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ORIGINAL ARTICLE

Compressive strength and environmental performance of blended cements with waste marble dust

Resistência à compressão e desempenho ambiental de cimentos com resíduo de serragem de mármore.

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Abstract: Replacing limestone filler by waste marble dust is one of the paths to explore, at a regional level, to stimulate circularity and reduce environmental burdens of ornamental stone processing. This study investigated six blended cements which combined supplementary cementitious materials (SCMs) and limestone filler or waste marble dust, with a clinker factor of 50%. Compressive strength was determined at 3, 7, 28 and 91 days and evaluated based on the criteria standardized in ABNT NBR 16697:2018. The cementing efficiency at 7 and 28 days of the set of mineral admixtures used and greenhouse gas (GHG) emissions intensity of each cement were also calculated. The synergy between the combinations reduced the clinker factor and cement GHG emissions, whilst preserving the mechanical performance required by the construction market.

Keywords: waste marble dust, limestone filler, supplementary cementitious materials, cementing efficiency, compressive strength and greenhouse gas emissions intensity.

Resumo: A substituição de filer calcário por resíduo de serragem de mármore é uma das alternativas, em escala regional, para estimular a circularidade e reduzir os impactos ambientais do beneficiamento de rochas ornamentais. Este artigo investigou seis cimentos compostos a partir da combinação de materiais cimentícios suplementares, filer calcário ou resíduo de serragem de mármore, com um fator clínquer de 50%. A resistência à compressão foi avaliada nas idades de 3, 7, 28 e 91 dias, com base nos critérios da ABNT NBR 16697:2018. A eficiência cimentícia foi determinada aos 7 e 28 dias de idade e a intensidade de emissões de gases de efeito estufa (GEE) de cada cimento também foi calculada. A sinergia entre as adições minerais possibilitou a redução do fator clínquer e das emissões de gases de efeito estufa dos cimentos estudados, ao mesmo tempo em que assegurou o atendimento de exigências de desempenho mecânico do mercado de construção.

Palavras-chave: resíduo de serragem de mármore, filer calcário, materiais cimentícios suplementares, eficiência cimentícia, resistência à compressão e intensidade de emissões de gases de efeito estufa.

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1 INTRODUCTION

The Brazilian ornamental stone sector has grown intensively since 1989, reaching the production of 154.5 million tonnes in 2019 [1]. Stone extraction activities have been traditionally concentrated in the states of Espírito Santo and Minas Gerais. About 40% of the mining output from the 1,500 fronts currently active [2] originates in Espírito Santo, mostly from the municipalities of Barra de São Francisco, Colatina, Ecoporanga and Nova Venécia, in the North of the state, and from Alegre, Cachoeiro do Itapemirim and Castelo, in the South [3].

Currently, the greatest challenge faced by the sector is the rational use of waste from stone mining and processing operations. The waste generated during sawing of stone blocks is approximately 40% of the block volume, which averages $10m^3$. Of this total, 26% is very fine waste [4]. Approximately 1.5 Mt of fine waste (rock dust) and 1 Mt of coarse waste are generated yearly in Brazil [5].

With technological advances and the global alignment for reducing environmental impacts of industrial activities, recycling efforts to divert stone waste from landfills have come to the forefront. Environmental policies have also encouraged replacing conventional sawing machines in the cutting units by multiwire diamond ones, favoring more uniform composition and size of ornamental stone waste. Among the feasible applications of stone dust waste, the production of crushed stone, sand and cement stand out [6], [7].

Given the major challenges faced in terms of energy demand [8], non-renewable raw materials consumption and greenhouse gas (GHG) emissions, the cement industry has explored environmental impacts mitigation strategies to comply with internationally agreed targets, such as those of the Paris Agreement [9], whilst meeting the demand for cement in the market. Among all the alternatives, partial substitution of clinker by mineral additions was highlighted in the Brazilian Cement Technology Roadmap [10], for its potential for a 69% reduction in cumulative carbon emissions by 2050. The use of limestone filler and supplementary cementitious materials (SCMs) in cement is practiced and already standardized worldwide by, for example, EN 197-1:2011 [11] in Europe, ABNT NBR 16697:2018 [12] in Brazil and ASTM C 595:2019 [13] in the United States.

Alternative carbonate materials have also been investigated as substitutes for limestone filler to reduce the environmental impacts of cement [14]. The limestone filler needs to be extracted, crushed and finely ground, with energy consumption related to those processes. Contrastingly, waste marble dust only embodies the recycling (basically, drying and clod breaking) and transportation impacts to enable its use as material input [15]. Hence, using waste marble dust to replace limestone filler can be explored at a regional level by the cement industry.

Waste marble dust can replace up to 15% of the clinker mass without compromising physical, chemical, and mechanical properties [16]. If waste marble dust and calcined clay are combined, the replacement fraction can increase to 45% [17], due to the chemical reactions enabled. The pozzolanic reaction between calcined clay and portlandite forms C-A-S-H [18]. The alumina in the calcined clay reacts with the carbonate supplied by the waste. The hydrated carboaluminate precipitates and increases the volume of the hydrated phase and matrix density, with mechanical strength benefits [19], [20].

The pozzolanicity of calcined clays is influenced by its kaolinite content and appropriate calcination conditions [21]. Clays with the highest kaolinite contents and calcined at the highest temperatures result in cements with increased mechanical strength. To maximize the pozzolanicity of calcined clays, Avet and Scrivener [22] recommend clays with at least 40% kaolinite content and a calcination temperature between 600 and 800°C. Blast furnace slag, another alumina-rich SCM, can also be combined with waste marble dust to explore synergistic effects on mechanical strength [23]. While the physical and chemical characteristics of the waste marble dust favor hydration at early ages, the blast furnace slag hydrates slowly [23] and provides alumina to form hydrated carboaluminate [24].

The Brazilian Cement Technology Roadmap [10] points out the trend of reduced availability of blast furnace slag and fly ash, which would probably drive the increased use of limestone filler and, to a lesser extent, calcined clay. Thus, given the regional issues described and the need for optimized cement formulations, this research investigates the synergistic effect between SCMs and limestone filler or waste marble dust. For each blended cement, the compressive strength (3, 7, 28 and 91 days) and the cementing efficiency - k-value (3 and 28 days) of the set of SCMs and limestone filler or waste marble dust were determined. The life cycle assessment of each blended cement estimated the corresponding GHG emissions (in CO_{2eq}), which were related to the compressive strength unit for calculating the GHG emission intensity.

2 MATERIALS AND METHODS

2.1 Materials

The materials used were Portland cement with High Initial Strength (CP V-ARI), equivalent to CEM I 42.5R of EN 197-1:2011 [11], blast furnace slag, calcined clay, limestone filler and waste marble dust. The marble waste used results from cutting in a multiwire diamond sawing machine and oven-dried at 100ºC ± 5ºC for 24 hours. The CP V-ARI cement is composed of approximately 89% clinker, 6% limestone filler and 5% gypsum, according with the cement manufacturer. The calcined clay was ground for 5h30min in the laboratory, using a ball mill with a 50 kg-load. The blast furnace slag and the limestone filler were ground in a cement plant until reaching the granulometry used in its production process.

Figure 1 shows the particle size distribution, determined by laser granulometry in CILAS equipment (*Compagnie Industrielle des Lasers*), model 1090 LD wet method. Table 1 shows the chemical composition, determined by X-Ray Fluorescence (XRF) in *NexGo–Rigaku* using pressed inserts and the main physical characteristics of the CP V-ARI cement (equivalent to CEM I 42.5R), calcined clay, blast furnace slag, limestone filler, and waste marble dust used.

The main constituents of limestone filler and waste marble dust are calcium and magnesium oxides. These carbonate fillers have a high fire loss, as carbonate decarbonation occurs at a temperature of approximately 700 °C [25].

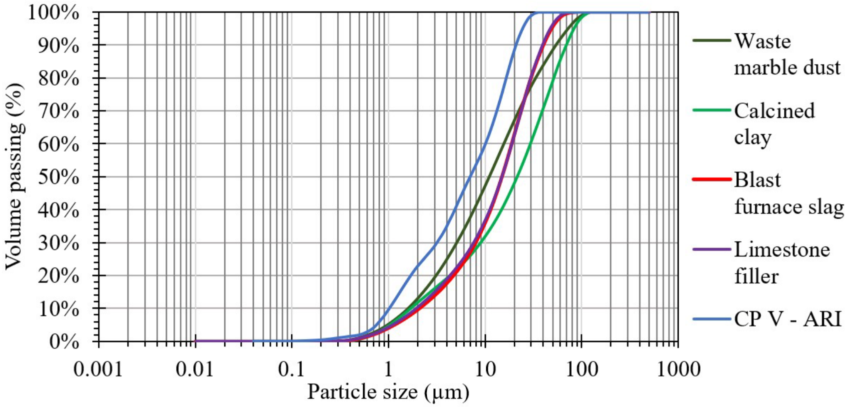

Figure 1. Particle size distribution of the waste marble dust, calcined clay, blast furnace slag, limestone filler and CP V-ARI cement.

Table 1. Chemical composition and physical characteristics of the CP V-ARI cement, calcined clay, blast furnace slag, limestone filler and waste marble dust.

	CP V-ARI	**Calcined clay**	**Blast furnace slag**	**Limestone filler**	**Waste marble dust**
SiO_2 (%)	18.1	59.30	37.90	1.50	8.22
Al_2O_3 (%)	4.68	31.90	10.50	0.33	1.52
Fe_2O_3 (%)	3.29	4.00	0.40	-	0.50
CaO (%)	58.0	1.20	48.8	47.50	47.20
MgO (%)	2.66	-	7.37	5.92	21.8
Na_2O (%)	-	-	0.10	-	-
K_2O (%)	0.41	0.31	0.41	0.08	-
SO_3 (%)	4.12	0.83	-	0.04	0.38
LOI	3.74	2.46	-	34.21	37.31
D10 (µm)	0.97	2	2	2	1.6
D50 (µm)	6.72	25	15	15	12
D90 (µm)	22.99	80	40	40	52
Blaine Fineness (m^2/kg)	485.78	1483.11	403.43	380.72	540.77

Figure 2 shows the minerals present in limestone filler, waste marble dust and blast furnace slag from X-ray diffraction with copper radiation.

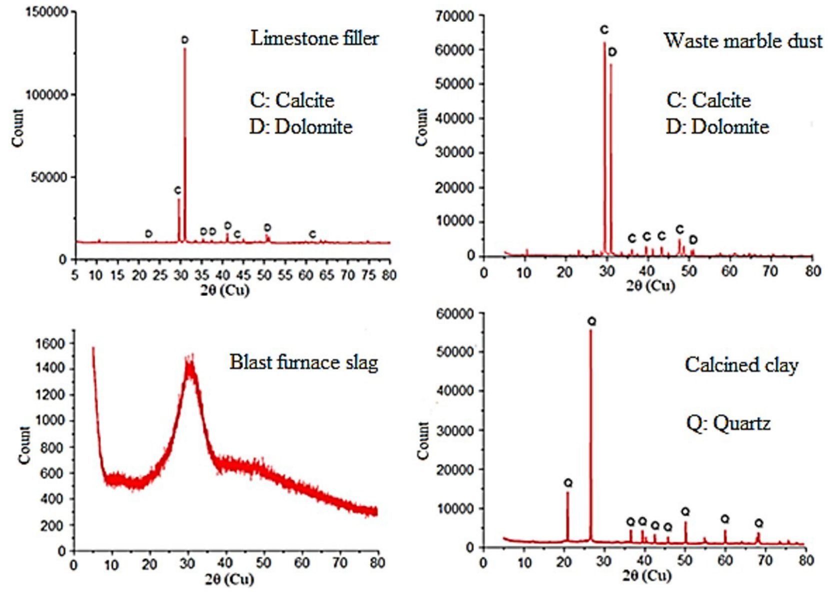

Figure 2. XRD diffraction results of the limestone filler, waste marble dust, blast furnace slag and calcined clay.

The CP V-ARI cement (2.66%), the SCM (blast furnace slag, 7.37%) and the carbonate fillers (limestone filer, 5.92%; waste marble dust, 21.8%) used in this research all contain MgO. The dolomite in the waste marble dust renders it unsuitable for clinker production, as the dolomite would decompose in the rotary kiln to form periclase (MgO), which would - later in cement hydration – undergo an expansive reaction to generate brucite ($Mg(OH)_2$). However, the marble waste can be used as a partial replacement of clinker in cement [16], [17]. In this way, the EN 197-1:2011 [11] only limits the MgO content in clinker and there is no limitation of MgO in blended cements. ABNT NBR 16697:2018 [12] follows the same logic and imposes no MgO limitations for blended cements, such as CP II-E, CP II-F, CP II-Z, CP III and CP IV.

According to the Brazilian cement manufacturer that supplied the calcined clay for this research, its kaolinite content is around 32% and the calcination temperature is between 450 and 500°C. Although these values are below those recommended by Scrivener et al. [26], this calcined clay is currently used for pozzolanic cement production in a plant in southern Brazil, for its pozzolanic activity (585.58 mg $Ca(OH)_2$/g), determined by the modified Chapelle method [27], complies with the standardized requirement for pozzolanic materials (minimum consumption of 436 mg $Ca(OH)_2$/g) [28].

2.1 Constituents selection for the blended cements

The constituents of each blended cement (% by mass) are shown in Table 2. Three cements were composed of waste marble dust and another three of limestone filler. The compositions were defined based on the regional characteristics presented in the Brazilian Cement Technology Roadmap [10]; in standards such as ABNT NBR 16697:2018 [12] and EN 197-1:2011 [11]; and in systematic literature review on ternary and quaternary cements [23], [26], [29]–[31].

Table 2. Percentage by mass of clinker, calcined clay, blast furnace slag, limestone filler and waste marble dust of each blended cement.

Series	Blended cement ID	Clinker (%)	Calcined clay (%)	Blast furnace slag (%)	Limestone filler (%)	Waste marble dust (%)
Serie A	BM	50%	-	30%	3.4%	11.6%
	BL	50%	-	30%	15%	-
Serie B	CM	50%	30%	-	3.4%	11.6%
	CL	50%	30%	-	15%	-
Serie C	CBM	50%	20%	10%	3.4%	11.6%
	CBL	50%	20%	10%	15%	-

The gypsum content can affect the hydration, phase assemblage, porosity, and strength of cementitious materials and varies according to clinker, SCM, and gypsum characteristics. All blended cements in this research contain 5% gypsum by mass of cement. This is in line with the recommendation for the LC^3 [26] and is also used in the Brazilian cement industry. The CPV- ARI cement, equivalent to CEM I 42.5R, supplied clinker to the formulations and extra gypsum was added until the 5% content, by mass, was achieved for each blend. The clinker proportion completes the total blend mass.

The synergy among the different mineral admixtures enables blends with clinker factors around 50% to achieve compressive strength at 7 and 28 days similar to that of ordinary Portland cement [22], [24], [32]. In order to explore the combination effect of blast furnace slag-waste marble dust and blast furnace slag-limestone filler (Series A); calcined clay-waste marble dust and calcined clay-limestone filler (Series B); calcined clay-blast furnace slag-waste marble dust and calcined clay-blast furnace slag-limestone filler (Series C). A clinker factor of 50% was used in each blended cement formulation.

The mixtures were named with letters that represent the main components used: "B" for blast furnace slag, "C" for calcined clay, "L" for limestone filler and "M" for waste marble dust. CM and CL cements replicate the proportions of the LC^3 studied by Antoni et al. [33], Berriel et al. [34] and Krishnan et al. [17]. BM and BL cements are standardized by ABNT NBR 16697:2018 [12] as Portland cement composed of granulated blast furnace slag (CP II – E). CBM and CBL cements were formulated to evaluate the interaction between calcined clay and blast furnace slag with, respectively, waste marble dust and limestone filler.

2.2 Test Methods

2.2.1 Compressive strength

The compressive strength of the mortars produced with the cements studied was determined for cylindrical specimens (diameter of 5 cm and height of 10 cm) at the ages of 3, 7, 28 and 91 days. The specimens followed the ABNT NBR 7215:2019 [35] standard mortar mix, with a water/binder ratio of 0.48 and a sand/binder ratio of 3. The specimens were kept in a humid chamber for 24 hours, then demolded and immersed in water saturated with lime [35].

Compressive strength results of six specimens per age for each cement were statistically treated by means of analysis of variance (ANOVA) with a significance level of 5% to identify (dis)similar groups. Tukey's post-test was used to compare means and identify groups with the highest and lowest compressive strengths. For each series and age, the influence of replacing limestone filler with waste marble dust on compressive strength was investigated. Subsequently, all cements were evaluated by age to identify those with the highest and the lowest compressive strengths and which were statistically similar.

2.2.1 Cementing efficiency

The equations of Bolomey [36] and Smith [37] were used to calculate the cementing efficiency, similarly to the studies of Yu et al. [38] and Yu et al. [39], which evaluated cements with high contents of supplementary cementitious materials. Although the use of approximate equations as proposed by Bolomey [36] can favor uncertainties propagation, the study of Kuder et al. [40] indicated an 86% correlation between experimental data and those obtained by the Bolomey [36] equation, considering different ages and replacement contents.

In Bolomey's [36] equation (Equation 1), the compressive strength (fc) of Portland cement-based concrete is a function of the water/cement ratio (w/c) and two constants (A and B) that are influenced by curing and by the type of cement.

$$Fc = A \times (w/c)^{-1} - B \tag{1}$$

The constants A and B were determined by non-linear regression from the results of the compressive strength of the CP V-ARI cement and the water/cement ratios of 0.40, 0.48, 0.60 and 0.70. The value of the effective water/cement ratio ($(w/c)_e$) required for the CP V-ARI cement to achieve compressive strength similar to that of the experimental cement is obtained by substituting the compressive strength results (fc) of the cements in Equation 1, (Figure 3).

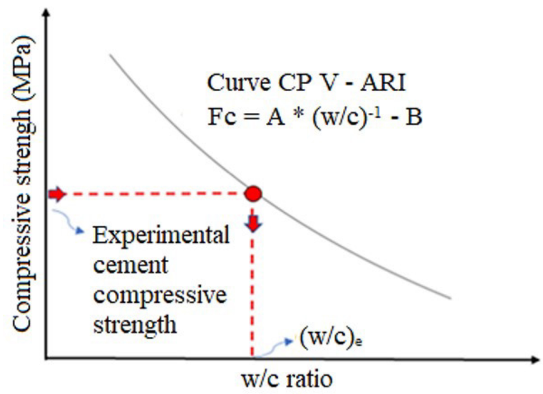

Figure 3. Method for obtaining the effective water/cement ratio ($(w/c)_e$).

In the equation proposed by Smith [37] (Equation 2), the parameters are water consumption (w), reference cement consumption (c), and SCM consumption in the cement (in kg/m^3 of concrete or mortar).

$$(w/c)_e = W/(C + K * SCM) \tag{2}$$

The design mix used is 1:3:0.48 (binder:sand:water/binder ratio, by mass) [35]. The cement consumption, considering all constituents (Cc) is given by Equation 3:

$$Cc = \frac{1000}{\frac{1}{\gamma_{blends}} + \frac{a}{\gamma_a} + x} \tag{3}$$

Where: γ_{blends} represents the specific gravity of the blended cements, in kg/dm^3, calculated according to the proportions of the blended cement constituents; γ_a represents the specific gravity of the standardized Technology Research Institute (IPT) sand (2.62 kg/dm^3); and x represents the water/cement ratio (0.48).

The CP V-ARI cement (equivalent to CEM I 42.5R) was used as reference for plotting the compressive strength *vs* water/cement ratio curve. As this cement contains approximately 89% of clinker, it was necessary to correct the constituents' proportions of the blended cements, to also consider the limestone filler and gypsum already present in the CP V-ARI cement. Figure 4 and Table 3 respectively show the constituents proportions in each blend in terms of percentages by mass and of kg/m^3.

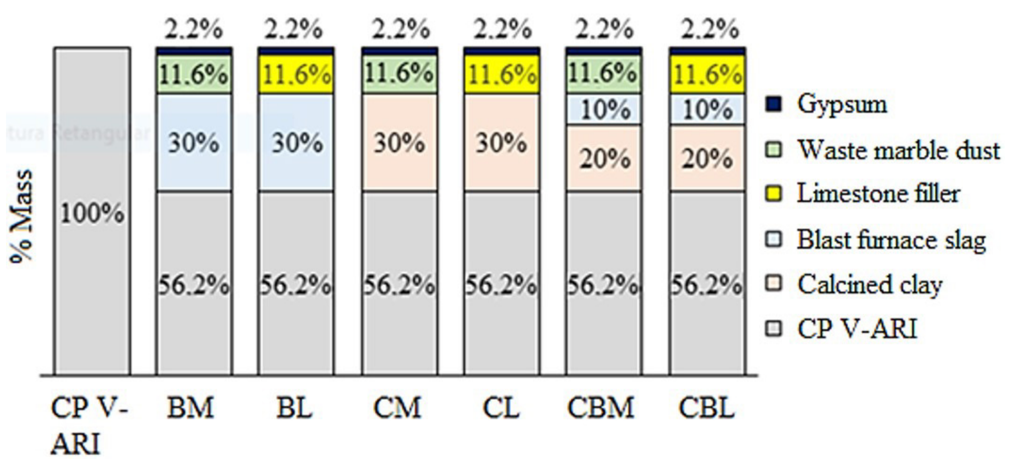

Figure 4. Percentages by mass of gypsum, waste marble dust, limestone filler, blast furnace slag, calcined clay and CP V-ARI cement in the studied blends.

Table 3. Consumption of each constituent of the blended cements (in kg/m^3).

Blended cement	CP V- ARI	Calcined clay	Blast furnace slag	Limestone filler	Waste marble dust	Gypsum	Water
BM	286.46	-	152.92	-	59.13	11.21	245.06
BL	286.42	-	152.89	59.12	-	11.21	245.02
CM	283.59	151.38	-	-	58.54	11.10	242.60
CL	284.54	151.89	-	58.73	-	11.14	242.56
CBM	284.54	101.26	50.63	-	58.73	11.14	243.42
CBL	286.42	101.93	50.96	59.12	-	11.21	243.38

With the effective w/c ratio ($(w/c)_e$), water consumption (w), consumption of the CP V-ARI cement (c) and consumption of supplementary cementitious materials (SCMs), it is possible to determine the cementing efficiency (k-value) of the set of supplementary cementitious materials and limestone filler or waste marble dust at 3 and 28 days by substituting Equation 1 in Equation 2.

2.2.3 Environmental performance

Life cycle GHG emissions cover a single environmental issue (global warming). When local data were unavailable, datasets from the Ecoinvent v. 2.2 database, adjusted to the Brazilian energy matrix, were used. Emissions were modeled from *"cradle to gate"*, which covers impacts from raw material extraction to the cement plant's gate. The life cycle impact assessment (LCIA) was performed using the CML baseline 2001 method.

Based on a directive from the European Union that differentiates waste from co-product [41], in the case of waste marble dust, only the GHG emissions resulting from drying the marble sludge dust (0.007 kgCO_{2eq}/t) for 24 hours in an oven (power of 3.96 kW) were considered. A clinker GHG emission factor ($Ef_{clinker}$) of 902.47 kgCO_{2eq} per tonne produced was retrieved from the Ecoinvent 2.2 database [42]. The GHG emission factor per tonne of each cement constituent is presented in Table 4.

Using the GHG emissions factors of Table 4, the GHG emissions embodied in each blended cement were calculated according to the Equation 4:

$$GHG\ Emission = \sum Pc \times Ef_{const} \tag{4}$$

where Ef_{const} is the emission factor (in CO_{2eq}) of each cement constituent, and Pc is its corresponding proportion (%) of the blended cement.

Table 4. GHG emission factors of the cement constituents.

Cement constituent	GHG emissions factor (kgCO_{2eq}/t)
Clinker	902.47[a]
Blast Furnace Slag	3.42[b]
Limestone filler	14.58[b]
Calcined Clay	276.27[b]
Gypsum	2.13[b]
Waste marble dust	0.007

Sources: [a]Ecoinvent 2.2 databases; [b]Silva et al. [42].

The GHG emissions intensity, an eco-intensity index used by Rodrigues et al. [43], relates the GHG emissions (in CO_{2eq}) for producing one tonne of cement to one unit of compressive strength at each testing age (MPa). This GHG emission intensity enables to jointly analyze environmental and mechanical performance and to identify optimal cement proportioning. The GHG emissions intensity can be reduced by increasing compressive strength and/or decreasing GHG emissions per tonne of cement. For comparison's sake, each blended cement GHG emission intensity was normalized relatively to the CP V-ARI cement (equivalent to CEM I 42.5R).

3 RESULTS AND DISCUSSION

3.1 Compressive strength

The influence of SCMs and waste marble dust on the compressive strength of cements is shown in Figure 5. All cements met the criteria for ABNT NBR 16697:2018 [12] class 40.

As age increased, the compressive strength also increased due to clinker hydration and reactions involving the components development, as indicated by Adu-Amankwah et al. [44].

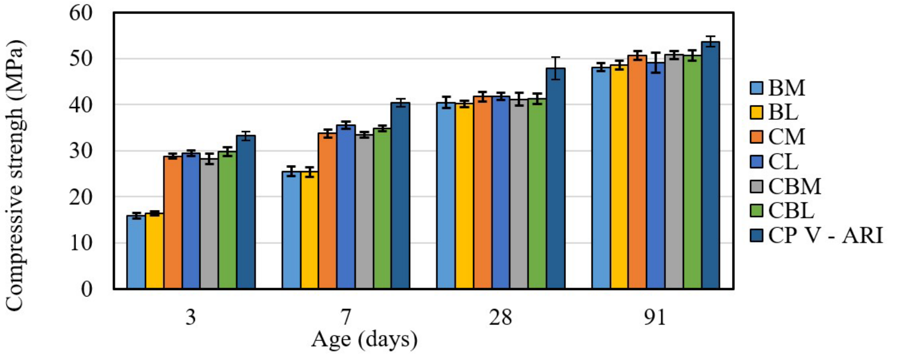

Figure 5. Compressive strength of blended cements at 3, 7, 28 and 91days.

The CP V-ARI cement (clinker factor of 86%) achieved the highest values of compressive strength at all ages, ratifying, as expected, the importance of clinker content on the compressive strength development [44]–[46]. However, some blended cements with clinker factor of 50% (CM, CL, CBM and CBL) showed a combination of effects resulting from the SCMs' characteristics. They also showed a similar trend: compressive strength at 3 and 7 days was higher than ternary blast furnace slag and limestone filler/waste marble dust, due to the addition of calcined clay that, in the presence of limestone filler/waste marble dust, contributes to the rapid hydration reaction.

The analysis of variance by series of Table 2 showed that the p-value was less than 0.05 for all ages, indicating that the replacement of limestone filler with waste marble dust does not significantly alter the compressive strength at the ages studied. On the other hand, the analysis of variance in all cements of Table 3 at the ages of 3, 7 and 91 days showed a p-value greater than 0.05, indicating that, for these ages, the interaction between the cement constituents and the compressive strength is significant.

At 3 and 7 days, Tukey's post-test indicates that the blended cements containing calcined clay (CM, CL, CBM and CBL) compose the subset with the highest strength. Compressive strength development at early ages occurs as a function of the fast chemical reactions between the alumina of the calcined clay and the carbonate of the waste marble dust and limestone filler, producing carboaluminate phases that densify the matrix [33], [17]. At 28 and 91 days, the Tukey post-test indicates no significant difference in the compressive strength of the blended cements.

The interaction between the blast furnace slag-waste marble dust and the blast furnace slag-limestone filler contributes to the formation of hydrated calcium monocarboaluminate (Mc) and hydrated calcium hemicarboaluminate (Hc) phases [29], [47]. However, the availability of alumina from blast furnace slag and calcined clay influences the compressive strength development: the higher alumina content in the calcined clay (Table 1) favors a faster compressive strength development for cements CM, CL, CBM and CBL relatively to that of BM and BL blends.

The compressive strength increases of BM and BL cements ranged from 144 to 155% between the ages of 3 and 28 days. The compressive strength development at more advanced ages is associated with the hydration of the blast furnace slag and the formation of hydrated products (C-S-H e C-A-S-H), as demonstrated by Parashar and Bishnoi [48].

3.2 Cementing efficiency

Figure 6 shows the curves relating the water/cement ratio to the compressive strength at the ages of 7 and 28 days for CP V-ARI cement (equivalent to CEM I 42.5R), obtained through non-linear regression of the experimental compressive strength results.

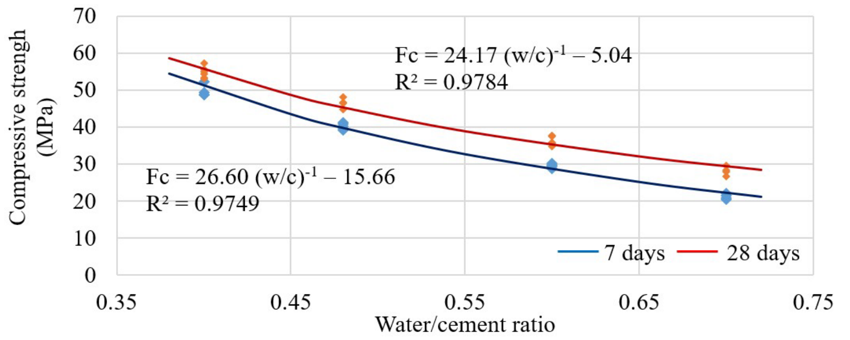

Figure 6. Compressive strength of CP V-ARI cement at 7 and 28 days.

Table 5 and Table 6 present the compressive strength values at 7 and 28 days (fc), the effective w/c ratio ($(w/c)_e$), calculated from the equations in Figure 6, the consumption of CP V-ARI cement (c), the consumption of the SCMs that replace the CP V-ARI cement, the water consumption (w), and the k-value, calculated as described in item 2.2.1.

Table 5. Smith's equation parameter input values and cementing efficiency (k-value) at 7 days.

Series	Blended cement	fc (MPa)	$(w/c)_e$	C (kg/m³)	SCM (kg/m³)	w (kg/m³)	k-value
Series A	BM	25.50	0.65	286.46	223.26	245.06	0.42
	BL	25.41	0.65	286.42	223.22	245.02	0.41
Series B	CM	33.75	0.54	283.59	221.02	242.60	0.76
	CL	35.53	0.52	284.54	221.76	242.56	0.82
Series C	CBM	33.49	0.54	284.54	221.76	243.42	0.75
	CBL	34.87	0.53	286.42	223.22	243.38	0.79

Table 6. Smith's equation parameter input values and cementing efficiency (k-value) at 28 days.

Series	Blended cement	fc (MPa)	$(w/c)_e$	C (kg/m³)	SCM (kg/m³)	w (kg/m³)	k-value
Series A	BM	40.52	0.53	286.46	223.26	245.06	0.79
	BL	40.17	0.53	286.42	223.22	245.02	0.77
Series B	CM	41.78	0.52	283.59	221.02	242.60	0.84
	CL	41.84	0.52	284.54	221.76	242.56	0.84
Series C	CBM	41.16	0.52	284.54	221.76	243.42	0.82
	CBL	41.29	0.52	286.42	223.22	243.38	0.81

At 7 days, the combination of calcined clay-limestone filler (CL) and the combination of calcined clay-blast furnace slag-limestone filler (CBL) presented the highest k-values, followed by the combination of calcined clay-waste marble dust (CM) and the combination of calcined clay-blast furnace slag-waste marble dust (CBM).

At 28 days, the combination of calcined clay-limestone filler (CL) and the combination of calcined clay-waste marble dust (CM) presented the highest k-value, followed by the combination of calcined clay-blast furnace slag-waste marble dust (CBM) and of the combination of calcined clay-blast furnace slag-limestone filler (CBL).

The combinations of blast furnace slag with waste marble dust (BM) and with limestone filler (BL) consistently showed the lowest k-values for the ages of 7 and 28 days. However, the combinations of blast furnace slag-limestone filler and blast furnace slag-waste marble dust showed the most significant k-value growth from 7 to 28 days (respectively 82% and 84%).

Analyzes of Series A, B and C of Tables 5 and 6 highlighted the similarity of the cementing efficiency of waste marble dust and limestone filler, which was also observed for the compressive strength results (item 3.1). However, direct comparison of the cementing efficiency obtained in this research and those of the literature review is impaired because the characteristics of the materials used can influence the k-value results [47], and the use of approximate equations to determine the compressive strength, as proposed by Bolomey [36], can favor the uncertainties propagation, as these curves were determined for different cements.

3.3 Environmental performance

Figure 7 presents the GHG emissions (in CO_{2eq}) per tonne of each blended cement. As expected, the CP V-ARI cement - equivalent to CEM I 42.5R - has the highest GHG emissions, due to its highest clinker content, and sets the reference for analyzes.

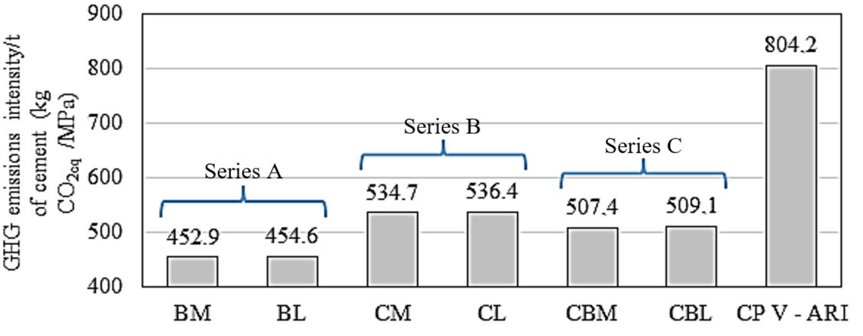

Figure 7. GHG emissions per tonne of blended cements and of CP V-ARI (in CO_{2eq}).

Series A cements reduce approximately 43% GHG emissions relatively to CP V-ARI, followed by cements in Series C (~36% reduction) and Series B (~33% reduction). Series B cements emit more GHGs due to their higher calcined clay content. Replacing limestone filler with waste marble dust does not significantly reduce GHG emissions.

Figure 8 presents the GHG emissions intensity (in CO_{2eq}) per unit of compressive strength of the cement (in MPa) at 3, 7, 28 and 91 days, normalized relatively to CP V-ARI cement (equivalent to CEM I 42.5R).

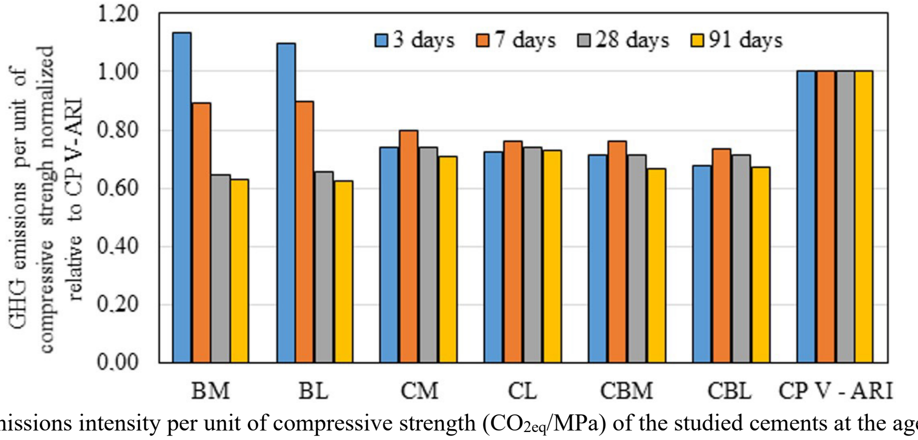

Figure 8. GHG emissions intensity per unit of compressive strength (CO_{2eq}/MPa) of the studied cements at the ages of 3, 7, 28 and 91 days, normalized relatively to CP V-ARI.

Series B and C cements confirmed the benefits that replacing clinker has on GHG emissions and compressive strength at all ages. Due to the slow development of compressive strength, cements containing blast furnace slag are not the best options in terms of GHG emission intensity at early ages. But after 28 days, cements in Series A outperformed all other combinations.

The GHG emissions intensities are similar for cements with waste marble dust or with limestone filler. Hence, although replacing limestone filler with waste marble dust has a negligible effect on GHG emissions, it can mitigate environmental liabilities of ornamental stone processing and encourage circularity within the cement industry.

4 CONCLUSIONS

Given the approaching scenario with limited availability of SCMs with adequate quality and the increasing environmental pressures on the cement industry, it is necessary to investigate new materials and formulations that reduce GHG emissions and other environmental impacts, while meeting regulatory and technical requirements. All blended cements herein studied met the compressive strength requirements for all classes normalized by ABNT NBR 16697:2018. Replacing limestone filler with waste marble dust does not significantly alter the compressive strength at the ages studied.

The blended cements with reduced clinker factor 50% showed synergistic effects resulting from the characteristics of the SCMs. The combination of calcined clay-waste marble dust (CM); calcined clay-limestone filler (CL); calcined clay-blast furnace slag-waste marble dust (CBM) and calcined clay-blast furnace slag-limestone filler (CBL) resulted in higher compressive strengths in the early ages, due to the rapid chemical reactions between the alumina from the calcined clay and the carbonate provided by the waste marble dust and the limestone filler.

Contrastingly, the combinations of blast furnace slag-waste marble dust and blast furnace slag-limestone filler showed low compressive strength at 3 and 7 days, but a considerable growth at the age of 28 days, which is associated with the hydration of the blast furnace slag and the formation of the hydrated products (C-S-H and C-A-S-H).

Cementitious efficiency follows the trend of compressive strength. The combination of calcined clay-limestone filler (CL); calcined clay-blast furnace slag-limestone filler (CBL); calcined clay-marble dust waste (CM) and calcined clay-blast furnace slag-waste marble dust (CBM) presented the highest cementing efficiency at 7 and 28 days. However, the combinations of blast furnace slag-limestone filler (BL) and blast furnace slag-waste marble dust (BM) showed the most significant cementing efficiency increase from 7 to 28 days. Analyzes of the blended cements in Series A (BM and BL), B (CM and CL) and C (CBM and CBL) showed no significant difference in cementing efficiency and in the compressive strength of cements with waste marble dust or limestone filler.

The GHG emission intensity offers a joint perspective of the environment and the mechanical performance that can support formulation development and selection. Series B (CM and CL) and C (CBM and CBL) cements demonstrated benefits at all ages. Cements containing blast furnace slag are not the best options in terms of intensity of GHG emissions at early ages, due to the slow development of compressive strength. But after 28 days, Series A cements (BM and BL) outperform all other combinations.

Although the GHG emissions and the GHG emissions intensities are similar for cements with waste marble dust and cements with limestone filler, the study showed the benefits of the synergistic effect of the combined SCMs and limestone filler/waste marble dust on GHG emissions and compressive strength. Thus, cements with waste marble dust and/or limestone filler combined with calcined clay and/or blast furnace slag favor the reduction of environmental liabilities of the ornamental stone processing and encourage circularity within the cement industry. These blends can reduce the clinker factor and GHG emissions intensity, while preserving the mechanical performance required by the civil construction market.

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