



The Reuse of Basic Oxygen Furnace Slag as Concrete Aggregate to Achieve Sustainable Development: Characteristics and Limitations

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Abstract: Basic oxygen furnace slag is considered a potential material for the replacement of natural aggregate in Portland cement concrete due to its similar physical characteristics. Therefore, in the present work, the slag was analyzed by Nitrogen sorption porosimetry, elemental analysis, mineralogical analysis, and volume stability. On the other hand, concrete mixtures were manufactured with basic oxygen furnace (BOF) slag aggregates and characterized mechanically and morphologically. The results showed superior compressive strength due to interfacial improvements in the slag aggregate. Statistically, there was no differentiation between the reference concrete and the slag concrete mixtures for tensile strength. Additionally, due to the porosity and low stiffness presented by the slag, the concrete elastic modulus showed a slight decrease. The replacement of sand with BOF slag as fine aggregate in Portland cement concrete presents itself as a sustainable alternative for the elimination and valorization of this environmental liability.

Keywords: BOF slag; Portland cement concrete; fine aggregate; steel



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1. Introduction

The world's production of crude steel has grown positively in recent years, with remarkable production figures seen in 2021, the year in which 1960 million tons of steel were produced around the world. The Brazilian steel industry remains among the tenth largest steel producers in the world and was responsible for the production of about 34 Mt of crude steel in 2022. In addition to iron and steel, the steel industry produces high levels of co-products. Therefore, the main solid co-products are slags, 90% by mass, and as steel production increases, so does slag production. Compared to Electric-Arc Furnace (EAF) slag, produced by high-power electric arcs, the BOF slag route is responsible for the production of, on average, a greater quantity of by-product, 400 kg/ton of steel, while EAF slag route is responsible for 200 kg/ton [1].

Despite presenting similar characteristics (the presence of free oxides, a chemical composition based on iron oxides, aluminum-silicon, and calcium), both materials present important differences: (1) CaO and free CaO contents in BOF slag are higher; (2) FeO content is lower in EAF slag; (3) EAF slag has a greater SiO_2 content; and (4) BOF slag has more hydraulic silicate minerals. Those are all relevant characteristics regarding the expansive potential of slag and the development of reactions between slag and the cementitious matrix, facts that make BOF slag a distinct material and imply the individual study of its properties and the impact of its use [1–4].

Studying ways to reuse slag is crucial, considering that recovered slag can be sold and used by other industries (i.e., the cement and construction industries), which generates revenues for steel producers and might prevent waste landfill development [1]. The

landfill's creation for slag storage has inflated costs and demanded the occupation of huge areas, as well as exposing the surrounding areas when the material is dragged due to weather conditions, bringing the risk of health contamination to the environment due to air, surface water, groundwater, and soil contamination by the deposition of an uncommon material [1,5].

Steel slags can be used in several fields according to their characteristics. Many studies have demonstrated the possibility of using BOF slag as construction materials, such as in Portland cement manufacturing (by replacing clinker, cement, or blast furnace slag) or using the slag as an artificial aggregate in concrete manufacturing. Considering the use of slag in construction, the main benefits to be achieved are the cost reduction of creating and maintaining landfills; boosting the production of cement plants; and the costs and environmental impact reduction concerning the extraction and the transportation of natural aggregates [6–11].

An effective way to encourage and increase BOF slag use in constructions is by the substitution of natural aggregates. The use of slag, as an aggregate, presents greater advantages, economic and environmental, when compared to its use in the replacement of clinker or cement. The first factor corresponds to the volume of material to be used; as the aggregates correspond to about 70% of the concrete volume, a higher content of aggregates of slag might be incorporated into the concrete. Furthermore, for clinker replacement, a possible reduction in mechanical properties can be observed in the concrete if the clinker content is abruptly reduced [10–13].

Another significant factor is the energy consumption related to the grinding of the slag to the granulometry of the cement (about 75 μ m). The grinding process is one of the most energy-intensive steps in cement production, with approximately one-third of the total energy being consumed. However, in the case of BOF slag, if the recovery of iron in the BOF slag has been carried out efficiently, the costs can be reduced [6].

As previously mentioned, BOF slag is an significant environmental liability produced by the steel industry, considering the volume of material produced and its specific physical and chemical characteristics. Although other studies have explored the theme of using BOF slag in civil construction, there is little information about the use of BOF slag as fine aggregate in the production of concrete. In addition, due to the complexity of the slag and its properties, the results observed in the literature are widely variable and slightly contribute to reducing the impact of the growing demand for solid waste disposal areas. This inhibits the technological progress that using slag can make possible by integrating slag processing technologies with production techniques with less environmental impact.

To further understand the advantages and limitations of BOF slag aggregate and to establish its role in the replacement of natural sand in Portland cement concrete, the slag was characterized in depth in terms of both its physical and morphological properties (specific gravity, water absorption, granulometric composition, porosimetry by nitrogen adsorption, and Los Angeles abrasion), and chemical and mineralogical aspects (X-ray Fluorescence, electron microprobe analysis, thermogravimetric analysis, and X-ray diffraction). To specifically verify the potential for expansion of the slag in an eventual interaction with the cement alkalis, a test to detect the alkali-aggregate reaction was performed in the field of durability. In order to achieve these goals, four samples of concrete with different contents of BOF slag were prepared with masses of: 0%; 25%; 50%, and 75%. The concretes were evaluated by compressive strength, splitting tensile strength, modulus of elasticity, and surface hardness. The results obtained show equivalent or superior properties to concrete without slag incorporation as aggregate. With these understandings, the article intends to promote discussions to subsidize the use of this residue as aggregates in the production of concrete to enhance the reuse of the slag.

1.1. BOF Slag Characteristics and Properties

BOF slag is generated during the blowing of oxygen inside a basic oxygen furnace (Figure 1) to remove the undesirable elements (silica, calcium oxide, magnesium oxide, and aluminum and iron oxides) and to reduce the carbon content in pig iron [7,8,14].



Figure 1. Schematic representation of the BOF process [2].

During the conversion process, lime $(CaO/Ca(OH)_2)$ or dolomite $(CaMg(CO_3)_2)$ is added to the furnace to increase the slag capacity to incorporate deleterious elements of steel and protect the refractory lining of the furnace, which makes the CaO content in BOF slag particularly high [1,10,12–14].

The main BOF slag components and the variation range are 34–55% of CaO, 8–20% of SiO₂, 1–7% of Al₂O₃, 14–32% of Fe₂O₃, and 1–10% of MgO, as presented in Table 1. The BOF slag presents great mineralogical similarity to raw materials for cement manufacture such as olivine [(Mg,Fe)₂(SiO₄)], merwinite [Ca₃Mg(SiO₄)₂], 3CaO·SiO₂ (C₃S), 3CaO·SiO₂ (C₂S), 4CaO·Al₂O₃·Fe₂O₃ (C₄AF), 2CaO·Fe₂O₃ (C₂F), CaO, and solid solutions of CaO-FeO-MnO-MgO (the RO phase). The presence of calcium silicates (C₂S and C₃S), tetracalcium aluminoferrite (C₄AF), and other mineral components provide cementitious properties to steel slag. However, the solidification process of BOF slags, normally slowly cooled under atmospheric conditions, leads to the transformation of most C₃S into C₂S + CaO and to the formation of a crystalline structure, which makes BOF slag far less reactive than Portland cement and more susceptible to expansion [15–20].

Table 1. Chemical composition of BOF slags according to the literature.

Reference	[2]	[4]	[7]	[9]	[10]	[11]	[14]	[15]	[1 8]	[20]
CaO	39.4	37.9	40.1-45	43.4	45.1	30–55	52.3	46.4	58.5	34.3-41.9
SiO ₂	11.9	15.5	8.6-13.1	13.5	11.3	8-20	15.3	11.1	10.7	13.6-17.3
Al_2O_3	2.16	4.3	1.7-2.1	3.3	1.1	1–6	1.3	1.2	1.71	3.4-6.9
MgO	9.7	3.5	4.5 - 7.5	5.8	6.1	5-15	1.1	8.3	4.3	2.7-7.1
Fe_2O_3	30.2	28.2	28.3 -3	28.6	29.6	14–2	16.2	23.2	15.3	20.1-36.2
SO_3	0.12	0.7	0.4-1.2	0.4	0.1	0.5-0.2	-	-	0.04	0.4-0.7
MnO	2.7	3.6	2-4.1	-	4.4	1–5	0.4	2.71	-	2.9-4
P_2O_5	1	1.8	1.4-2.4	1.5	1.5	0.2–2	3.1	2.1	-	1.1-1.9
Na ₂ O	0.25	0.2	-	-	0.2	-	-	0.09	-	0.13-0.15
K ₂ O	0.05	0.09	-	0.05	-	-	-	0.0	-	-
TiO ₂	0.4	0.8	0.5 - 0.9	0.6	0.45	0.4–2	-	0.4	-	-

The slow cooling process forms an overly complex mineralogical composition crystalline structure and, in general, the mineralogical composition of BOF slag is highly variable, as summarized in Table 2, with many overlapping peaks of crystalline phases, which makes its characterization a difficult task [2,7,15–17].

Reference	Mineral Composition
[2]	$Ca(OH)_2$, $Ca_2Fe_2O_5$, $Ca_3Mg(SiO_4)_2$, Ca_2SiO_4 , $CaCO_3$, CaO , $Mg(CO_3)_2$,
	$CaSiO_3$, MgO, MgSO ₄ ·5H ₂ O, CaMgSiO ₄ , Fe ₂ O ₃ , MgCO ₃
[3]	CaO, Ca(OH) ₂ , Mg(OH) ₂ , CaCO ₃ , 2CaO·SiO ₂ , FeO, SiO ₂ , Fe ₃ O ₄ ,
[0]	Ca ₂ SiO ₄ ·H ₂ O,4CaO·Al ₂ O ₃ Fe ₂ O ₃ , CaSiO ₃
[4]	Ca(OH) ₂ , C-S-H, SiO ₂ , CaCO ₃ , 3CaO·SiO ₂ , 3CaO·Al ₂ O ₃ ,MgFe ₂ O ₄ , RO, FeO,
[4]	$2CaO \cdot SiO_2$
[7]	CaCO ₃ , Ca(OH) ₂ , 2CaO·SiO ₂ , FeO, 2CaO·Fe ₂ O ₃
[10]	2CaO·SiO ₂ 2CaO·Fe ₂ O ₃ MgO, AlFe ₂ O ₄
[11]	3CaO·SiO ₂ , 2CaO·SiO ₂ , CaO, CaCO ₃ , FeO, MgFe ₂ O ₄ , 2CaO·Fe ₂ O ₃
[1 4]	CaO·SiO ₂ , 2CaO·Fe ₂ O ₃ , 3CaO·SiO ₂ , Fe ₂ O ₃ , (Ca, Mg, Si, Fe)O
[14]	2CaO·Fe ₂ O ₃ , 3CaO·P ₂ O ₅ , 2CaO·SiO ₂ , CaO
[4]]	2CaO·SiO ₂ 4CaO·Al ₂ O ₃ .Fe ₂ O ₃ , Ca(OH) ₂ , 3CaO·SiO ₂ , 2CaO·Fe ₂ O ₃ , Fe ₃ O ₄ ,
[17]	2, 2 0 2 0, 12 2 2 2 0, 0 1
[18]	Fe, FeO, Fe ₂ O ₃ , (Ca, Fe, Mg, Si)O, 2CaO·Fe ₂ O ₃ , 2CaO·SiO ₂
[19]	CaCO ₃ , CaO, Ca(OH) ₂ , 2CaO·SiO ₂ , FeO, 2CaO·Fe ₂ O ₃ , SiO ₂
[20]	3CaO·SiO ₂ , 2CaO·SiO ₂ , 2CaO·Fe ₂ O ₃ , C ₁₂ A ₇ , Fe ₃ O ₄ , CaO, MgO, Ca(OH) ₂ , (Ca,
[20]	Al, Si)O
[21]	Ca ₃ SiO ₅ , 2CaO·SiO ₂ , CaO, Ca(OH) ₂ , 4CaO·Al ₂ O ₃ ·Fe ₂ O ₃ , MgO, Ca ₂ SiO ₄

Table 2. Mineralogical composition of BOF slag according to the literature.

The slow cooling justifies the possibility of using slag as aggregates in roads, parking lots, buildings, and other constructions due to developed characteristics such as particle rough texture, high angularity, high surface area, high hardness, and wear resistance, all fairly influenced by the cooling process due to the crystalline structure and the mineral phases formed, which consequently determine its application. Figure 2 shows the relationship between the characteristics of steel slag and its potential application [8,15–19,22].



Figure 2. Relationship between the steel slag characteristics and its potential application.

The limiting factor for the use of BOF slag aggregates corresponds to the slag volumetric variation, due to the presence of unstable mineralogical phases. One of those unstable phases is the dicalcium silicate (C₂S), commonly present in BOF slag in content higher than the C₃S phase. The C₂S presents four polymorphic forms: α , α' , β , and γ , with β C₂S being widely observed in BOF slag. As a metastable phase, during the cooling (500–530 °C), β C₂S is converted to γ C₂S and this transformation leads to a specific gravity reduction from 3.28 to 2.97 g/cm³, and a volume increase of approximately 10% [2,12].

Less intensely, the BOF slag expansion may also be related to the oxidation and corrosion of metallic iron (FeO) present in its composition. However, the main expansive agent is the free CaO, followed by free MgO. The main reason for their occurrence in the slag results from the incomplete dissolution of lime or dolomite added in the converter, which leads to the formation of agglomerates of unreacted oxides. Consequently, during slow cooling, free CaO and free MgO are retained inside the crystalline slag particles, formed in different sizes, which makes the hydration of the inner layers of the slag extremely difficult, resulting in a high content of unreacted CaO and MgO [17,23–25].

The disadvantage of the presence of those free oxides is the fact that their hydration causes significant volumetric expansion during the formation of portlandite (Ca(OH)₂—a volumetric increase of almost 100%), and brucite (Mg(OH)₂—about 120%), and occurs

after the cement hydration, consequently, in confined environments. This may lead to the slag disintegration and to the mortar or concrete cracking due to the high internal pressures, reducing its mechanical strength and its resistance to chemically aggressive environments [10–12,17–25].

In this context, Wang et al. [26] confirmed that free lime is the main expansion agent, even after four years of concrete curing, compared to free MgO. The authors found that the content of f-CaO about 5%, provided to the steel slag's bad soundness, which caused the concrete cracking. According to the authors, many cracks were visible on the surface of the concrete specimens, and the concrete lost its rigidity, but, in extreme cases, concrete specimens were completely fractured due to the f-CaO expansion, and then the strength tests were impossible to be carried out.

1.2. Slag Concrete Properties

For the replacement of natural aggregates, the most important properties required for a satisfactory concrete performance correspond to specific gravity, shape, resistance to fragmentation, water absorption, wear resistance, and volumetric stability. A comparison of the properties of natural and BOF slag aggregates is shown in Table 3.

BOF slags present high specific gravity, generally between 3.1 and 3.7 g/cm³, due to their high iron-oxide content and iron particles, which can result in longer crushing and grindability time for BOF slag (i.e., grindability index of steel slag: 0.7, blast furnace slag: 0.96 and sand: 1.0) [4,23,27]. As shown in Table 3, BOF slag water absorption is generally higher than that of sand and coarse aggregates, especially for finer particle sizes, which can lead to increased water demand to maintain the consistency of the concrete in the fresh state and, consequently, increase the concrete porosity.

In terms of morphology, BOF slag presents an irregular and angular shape, which generates better interlocking between the aggregates and the cement paste, characteristics attributed to the crushing process and to the mechanical strengths resulting from slow cooling [9,10,16]. On the other hand, the very sharp edges can have a negative effect, reducing the workability of the mixtures of concrete with high substitution content due to a reduction in concrete mobility as well as possible particle segregation in the mixture [9,17,22,28–33].

BOF slag has a rougher surface compared to natural aggregates, resulting in a higher bonding strength with cement paste. Higher bonding may also be a result of the porous slag particles due to the ability of cement paste to penetrate the porous structure, which increases the bond, or to asphalt concrete, as observed by Kambole et al. [23]. They studied the use of BOF slag aggregates for asphalt concrete and found that the high-water absorption of slag allowed the asphalt binder to be properly absorbed, ensuring the required grip and satisfactory pavement performance compared to less absorbent aggregates [9,17,22,29].

According to the literature, the positive results of BOF slag aggregates use in concrete manufacturing are mainly justified due to the angularity of the slag particles, and the rough surface texture that promotes interlocking and higher mechanical bonding [9,11,17,29,30]. Nevertheless, the nullifying effects observed by the authors are the differences in the particle size distribution of slag compared to natural aggregates. This factor has a great influence on the packing degree of the mixture, which may increase the voids' proportion and porosity. Moreover, the high angularity of slag particles might hamper the particles' ability to roll and densify, which may require more water or cement paste to achieve the desired workability [9,11,15,17,22,29,30].

Few results concerning the effects of slag use on the tensile strength and elastic modulus of concrete are found. Tensile strength is sensitive to the amount of f-CaO and f-MgO present in the slag due to the generation of cracks from the expansive process of late hydration, derived from the tensile stress sensitivity of the hardened cement paste. Concerning the elasticity modulus of concrete, it is largely affected by the modulus of concrete constituent aggregates, since a significant difference between the modulus of elasticity promotes the development of fissures in the cementitious aggregate-matrix interface. Regarding the effect of BOF slag use, the greater slag's porosity might reduce the concrete strength of elastic deformation, compared to some natural aggregates. Thus, the aggregate porosity determines the stiffness and controls the ability of the aggregate to restrict the deformation of the cementitious matrix, which, consequently, leads to the reduction of concrete elasticity modulus in the function of the boost in BOF slag aggregate content [9,17].

Aggregates	Ap. Specific Gravity [g/cm ³]	Water Absorption [%]	Los Angeles Abras. Value [%]	f-CaO [%]	Reference	
BOF SLAG	3.4	2	18	5.3		
Granite	2.7	0.5	20	N/A	[9]	
Sand	2.6	0.8	-	N/A	-	
BOF SLAG	3.5	2.3	-	3.4		
Dolomite	2.8	1.8	-	N/A	[11]	
Sand	2.7	1.5	-	N/A	-	
BOF SLAG	3.4	2.4	12.7	-	[15]	
Gravel	2.7	0.5	20.8	N/A	[15]	
BOF SLAG	3.4	1–1.7	14–16	1.1	[1 (]	
Dolerite	2.8	0.3	14.8	N/A	[16]	
BOF SLAG	3.1–3.7	0.2–1).2–1 17		[1]	
Granite	2.6–2.8	0.3–1.2	0.3–1.2 -		[1/]	
BOF SLAG	3.1–3.7	0.2–1	9–18	<10	[20]	
Basalt	2.6–2.8	0.3–1.2	15–20	N/A	[28]	
BOF SLAG	3.3	1	22	<10		
Granite	2.5	<0.5	12	N/A	[29]	
Gravel	2.6	<0.5	21	N/A	-	
BOF SLAG	3.4	1.9	12.9	1.9		
Limestone	2.7	0.9	-	N/A	[30]	
Basalt	2.8	0.3	17.1	N/A	-	
BOF SLAG	3.4	1.8	11	-	[01]	
Limestone	2.6	1.1	25	-	[31]	
Fine BOF SLAG	3.58	4.2	-	6.5		
Sand	2.56	4.0	-	N/A	[22]	
Coarse BOF SLAG	3.42	3.3	-	6.5	[32]	
Limestone	2.49	1.7	-	N/A	-	

Table 3. Properties of BOF slag aggregate, according to the literature.

In general terms, comparing the replacement of the coarse aggregate with the fine aggregate in concrete manufacturing, it was observed that the substitution of sand introduced more benefits at different levels of replacement, while the increase in slag coarse aggregate content in the composition of some concretes led to a decrease in mechanical strength. A reduction in the workability of some concretes with high slag content was also observed with the replacement of coarse aggregate due to the angular shape of the aggregates and the higher water absorption of slag aggregate, which reduced the mobility of the cementitious matrix. Additionally, in some cases, a tendency towards slight bleeding and segregation was observed [9,17,22,26,29,32–36]. Concerning the composition, the formation of a thin layer of calcite on the slag aggregate surface was identified in some studies. The formation of this layer generated poor adhesion at the interface between the aggregate and cement paste, and a reduction in the concrete's mechanical properties. In addition, it is usual to observe that the strength of the BOF slag concrete continues to grow after 90 days of curing due to the low rate of BOF slag hydration. Along the same lines, the literature reports that the expandability increases with the curing time, due to free CaO and MgO presenting slow hydration rates. This fact might promote the formation of fissures and crack propagation in the concrete structure, which exposes the slag aggregate and in severe cases, might lead to the concrete rupture caused by the decrease in split tensile and flexural strengths. Nonetheless, the occurrence and the age of the failures induced by the expansion might vary for several reasons, such as the function of the steel slag contents, the f-CaO and f-MgO contents in the slag, and the concrete's water-to-binder ratios [9,22,26,29,30,35–38].

2. Materials and Methods

2.1. Materials

In the present study, for the reference concrete, natural sand was used as the fine aggregate and crushed granite as the coarse aggregate. The basic oxygen furnace (BOF) slag obtained by Companhia Siderúrgica Nacional—CSN (Brazil) was used with particle sizes between 150 μ m and 4.75 mm, and a time of weathering in outdoor conditions greater than 12 months to increase its stability. In addition to these materials, for the manufacture of concrete, CEM I was used as the binder, the Sikament[®]RM-320 as the superplasticizer admixture, and, for the alkali-aggregate reaction tests, Silica Fume (specific gravity: 2.27 g/cm³, specific surface area: 22.02 m²/g) and the cement CEM IV were used.

2.2. Methods

The chemical composition of BOF slag was obtained by X-ray Fluorescence (XRF), using Axios MAX equipment manufactured by Malvern Panalytical, for which the sample was prepared by pressing with boric acid. The content of free lime in BOF slag was obtained according to the ethylene glycol extraction method, and X-ray diffraction (XRD) was applied to determine its mineralogical composition. A diffractometer D8-XRD with CuK α radiation, manufactured by Bruker, was used, equipped with a Dectris detector, model Pilatus 3R 100K-A, using a scan range from 5 to 70°, step angle of 0.005°, and a scan speed of 0.6 degrees per second. The quantitative results were obtained by adopting the Rietveld method.

The chemical analysis of the phases present in the slag was obtained by an Electron Microprobe Analysis (EMPA)JXA-8230, manufactured by JEOL, combined with EDS analyses. The thermogravimetric analyses (TGA/DTGA) were performed to determine the hydroxide and oxide calcium content on a thermobalance Labsys Evo, produced by SE-TARAM Instrumentation, from 25 to 1000 °C and a heating rate of 10 °C/min, and Argon flux. All the analyzes mentioned were carried out in Goiania, Brazil.

The tested aggregate physical properties included the determination of the particle size distribution [39], apparent specific gravity, water absorption [40], and Los Angeles Abrasion value [41]. The potential reactivity of the slag for the alkali-aggregate reaction was evaluated using the accelerated method [42]. In terms of the absorption and pore size distribution, water absorption and nitrogen sorption porosimetry tests (in Autosorb-iQ equipment—Quatachrome Instrument) were performed. The concrete specimens were subjected to curing in a humid chamber (relative humidity of $(95 \pm 5)\%$ and room temperature of (24 ± 2) °C)) until the test time (28 and 180 days) to evaluate concrete mechanical performance in order to understand whether the use of BOF slag aggregates would result in less resistant concrete, which could lead to problems such as excessive deformation and cracking. For this purpose, mechanical tests were performed, including compressive strength [43], splitting tensile strength [44], modulus of elasticity [45], and surface hardness, using the Rebound Hammer Method with Original Schmidt N/L, PROCEQ.

2.3. Mix Proportion and Specimen Details

The concrete was designed to achieve compressive strength in the range (30 ± 5) MPa after 28 days of curing, with a slump of (120 ± 20) mm, with a water/cement ratio of 0.54. With the aim of evaluating the partial replacement of the fine aggregate by BOF slag on the concrete properties, the following contents of BOF slag were used: 0% (CR); 25% (CE25); 50% (CE50), and 75% (CE75) by mass. The specifications of the concrete mix proportions are listed in Table 4.

	CR	CE25	CE50	CE75	
	Portland cement—CEM I (kg/m ³)	299	303	305	309
	Water (kg/m ³)	162	164	165	167
Composition	Natural sand (kg/m ³)	754	574	384	195
data	BOF slag (kg/m ³)	0	191	384	586
	Granite crushed stone (kg/m ³)	1180	1197	1203	1222
	Admixture (%)	0.8	0.8	0.8	0.8
	water/cement ratio	0.54	0.54	0.54	0.54
Fresh	Slump (mm)	120	120	90	55
concrete	Air content (%)	2.12	1.37	1.72	1.31
properties	Unit weight (kg/m ³)	2397	2432	2441	2480

Table 4. Concrete mixture and test data for fresh concrete.

3. Results and Discussion

3.1. BOF Slag Characterization

3.1.1. Granulometric Analysis of Fine Aggregate

The BOF slag and sand cumulative percent passing rates are considerably different, as graphically shown in Figure 3A. The slag presented a coarser grain size distribution, which extrapolates the limits of the usable zone and optimal zone presented graphically, according to the Brazilian Standard [46]. Nonetheless, the slag and sand granulometric compositions were finer than the isolated particles of BOF slag, and as observed in Figure 3B, those three compositions are within the usable zone limits, which is a commonly used particle size distribution in the production of concretes. The limits are in accordance with the Brazilian standard [46]. Hence, a reduction in negative effects on the packaging and workability of the mixture with those compositions was expected in the fresh state.



Figure 3. Granulometric analysis. (**A**) Isolated gradations of BOF slag and sand; (**B**) gradations of BOF slag and sand compositions.

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3.1.2. Physical Properties of Fine Aggregate

Due to the high iron content, BOF slag has higher specific gravity than natural sand, as shown in Table 5, which may be up to 30% higher than the natural aggregate [24], opposing the concept that porous and absorbent materials have a lower specific mass due to the voids that constitute them. The Los Angeles abrasion value demonstrates that the ability of the slag aggregate to resist wear due to the attrition, impact, and crushing action in the test was relatively poor, with a mass loss of 33%. This result was considerably influenced by the high porosity of the material, thus justifying the presence of pulverulent material content, generated by the breaking and abrasion of the slag grains during the test.

Table 5. Aggregate physical properties.

Test	BOF Slag	Sand
Water absorption (%)	4.8	0.3
Fineness modulus	3.3	2.6
Specific gravity (g/cm ³)	3.1	2.7
Los Angeles abrasion value (%)	33	-
Pulverulent material content (%)	2.83	-

The water absorption of the slag aggregates is significantly higher than that of the sand (Table 5), and it is a relevant factor regarding the workability of the concrete in the fresh state and the mechanical strength of the hardened concrete, resulting in the water demand increment to maintain the consistency of fresh concrete. This tendency towards higher absorptivity was also observed by several authors and, as pointed out by Kambole et al. [23], the water absorption of BOF slag was generally higher, especially for finer particles. This was because the magnitude of the water absorbed by the aggregates may indirectly indicate the number of interconnected voids.

3.1.3. Pore Size Distributions of Fine Aggregate

The pore size distributions are plotted in Figure 4. Nitrogen sorption porosimetry of the aggregate samples gave an indication of the pores' radius, the values of which resulted in ranges of 1.5 to 120 nm for slag, and of 1.7 to 140 nm for natural sand. The results demonstrated that although the BOF slag and the sand have a similar pore radius, the natural sand has a smaller overall pore volume than the BOF slag, Figure 4B, with the highest pore volume observed at about 2 nm.





3.1.4. Elemental Analysis of BOF Slag

The chemical composition of BOF slag may be highly variable and is presented in Figure 5. The major chemical constituents are SiO₂, CaO, Fe₂O₃, and MgO, which constitute more than 80% of the total chemical composition. It was noted that the alkali content (Na⁺, K⁺) present in the slag is low, not acting as an external input of alkalis in the concrete for the development of the alkali-aggregate reactions.



Figure 5. BOF slag chemical composition.

From the ethylene glycol extraction, the total free lime [CaO and Ca(OH)₂] correspond to 3.8%. Although the ethylene glycol extraction test does not establish a normative limit, the literature reports that the free lime content in the BOF slag can reach levels higher than 12% [2] and that from 4.9% it may cause concrete rupture [26]. The ethylene glycol test was complemented by thermogravimetric analysis with the aim of distinguishing CaO and Ca(OH)₂ contents. The TGA curve of BOF slag is presented in Figure 6 and the results of CaO and Ca(OH)₂ contents are given in Table 6. Considering the loss of mass in the temperature range of 200–800 °C, contents of CaO, Ca(OH)₂, and CaCO₃ were calculated by mass balance based on the dehydration (about 420 °C) and decarbonization (about 760 °C) reactions.



Figure 6. Thermogravimetric curve of BOF slag.

	Total <i>f</i> -CaO (Ethylene		
Decarbonization	Total Fre	Glycol Extraction)	
Decarbonization	Dehydration	f-CaO	
CaCO ₃	Ca(OH) ₂	j euc	~3.8%
~4.0%	~1.7%	~2.1%	0.070

Table 6. Free CaO content by ethylene glycol analysis and Ca(OH)₂, CaO and CaCO₃ contents by TGA analysis.

3.1.5. Mineralogical Analysis by XRD

In terms of mineralogy (Figure 7), the main mineralogical phases identified in the slag are as follows: (a) calcium aluminate-sodium silicate (labradorite), a phase that may have originated from the slow cooling process of molten slag; the content of this phase corresponds to ~17.6%; (b) larnite (C_2S) (~16.9%), from the slow cooling that transforms the C_3S into C_2S and CaO; and (c) tricalcium silicate (C_3S) (~15.8%), which was also identified. The other phases present were quartz, hematite, and wustite, as well as traces of magnetite, goethite, and metallic iron.



Figure 7. Mineralogical phases obtained for BOF slag by XRD.

In addition, the free lime was identified by Rietveld analysis, with content corresponding to ~2%, similar to the result obtained by TGA. The MgO (periclase), portlandite, and calcite were detected, and calcite occurred in higher quantity (~4.6%) due to the weathering period by which the slag was submitted. The natural sand was also characterized by XRD, essentially indicating quartz.

3.1.6. Chemical Compositions of Slag Phases by Electron Microprobe Analysis

From the Electron Microprobe Analysis, the specific chemical compositions of slag phases were obtained, distinguished by the contrast of the gray color, which allowed verifying the heterogeneous composition of BOF slag, as seen in Figure 8. It was verified that there were few independent phases within the BOF slag grains; instead, most phases were mixed and not free of impurities. The FeO was identified as a homogeneous and independent phase, which corroborates the XRD results. The metallic iron particles in Figure 8C surrounded by calcium and aluminum silicate (CaAl₂Si₂O₈) and calcite, and in



Figure 8D surrounded by dicalcium silicate and a magnesium aluminum silicate phase, were also identified.

Figure 8. Electron Microprobe Analysis performed in different grains of BOF slag (**A**–**D**), in which: 1-FeO; 2-C₂F; 3-MgFe₂O₄; 4-MgAl₂O₄; 5-C₂S, 6-Fe⁰; 7-C₄AF.

From the analysis, the presence of crystals whose main composition corresponds to CaO and SiO₂, associated with a high amount of FeO (23% by mass) and Al₂O₃ (14% by mass), were identified [Figure 8A]. As shown in Figure 8A, the presence of calcium and aluminum silicate particles was also found; the calcium ferrite (Ca₂Fe₂O₅-C₂F), containing Mg and Mn in variable contents and, additionally, in Figure 8B, the magnesium ferrite (MgFe₂O₄), were not identified by XRD. The same Figure presents a calcium silicate phase with a "C/S" ratio of 1.5 (average value), indicating the presence of dicalcium silicate (C₂S), with the impurity content of magnesium, as well as the spinel phase (MgAl₂O₄) associated with iron content. In Figure 8D, brownmillerite (C₄AF) was also observed.

Evidence was found of the occurrence of bubbles in the slag particles, filled with a black resin as a result of the sample preparation for microprobe analysis, and some cracks along the particles. The bubbles present in the slag originate from the gases released during slag solidification and are important porosity indicators, as shown in Figure 8(D), in which several bubbles are surrounded by a phase majority composed of CaO and SiO₂, associated with MgO and Al₂O₃ contents.

3.1.7. Volume Stability Using the Mortar Bar Method

The volume material stability used as aggregates in concrete was evaluated using the mortar bar method [42] in the presence of alkali hydroxide for 30 days, obtaining daily measurements of the linear expansion of mortar bars composed by the BOF slag aggregate and CEM I. The mortar bars showed expansion equivalent to 0.15% after 30 days of curing (Figure 9), which characterizes the material as potentially reactive for concrete use and indicates potential expansion. It should be emphasized that the expansion behavior presented by the slag differs from the behavior commonly presented by reactive aggregates [47,48], since those show expansions higher than 0.2% in the first few days of the test. In contrast, low expansion percentages, less than 0.2%, were observed throughout the evaluation period.

Figure 9. Alkali-aggregate reaction expansion of mixtures containing BOF and silica fume.

Afterwards, to achieve the mitigation and the prevention of the development of alkaliaggregate reaction (AAR), tests were carried out by using the cement CEM IV/A and the addition of 8 and 15% of silica fume (SF) in relation to the cement mass (Figure 9).

It is known that the addition of silica fumes up to 15% in relation to the cement mass leads to alkali-aggregate reaction inhibition. This due to (1) by the reduction in reactive silicon due to the development of pozzolanic reactions, and, consequently, (2) by the increase in pozzolanic C-S-H formation, which presents a fine pore structure. The finer pores imply a reduction in the diffusion and permeability coefficients of the cement paste, reducing the migration of alkalis towards the reactive aggregates. The cement exchange by a pozzolanic material also reduces alkalis input and contributes to the development of pozzolanic reactions [48].

The results show that the replacement of CEM I (by CEM IV/A) and the addition of a silica fume (8% and 15%) were able to reduce expansion to values below the standard limit (0.2%). For up to 16 days, both showed similar behavior; the expandability was about 0.03%. However, from this age, the expandability was more pronounced for the mixture with lower silica fume content; however, the values were lower than the limit (0.15%). Considering the slag mineral phases and the results obtained by the mitigation test, it can be assumed that the expansion presented by the bars did not correspond to the alkali-aggregate reaction, which would have been suppressed by the cement exchange and the addition of a silica fume. Accordingly, the developed reactions were characterized as expansive reactions of the BOF slag constituents.

Similar results were obtained by Ozkan and Saribiynk [18] and Lu et al. [3]. Ozkan and Saribiynk [18] combined steel slag with blast furnace slag (BFS) to produce blended Portland cement, which allowed the development of pozzolanic reactions between the SiO₂ of BFS and the CaO of BOF slag, reducing the material expansion, which was higher than 0.2% at 14 days. Lu et al. [3] observed that the linear expansion of the mortar bar prepared with fine aggregate of BOF slag continuously increased from the beginning up to 22 days, and at 16 days corresponded to 0.17% (greater than the limit, 0.10% at 16 days).

3.2. Concrete with BOF Slag Aggregates Mechanical Properties

The compressive strength of the BOF slag concrete relative to the reference concrete was statistically higher in the two evaluated ages (28 and 180 days), with 95% confidence, as shown in Figure 10, and the results of univariate tests of significance can be found in Table 7. Based on the mean multiple comparisons performed by Duncan's test [49], for the two ages, the concretes were divided into two distinct groups, which indicates the statistical differences and similarities among concretes.

Figure 10. Compressive strength of concretes.

Effect	SS	Degree of Freedom	MS	F	p	Result
Intercept	33,065.75	1	33,065.75	14,662.68	0.000000	statistically significant
Age	127.10	1	127.10	56.36	0.000001	statistically significant
Substitution content	59.05	3	19.68	8.73	0.001164	statistically significant
Age*Substitution content	2.41	3	0.80	0.36	0.784863	not significant
Error	36.08	16	2.26			

Table 7. Univariate Tests of Significance for compressive strength.

Note: SS = sum of squares, MS = Mean squares, F = Fisher's parameter for the effect significance test, <math>p = p-value.

The increase in the compressive strength can be justified due to different mechanisms: (1) Physical mechanism: changes in the mechanical bond of the concrete as a result of the high angularity of the slag particles, associated with their rough surface texture, allowing the boost of the particles interlocking (slag and cement). On the other hand, as previously discussed, these same factors interfere in the workability of the mixtures and reduce the fluidity of the fresh concrete with the increase in the replacement content, as presented in Table 4 (slump test). (2) Interface modification mechanism: based on the water absorption and the mineralogical composition of BOF slag. From the absorption, the higher slag water absorption (compared to natural sand) might lead to the local absorption of adsorbed water

on the surface of the slag grains, reducing the thickness of water films adsorbed around the fine aggregate, which leads to the local reduction of the cement paste porosity in the interfacial transition zone of the fine aggregate of slag. The consequence is a boost in the concrete mechanical strength. In relation to the mineralogical composition, it is possible to infer that due to the presence of C_2S , C_3S , and C_4AF phases in the slag, reactions on the surface of slag aggregates led to the formation of hydrated silicates and aluminates, which increased the adhesion of the aggregate to the cement matrix.

Regarding the tensile strength, no significant difference was found between the concretes (Table 8); therefore, the percentage variation among the means is not relevant, and a single group of concretes was obtained for both ages (Figure 11).

Effect	SS	Degree of Freedom	MS	F	p	Result
Intercept	294.51	1	294.51	3269.57	0.00000	statistically significant
Age	0.71	1	0.71	7.9	0.01000	statistically significant
Substitution content	0.42	3	0.14	1.56	0.24000	not significant
Age*Substitution content	0.08	3	0.03	0.28	0.84000	not significant
Error	1.35	15	0.09			

Table 8. Univariate Tests of Significance for splitting tensile strength.

Note: SS = sum of squares, MS = Mean squares, F = Fisher's parameter for the effect significance test, p = p-value.

Figure 11. Splitting tensile strength of concretes.

The multiple comparison analysis of the means for the results of modulus of elasticity performed by Duncan's test, categorized the concretes into three distinct groups based on the statistical significance of the results (Table 9), so that none of the concretes containing slag was grouped with the reference concrete (Figure 12).

Effect	SS	Degree of Freedom	MS	F	p	Result
Intercept	33,265.75	1	33,265.75	19,900.30	0.00000	statistically significant
Age	34.37	1	34.37	20.56	0.000339	statistically significant
Substitution content	56.51	3	18.84	11.27	0.00032	statistically significant
Age*Substitution content	1.68	3	0.56	0.33	0.800299	not significant
Error	26.75	16	1.67			

Table 9. Univariate Tests of Significance for modulus of elasticity.

Note: SS = sum of squares, MS = Mean squares, F = Fisher's parameter for the effect significance test, p = p-value.

As observed, all concrete with substitution showed a reduction in the modulus of elasticity due to the lower rigidity of the slag aggregate when compared to the natural aggregate, which led to variation in the elastic properties, promoting more severe deformations and reducing the deformation resistance of slag concrete. Similar results were observed by Palankar et al. [9] and Guo et al. [17] for high-strength and normal-strength concrete, for which the increase in steel slag content above 10% by weight led to the decrease in the elastic modulus of concrete.

Certain properties observed in the slag aggregate characterization support the understanding of these outcomes, such as the slag aggregate porosity, characterized by high-water absorption (Tables 3 and 5), which demonstrates the open and interconnected porosity presented by the material. Additionally, the high content of disaggregated material, determined by the Los Angeles abrasion test, exposed the lower surface wear resistance of BOF slag. Both slag characteristics reinforce its low rigidity, which in turn explains the effect of this material in reducing the modulus of elasticity of concrete.

Corroborating the results obtained by the modulus of elasticity, the Rebound Hammer Method was performed on concrete elements to determine the surface hardness of the concrete. As shown in Figure 13, there was a decrease in the number of rebounds (RN) with the increase in the slag content due to a reduction in the wear resistance of concretes with slag incorporation. It was observed that the analysis of the modulus of elasticity and the number of rebounds showed a high correlation, with a coefficient of determination (R^2) of 0.98, demonstrating that the lower stiffness of the slag aggregates reduced the resistance to the mechanical stresses imposed on the concrete for all replacement contents.

Figure 13. Correlation between elasticity modulus (E) and hammer rebound (R) of concretes.

4. Conclusions

In this study, different properties of BOF slag were analyzed for its use in the production of concrete as fine aggregate. Therefore, concretes with different slag contents replacing sand with BOF slag aggregates (25%, 50%, and 75%, by mass) were prepared, which demonstrated satisfactory mechanical behavior.

For BOF slag characterization, an ethylene glycol extraction with complexometric titration was utilized to determine the total free calcium oxide content, which was 3.8%, and f-CaO equal to 2.2% by TGA. Based on the accelerated test of the alkali-aggregate reaction, the potential expansion of BOF slag in an alkaline solution was observed, which was related to the f-CaO and f-MgO contents in the slag aggregates. In accordance with these results, the content of f-CaO greater than 3.8% shows that the steel slag may present poor soundness, which can lead to the failure of concrete when used as an aggregate. This fact emphasizes the importance of prior stabilization of the slag.

Regarding the mechanical behavior of concretes, the concrete compressive strength increased for all replacement levels. The splitting tensile strength of concrete was statistically similar to the reference concrete; however, a slight reduction in the modulus of elasticity was noted as a function of the slag porous structure. It is possible to conclude that considering that the development of expansion reactions leads to the formation of cracks and a reduction in mechanical strength, the mechanical strength evaluation of slag cementitious materials must be performed at older ages, preferably over 180 days since, in the face of sustainable development, the durability of a structure is an important factor, considering there is no sustainability if a structure does not have longevity.

In general, the incorporation of slag in concrete brings benefits to the mechanical properties; however, it is greatly influenced by the heterogeneity of its properties and its expansive behavior. Additionally, creating technologies to remove the potentially expansive behavior of natural BOF slags might lead to economic and technical benefits, boosting the incorporation of slag in construction and reducing the environmental impacts of this residue.

5. Future Directions

Considering all outcomes previously discussed, in addition to statements from the literature, it is understood that environmental, economic, and technical viabilities are the main factors in BOF slag utilization. The use of BOF slag aggregates as a substitute for

natural materials in concrete manufacturing mainly brings environmental and economic benefits to sustainable development concerning natural resources conservation and waste storage reduction. However, there are concerns that this material may potentially lead to crack formation due to the presence of free oxides, which can cause BOF slag expandability. Limited by the expansive oxide content, the utilization of natural BOF slag can be extremely harmful due to its low volume stability even after the slag ages in curing yards for periods that are considered suitable since the stabilization of free oxides might not be achieved for all aged particles homogeneously.

On the other hand, it is clearly known that the BOF slag aggregates may enhance the mechanical properties of concrete, as verified in several studies (Table 3), mainly if used as fine aggregates, which leads to considering the utilization of a preparation step integrated into the aging process. Although the industry aims to directly use aged and ground slag, the real demand for using BOF slag implicates the stabilization process in addition to the aging process. The following stabilization techniques may be possible:

- (1). Air oxidation: By the reheating and controlled temperature cooling of the slag at oxidizing conditions (air atmosphere) under high oxygen partial pressure, in order to oxidize wustite to hematite, which might react with the free lime to form brownmillerite [Ca₂(Al,Fe)₂O₅] during slag solidification in the air [50].
- (2). Water immersion: Research has been frequently conducted on the use of BOF slag in cement manufacturing. The technique involves the quenching of melted slag in water, which might form a vitreous and reactive slag structure and prevent the transformation of C_3S into C_2S and free CaO [10,14,51–53].
- (3). Chelating process: By the reaction between a chelating reagent, the oxalic acid (H₂C₂O₄), and the free lime present inside the slag to precipitate calcium oxalate monohydrate (CaC₂O₄.H₂O). The oxalate produced is relatively insoluble and stable over a wide range of conditions, including high pH conditions. As an additional effect, the precipitation of the mentioned salt can close the capillary porosity of the cement matrix [54,55].
- (4). Accelerated carbonation: BOF slag has great carbonation reactivity; in this way, the natural weathering process is replicated by the CO₂ and the slag particle's reactions, under controlled conditions to accelerate the carbonation reaction and to form stable and insoluble carbonates mainly from CaO and MgO (CaCO₃ and MgCO₃), although also from hydraulic calcium silicate (C₃S, β -C₂S), non-hydraulic calcium silicate (γ -C₂S, CaSiO₃), and hydrated compounds [CaO·SiO₂·nH₂O, 2([Ca₂Al(OH)₆]⁺OH⁻·3H₂O)]. Additionally, the formation of carbonates leads to microstructure densification due to a controlled volume increase [27,32,54–57].

Clearly, the introduction of a stabilization step generates extra costs for the process. However, it also determines the setting up of a new production chain, committed to reducing waste generation and transforming it into new products.

Within this context, the accelerated carbonation process stands out from the other techniques mentioned. Apart from the elimination of the contents of expansive oxides, after the carbonation reaction, significant differences are observed in the physical properties of the slags, including the density, particle size, and formation of finer pore structures, since the CO_2 gas will penetrate the interior of slag particles through surface imperfections such as pores and cracks to form denser particles, which are interesting in terms of their aggregate properties. This whole process also involves the sequestration of considerable amounts of carbon dioxide. The higher the content of free oxides in BOF slag, the more relevant the CO_2 sequestration is, despite the complex mineralogical and chemical composition of BOF slag, which does not allow the prediction of its CO_2 sequestration ability [27,32,56–59].

During the pig iron refining process, one of the main reactions that occurs inside the oxygen converter is carbon oxidation to carbon monoxide or carbon dioxide. Consequently, it leads to the release of high levels of CO_2 into the atmosphere and to the potential of the industry as a source of CO_2 , whose released gas must be used in the accelerated carbonation process. From the recovery of the generated gas during the production of steel in the

converter to be burned or released into the atmosphere, it would be possible to redirect it to a carbonation chamber since the current laboratory process is achieved by CO_2 curing in a sealed chamber with high CO_2 concentration and controlled conditions, e.g., gas pressure, temperature, humidity, to obtain CO_2 sequestration in steel slag [56,59]. Moreover, the CO_2 curing process is consolidated and resembles the carbonation techniques of cementitious materials, or it might be obtained by the high gravity carbonation method, using a rotating packed bed, to achieve a high conversion in a short period of time [56,57,59].

Despite the necessity of technological development to recover and store the produced CO_2 , steelmaking is a contiguous and valuable source of CO_2 , and the steel process already presents favorable conditions for slag-accelerated carbonation. Currently, the slag is aged under environmental conditions and with constant humidification to accelerate the oxides hydration, which facilitates the accelerated carbonation process by providing the initial slag moisture as a pre-curing step.

The accelerated carbonation of steelmaking slags might be viable given that slags are ideal for CO_2 sequestration due to their oxide composition, their favorable reaction with CO_2 , their availability, low cost, and closeness to large CO_2 emission sources. These conditions encourage their reuse in a large variety of applications through the improvements in their chemical, mineralogical, and physical characteristics as concrete and asphalt aggregates and their ability to reduce CO_2 emissions to the atmosphere.

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