Development of Concretes with Different Strength Classes and Variations of Untreated Bauxite Residue Consumption

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Abstract

Most (estimated > 90 %) of the world's concrete exhibits compressive strength of up to 50 MPa, with the remaining percentage divided between high- and ultra-high-strength concretes. These different compositions find applications in monolithic floors, paving blocks, ordinary concrete for columns and beams, urban furniture, and more, and are of great practical interest. It's important to note that a solution designed for a specific application and using certain raw materials may not be suitable for another purpose, particularly in highly demanding situations. Therefore, development efforts must be driven by the requirements of both fresh (before hardening) and hardened (aged) state properties. The combination of bauxite residue (BR) with Portland cement (PC) is closely linked to the key sustainability challenges facing the aluminium and cement industries. This integration is in line with the roadmap outlined by the International Aluminium Institute, which recognizes it as one of the most impactful applications for this residue. The focus of this project is to develop large-scale and low-cost applications for BR without the need for additional treatments, such as energy-intensive calcination or environmentally harmful additives. The research presented here, explores alternatives for incorporating a significant amount of BR into cementitious compositions (up to 116 kg/m³ of concrete, equivalent to 5.4 % of total solid mass or 27 % of cement mass) with compressive strengths in the range of 30 to 50 MPa. These compositions are intended to produce paving or monolithic concrete, suitable for various cementitious building components. In particular, the study used raw materials from the northern region of Brazil, considering logistical considerations. The material was easy to mould, showed satisfactory fresh and hardened (aged at 28 days) performance, and, as a considerable amount of residue was incorporated without compromising performance, it resulted in more environmentally friendly products.

Keywords: Normal strength concrete, Bauxite residue, Rheological properties, Mechanical properties, Colour.

1. Introduction

The bauxite residue (BR) generated by the Bayer process for alumina production is accumulating worldwide, posing costs and risks for the aluminium industry in terms of handling and storage. Despite research efforts, applications on a relevant scale remain limited. Any proposed solution should not only consume significant volume, but also demonstrate adequate technical performance considering quality, cost, and risk of environmental contamination [1, 2].

The roadmap developed by the International Aluminium Institute [3] recognizes the use of BR in cementitious products as one of the most promising applications. Various applications can be envisaged, such as monolithic floors (roads, logistics warehouses, sidewalks), interlocking pavers, general-purpose concrete for structures (columns, beams, slabs, and walls), urban furniture, etc., which show significant practical interest.

Different routes have been proposed for the use of BR in cementitious materials, some of which involve the beneficiation of BR. As these stages usually involve significant energy consumption (grinding, calcination) or the use of special additives, they result in higher costs and potential environmental impacts. However, studies indicate that it is possible to use the residue in its natural state, without any additional process [4–7].

The objective of this work is to assess the impact of adding BR without additional processing (only filter pressed) for use in common concrete applications with strengths of up to 50 MPa. As these applications include most - estimated > 90 % [8, 9] - of the concrete volume produced currently, the aim is to reach a relevant potential market share. In addition, local aspects of material availability have been considered, integrating the use of BR into the regional consumption logic.

2. Experimental

The study started with the development of reference concretes (without BR) with cement consumption ranging from 318 to 429 kg/m³ and water/cement ratios between 0.48 and 0.65 to obtain concretes for different applications. Table 1 presents these mix designs.

A cement type CPV-ARI RS (Sulfate Resistance High Early-Strength Portland Cement) was used, which meets the requirements of the Brazilian standard ABNT NBR 16697:2018. As aggregates, river sand and gravel were used, both of which are common in the region where the BR is generated. A water-reducing plasticizer admixture (Miraset® 63 from GCP) was also employed, in a proportion of 0.9% by mass of cement.

The water and admixture consumption were adjusted to achieve suitable workability to produce various products, such as moulding pieces in moulds applying vibration through vibrating tables, or for use in ready mix concrete and vibration through immersion vibrators. A target slump of 100 ± 20 mm was adopted.

Table 1. Compositions of reference concretes (mass in kg/m³).

ID	Cement	Sand	Gravel	Water
REF_L	318	785	980	209
REF_M	374	750	997	201
REF_H	429	710	1010	204

For each reference concrete, three additional concretes containing BR were produced, in proportions of 10, 20, and 30 % relative to the volume of cement (mix design presented in Table 2). This addition of BR was done by replacing part of the sand, therefore not changing the cement, gravel and total (added + water in BR) water consumption. So, the total water/cement ratio was kept unchanged.

The bauxite residue (BR) was obtained from Alunorte/Hydro in Barcarena (north of Brazil), from the production of alumina by the Bayer process, directly from the filter presses. It was delivered with a moisture content of 26 % and used in this condition for the production of concrete. Table 3, Figure 1 and Table 4 present the physical and chemical properties of the materials.

Table 2. Compositions of concretes with BR.

Cement (kg/m³)	BR(1) (% of cement volume)	BR(1) (kg/m³)	Sand (kg/m³)
318	10	29	754
	20	59	727
	30	88	698
374	10	35	715
	20	70	684
	30	103	646
429	10	40	666
	20	79	631
	30	116	584

(1) Expressed on dry basis

Table 3. Physical properties of the materials.

Material	Density (g/cm³) (1)	SSA (m²/cm³) (2)	D_{10} (µm) (3)	D_{50} (µm) (3)	D_{90} (µm) (3)
BR	2.98	7.74	1.6	7.5	85
Cement	3.19	1.56	4.9	17	45
Sand	2.66	-	182	327	926
Gravel	2.65	-	5,751	11,076	16,062

(1) by helium gas pycnometry (AccuPyc II 1340/Micromeritics);
(2) Specific Surface Area: by N_2 adsorption and BET-model (Belsorp max/Bel Japan);
(3) characteristic particle sizes: Laser diffraction (Helos/Sympatec) for fines; Dynamic Image Analysis (Qicpic/Sympatec) for aggregates.

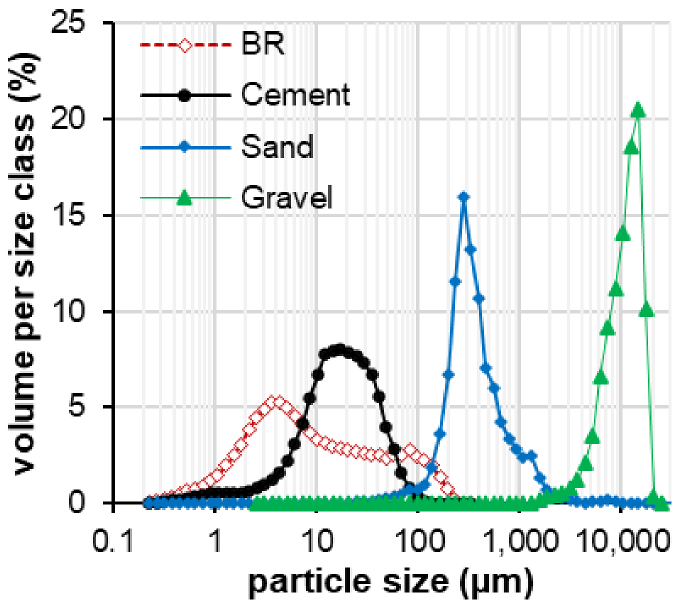

Figure 1. Particle size distribution of the materials.

Table 4. Chemical composition of cement and bauxite residue (by XRF).

Oxide	Cement	BR	Oxide	Cement	BR
SiO_2	17.8	16.3	SO_3	2.29	n.m.
CaO	61.8	1.15	Na_2O	0.06	9.72
MgO	3.18	< 0.10	K_2O	0.34	0.03
Fe_2O_3	4.08	36.2	P_2O_5	0.14	0.05
Al_2O_3	4.35	22.1	ZrO_2	n.m.	0.62
MnO	0.17	0.07	Cr_2O_3	n.m.	n.d.
TiO_2	0.24	5.27	LOI	4.85	8.40

n.d.: not detected; n.m.: not measured

The concretes were mixed using a PHESO (Calmetrix) rheometer with an attritor impeller to evaluate the effect of introducing BR during the mixing process. Mixing was conducted in two stages: first, the mortar was mixed, and the rheological properties measured; then coarse aggregates were added to obtain the concrete. This mixing procedure was adopted to study different products. In this paper, as the focus is on concrete, mortar data will not be presented.

After the final homogenization, other tests were carried out (rheometry, slump test, and density, to evaluate the air content). Shear cycle tests were carried out to determine the rheological behaviour of the mortar and concrete. Each cycle consists of an acceleration phase (8 steps of 8 seconds each, with speeds increasing from 6 to 250 rpm) followed by a similar deceleration phase. Initially, all concretes with BR were tested with the same amount of water as the respective reference concrete to assess the effect of the introduction of BR on the rheological properties. Then, for those concretes that showed slump values below the specified minimum, adjustments were made by adding extra water, as is often done in concrete production practice.

Concrete test specimens were moulded using a vibrating table to ensure proper compaction. After 24 hours of curing in the moulds, the specimens were demoulded and stored in a wet chamber to cure for 28 days. Cylindrical specimens (10 × 20 cm) were prepared for compression testing, while prismatic specimens (10 × 10 × 35 cm) were prepared for flexural testing. Additional tests (abrasion and water absorption) were carried out on the same specimens after the flexural test. These last two properties were evaluated in accordance with the Brazilian standard (ABNT NBR 9781:2013) with the aim of assessing their applicability for use as concrete interlocking concrete pavements exposed to pedestrian traffic, vehicles with pneumatic tires, and storage areas.

3. Results and Discussion

3.1 Fresh State

Figure 2 shows the slump obtained after the first mixing. Initially, an increase in slump can be observed for all concretes with low BR contents (up to 10 to 20 % of the cement consumption). In the case of higher cement consumption, high BR contents (30 %) result in BR consumptions above 100 kg/m^3, in which case a reduction in slump, to values below the acceptable minimum, is observed.

The improvement in workability at lower BR contents can be attributed to the enhancement in the granular packing of the paste due to the BR being finer than the cement (see Figure 1). However, due to the high specific surface area of BR (5 times higher than that of cement, see Table 3), this improvement is overshadowed at higher BR content by the greater demand for water required to cover the entire particle surface area, particularly in those cases where BR substitutes part of the sand.

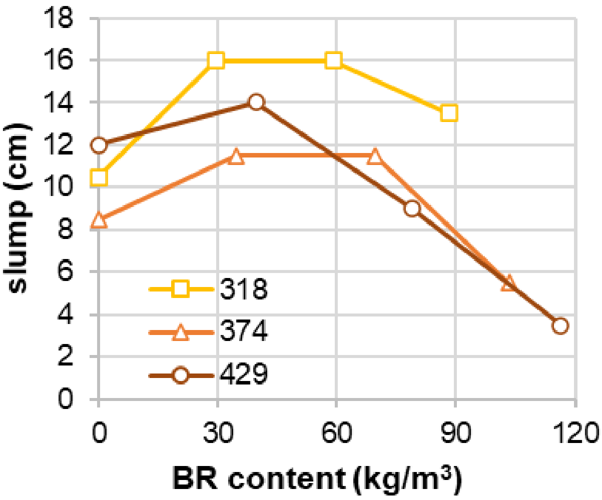

Figure 2. Slump in relation to BR content (labels refer to cement content in kg/m^3)

The rheological parameters obtained from the shear cycles rheometry show that similar trends can be observed for the yield torque (Figure 3-left) and apparent viscosity (Figure 3-right). For lower cement contents, the torques are even lower than for higher cement contents, indicating that the introduction of the BR has resulted in an additional paste volume that has increased the mobility of the aggregate skeleton. Up to 90 kg/m^3 of BR, there are no significant changes in rheological properties, whether in low or high shear rate applications.

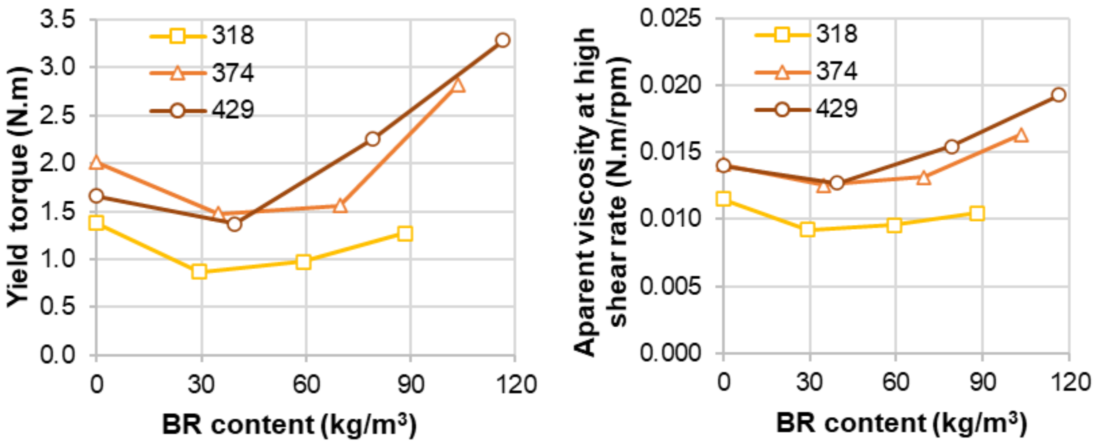

Figure 3. Rheological properties. Left: Yield torque, modelled according to Bingham model, Right: Apparent viscosity at high shear rate (250 rpm).

At higher BR consumptions, more significant changes in the rheological properties were observed, which may affect the applicability. For example, two concretes with the highest cement consumption and 30% BR required an increase in water consumption: 12 and 19 litres, respectively, for the intermediate and higher cement consumptions. After these adjustments, these concretes exhibited slumps (9 cm for intermediate cement consumption and 12 cm for higher consumption), which are both above the minimum acceptable limit.

3.2 Hardened State

Formulation decisions made to meet fresh state requirements affect the microstructure of concretes and can influence their properties in the hardened state. Maintaining the water/cement ratio is a common strategy aimed at preserving the porosity of the hardened material, thereby enhancing strength and durability. However, as observed in the previous step, introducing BR without adjusting the water can potentially affect the feasibility of concrete applications, necessitating modifications.

The effect of introducing BR on certain properties in the hardened state was assessed after a curing period of 28 days. Parameters such as compressive strength, flexural tensile strength, water absorption and abrasion resistance were measured and are detailed in Table 5.

In the sequence, Figure 4-left illustrates the results of compressive strength. Due to the variation in cement and water consumption (including that required for rheological adjustment), the analysis is carried out in relation to the final water/cement ratio (w/c). The reduction in strength as the w/c ratio increases is expected. However, the presence of BR resulted in a slight increase in strength under the same w/c ratio conditions. This suggests that BR, even in untreated condition, promotes an improved microstructure. Part of this improvement may be due to a slight pozzolanic effect (in the Chapelle test, a consumption of 453 mg of $Ca(OH)_2$ per gram of BR was obtained, indicating some pozzolanic reactivity). The enhancement in paste packing due to the introduction of finer particles, which also act as nucleation sites for hydration products or reduce micro-bleeding at the aggregate-paste interface are other mechanisms to be considered. The contribution of each of these mechanisms needs to be evaluated in a future complementary study.

Table 5. Concrete properties in the hardened state (28 day aged).

Cement (kg/m^3)	% BR	f_c (MPa) (1)	$f_{t,f}$ (MPa) (2)	w (%) (3)	Abrasion (mm)(4)
318	0	32.6	4.93	5.8	19.5
	10	30.6	4.89	5.0	20.8
	20	34.0	5.52	4.9	20.9
	30	37.6	6.56	4.7	21.3
374	0	43.0	5.13	4.7	20.2
	10	45.1	5.80	4.5	20.5
	20	44.8	7.42	4.4	20.8
	30	46.3	6.79	4.6	19.9
429	0	47.5	6.02	4.6	20.9
	10	52.2	6.45	4.0	20.2
	20	51.8	7.47	4.1	19.8
	30	47.2	6.70	4.8	20.3

(1) Compressive strength; (2) Tensile strength in third-point loading test; (3) Water absorption; (4) Abrasion wear width, according ABNT NBR 9781:2013

It is important to note the effect of rheological adjustment with water on strength. This effect was most evident in the concrete that required the greatest adjustment (30 % BR in concrete with 429 kg/m3 of cement). Extrapolating the curves, it would be possible to obtain a strength of approximately 55 MPa if the w/c ratio could be maintained at 0.48. However, the required addition of 19 l/m^3 of water resulted in an increase in w/c to 0.52 and a reduction in strength to 47 MPa.

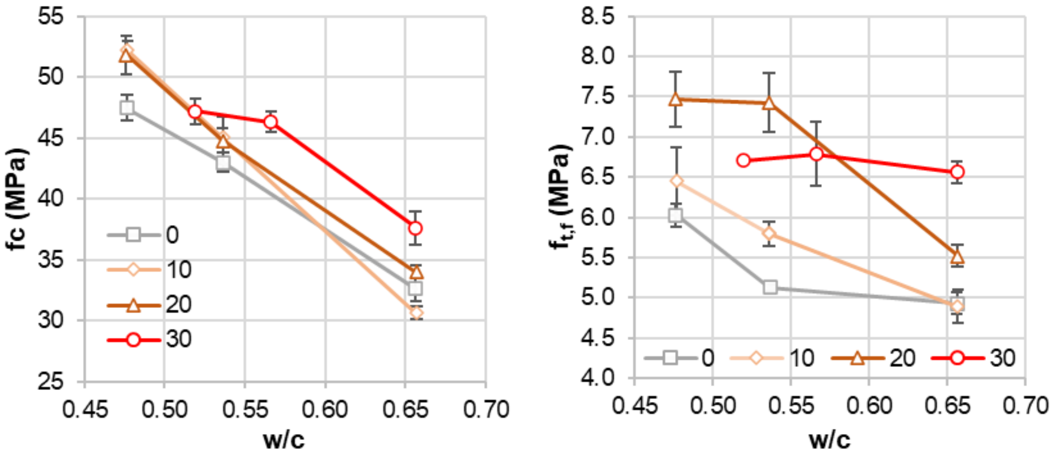

Figure 4. Mechanical properties versus water/cement ratio. Left: Compressive strength, Right: Tensile strength in flexural test. Labels represent BR content in % cement volume; error bars represent ± standard deviation.

Figure 4-right shows the flexural strength results analysed as a function of the w/c ratio. A surprising increase is observed with the introduction of BR. This effect is much more pronounced than the increase in compressive strength. It is possible that the increase in the paste volume with a higher solid concentration, contributed to the reduction of critical defects (possibly improving the paste-aggregate interface zone), leading to fractures at higher loads. It is also worth noting the negative effect on strength (concretes with 30% BR and lower w/c ratios) when it is necessary to increase water consumption to achieve the specified workability. It should be noted that a minimum strength of 4.5 MPa is generally specified for concrete pavements. The introduction of BR therefore makes it favourable to achieve adequate strength for this type of application.

Water absorption is a parameter related to durability, as it assesses the availability of microstructural porosity through which aggressive agents can penetrate. The results (Figure 5) show a reduction in water absorption as the BR and cement content increases. In the case of concretes that required an increase in water to adjust the workability (concretes with cement contents of 374 and 429 kg/m^3 and 30% BR content), there is again an increase in water absorption.

It is worth noting that for concrete paver flooring, according ABNT NBR 9781:2013, a water absorption of less than 6 % is considered suitable for use, and the reduction achieved with the introduction of BR can be considered favourable for the purpose of improved durability.

The wear obtained in the abrasion test, as illustrated in Figure 6 (with error bars representing a 90 % confidence interval), indicates little influence of cement and BR content. The Brazilian standard (ABNT NBR 9781:2013) which prescribes this test for interlocking paving units classifies the range between 20 and 23 mm as suitable for light traffic and below 20 mm for heavy traffic.

Both the cement paste and the aggregates are relevant in defining abrasion resistance. In the case of more porous pastes with lower resistance (concretes with 318 kg/m^3 of cement), a slight increase in wear is observed when replacing river sand (mainly quartz) by BR. However, in other concretes where the paste has a higher resistance, this effect is less pronounced and even reversed in the case of concrete with 429 kg/m^3 of cement. Nevertheless, given the variability of this test, as shown by the size of the error bars in the graph in Figure 6, it is not possible to state that the introduction of BR resulted in statistically relevant differences in the concretes tested.

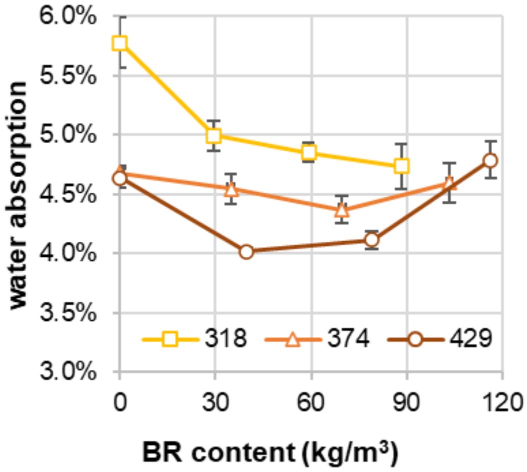

Figure 5. Water absorption versus BR content.
(labels represent cement consumption; error bars represent ± standard deviation)

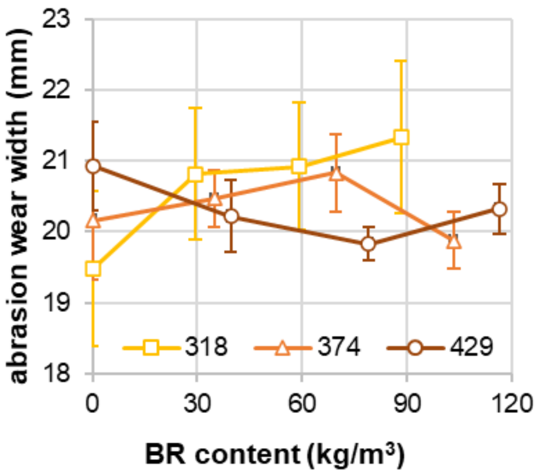

Figure 6. Abrasion wear versus BR content.
(labels represent cement consumption; error bars represent confidence interval of 90 %)

Figure 7 illustrates the colour of the concrete after 28 days of curing and superficial drying. The images refer to concrete moulded in contact with the formwork. Minor variations should not be considered as actual results, since precise lighting control was not conducted, and there is variability among test specimens. Even at lower BR contents, a change in colour from grey to reddish occurs. Colour is an issue that needs to be considered in applications. If it is not possible to incorporate the obtained colour into the architectural proposal, which varies according to the application type and even due to geographical-cultural reasons, its use should be restricted to non-apparent applications, where there will be subsequent finishing with another material to achieve the desired colour.

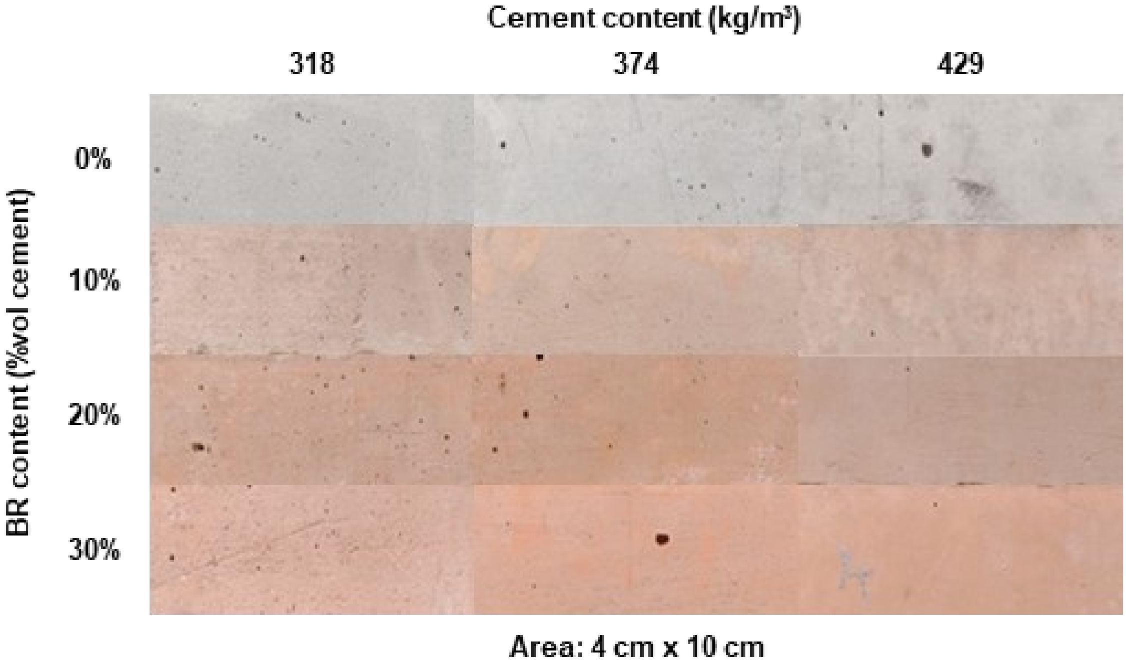

Figure 7. Colour evaluation, based on the cement and BR content.

3.3 Simplified Eco-efficiency Analysis

As an eco-efficiency indicator, binder index (bi) [10] was calculated according to Equation (1).

$$bi = \frac{B}{f} \tag{1}$$

Where

bi Binder intensity, $kg.m^{-3}.MPa^{-1}$

B Binder content, not including non-reactive fillers in cement, kg/m^3

f Concrete strength (compressive or tensile), MPa

In Figure 8-left, the efficiency of cement use in compressive strength is analysed in relation to the addition of BR. It is observed that for the same application (same strength), the introduction of BR provides a potential reduction in cement consumption (less cement is needed to achieve 1 MPa in compression).

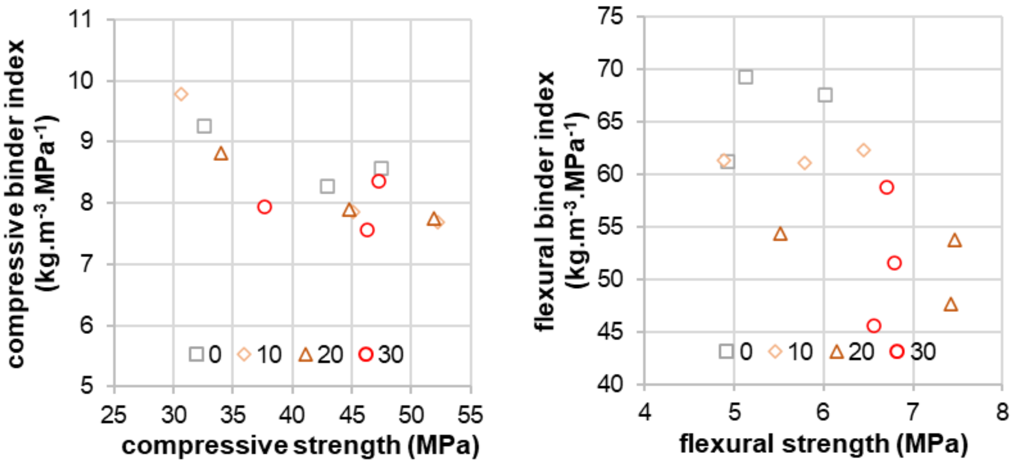

Figure 8. Binder index. Left: For compressive strength, Right: For flexural strength. (Labels represent BR content in % cement volume)

In applications where concrete is designed for flexural stress, such as paving, there is an even more explicit advantage in adding BR. As shown in Figure 8-right, there is a considerable reduction in the cement required to achieve 1 MPa of flexural tensile strength.

Since cement is the component with the highest CO_2 emissions in concrete, this reduction signifies a decrease in environmental impact regarding greenhouse gas emissions. Additionally, there is an environmental gain from the reuse of a material considered waste and a reduction in the use of a natural material (river sand), which also has environmental impacts associated with its extraction.

4. Conclusion

The study demonstrated the feasibility of developing concretes for different applications and strength levels (30 to 50 MPa) with the introduction of BR in significant quantities (up to 116 kg/m³ of concrete, equivalent to 5.4 % of total solid mass or 27 % of cement mass). Although the annual amount of BR generated is significant (estimated to be over 165 Mt in 2023), it is important to note that annual cement production reaches 4.2 Gt, meaning that BR production represents only about 4 % of this total. The main challenge will be to overcome local logistical barriers to find diversified applications. Therefore, higher BR content relative to cement and a variety of applications are strongly encouraged.

Regarding the various technical aspects evaluated, it can be concluded that:

- The mechanical performance obtained was at least equivalent, and in the case of flexural stress applications such as pavements, considerably superior to reference concretes without BR.
- There are indications of possible improvement in resistance to the ingress of aggressive agents, as evidenced by a reduction in water absorption (lower porosity).
- In applications involving surface abrasion processes, the effect is small and depends on the compressive strength. In lower-strength concretes, there tends to be a slight increase in abrasion, while in higher-strength concretes, the addition of BR tends to slightly reduce it.
- At low levels (up to about 60 kg/m³), the introduction of BR improved workability, potentially allowing for a reduction in water consumption. For higher BR contents, above

100 kg/m^3 of concrete, an increase in water content was necessary to adjust workability. The decision to make the adjustment with water resulted in impacts on properties in the hardened (aged) state, reducing strength and increasing water absorption. This highlights the need to consider applicability criteria (workability) when developing an application and seeking higher BR incorporations.

- The introduction of BR in concretes enables a reduction in environmental impact. Firstly, due to the possibility of reducing cement consumption, which reduces the CO_2 footprint of concrete, especially in applications designed for flexural stress. Secondly, by offering a destination for an industrial waste. And finally, by reducing the consumption of natural sand, requiring less extraction of this natural resource.

Regarding colour, even with lower BR contents, there's a shift from grey to reddish. If the obtained colour cannot be integrated into the architectural proposal, its use can be limited to non-visible applications, with subsequent finishing using another material for desired colour. But it is worth to remember that colour is a matter involving the type of application as well as regional and cultural considerations.

Lastly, it is necessary to consider that besides technical adequacy, environmental safety evaluations are still necessary, especially regarding leaching and solubilization of elements that may be harmful to the environment.

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