHs as the input data, suppressing precipitation of all solids; (2) potential solubility-controlling minerals were identified among those displaying saturation indices (SI) between -1.5 and +1.5, and on the basis of likelihood of formation in steel slag leachates; and (3) the predicted concentrations of each element in equilibrium with the mineral phases were empirically derived as  $C_{eq} = C_{meas} (10^{-SI})^{1/n}$ , where  $C_{eq}$  and  $C_{meas}$  are the theoretical and measured concentrations, and  $n$  the stoichiometric coefficient of the element in the mineral [38]. For redox-sensitive elements including Cr, Fe and V different modelling runs were conducted at varying redox states; the results reported later in the manuscript correspond to those redox states that gave the best fit of the experimental leaching data by the model-predicted solubility curves of mineral phases.

## 3. Results and discussion

### 3.1. Chemical and mineralogical characterisation

The chemical analysis of the slag (Fig. 1) revealed significantly higher Ca concentrations (21 and 23% for the BOF1 and BOF2 samples) compared to Mg (3.3 and 3.8%). Other elements detected at appreciable concentrations were Fe (25 and 20%), Si (5.6 and 4.1%), Mn (3.8 and 2.5%), Al (10800 and 6900 mg/kg), V (9300 and 6900 mg/kg) and Cr (1800 and 1300 mg/kg). In addition to Cr and V, other metals of potential environmental concern in the BOF slags included Zn (35 and 88 mg/kg), Pb (~60 mg/kg) and Cu (8 and 16 mg/kg). The initial carbonate content of the material was 0.8 and 6.7%, respectively; the significantly higher concentration detected for BOF2 was ascribed to natural carbonation related to exposure to atmospheric CO<sub>2</sub> in the storage site.

Since the present study was specifically focused on the investigation of the leaching behaviour of steel slag as affected by accelerated carbonation, only a brief summary of the relevant results of the carbonation treatment in terms of the achieved CO<sub>2</sub> uptakes and modifications in slag mineralogy is reported here. For a more detailed discussion of the CO<sub>2</sub> sequestration capacity of the

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