



## Laboratory investigation of carbonated BOF slag used as partial replacement of natural aggregate in cement mortars



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### ABSTRACT

Direct mineral carbonation produces a material rich in carbonates and with reduced quantities of free oxides. The aim of this work was to show that such materials can be used in the construction domain. Basic Oxygen Furnace (BOF) slag from the steelmaking process has been traditionally seen as unfit for bounded applications due to its propensity to swelling, resulting from hydration of its high free lime content. Here, BOF slag was crushed to suitable particle sizes, carbonated in an aqueous solution of carbonic acid, and utilized to replace 50% of natural sand aggregate in cement mortars. The mechanical and chemical properties of these mortars were compared to mortars containing non-carbonated slags, and a standard cement mortar as a reference. Tests were conducted to determine mortar paste consistency and soundness, and cured mortar compressive strength and leaching tendencies. The results showed a satisfactory performance for all considered aspects (comparable with the reference) of the mortar sample containing 37.5 wt% (1.5 in 4 parts solids) carbonated BOF slag of <0.5 mm particle size.

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### 1. Introduction

The development of various treatment routes for the valorization of steelmaking slags has resulted in these residues being withdrawn from the European Waste Catalogue [1]. With the purpose of using steelmaking slags in the construction domain, and after much research conducted internationally, different standards have been implemented regarding the conditions that should be met by these industrial wastes before utilization. When intending to use slags as binders, the hydraulic activity of the material is very important [2]. Several researches have shown that finely milled Basic Oxygen Furnace (BOF) slag can be used as a cement replacement in mortars and concretes [3–5]. Others have combined BOF slag with Blast Furnace (BF) slag [6,7], where the BOF

slag acts as basic activator due to its free lime content. However, with the use of BOF slag as a binder requiring intense grinding, and with the high metallic iron content of BOF slag, this processing step represents a high energy demand, and consequently a high cost.

Lower processing costs (due to at least halving the specific grinding energy requirement [8]) can be achieved when using BOF as an aggregate, as the extent of particle size reduction (from monolith to millimeter-sized grains, rather than to micron-sized powder) can be reduced. For this application, the main parameters that must meet stringent specifications are the volume stability, referred to as soundness, the basicity, and the leaching of regulated toxic heavy metals and metalloids [9]. Wang et al. [10] proposed a calculation method for volume expansion and the expansion forces of slag used, respectively, as an unbound or bound aggregate. A usability criterion was developed based on the free lime content, the specific gravity and the bulk relative gravity of the slag. The criteria can be used as guidance for the selection of steel slags as granular materials for use in highway construction, a field

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of extensive study [11–15]. Xie et al. [14] found that the failure mechanism in BOF dense-graded asphalt mixtures in moist environment is determined by the formation of portlandite ( $\text{Ca}(\text{OH})_2$ ) and calcium-silicate-hydrates (C–S–H), which leads to partial structural failure and facilitates water infiltration.

To obtain acceptable values of hydraulicity and soundness, one solution has been to control the slag's cooling rate from the hot to cold stage. Gautier et al. [16] demonstrated the possibility to obtain low free lime (CaO) content in BOF slag (the main mineral component responsible for volume instability) with slow cooling (72 h), and to obtain high  $\text{C}_3\text{S}$  (tri-calcium silicate, the mineral responsible for hydraulicity) content by rapid cooling (3–5 s), thus obtaining materials potentially suitable for application in the construction domain. This approach, however, is less effective for slags containing high amounts of free lime, as much of this free lime will be present as residual (undissolved) rather than precipitated (originating from the decomposition of  $\text{C}_3\text{S}$  to dicalcium-silicate ( $\text{C}_2\text{S}$ )) lime [17], and thus will be inert to changes in cooling rate. Another hot-stage approach is to modify the slag's chemical composition prior to cooling. Li et al. [18] showed that by adding EAF and coal bottom ash to BOF slag, it is possible to reduce free lime content and improve soundness and cementitious properties. A disadvantage of this approach is the added process complexity of controlling slag chemistry (typically undesirable as this is a secondary product of steelmaking) and ensuring sufficient residual process heat to drive the dissolution and reaction of the additives (waste heat from the BOF process is limited [1]).

An alternate route is the reaction of the alkaline oxides in BOF slag with  $\text{CO}_2$ , a process called mineral carbonation that leads to the formation of geochemically stable carbonates (e.g.  $\text{CaCO}_3$ ), thus preventing the formation of expansive hydroxides and implicitly obtaining a stable material [19]. An advantage of this method over hot-stage treatments is that it is applicable to the existing storage heaps of these wastes, which are significant worldwide (e.g. in Turkey [20]). Another advantage is that both forms of free lime found in BOF slag (residual and precipitated) can be made to react with  $\text{CO}_2$ , thus substantially reducing basicity [9] and likely propensity for swelling, although the latter has not been reported in literature. The noble purpose of sequestering carbon dioxide, however, is insufficient for extending the process of mineral carbonation to an industrial scale if the economical balance is not a positive one. For this reason, the materials obtained after mineral carbonation must be valorized by utilizing them in large-scale commercial applications. Recently, Kirchofer et al. [21] calculated the  $\text{CO}_2$  sequestration capacity through mineral carbonation of alkaline industrial wastes that can be subsequently used as aggregates in the construction domain at 7.6 Mt,  $\text{CO}_2$ /year (for the USA). Out of that quantity, 0.6 Mt,  $\text{CO}_2$ /year represents the amount of emissions avoided by reducing the exploitation of natural sand, gravel, granite and limestone.

Besides capture of  $\text{CO}_2$ , desirable for emissions reduction, mineral carbonation has also been reported to yield positive effects in terms of basicity and the leaching behavior of alkaline earth metals, heavy metals and metalloids from steelmaking slags [22]. Van Gerven et al. [23], however, observed that the leaching evolution of some metals, from blocks obtained by mixing cement with municipal solid waste incineration ashes, increases at high carbonation degrees, compared to lower leaching levels for partially carbonated specimens. This was suggested to result from leachate-induced transformation of carbonates into more soluble bicarbonates once the pH drops below 10. This means that an optimal level of carbonation might exist that minimizes leaching.

In the case of carbonated Basic Oxygen Furnace (BOF) steel-making slag, vanadium and chromium leaching are reported to be the most problematic elements at low pH values. Chaurand et al.

[24] found that chromium is present at octahedral coordination in the trivalent form, the less mobile and less toxic form of Cr, and that its speciation does not evolve during natural ageing (the reaction of slags with  $\text{CO}_2$ -containing air and moisture). Vanadium, on the other hand, was found to be highly mobile; it was found to be present predominantly in the +4 oxidation state, and became oxidized to the pentavalent form (the most toxic form) during natural ageing. After carbonation, Santos et al. [9] observed increased leaching of chromium and vanadium from BOF slag, and like Van Gerven et al. [23] observed a minimum to occur at intermediate levels of carbonation extent and basicity reduction.

Monkman et al. [25] demonstrated that mortars with carbonated BOF slag as fine aggregate can reach 28-day strengths comparable to the strengths of typical river sand mortars, though this study did not present results regarding soundness nor heavy metals leaching. Saikia et al. [26,27] tested heavy metal leaching from mortars containing different industrial wastes as partial replacement of natural fine aggregate, and found that the leaching effect is considerably reduced in comparison to leaching from the unbound industrial wastes. Formation of expansive products like ettringite, aluminum hydroxide and  $\text{H}_2$  gas, due to the reaction of some constituents of the wastes with alkali, created cracks in the pastes and the cement mortars, which lowered the compressive strength of those mortars. Manso et al. [28] termed as 'cloistering' the effect of the cementitious matrix on the leaching reduction of contaminant elements from non-carbonated Electric Arc Furnace (EAF) slag aggregates bound in concrete.

The purpose of this work was to assess the mechanical and chemical properties of cement mortars containing non-carbonated or carbonated BOF slag as a fine aggregate. This work complements those previously described by applying a more complete set of testing methods, including consistency of fresh mortar pastes, soundness, compressive strength, basicity and contaminant leaching. This is the first work to report values of soundness, determined by the Le Chatelier test, for carbonated BOF slag-containing mortars. This work also aimed at evaluating the effect of slag particle size on the carbonation extent, and the effects of slag particle size and carbonation degree on the mortar properties.

## 2. Materials and methods

### 2.1. BOF slag characterization

The BOF slag used in this study was acquired from a steelmaking plant in Belgium. Prior to analyses or experimentation, the material was first crushed using a jaw-crusher (Retsch BB100) to obtain particles smaller than 1.6 mm. The obtained material was further divided into three particle size classes using standard sieves: <0.08 mm, 0.08–0.5 mm, and 0.5–1.6 mm. These three particle size classes were chosen based on the values reported in literature for BOF slag dry granulation [29] and in accordance to our group's previous researches regarding BOF slag carbonation [9,30].

The chemical composition of the slag was determined by X-ray Fluorescence (XRF, Panalytical PW 2400) analysis; results are presented in Table 1. High concentration of calcium was detected, which imparts this material its alkaline properties and high reactivity towards mineral carbonation. Also, BOF slag contains large quantities of iron, an undesirable loss of product from the steel-making process, and a significant quantity of silicon, important for the formation of silicate minerals.

The mineralogical composition of the slag was determined by Quantitative X-Ray Diffraction (QXRD) on a Philips PW1830 equipped with a graphite monochromator and a gas proportional detector, using Cu  $K\alpha$  radiation at 30 mA and 45 kV, step size of  $0.03^\circ$   $2\theta$  and counting time 2 s per step, over  $5$ – $70^\circ$   $2\theta$  range.

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