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Effective Contribution of Brazilian Rice Husk Silica (RHS) as Eco-Friendly Silica in Concrete Plants

Carlos Marmorato Gomes[1] and Antonio de Paulo Peruzzi[2]

1. School of Civil Engineering, Architecture and Urban Design, University of Campinas, Campinas 13083-970, Brazil

2. Faculty of Civil Engineering, University Federal of Uberlandia, Uberlandia 38408-100, Brazil

Abstract: Due to growing consumption of Portland cement and high levels of CO_2 emissions in its production process, this study examined the use of Brazilian RHS (rice husk silica) obtained by FBC (fluidized bed combustion) as an alternative material in concrete production; the principal cause is the inappropriate disposal of carbonized or *in natura* rice husk in southern Brazil. To analyze its feasibility, concretes made with several types of cements were examined combined with RHS, and also concretes with different strength classes employing a slag-based binder featuring a smaller amount of clinker in its composition. Mechanical and scanning electron microscopy assays were carried out to verify formation of C-S-H and calcium hydroxide in the cement pastes. This study concludes that replacing 10% of Portland cement (CEMIII/A) by 3% of RHS can result in a cost reduction of around 5% and allows a reduction of 4% in CO_{2eq} levels. In this way, RHS from renewable sources can be a highly impactful sustainable alternative in civil engineering work, allowing the concrete industry to produce environmentally-sound concrete mixtures with lower CO_2 emissions.

Key words: Sustainable concrete, eco-friendly silica, RHS, cement, geopolymer.

1. Introduction

Rice husk has become a serious environmental problem due to increasing agricultural production and the amount of methane gas generated by incorrect disposal of this waste [1, 2]. According to the literature, for every metric ton of rice around 200 kg of *in natura* husk is produced [3, 4]. These figures become even more critical when we consider Brazilian production, especially in the state of Rio Grande do Sul, which is around 8 million metric tons per year. Production of RHS (rice husk silica) requires the burning of the husk to be performed in controlled conditions. In Brazil, an innovative system of FBC (fluidized bed combustion) has been very efficient in the production of this pozzolana. FBC not only improves combustion efficiency, but also allows extraction of silica from rice husk with consistent quality, high degree of amorphicity and consequentially increased pozzolanacity, allowing its use in concrete and mortar dosing, to complement or partially replace cement. Besides, Portland cement consumption is currently high throughout the country. Carbon dioxide emissions in clinker production are also elevated. As shown in Fig. 1, the major Portland cement consumers are China, with 2,229 Mt (million of tonnes); followed by India, 237 Mt; USA, 78 Mt; Brazil, 69 Mt; and Russia 65 Mt. In addition, data from 2012 show China as the biggest per capita consumer, to the sum of 1,650 kg/inhab, while the USA had a consumption of 245 kg/inhab and Brazil 348 kg/inhab. It is estimated that every metric ton of clinker produced releases up to one metric ton of CO_2 to the atmosphere. Due to additions made by the cement industry, the Brazilian average is about 650 kg of CO_2 per metric ton of cement. This average has remained virtually constant throughout recent years (Fig. 2).

Concrete production in the current environmental scenario requires measures to lower consumption of raw materials, while maintaining desired performance parameters. In this sense, addition or partial replacement of Portland cement with agri-industrial residues, such

Corresponding author: Antonio de Paulo Peruzzi, Ph.D., professor, research fields: industrialized construction systems, construction materials, geopolymers and fiber concretes

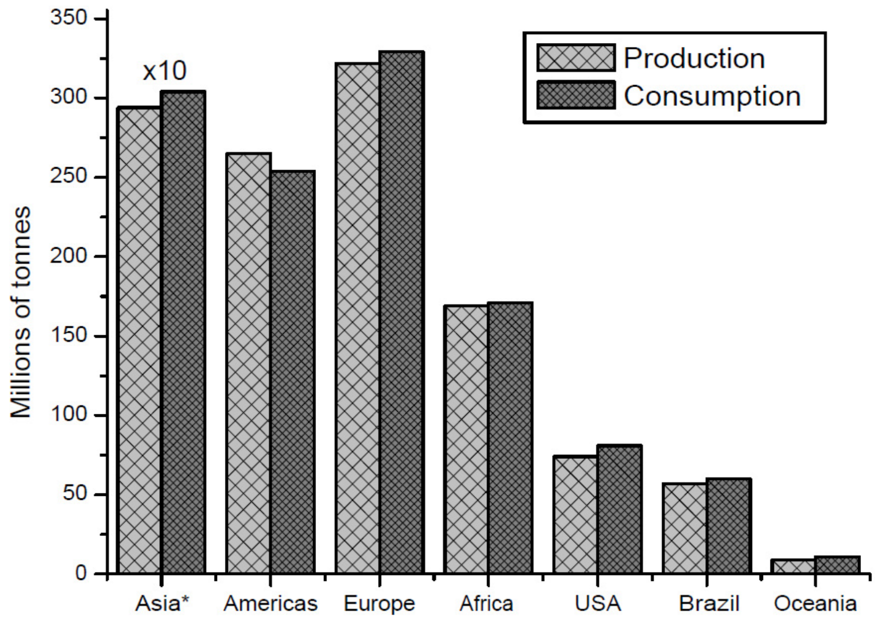

Fig. 1 Principal producers and cement consumers.

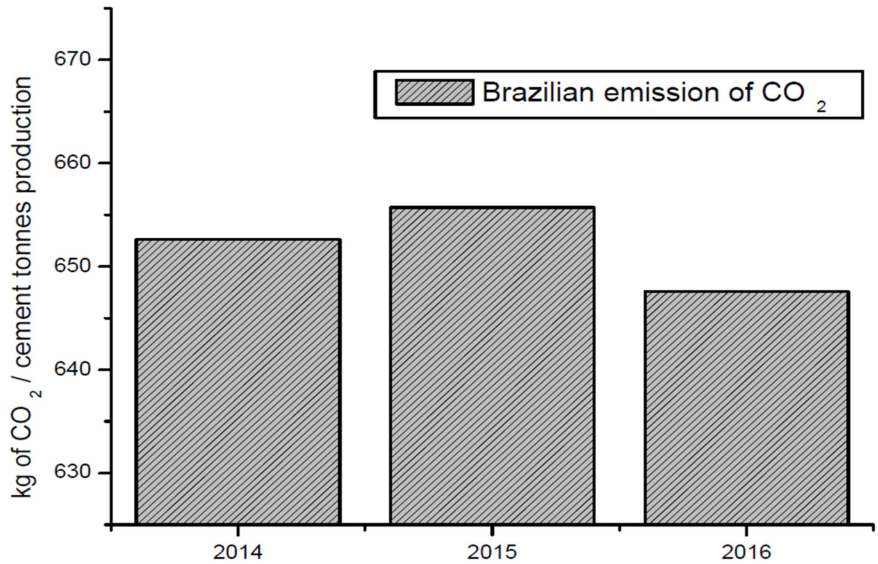

Fig. 2 Brazilian CO: emission at cement industry.

as RHS, emerges as a viable method to produce more sustainable, economical, sturdy and durable concretes.

2. RHS

Research of rice husk ash as a pozzolanic material is not new and has been going on for over 40 years [5]. This technology received an impulse from the development of HPC (high-performance concrete) and the employment of silica derived from the fabrication of iron-silicon alloys and metallic silicon. Rice husk combustion through the fluidized bed process has been regarded as a technological innovation, resulting in amorphous silica (Fig. 3) compatible with the needs of the construction industry.

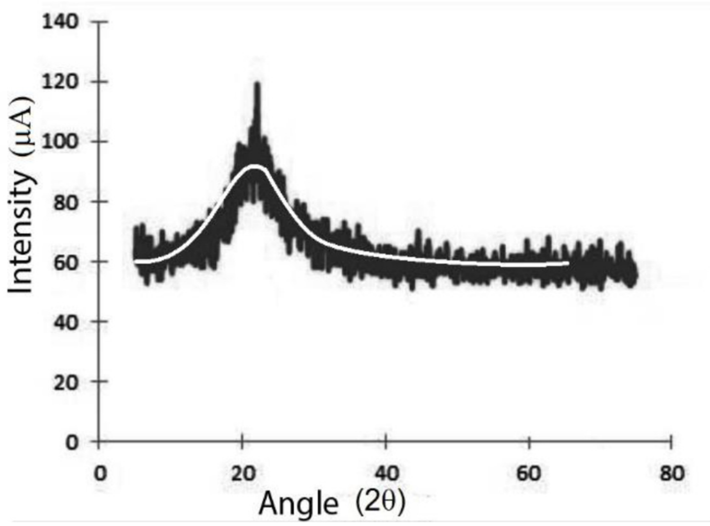

Fig. 3 X-ray diffraction of RHS.

Addition of RHS to Portland cement influences its properties in both fresh and hardened states. In fresh state cement, RHS reduces segregation and exudation. In hardened state cement, small additions between 3% and 8% provide increased mechanical strength, lower permeability, especially against aggressive agents (sulfates, chlorides, CO_2 etc.) [5], and greater durability, the latter a fundamental sustainability parameter. As the literature shows, it is even possible to replace much of the ligand (replacements of up to 70% total mass have been reported). Although the final product may sometimes have a slightly higher cost than Portland cement, due to the high pozzolanacity of silica, a lower cost/compressive strength ratio can be obtained by fine-tuning ligand amounts. Of course, for definition of the optimal cost vs. benefit curve, it is important to consider the attributes of materials, such as aggregates and cement type. Higher mechanical resistance and lower permeability both result from characteristics of RHS, such as increased yield of calcium silicate hydrates (C-S-H) and greater Portlandite (CH) consumption.

3. Sustainability Context of the Research

Electrical energy shortages and the need to compound new energy sources to the current energy matrix has increased burning of rice husk by thermal power plants, especially in the southern region of Brazil. Biomass can be used not only to increase energy supply, but also to decentralize the energy supply system. Additionally, partial substitution of energy generated from fossil fuels (coal and natural gas) results in a proven decrease of greenhouse gas total emission. The use of biomass makes environmental sense, because CO_2 released through burning is offset by photosynthesis, thus maintaining a certain balance. According to the literature, there are several ways to convert biomass into energy. Direct combustion, gasification and pyrolysis are the most used.

RHS used as an addition to or partial substitute for Portland cement can be considered of great sustainability impact, since it can be beneficial not only to alternative energy generation, but also to decreased Portland cement consumption and increased durability of concretes and mortars. Specific sustainability parameters are shown ahead during the discussion of the results. In fact, the cement industry has been seeking ways to reduce CO_2 emissions during the production of clinker by its increasing use of mineral additions. Besides it, another noteworthy environmental action is the co-processing of cement industry waste and consequent decrease in consumption of fossil fuels. Regardless, the construction industry should pursue better performance and durability of concretes, and the adoption of new concepts about materials. As such, this study employed RHS with a view not only to the optimization of Portland cement levels, but also to the production of sustainable concretes. The experimental procedure had two stages. In the first stage (A), a reference mix made with approximately 350 kg/m^3 cement was used to verify possible mechanical strength gains in concretes made with different types of Portland cement and 10% RHS.

In the second stage (B) the Portland cement with the lowest amount of clinker (CP III-40) was selected, which allowed for the most reduction in the CO_2/kg ratio. Thus, in stage B concretes with the following mechanical resistance classes were produced: f_{ck} 18, f_{ck} 25, f_{ck} 30 and f_{ck} 40 (MPa). A maximum slump about 10 cm (1 h of the test duration) was adopted, in accordance to common specifications of the cement industry (due to the logistics involved in the delivery of the material).

4. Materials and Methods

The materials used in the present study are:

(a) Portland cements classified in conformity to Brazilian regulations as CP II-E-32 (3.01g/cm^3), CP III-40 (2.99g/cm^3), CP IV-32 (2.86g/cm^3) and CP V-ARI (3.13g/cm^3). Table 1 shows similar cements according to International Standards.

(b) RHS with SiO_2 amorphous up to 93.2%, Pozzolanic activity up to 1,300 mg $Ca(OH)_2$/g, surface area about 21,150 m^2/kg, average diameter about 14 µm, density about 2.17 g/m^3, particle size distribution according to Fig. 4, and morphology verified by SEM (scanning electron microscopy) illustrated in Fig. 5.

(c) Quartzite natural fine aggregate with 4.8 mm NMS (nominal maximum size), 2.80 fineness modulus, 2.66 g/cm^3 of density (measurements conforming to Brazilian standards NBR 7211 and NBR 9776).

(d) Limestone artificial fine aggregate with 1.2 mm NMS, 1.36 of fineness modulus, 2.65 g/cm^3 density (measurements conforming to Brazilian standards NBR 7211 and NBR 9776).

(e) Basaltic coarse aggregate with 25 mm NMS, 6.91 of fineness modulus; 2.76g/cm^3 of density (measurements conforming to Brazilian standards NBR 7211 and NBR 9776).

(f) SP (Superplasticizer) admixture, employed as a water reducer, polycarboxylate-based; 0.6% (percent by mass of cement) dosed to all mixtures.

Table 1 Cement types applied.

CP II-E-32	CP III-40	CP IV-32	CP V-ARI
CEM II/B-S (EN)	CEM III/A (EN)	CEM II/B-Q (EN)	Type III (ASTM)

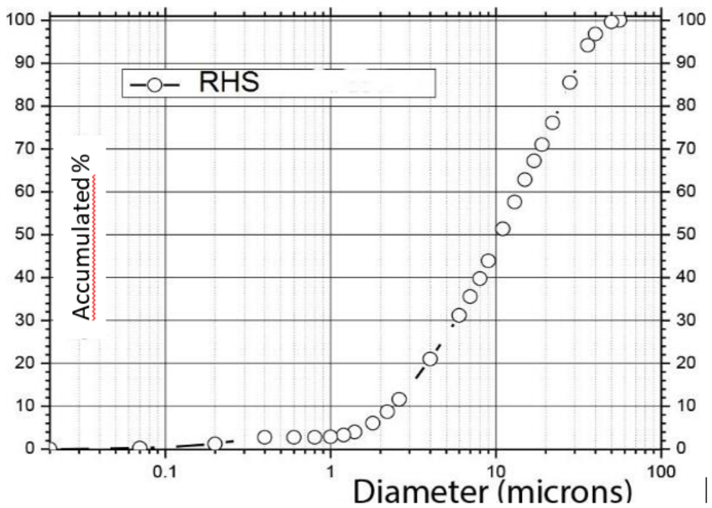

Fig. 4 RHS particle size distribution.

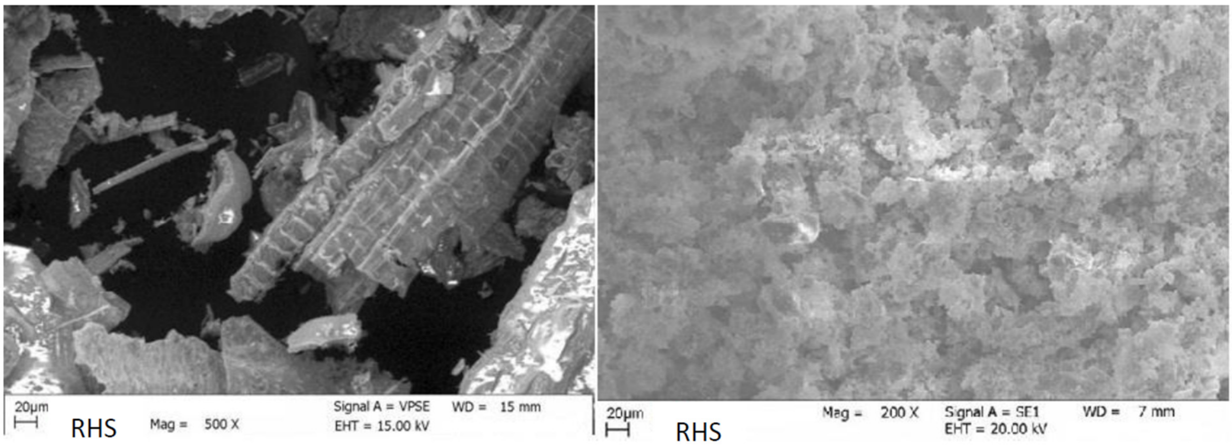

Fig. 5 RHS SEM images.

4.1 Stage A

Concrete mixtures analyzed during the Stage A had focus to verify the RHS efficiency towards many types of cements commonly found in the Brazilian market, then the types of cement were tested: CP II-E-32, CP III-40, CP IV-32 and CP V-ARI. For each type of cement studied, we made two kinds of samples of cements: "Reference" and "RHS" and Table 2 presents its formulations.

For each mixture, 8 cylindrical specimens (10 cm × 20 cm) were molded, 4 for compressive strength tests and 4 for tensile strength tests and were tested 28 days after molding and wet-cured at 25 °C and 95% of relative humidity.

4.2 Stage B

In the Stage B, one type of Portland cement was fixed; CPIII-40 specific mixtures were analyzed just with Portland cement type CP III-40 and different proportions of concrete (A, B, C and D) were mixed, resulting in different classes of resistance (18, 25, 30 and 40 MPa). The concrete formulations of this stage are presented in Table 3. CP III-40 was chosen specially because it has minimum clinker content compared to other types.

In sequence, mixes were obtained by replacing 10% cement with 3% silica (by mass) to assess the efficiency of substituting RHS instead of Portland cement, resulting in what can be called "alternative concretes" (A1, B1, C1 and D1). Table 4 shows estimates of RHS and Portland cement consumption per concrete cubic meter in the alternative concretes.

4.3 Maintenance of Fresh State

Concrete consistency in fresh state was verified according to ASTM C143 by Slump test. It was admitted a difference about 10 cm after 1 h in it consistence, if necessary water was added. Table 5 shows final water/cement and water/(cement + RHS) ratios.

Table 2 Concrete dosage (by mass) to all cement types (CP II-E-32, CP III-40, CP IV-32, CP V-ARI).

Type of concrete	Cement	Fine aggregate	Coarse aggregate	RHS	w/c (water/cement)
Reference	1	2.15	3.15	-	0.60
RHS	1	2.15	3.15	10%	0.60

Table 3 Concrete dosage (by mass) to all cement types (CP II-E-32, CP III-40, CP IV-32, CP V-ARI).

Type of concrete	Proportion	f_{ck} (MPa)	Estimated cement consumption
A	1:1.15:0.98:2.67:0.48	40	400 kg/m^3
B	1:1.67:1.41:3.29:0.59	30	312 kg/m^3
C	1:2.13:1.81:3.73:0.70	25	264 kg/m^3
D	1:2.78:2.34:4.35:0.85	18	218 kg/m^3

Table 4 Alternative concretes and the reduction of Portland cement content and RHS consumption.

	Estimated cement consumption	Estimated silica consumption
A1	360.0 kg/m^3	12.0 kg/m^3
B1	280.8 kg/m^3	9.4 kg/m^3
C1	237.6 kg/m^3	7.9 kg/m^3
D1	196.2 kg/m^3	6.5 kg/m^3

Table 5 Maintenance of the unhardened concrete consistency.

Mixture	w/c initial		Slump (cm)		Additional water	w/c final		Final slump (cm)
	Cement	Cement + RHS	Initial	Next 60 min		Cement	Cement + RHS	
A1	0.48	0.46	9.0	5.0	Yes	0.51	0.49	9.0
B1	0.59	0.57	9.5	7.0	Yes	0.62	0.60	8.0
C1	0.70	0.67	15.0	11.0	No	0.70	0.67	11.0
D1	0.85	0.83	10.0	8.5	No	0.87	0.83	8.5

5. Results and Discussion

5.1 Stage A

Table 6 compares consistencies via slump test of reference cements and RHA-added cements (measured during stage A). There is loss of consistency due to the greater surface area of the RHS, except in the CP V ARI mixture, which has the highest Blaine value of all employed cements (above 300 m^2 per kg of cement). During this stage there was no attempt to maintain consistency through addition of water or plasticizer.

Fig. 6 presents compressive strength and diametral tensile strength results at 28 days, for each employed cement. It should be noted that, according to Brazilian regulations, clinker contents can range from 56% to 94% for CP II-E-32, 25%-65% for CP III-E-40, 45%-85% for CP IV-32 and 95%-100% for CP V ARI. When comparing mixtures A (reference) and B (addition of 10% RHS over the cement mass), the below mechanical increments were observed for each type of cement. Axial compressive strength increments: CP II—20%; CP III—10%; CP IV—27%; CP V-ARI—18%, and diametral tensile strength results: CP II—27%; CP III—17%; CP IV—29%; CP V-ARI—28%. The results obtained in all the formulations containing silica from rice husk conformed its effectiveness as pozzolanic material, revealing significant macrostructural improvements at 28 days. This is in conformity with data via the modified Chapelle method, showing a 1,379.37 mg consumption of $Ca(OH)_2$/g of RHS.

SEM of the 10% RHA pastes shows besides formation of the main constituents of Portland cement, such as C-S-H and ettringite great availability of $Ca(OH)_2$. This is congruent with the literature and indicates it is possible to employ levels of silica above the 10% practiced in this study (Fig. 7).

5.2 Stage B

Thus, in stage B concretes with the following mechanical resistance classes were produced: f_{ck} 18, f_{ck} 25, f_{ck} 30 and f_{ck} 40 MPa. A maximum slump of 10 cm (test duration: 1 h) was adopted, in accordance to common specifications of the cement industry (due to the logistics involved in the delivery of the material). Fig. 8 shows the mechanical strength gains for alternative mixtures 1, 7, 14 and 28 days old, obtained via axial compression assays. As shown in Fig. 9, for all resistance classes and concretes the addition of RHA resulted in better performance. In addition to greater mechanical resistance, addition of RHA resulted in increased modulus of elasticity and better porosity. This occurs due to the greater compactness of the concretes, especially through higher volume of C-S-H and reduction of open porosity, important factors in obtaining higher performance in comparison to reference concretes, as evidenced by microscopy images.

Table 6 Consistency of unhardened concrete—Slump test (cm).

Cement type	Reference	RHA
CP II-E-32	7.0	4.0
CP III-40	3.0	3.0
CP IV-32	3.5	2.0
CP V ARI	0.5	1.0

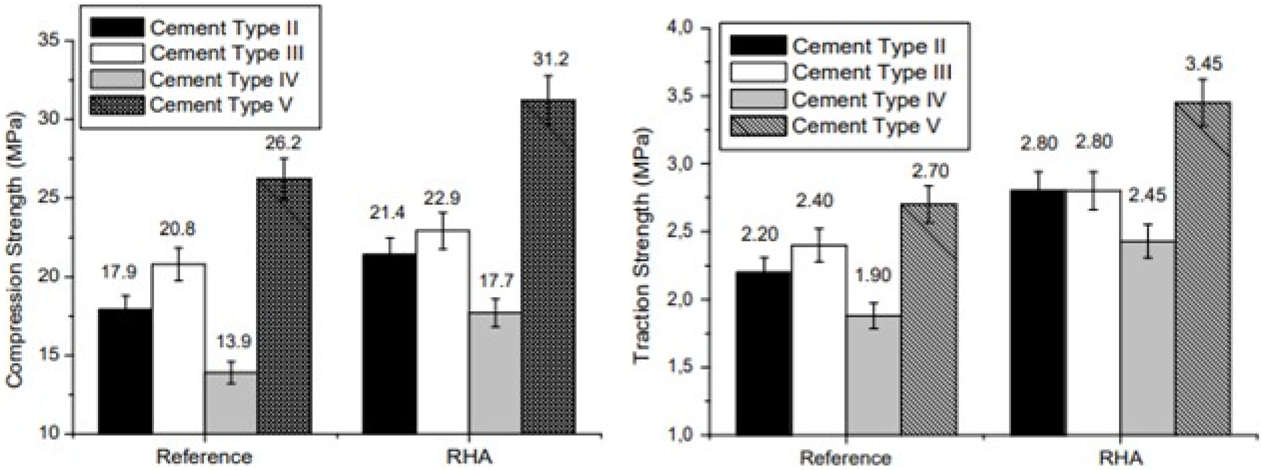

Fig. 6 Compressive strength and diametral compressive strength of samples at 28 d.

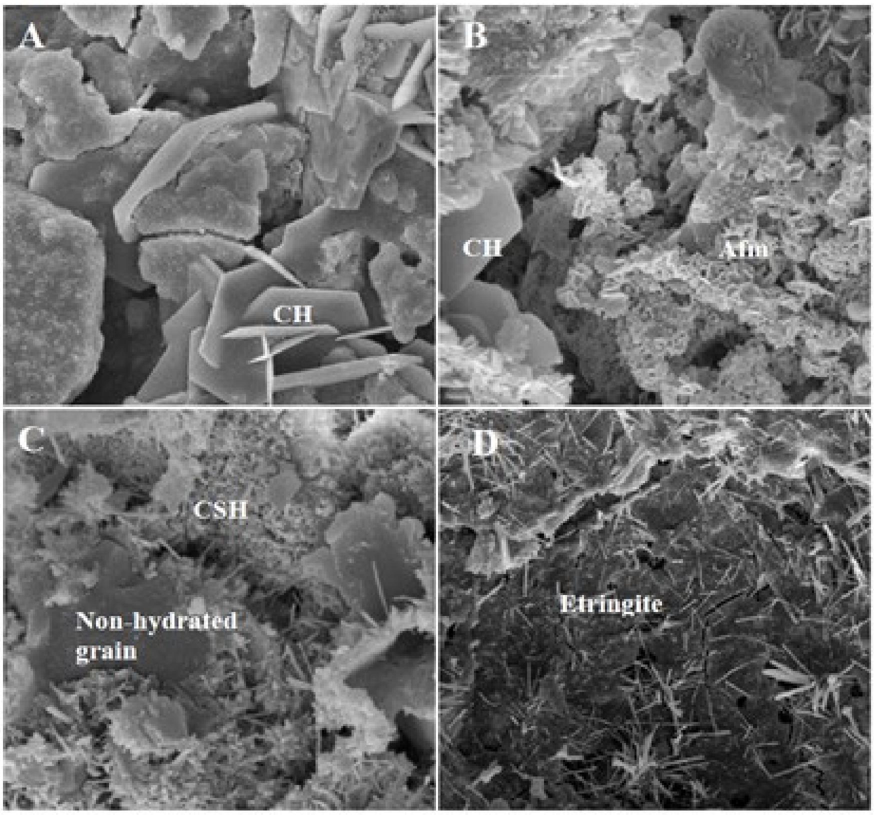

Fig. 7 SEM images of samples: (a) CP II magnification 5,000×; (b) CP III magnification 10,000×; (c) CP IV magnification 10,000×; (d) CP V ARI magnification 3,000×.

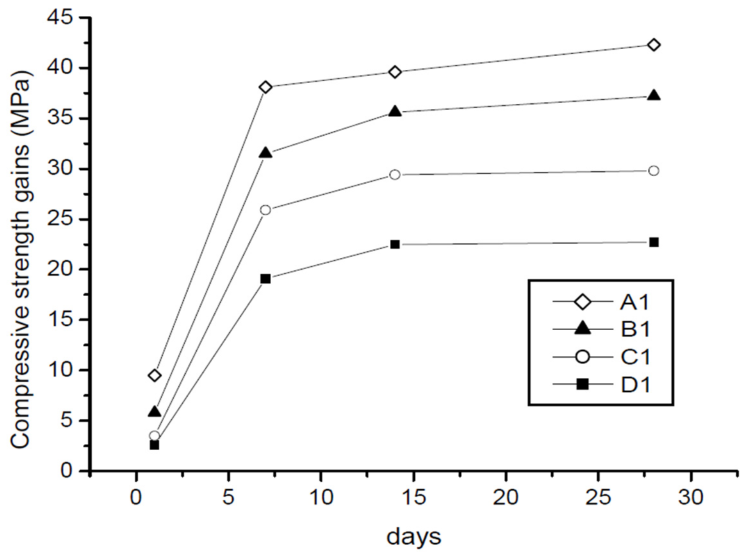

Fig. 8 Compressive strength evolution.

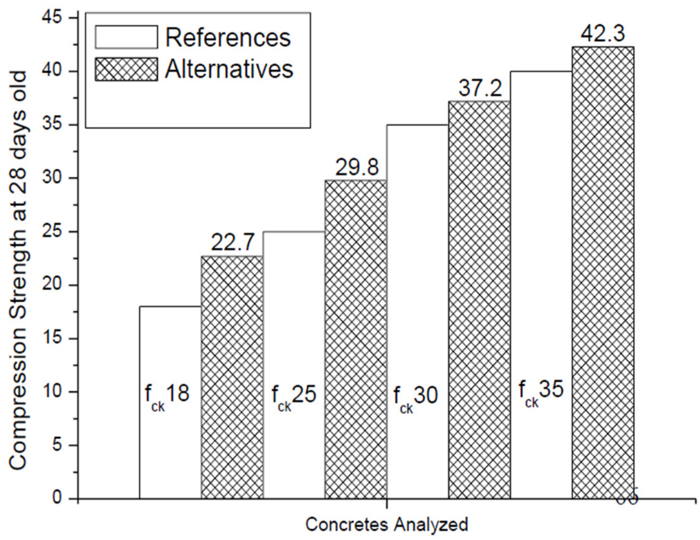

Fig. 9 Compressive strength comparation.

Microscopy images reveal the presence of $Ca(OH)_2$, indicating the possibility of employing higher contents of RHS (Fig. 10).

Results demonstrate the efficiency of RHS as pozzolan, evidenced by the significant mechanical resistance increase of the alternative concrete mixtures. In comparisons between reference and alternative mixtures, in Fig. 9 in particular, there is an increase in axial compressive strength of approximately 15%, for all mixtures. Considering RHS has around two times the cost of Portland cement, adoption of the proposed levels allows for a final cost reduction of approximately 5%. According to the literature, each tonne of rice generates around 200 kg of *in natura* husk. When burned in boilers or furnaces, this amount can produce approximately 40 kg of silica. Estimated reductions in CO_2 emission rates are shown in Table 7 below. For this assessment, the amount of methane (CH_4) a ton of rice deposited in landfill can release into the atmosphere was estimated to be 119.2 kg. Knowing the greenhouse

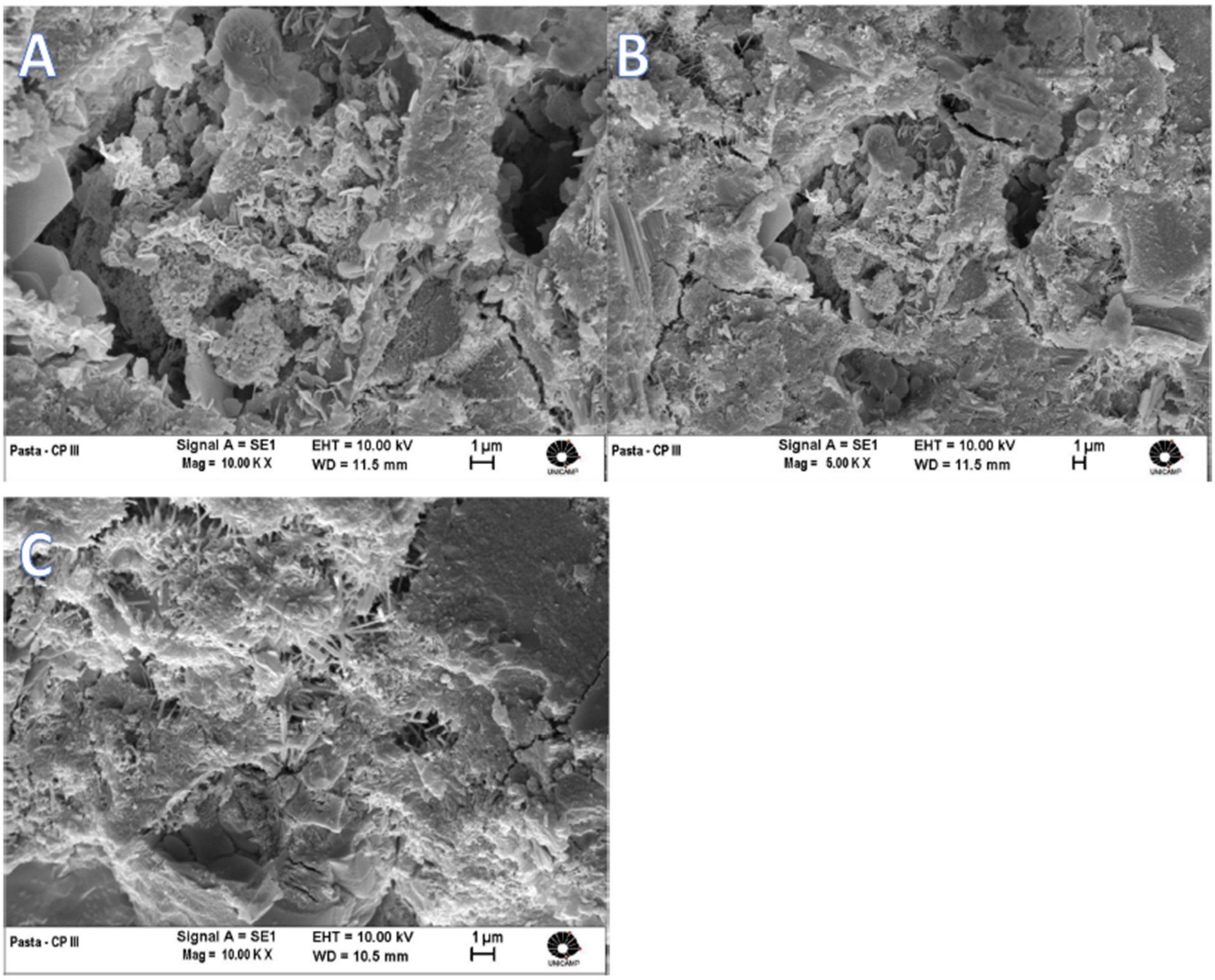

Fig. 10 SEM images of the CP II + RHA paste.

Table 7 Effective contribution of Brazilian RHS as eco-friendly silica.

References concretes	Estimated cement consumption kg/m^3	Reference binders kg CO_{2eq}/m^3	
(A)	400.0	180.0	
(B)	312.0	140.4	
(C)	264.0	118.8	
(D)	218.3	93.1	
Alternatives concretes	Estimated cement consumption kg/m^3	Estimated silica consumption kg/m^3	Alternative binders kg CO_{2eq}/m^3
(A1)	360.0	12.0	180 - 7.2 = 172.8
(B1)	280.8	9.4	140.4 - 5.6 = 134.8
(C1)	237.6	7.9	118.8 - 4.7 = 114.1
(D1)	196.2	6.5	93.1 - 3.8 = 89.3

effect of methane gas is approximately 21 times stronger than that of carbon dioxide, a rate of 2,503.2 kg CO_{2eq} per metric ton of composted husk was calculated. Also the 50% average clinker content of CP III-40 was considered. According to Brazilian regulations, however, this value can be as low as 31%.

6. Conclusions

RHS obtained via FBC is an excellent alternative to active silica derived from iron-silicon or metallic silicon manufacturing, especially in a large productive center such as Brazil. RHS allows for a reduction in binder consumption and optimization of concrete mixtures, resulting in mechanical resistance gains with no adverse effect on workability. As such, replacement of cement with RHS is an excellent alternative for concrete mixing factories in the production of economical and sustainable concretes. Replacing 10% of Portland cement type III with 3% RHA can result in a cost reduction of around 5%, and allow a reduction of 4% in CO_{2eq} levels, pointing to RHA as a feasible alternative for sustainability in the civil engineering sector.

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