



Carbonation of stainless steel slag in the context of in situ Brownfield remediation



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ABSTRACT

The main aim of this work was to assess the potential of in situ carbonation as a treatment to modify the properties of alkaline materials such as industrial soil in terms of leaching behaviour and mineralogy and to store the CO₂ generated by specific treatments applied in the context of Brownfield regeneration. The process was investigated through lab-scale column carbonation experiments, in which 100% CO₂ was fed through humidified stainless steel slag under ambient temperature and pressure for set reaction times. The reaction kinetics and the maximum CO₂ uptake attained (5.5%), corresponding to a Ca conversion yield of 15.6%, after 4 h treatment proved slightly lower than those resulting from batch tests carried out on the same particle size fraction at enhanced operating conditions. The mineralogy of the material showed to be affected by column carbonation, exhibiting a higher calcite content and the decrease of Ca hydroxide and silicate phases. As a result of carbonation, the material showed a decrease in pH and Ca release as well as an increase in Si mobility. Furthermore, a reduction of Cr and Ba leaching, up to 63% and 96% respectively, was achieved after 2 h of reaction. However, carbonation was observed to lead to an increased leaching of V and Mo. The effects of carbonation on the leaching behaviour of the material were also investigated performing pH-dependence leaching tests and the results indicated that in situ carbonation appears to be a promising treatment to improve the properties of alkaline materials in view of their reuse on-site.

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1. Introduction

The modification of land use and the subsequent dismantling of several industrial activities has led to the formation of a number of derelict or underused sites, i.e. Brownfields, all across Europe (Oliver et al., 2004) and worldwide (Marker et al., 2007) that are waiting to be reintroduced within the so called Land Management Cycle. One of the main factors that hinders the revitalization of Brownfield sites, such as former metallurgical plants or mining areas, is the need to address soil contamination issues, which may arise from the past land activities as well as from the interim uses of the site. Partly also as a consequence of the lack of a clear requalification pathway, traditional remediation strategies, e.g. excavation and landfilling, are generally applied for the management of the contaminated soil, implying off-site transport and leading to further land consumption. The poor acceptance of landfilling and the environmental impact of this management practice, together with its excessive costs in the case of large volumes of material to be disposed of, drive the need of identifying alternative

integrated approaches for the management of contaminated soil within a Brownfield regeneration framework. In this type of framework, the approach to the Brownfield environmental problems should shift from considering the individual contamination issue towards a wider perspective accounting for integrated strategies in the context of land management. The clean-up of the site, which is mandatory for the reintroduction of a Brownfield within the land cycle, should be fulfilled by (re)using material and exploiting resources already present at the site and/or produced as a result of the regeneration activities themselves (Baciocchi et al., 2012). Namely, contaminated soil and industrial residues with specific properties could be reused within the site as aggregates (Manso et al., 2006; Scanferla et al., 2009), filling material (Scanferla et al., 2009) or pozzolanic material for cement substitution (Pan et al., 2008).

In a Brownfield regeneration context, soil is commonly referred to as industrial soil, since it may be characterized by the presence of material of anthropogenic origin arising from the historical use of the site that may cause detrimental environmental effects related to the release of toxic metals and metalloids (Voglar and Leštan, 2010). For instance, the profile of an industrial soil may be characterized by the stratification or the heterogeneous mixture

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of native soil and industrial by-products, such as demolition waste and slag from steel making processes (Renforth et al., 2009) for which the leaching of specific elements may be significant, e.g.: Cr from stainless steel slag (Shen et al., 2004). Furthermore, Renforth et al. (2009) showed that some kinds of alkaline soils present in Brownfield sites may act as carbon sinks by reacting with atmospheric carbon dioxide. Namely, they found that the 20 cm-layer of the industrial soil pertaining to a former steelmaking company was characterized by a variable CaCO_3 content, ranging between 0% and 38% but presenting a poor correlation with sample depth. A step forward aimed at employing this type of industrial soil as alkaline source could be represented by the application of accelerated carbonation, using far more concentrated carbon dioxide sources than atmospheric air. So far, accelerated carbonation, has been developed as an ex situ CO_2 storage option, by which a material rich in alkaline earth metal oxides and/or silicates is contacted with carbon dioxide at optimized operating conditions (i.e. temperature, pressure and pH), thus leading to the formation of the corresponding thermodynamically and chemically stable carbonate phases (Lackner et al., 1995). Several reaction routes (e.g. indirect, direct gas–solid or direct aqueous) have been investigated with the aim of maximizing the CO_2 uptakes achievable by ex situ carbonation of minerals and alkaline residues such as steel slag (see e.g. Doucet, 2010; Bobicki et al., 2012).

In addition, ex situ carbonation processes carried out in aqueous conditions, applying liquid to solid (L/S) ratios above 2 l/kg (slurry phase route) or below 1 l/kg (wet route) and generally mild operating conditions have been tested as treatment strategies to reduce the release of metals and metalloids from alkaline industrial residues, such as incineration bottom ash and steelmaking slag (e.g. Van Gerven et al., 2005; Baciocchi et al., 2010a, 2010c; van Zomeren et al., 2011). Furthermore, accelerated carbonation has been tested in combination with cement as a stabilization treatment for contaminated industrial soil (Antemir et al., 2010) and also as a technique to increase the compressive strength of compacts formed by residues rich in Ca silicates such as steel slag (Johnson et al., 2003).

Hence, the application of accelerated carbonation in a Brownfield regeneration context appears to be of particular interest since it may allow the achievement of multiple benefits. In Brownfield sites primary CO_2 sources, such as combustion or power plants or other industrial processes, may be no longer available; however, CO_2 emissions may result as a consequence of treatments aimed at the remediation of groundwater contaminated by organic compounds such as oxidation or CO_2 stripping (Nelson et al., 2009). Besides, CO_2 may evolve from innovative treatments for the improvement of the structural properties of the subsoil in view of Brownfield sites redevelopment (Hartog et al., 2013). Therefore, in these types of contexts, CO_2 upward flows through the subsoil may be exploited to induce in situ carbonation reactions in existing or specifically prepared layers of alkaline residues and/or industrial soil to improve the environmental and technical properties of the materials as well as to permanently store CO_2 . A schematic representation of the proposed in situ carbonation process as a treatment for industrial soil in Brownfield sites coupled with in situ stripping of volatile organic contaminants (VOCs) from groundwater is depicted in Fig. 1. In this application the injected CO_2 can first serve to strip VOCs from groundwater before stimulating the carbonation of the slag material in the overlying industrial soil. As shown in Fig. 1, the reagents are injected into the subsurface, resulting in the evolution of a CO_2 upward flow as well as the volatilization of organic contaminants affecting groundwater quality. Once the CO_2 reaches the layer of alkaline industrial soil, carbonation occurs, resulting in the improvement of the environmental properties of the carbonated material and CO_2 storage. Extraction wells are also foreseen in order to collect the unreacted carbon

dioxide and the volatilized organic compounds to submit to proper treatment.

The main aim of this work was to assess the feasibility of the proposed in situ carbonation process by means of a column carbonation experimental method in terms of the effects exerted on the environmental behaviour of the treated material as well as on the achieved CO_2 uptakes. Specifically, this paper reports the results of laboratory scale column carbonation tests performed flowing 100% CO_2 through stainless steel (SS) slag and applying operating conditions expected at Brownfield sites. In addition, to assess the reactivity of the material with CO_2 , accelerated carbonation tests were also carried out under enhanced conditions in a stainless steel batch reactor. All tests were performed on a mixture of freshly produced slags provided by a stainless steelmaking plant. Despite this sample may seem not adequately representative of aged slags typically found in Brownfield sites, it is worth pointing out that the effect of aging on the extent of carbonation is often limited to the top layer of an industrial soil. For instance, Suer et al. (2009) and Arm et al. (2011) reported upon weathering a pH shift of steel slags sampled from a pile, whereas for instance those buried beneath a road did not show remarkable changes of pH. Similarly, also Renforth et al. (2009) found the extent of carbonation of an industrial soil collected in a former steelmaking plant below a depth of 20 cm to be negligible. Therefore, this evidence seems to suggest that the use of fresh slag samples adopted in this paper may provide meaningful results also for slags present in Brownfield sites. As for the effects of carbonation on the properties of the residues, the modifications occurring in the mineralogy, acid neutralization capacity and release of major elements and trace components from the material both at its native pH and as a function of pH are analyzed.

2. Materials and methods

2.1. Materials

The slag sample tested in this study, provided from a stainless steel production plant, is representative of the residues mix that is sent out of the plant for disposal. As the main residues produced by the plant are those generated from the Electric Arc Furnace (EAF) and from the Argon Oxygen Decarburization (AOD) converter units, the sample may be expected to be a mixture of these types of slag. After determining its particle size distribution (ASTM D422), the coarser fraction ($d > 0.84$ mm) was discarded as it was not considered suitable for performing reproducible lab-scale experiments. The remaining material was divided into a fine ($d < 0.177$ mm) and an intermediate (0.177–0.84 mm) fraction. Both fractions were characterized in terms of their elemental composition and mineralogy.

The elemental composition was determined by alkali fusion of the slag samples with $\text{Li}_2\text{B}_4\text{O}_7$ at 1050 °C followed by dissolution with 10% HNO_3 of the molten material and analysis of the solutions by inductively coupled plasma optical emission spectrometry (ICP–OES). The mineralogy of the as received and carbonated slag was evaluated by X-ray diffraction (XRD) analysis with $\text{Cu K}\alpha$ radiation using a Philips Expert Pro diffractometer (equipped with a copper tube operated at 40 kV and 40 mA) with an angular step of 0.02° held for 2 s with 2θ spanning from 5° to 85° .

The intermediate fraction was also characterized in terms of its leaching behaviour before and after column carbonation tests, in order to evaluate the influence of this treatment on the mobility of major and trace elements. Specifically, the EN 12457-2 compliance test and the CEN/TS 14429 pH-dependence leaching test were applied both on as received and carbonated samples and the eluates were analyzed by ICP–OES. For the latter type of test, in order to span a wide pH range, acid additions were performed employing

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