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Performance of Pre-Carbonated Basic Oxygen Furnace Slag as a Replacement for Natural Coarse Aggregates in Concrete

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Abstract. Basic Oxygen Furnace Slag (BOFS) is a significant by-product of the steel-making process, where over 250 million tonnes is produced annually around the world. However, the utilisation rate of the steelmaking by-product remains low due to the expansive hydration products formed by the BOFS. To support the potential high-volume use of the by-product as coarse aggregate in concrete, this study aimed to investigate the use of pre-carbonation over 7 days to pre-react the constituents that cause expansive behaviour. For this purpose, five concrete mixes were designed with a volumetric replacement of the Natural Coarse Aggregates (NCA) with the BOFS and the pre-carbonated BOFS (BOFC) at 0, 50 and 100% replacement percentages. X-ray diffraction (XRD) and water absorption analyses were conducted on both types of BOFS aggregates to support the compressive strength, prism expansion, and oxygen permeability tests, which were also performed in the study. The results from the compressive strength test showed a 10.7% and 1.4% higher 56–day compressive strength result for the concrete made with BOFC and BOFS, respectively, compared to the natural aggregates. Similarly, the oxygen permeability test showed that the concrete made with 50% BOFC was 2% less permeable than the untreated BOFS and at 28-days of curing, exactly the same compared to NCA mixes.. Lastly, the prism expansion test indicated that pre-carbonating the BOFS improved the volumetric stability of concrete made with BOFS over the 84-day curing regime. Therefore, the above results suggest that pre-carbonation can increase the utilisation rate of BOFS as a coarse aggregate in concrete.

Introduction

Economic development closely aligns with industrial growth, with steel usage being a key indicator [1]. Countries in the Brazil-Russia-India-China-South Africa (BRICS) forum account for 68% of global steel production [2], with South Africa producing 80–100 kg of steel per capita, which is comparable to India, the world's second-largest producer [3]. Roughly 70% of global steel is produced via the Basic Oxygen Furnace (BOF) process, which generates 100–200 kg of Basic Oxygen Furnace Slag (BOFS) per ton of steel [3]. In this process, oxygen is blown into molten scrap and pig iron to oxidise impurities, followed by limestone addition. The limestone decomposes to calcium oxide (CaO), which combines with impurities, forming liquid BOFS [4]. Though BOFS is used in agriculture, cement kilns, and road construction [4, 5], a significant portion of the more than 250 million tons of BOFS produced annually in the world is not utilised, where potential large-scale utilisation lies in the construction sector [4]. The predominant concern over the utilisation of the by-product as a construction material, specifically an aggregate due to its coarse nature, is the volume instability of the BOFS during hydration. The unreacted CaO remaining in the BOFS after steelmaking hydrates into calcium hydroxide [$Ca(OH)_2$], which has lower density relative to CaO and thus a larger volume for the same mass, which results in expansion of the BOFS in the process [6, 7].

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Materials Research Proceedings 63 (2026) 175-185 https://doi.org/10.21741/9781644904053-20

Various treatments have been explored to mitigate BOFS's expansion, minimising concrete cracking and spalling [8]. Sun *et al*. [9] evaluated pre-treatment methods, such as soaking in water, acetic acid, and silane coatings, which all reduced expansion. Silane proved most effective (0.5% expansion after 10 days) but is costly and industrially complex. Alkaline activation with sodium silicate improves the cementitious properties of BOFS by forming calcium silicate hydrate (C-S-H) gels [10], but low grindability of the BOFS raises energy costs. As BOFS has a vitreous structure, using it as aggregate instead of binder may offer better scalability, particularly given concrete's 60 to 80% aggregate content [11] which would promote high volume usage.

Early carbonation is another promising method, converting the accessible unreacted CaO to solid $CaCO_3$ in the pores and on the surface of the BOFS, thus limiting hydration and subsequent expansion [12]. While this method has been tested under ideal lab conditions (i.e. high CO_2 concentration and temperature), minimal research exists on ambient conditions. Additionally, limited studies assess "worst-case" BOFS (unweathered and untreated), highlighting the need for more practical assessments across variable feedstock. As such, this study investigates early carbonation of unweathered BOFS under ambient conditions (room temperature, atmospheric pressure, 20% CO_2, and 65% humidity) before its use as coarse aggregate as a means of investigating the adequacy of pre-carbonation as a solution to improving the volume stability of BOFS in concrete.

Pre-carbonation process

The following section describes the pre-carbonation process along with the procedure followed and the findings from the treatment process.

The principle: Mineral carbonation is a method of carbon sequestration in which carbon dioxide readily reacts with natural or synthetic mineral phases, such as CaO and MgO, to form various carbonates, effectively permanently trapping CO_2 in stable mineral phases [13]. This carbon sequestration method is considered permanent since it would require high energy levels through thermal decomposition to separate the CO_3^{2-}ions. Furthermore, the resulting products are known to be stable insoluble carbonates [13]. Given that a major limitation to the utilisation of BOFS is the presence of free CaO, in the early carbonation approach, the CaO and MgO in the solid BOFS feedstock readily react with the gaseous CO_2 to form the respective insoluble carbonates on the aggregate's surface and internal pores for further use in concrete. In the case where CaO has already hydrated to form $Ca(OH)_2$, the carbonation reaction still occurs without any major changes to the environmental conditions. The early carbonation approach aims to pre-react this compound, forming a stable and insoluble carbonate on the aggregate's surface and internal pores for further use in concrete.

It was reported by Zhao *et al.* [14], that the reaction of CO_2 with $Ca(OH)_2$ actually occurs at faster rates relative to the reaction of CaO and CO_2 where it was found that the former, also experienced higher conversion degree to $CaCO_3$. Furthermore it was found by Dostie *et al.* [15], that the $Ca(OH)_2$ was consumed highly to produce $CaCO_3$ in high humidity environments (60-95%) as it was found that the dissolution of $Ca(OH)_2$ and the precipitation of $CaCO_3$ are greater at higher RH where the most optimal was found to be 80%. The reactions which are anticipated to occur during the carbonation process are the following Eq. 1 – Eq. 3, where the source of the CaO is the BOFS feedstock.

$$CaO_{(s)} + CO_{2\,(g)} \rightarrow CaCO_{3\,(s)} \quad (1)$$

$$CaO_{(s)} + H_2O_{(g)} \rightarrow Ca(OH)_{2\,(s)} \quad (2)$$

$$Ca(OH)_{2\,(s)} + CO_{2\,(g)} \rightarrow CaCO_{3\,(s)} + H_2O_{(l)} \quad (3)$$

The Process: In a system where there is sufficient CO_2 gas available, reactions in Eq. 1 – Eq. 3 above are expected to occur within the system until the CaO which is accessible to fluids in the BOFS has been consumed. The various parameters which can be altered to make the process of CaO consumption occur faster are well investigated and have been reported to be: the concentration and pressure of the CO_2 gas in the system, the air pressure of the system temperature and relative humidity of the system and the addition of any admixtures to the system [16]. In the current study, a focus on selecting parameters which would require the least external inputs, thus improving the economic feasibility of the treatment process. As such, the process parameters which were selected in the current study are: Air with a 20% concentration of CO_2 gas in the chamber, relative humidity of 65 ± 2 %, room temperature of around 18ºC, and atmospheric pressure.

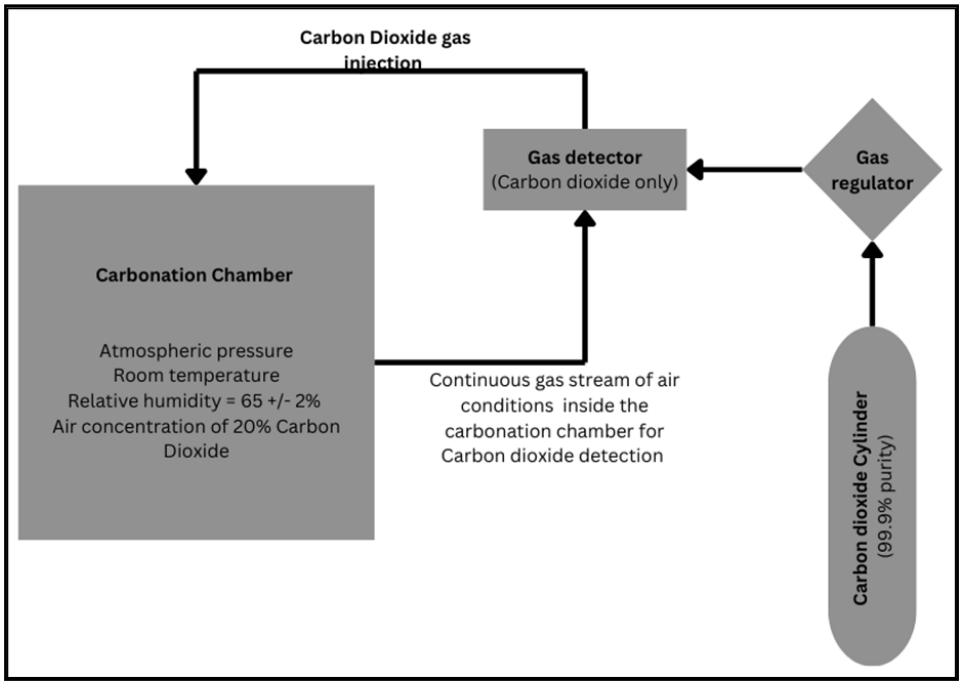


Figure 1: Schematic Diagram of the carbonation chamber

The carbonation process was carried out in a chamber which was kept closed, where all openings were covered with petroleum jelly to adequately seal the openings of the chamber. An air circulation fan was used to ensure uniform air conditions throughout the chamber. Figure 1 above shows a schematic diagram of the system used.

The Result: Using the selected process parameters, as mentioned above, a pre-investigation was required to establish the adequate period which the BOFS would have to remain in the chamber to ensure that the accessible CaO has reacted as per Eq. 1 – Eq. 3. Using mass as an indication of ongoing reactivity, a BOFS sample of 1472.4 g was placed in a 4.75mm aperture weighed sieve, then placed into the carbonation chamber. The sieve was used to ensure adequate gas circulation around the specimen. This weighed sieve was then removed from the chamber and weighed every 24-hours, with the change in mass recorded, where this change in mass is assumed to be due to the formation of $CaCO_3$ from the reaction of either the CaO or $Ca(OH)_2$, as per Eq. 1 or Eq. 3. This mass reading is shown in Figure 2, where the most reactivity occurs within the first four days of exposure in the chamber. After four days of exposure, the change in mass becomes negligible,

where it can be assumed that the accessible CaO in the BOFS has been converted to $CaCO_3$ at this period of treatment.

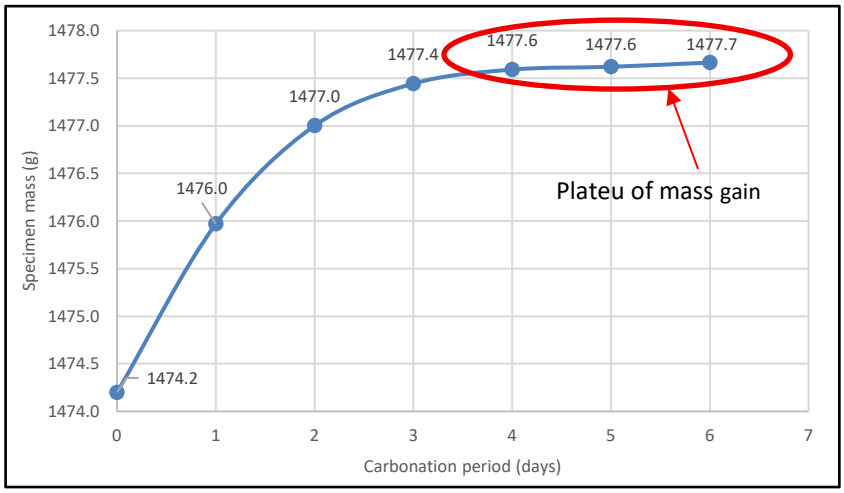


Figure 2: Results of carbonation pre-investigation

A period which was selected based on this set of results was a treatment period of 7 days. This is on the basis that after 5-days the gain in mass was negligible, but to account for any variations in treatment on a larger volume of BOFS, a 7-day treatment period was selected for certainty.

Characterisation of the material used in the study

The natural aggregate used in the study is a 22 mm Andesite stone and crusher sand from the Eikenhof quarry and the BOFS was from the Vanderbjilpark workings steel plant. The binder used was CEM I 52,5R Portland Cement (PC).

Physical properties of the aggregates used in the study: Table 1 below compares the specific density, water absorption and Aggregate Crushing Value (ACV) of the aggregates used in the study. The specific densities were determined using a helium gas pycnometer, the water absorption values were determined using ASTM C127-15 and the ACV was determined using SANS 3001-AG10:2012 as a guideline. Table 1 shows that the specific densities of the BOFS and BOFC are slightly higher than that of the NA. This difference will be factored into the study when considering the concrete mix design, where the replacements of NA are carried out on volumetric quantities. During this process, it was ensured that the particle size distribution curves of the replacing BOFS, matched that of the replaced NA. Additionally, it was observed that the ACV of the BOFS and BOFC is higher than that of the NA, which means that the NA is more resistant to crushing under a compressive load than the BOFS and BOFC. SANS 3001-AG10:2012 was used as a guide to determining the ACV.

Table 1: Physical properties of the aggregates used in the study

	Natural Coarse Aggregate	BOFS coarse aggregate	BOFC coarse aggregate
Specific Density [kg/m^3]	2893.1	3274.5	3274.5
Water Absorption [%]	0.38	1.45	1.08
Aggregate Crushing Value (ACV) [%]	5.64	11.17	11.17

Additionally, the NA exhibited the lowest water absorption, followed by the BOFC, while the untreated BOFS showed the highest water absorption at 1.45%. The difference between the BOFS and BOFC is attributed to the early carbonation process which resulted in the formation of calcite in the material's pores. This effect effectively clogs the internal pores of the BOFS and makes the aggregate less porous, thus less water is trapped internally, leading to lower water absorption [17].



Figure 3: Visual representation of the coarse aggregates used in the study

As seen in Figure 3, the surface roughness and aggregate shape and of the different coarse aggregates used is highly comparable. It should be noted that the early carbonation process does not change the visual aspects of the BOFS aggregates.

Crystal composition of the BOFS: An analysis of the crystal composition of the BOFS and BOFC is used to provide evidence of treatment adequacy and indicate whether any additional reactions occurred during the treatment process. This analysis was carried out using an X-Ray Diffraction (XRD) analysis. The equipment used to obtain the diffraction pattern was the D2 Phaser diffractometer where each sample was exposed to incoming X-rays for 10 minutes using a pre-programmed setting, where peak identification was done using the Diffrac Eva database, and 2Theta locations confirmed from literature [18, 19]. Two different BOFS (Named BOFS A and BOFS B) and two different BOFC (named BOFC A and BOFC B) single 22 mm aggregates were randomly selected and pulverised to powder form, such that XRD analysis can be undertaken, with the diffractograms shown in Figure 4 below. Two random aggregates were used to investigate whether there is a distinct difference in the mineralogy of the BOFS batch of aggregates used in the study, as the production of BOFS is not regulated, and a difference could exist. The same approach was considered for the BOFC aggregate batch. The diffractograms for BOFS A and B were plotted to identify any differentiation in the observed peaks, and the same approach was taken for the BOFC A and B diffractograms, though on a different vertical axis.

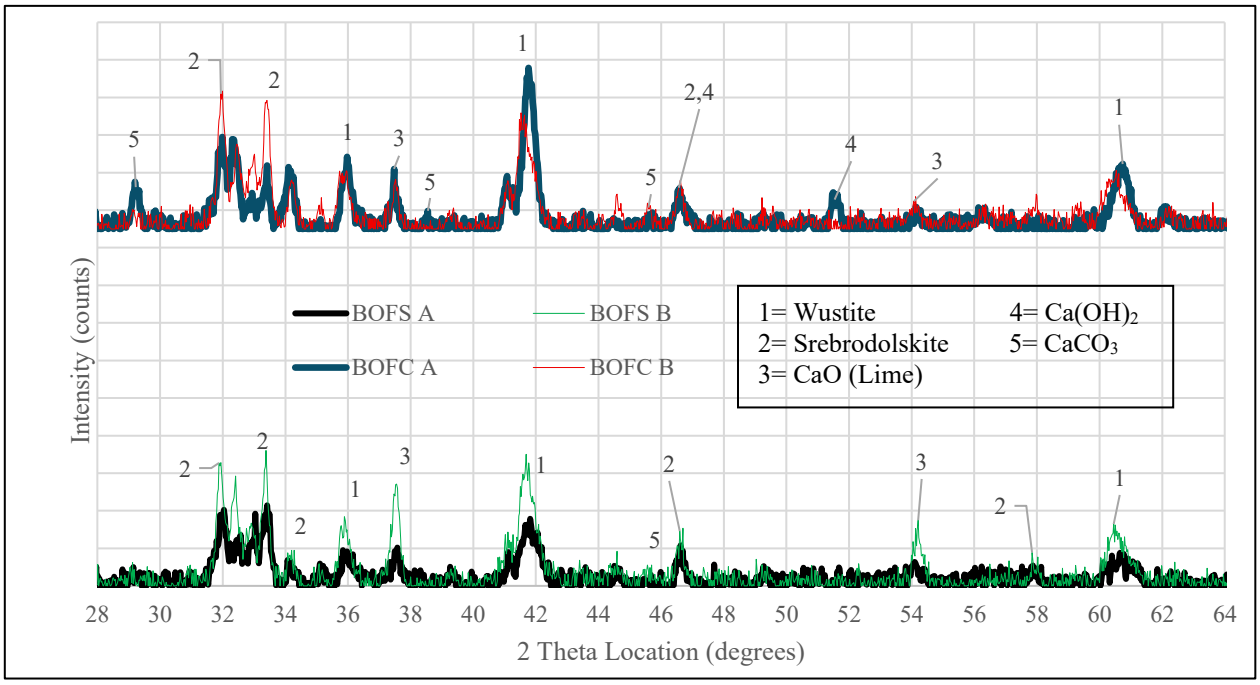


Figure 4: XRD for BOFS and BOFC

The combined diffractograms showed that despite the differences in the recorded peak intensities for the various crystals in both samples of both BOFS and BOFC the prevailing dominant phases in both BOFS and BOFC include Wustite (FeO) and Srebrodolskite ($Ca_2Fe^{3+}_2O_5$), which appear quite similar. A notable difference between BOFS and BOFC is the presence of calcite and portlandite peaks in the BOFC samples, alongside a significant reduction in the intensity of the CaO peaks relative to the BOFS samples. The CaO peak is not expected to disappear entirely after early carbonation, as CO_2 gas can only react with the free CaO content that is accessible to fluids in an unpulverized aggregate. However, not all lime pockets are accessible to fluids, as some are located within the internal matrix of the BOFS [20]. Therefore, due to the pulverisation of the BOFC aggregate following treatment for XRD analysis, the free CaO that remained inaccessible to fluids will be released, resulting in an expected CaO peak. It can be appreciated from Figure 4, that the reactions which occur during the treatment process, are those identified by Equation 1 to 3.

Experimental Procedure

Concrete Mix design: A single PC-binder system was used in the current study, where the fine aggregate quantity and water content were kept constant throughout all the mixes. The natural coarse aggregate was replaced, by volume, at 50% and 100% with either BOFS or BOFC. The 100% replacement percentage were selected to focus the study on the complete replacement and 50% was used to confirm any trends as an midway replacement. Each mix design used in the study had a unique blend of coarse aggregate content, where blends either consisted of NA and BOFS aggregates or NA and BOFC aggregates. Table 2 below shows the various unique mix designs used in the current study. The nomenclature used to identify the mixes includes the BOFS or BOFC name and the number after the dash represents the percentage which the NA has been replaced by in that mix i.e. BOFS – 50 / PC, means that 50% of the coarse aggregates BOFS and 50% is the NA.

Materials Research Proceedings 63 (2026) 175-185 https://doi.org/10.21741/9781644904053-20

Table 2: Concrete mix Designs used in the current study

MIXES	Mix Proportions [kg/m^3]						Total Density [kg/m^3]
	PC	WATER	FINE AGGREGATE	NATURAL COARSE AGGREGATE	BOFS	BOFC	
NA / PC	388.9	175	950.5	1073.3	0	0	2587.7
BOFS - 50 / PC	388.9	175	950.5	536.6	685.2	0	2736.2
BOFC - 50 / PC	388.9	175	950.5	536.6	0	685.2	2736.2
BOFS - 100 / PC	388.9	175	950.5	0	1370.3	0	2884.7
BOFC - 100 / PC	388.9	175	950.5	0	0	1370.3	2884.7

Testing Regime: The current study used concrete compressive strength to investigate the effect which precarbonation of BOFS coarse aggregates has on the performance of concrete. Testing was conducted at 3, 7, 28, 56 and 90-days of curing in a water (22 ± 2 ºC), where SANS 5863 was used as a guideline for this. To assess the effect of precarbonation of the BOFS on the volume stability of the concrete, the concrete prism expansion test was conducted, as per the guidance of ASTM C157. Additionally, to assess the effect of precarbonation of the BOFS on the gas permeability of concrete, the Oxygen Permeability Index (OPI) was determined for the concrete mixes, as guided by the Durability Index Manual and SANS 3001-CO3-1:2015.

Results and Discussion

Compressive strength test results: Figure 5 shows that the average compressive strength of concrete made with either BOFS or BOFC is higher than that of concrete made with NA (at 56-days, 100% BOFS mix is 1.5% stronger and BOFC mix is 5% stronger than NA mix). Furthermore, these results indicate that the use of early carbonation, to produce BOFC, results in a slightly stronger concrete at all curing days, relative to untreated BOFS. Given that all the physical properties of the materials which were characterised were similar, except for the water absorption and the ACV, it is believed that these properties could be the reason for the difference. However, a higher ACV value for the BOFS or BOFC, wouldn't positively affect the compressive strength of concrete, and if there had to be an effect, it would result in a decrease in the strength of concrete due to the aggregate cracking before the paste [21]. This leaves the different water absorption values as the physical property of the aggregates which could be the reason for the difference in concrete strength. Given that the NA has a lower water absorption and results in a lower compressive strength and the inverse is the case for BOFS, a relationship could be drawn between the water absorption and the strength. However, the BOFS has a higher water absorption relative to the BOFC, yet BOFC results in a higher concrete strength. This introduces the possibility of another property of the BOFS and BOFC aggregates also affecting the concrete strength, such as the volumetric expansion.

Another observed difference when characterising the BOFS and BOFC aggregates, are the peaks of $Ca(OH)_2$ and $CaCO_3$ in the BOFC aggregates which formed during the treatment process where the hydratable CaO was pre-reacted.

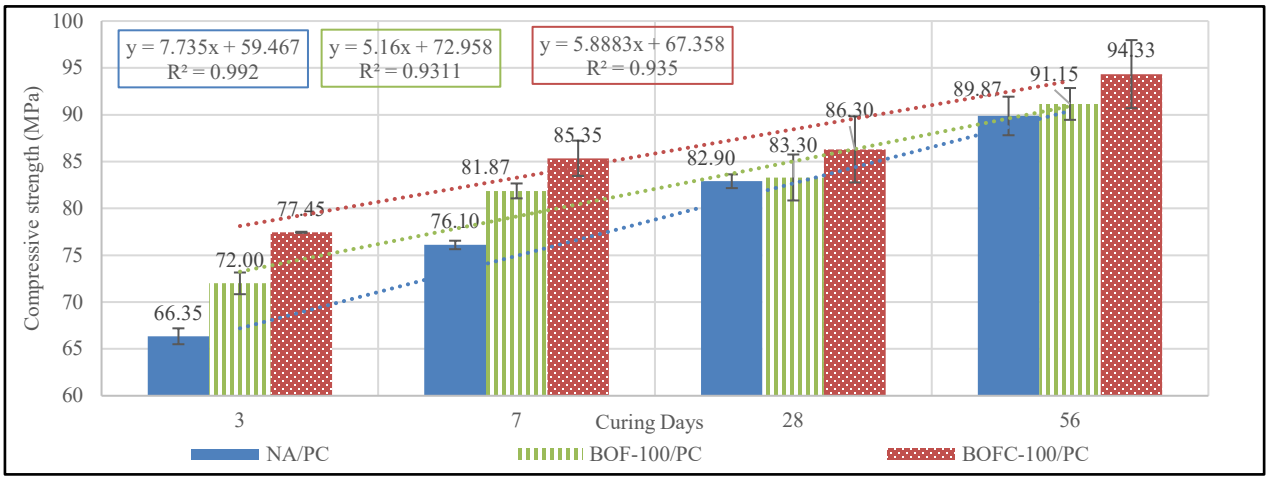


Figure 5: Average compressive strength of concrete mixes

The CaO in the BOFS is known for resulting in volumetric instability of the BOFS during hydration, due to the crystal formation of the hydration product [$Ca(OH)_2$] which has a larger crystal, relative to CaO. This expansive reaction causes a tensile force on the surrounding concrete paste, which induces additional cracking of the concrete. This cracking is believed to make crack propagation easier under loading, and as such, result in a lower bearing capacity of the concrete [22]. This phenomenon could explain why the relationship between the water absorption value and compressive strength of the concrete made with the BOFS and BOFC was unclear, as the BOFS is more volumetrically unstable, as seen in Figure 6, which would induce more internal cracking, and thus negatively affect the compressive strength of concrete. Figure 6 shows the results from the concrete prism expansion test, where it is seen that at 100% replacement of NA with BOFS, a 177% higher length expansion is recorded for the concrete . Furthermore, the opposite is recorded where a 71% reduction in length expansion is recorded in the concrete mixes where 100% NA is replaced with BOFC (improved volume stability). This indicates the effect of the pre-carbonation process on improving the volume stability of concrete made with the steelmaking by-product.

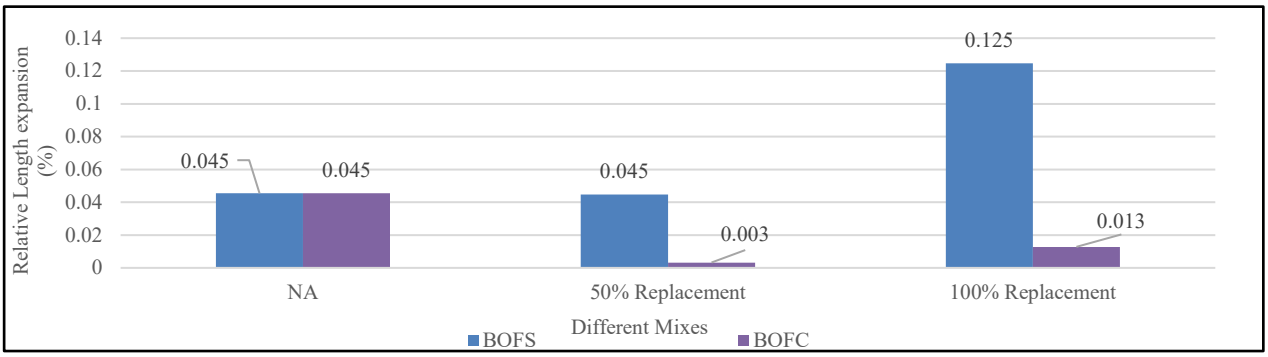


Figure 6: Results from the concrete prism expansion test

Additionally, another positive effect of the pre-carbonation process on the BOFS is the improved resistance to oxygen permeability of the resultant concrete. This is seen in Figure 7 where the OPI of concrete made with BOFS decreases more than that made with BOFC, which is represented by less steep (-0.1018 < -0.1579) trend line of the BOFC concrete mixes, relative to the BOFS mixes. Additionally, the OPI values for the 50% BOFS mix is 2.88% lower than of the 50% BOFC mix. This lower OPI occurs at all replacement percentages of NA with BOFS. The inclusion of both BOFS or BOFC however makes the concrete more permeable by gases as the OPI values are generally lower than those of the NA concrete mixes, however this can not be said

with a 95% confidence, as the error bars of the NA and BOFC concrete mixes overlap at both 50% and 100% replacements. The 50% replacement of NA with BOFC has an identical OPI value to the NA mix. The superior performance of the BOFC, relative to the BOFS is potentially due to the $CaCO_3$ formation within the pores of the BOFS after the pre-carbonation process [23]. A look back at Figure 4, shows that the BOFC aggregates had a $CaCO_3$ peak after the carbonation, which supports this statement. Additionally, the lower water absorption rate which was determined for the BOFC aggregates, relative to the BOFS is also indicative of the $CaCO_3$ formation occurring within the pores of the aggregates which clogged the pores and reduced the permeability of gases.

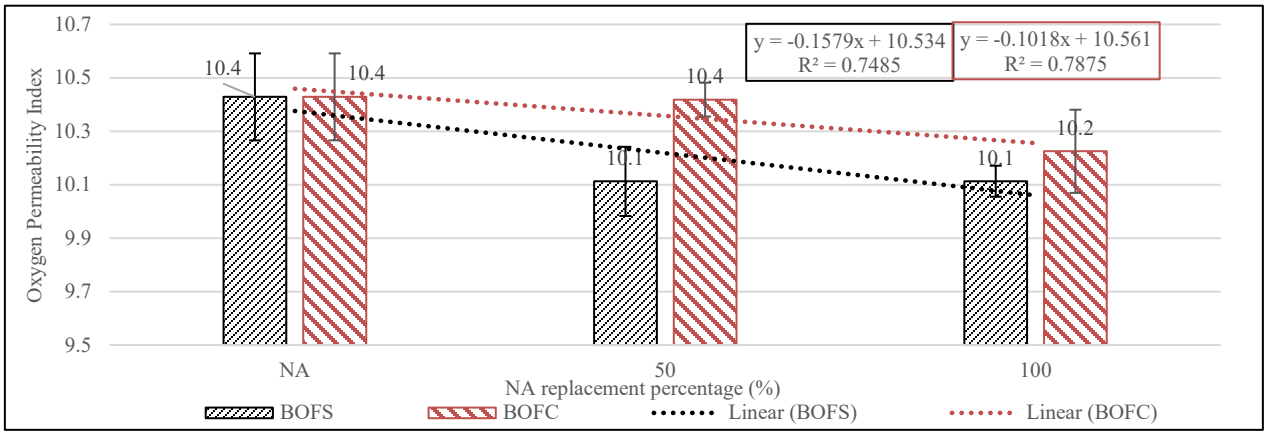


Figure 7: Average OPI test results of the concrete mixes

Conclusion

This study demonstrates that carbonation of BOFS under ambient conditions significantly enhances its performance as a coarse aggregate in concrete. The treatment effectively reduced the water absorption of BOFS and volume instability of the concrete, while improving compressive strength and gas impermeability. The formation of stable $CaCO_3$ within the BOFS microstructure played a key role in these improvements. These findings suggest that carbonation is a practical and scalable approach to increasing the utilisation of BOFS in concrete applications, supporting both sustainability and circular economy objectives in the construction and steel industries. The BOFC-100/PC showed superior compressive strength and volume stability, relative to all other concrete mixes and the oxygen permeability performace was only around 1% less than the NA mix, thus sugesting very insignificant differences in performance. As such the BOFC-100/PC would be the favourable mix for further investigations.

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