



Effects of thin-film accelerated carbonation on steel slag leaching



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HIGHLIGHTS

- Thin-film accelerated carbonation of EAF and AOD steelmaking slag was investigated.
- The leaching behaviour of untreated and carbonated slag samples was studied.
- Carbonation affected the leaching of both major elements and metal contaminants.
- Changes in pH and controlling solids explained the observed metal leaching.

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ABSTRACT

This paper discusses the effects of accelerated carbonation on the leaching behaviour of two types of stainless steel slags (electric arc furnace and argon oxygen decarburisation slag). The release of major elements and toxic metals both at the natural pH and at varying pH conditions was addressed. Geochemical modelling of the eluates was used to theoretically describe leaching and derive information about mineralogical changes induced by carbonation. 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; geochemical modelling indicated that the Ca/Si ratio of Ca-controlling minerals shifted from ~1 for the untreated slag to 0.5–0.67 for the carbonated samples, thus showing that the carbonation process left some residual Ca-depleted silicate phases while the extracted Ca precipitated in the form of carbonate minerals. For toxic metals the changes in leaching induced by carbonation appeared to be mainly related to the resulting pH changes, which were as high as ~2 orders of magnitude upon carbonation. Depending on the specific shape of the respective solubility curves, the extent of leaching of toxic metals from the slag was differently affected by carbonation.

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1. Introduction

Several types of alkaline residues are produced during steel manufacturing. The quantities generated worldwide are quite significant and are estimated to amount to roughly 10–15% by weight of the produced steel [1]. From current steel production data [2], a total crude steel production of 1.58×10^6 Mt was estimated for the year 2013 worldwide, with a main contribution of Asia accounting for 1.06×10^6 Mt (mainly due to China: 74% of the overall Asian production). According to the World Steel Association [3], about 69.5% and 29.4% of the steel produced worldwide is obtained from the basic oxygen furnace (BOF) and the electric arc furnace (EAF) processes, respectively. In Italy, these percentages are different, with a prevalence (~65.6%) of the EAF technology [4].

Steel manufacturing slags are generated either during the conversion of iron ores into crude steel in basic oxygen converters (BOF slag) when the integrated steel manufacturing cycle is adopted, or during iron scrap melting in electric arc furnaces (EAF slag from carbon steel [EAF-C] and stainless/high alloy steel production [EAF-S]) [1,5,6]. Secondary metallurgical slags are also part of the solid residues of the steel industry, and include argon oxygen decarburisation (AOD) slag (produced during the refining of stainless steel) and ladle furnace (LF) slag (produced during secondary processing of crude steel) [1,6]. In most plants, the slag after cooling is ground and subjected to magnetic separation in order to recover metal components to recycle in the steel production furnace [1].

On average, the steel industry generates about 125.8 kg of basic oxygen furnace (BOF) slag and 168.6 kg of electric arc furnace (EAF) slag per t of crude steel produced [2]. In Europe, the latest available statistics [5] report a production of 21.8 Mt of steelmaking slag in 2010 (of which 10.5 Mt of BOF slag, 6.8 Mt of EAF-C slag, 1.7 Mt of EAF-S slag and 2.8 Mt of secondary slag).

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All these types of residues are alkaline materials characterised by a prevalence of Ca-containing silicate phases and generally also display relevant amounts of Mg, Al, Fe and Cr, among others, in the form of oxides and/or silicates [6]. The presence of such minerals provides the background for recent research in which steel slag is used as a source of reactive elements for the purpose of CO₂ sequestration using accelerated carbonation whereby the material reacts with gaseous CO₂ and the alkaline elements are converted into thermodynamically stable carbonate minerals [7–20].

The slag generated during steel manufacturing is usually regarded as a non-hazardous waste owing to its limited release of trace contaminants, as mirrored by compliance with standard leaching criteria (see e.g. [1]). Nevertheless, depending on the leaching conditions some types of slag may in fact still release non negligible amounts of toxic metals, such as typically Ba, Cr, V and Mo [21–28]. As for the environmental behaviour of steel slag, surprisingly only limited literature studies have specifically focused on the mechanisms underlying the leaching process and governing the potential release of toxic elements. The lack of systematic information about the environmental behaviour of steel slag is also evident when accelerated carbonated slags are concerned, with only relatively few studies investigating the mobility of elements of environmental concern in carbonated steel slag [13–15,29–35]. In particular, the mechanisms through which accelerated carbonation affects the extent of leaching of major constituents and toxic elements from the solid matrix are still unclear and thus need to be further elucidated. In the present work, an attempt was made at filling the existing gaps in the knowledge of the leaching-controlling phenomena for accelerated carbonated steel slag. To this aim, an experimental investigation of pH-dependent leaching of untreated and carbonated steel slag, along with geochemical modelling of the resulting leaching solutions, was carried out.

2. Materials and methods

Slag samples were collected from an Italian stainless steel manufacturing plant after metals removal downstream of the electric arc furnace (EAF slag), and at the outlet of the desulfurisation unit subsequent to the argon oxygen decarburisation section (AOD slag). The EAF slag was quite heterogeneous in particle size and had the following grain size distribution (AASHTO classification): 18% gravel, 31% coarse sand, 30% fine sand, 18% silt and ~3% clay. The AOD slag was considerably more homogeneous and finer in grain size (90% of particles below 0.150 mm). Considering the critical influence exerted by particle size on carbonation [13], the EAF slag was milled to a particle size below 150 μm, while the 150 μm-oversize fraction was simply discarded from the bulk of the AOD slag.

Slag characterisation involved the determination of elemental composition, calcite content, mineralogy, acid neutralization capacity (ANC) and leaching behaviour. The elemental composition was determined by alkaline digestion with Li₂B₄O₇ at 1050 °C, followed by dissolution in 10% HNO₃ and measurement of element concentrations using atomic absorption spectrometry (AAS). The carbonate content was evaluated by calcimetry analysis using a Dietrich-Frühling calcimeter. The mineralogical composition was evaluated by powder XRD analysis with Cu Kα radiation using a Philips Expert Pro diffractometer equipped with a copper tube operated at 40 kV and 40 mA. The leaching behaviour was investigated through the EN 12457-2 and the CEN/TS 14429 (ANC) leaching tests. Eluate concentrations were determined by AAS analysis, whereas anion concentrations were measured by ion chromatography. All chemical analyses were performed in triplicate; the batch compliance leaching test was run in duplicate, while the ANC test was not replicated.

Batch carbonation tests were performed as detailed in Reference [15] using the direct thin-film (L/S ratio = 0.41/kg) carbonation route at an operating temperature of 50 °C and a CO₂ pressure between 1 and 10 bar. After carbonation, the samples were oven-dried at 50 °C and analysed by calcimetry testing. The mineralogy and leaching behaviour of the carbonated samples were also investigated as described above on samples carbonated under the following conditions: L/S = 0.41/kg, T = 50 °C, t = 1 h, pCO₂ = 3 bar (EAF slag) or 1 bar (AOD slag). The two samples selected to explore the leaching behaviour were chosen as those that, while keeping the carbonation conditions as mild as possible in view of a potential full-scale application of the process, still allowed to attain a significant carbonation yield.

To derive information on the mechanisms governing the leaching of contaminants from the slag, geochemical modelling of the ANC eluates was conducted using Visual Minteq. The standard thermodynamic database included in the modelling code was extended including the stability constants of several phases (see [36] for details). The application of the geochemical speciation code followed a three-step procedure: (1) the program was run using the measured eluate concentrations and pHs as the input data and suppressing precipitation for all solid phases; (2) potential solubility-controlling minerals were chosen among those displaying saturation indices (SI) in the range $-1.5 \leq SI \leq +1.5$ and on the basis of likelihood of formation in steel slag materials; and (3) the predicted concentrations of each element in equilibrium with the selected mineral phases were derived according to the empirical equation $C_{eq} = C_{meas} (10^{-SI})^{1/n}$ [37], where C_{eq} and C_{meas} are the theoretical and measured element concentrations, respectively, and n is the stoichiometric coefficient of the element in the mineral under concern.

3. Results and discussion

3.1. Chemical and mineralogical characterisation of the slag

The chemical composition of the EAF and AOD slag is reported in Table 1. In view of carbonation, the Ca and Mg content of the slag is particularly relevant: in both types of residues the Ca concentration (35% and 40% for the EAF and AOD samples, respectively), was significantly higher than the Mg content (2.4% and 1.8%, respectively). Appreciable contents of Cr, Fe, Al, Mo and V were also measured in the EAF slag, in particular. The initial calcite content accounted for 3.5% and 4.0% of the EAF and AOD slag mass, respectively.

Fig. 1 shows the results of the XRD analysis of the two types of slag. The most relevant phases present in the EAF slag were found to include different silicate minerals such as dicalcium silicate (Ca₂SiO₄, in its γ-type polymorph), akermanite (Ca₂MgSi₂O₇), cuspidine (Ca₄Si₂O₇(F,OH)₂) and gehlenite (Ca₂Al(AlSi)O₇); other identified minerals included oxide phases (periclase [MgO], a 0.86:1Ca–Al oxide with the formula (CaO)₁₂(Al₂O₃)₇, a 2:1Cr–Mg oxide having the composition MgCr₂O₄, the Fe(II)–Fe(III) oxide magnetite [FeO·Fe₂O₃] and quartz [SiO₂]); fluorite (CaF₂) and small amounts of calcite (CaCO₃) were also detected.

The XRD pattern of the AOD slag revealed a lower number of peaks, indicating a simpler mineralogical composition. The main phases identified included γ-dicalcium silicate, fluorite and magnetite, with traces of calcite. The results obtained from the mineralogical analysis confirm the findings of previous investigations on stainless steel slag [21,38].

3.2. Carbon sequestration capacity and degree of carbonation

The carbonation yield attained by the two slag samples is reported in Table 2 in terms of CO₂ uptake as a function of pressure

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