Marble and granite waste as mineral addition in mortars with different water-cement ratios

Resíduo de mármore e granito como adição mineral em argamassas com diferentes relações água-cimento

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Abstract

Civil construction is associated with several environmental impacts, such as CO_2 emissions during cement production and waste generation. In this context, aiming to contribute to the sustainable development of the sector, this article aims to study the effect of replacing cement by marble and granite waste (MGW) in different water/cement ratios (w/c). To do this, the waste was characterized and applied in 1:3 mortars, replacing the cement at 20% content. Mortars with and without plasticizing additives were produced, measuring the amount of water by fixing the workability and the additive content. Water demand, compressive strength, dynamic modulus of elasticity, water absorption by immersion and capillarity, electrical resistivity and accelerated carbonation were evaluated. The results showed that the MGW acted as a filler, promoting refinement of the porous structure and maintenance of compressive strength due to better particle packing. It was observed that MGW made the mortar structure more homogeneous. However, the filler effect of the waste (inert) did not compensate for the reduction in the cement content of the mortars for strength to carbonation. In general, it was observed that waste is more efficient in lower water/cement ratios.

Keywords: Marble waste. Granite waste. Waste recovery. Cementitious composite.

Resumo

A construção civil está associada a diversos impactos ambientais, como a emissão de CO_2 durante a produção de cimento e a geração de resíduos. Neste contexto, visando contribuir para o desenvolvimento sustentável do setor, este trabalho teve como objetivo estudar o efeito da substituição de cimento por resíduo de mármore e granito (RMG) em diferentes relações água/cimento (a/c). Para isso, o resíduo foi caracterizado e aplicado em argamassas de traço 1:3, substituindo o cimento no teor de 20%. Argamassas com e sem aditivo plastificante foram produzidas, dosando-se a quantidade de água pela fixação da trabalhabilidade e do teor de aditivo. Foram avaliados a demanda de água, resistência à compressão, módulo de elasticidade dinâmico, absorção de água por imersão e por capilaridade, resistividade elétrica e carbonatação acelerada. Os resultados mostraram que o RMG atuou como fíler, promovendo refinamento da estrutura porosa e manutenção da resistência à compressão devido ao melhor empacotamento de partículas. Observou-se que o RMG tornou a estrutura da argamassa mais homogênea. Contudo, o efeito fíler do resíduo (inerte) não compensou a redução do teor de cimento das argamassas para a resistência à carbonatação. De forma geral, observou-se que o resíduo é mais eficiente em relações água/cimento mais baixas.

***Palavras-chave**: Resíduo de mármore. Resíduo de granito. Aproveitamento de resíduo. Materiais cimentícios.*

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Recebido em 04/11/21
Aceito em 19/04/22

ALMADA, B. S.; SANTOS, W. J. Dos; SOUZA, S. R. Marble and granite waste as mineral addition in mortars with different water-cement ratios. **Ambiente Construído**, Porto Alegre, v. 22, n. 4, p. 7-22, out./dez. 2022.
ISSN 1678-8621 Associação Nacional de Tecnologia do Ambiente Construído.
http://dx.doi.org/10.1590/s1678-86212022000400624

Introduction

Civil construction is one of the industries that produces the most waste and disposes of it in the environment and, regarding the increased demand for concrete, is also related to the depletion of natural resources (MAGALHÃES; DANILEVICZ; SAURIN, 2017; HABERT *et al*., 2020; MEDEIROS *et al*., 2021). Thus, using alternative materials can contribute to sustainable development, reducing the environmental degradation caused by waste and prolonging natural material reserves. According to SNIC (SINDICATO…, 2022) – National Cement Industry Union, in 2021, Brazil produced almost 61 million tons of cement, contributing negatively to CO_2 emission into the atmosphere. Taking this into account, it is noteworthy that the country ranks high in the production of ornamental stones, whose extraction and processing process generates a large volume of waste, estimated at about 20 to 30% of the raw material (ALIABDO; ELMOATY; AUDA, 2014; ULUBEYLI *et al*., 2016; BUYUKSAGIS; UYGUNOGLU; TATAR, 2017). During the wet cutting process, ornamental rock slabs, such as granite and marble, experience wear, resulting in the formation of a grayish sludge, also called abrasive sludge, which, when dry, generates a fine powdery waste. Sousa (2007) presents soil and groundwater contamination, an alteration of the biological chain of living beings, and river silting as the main impacts generated by inadequate disposal of this sludge.

In this context, many studies have been carried out focusing on reusing marble and granite waste (MGW) in cementitious materials (ALIABDO; ELMOATY; AUDA, 2014; SARDINHA; BRITO; RODRIGUES, 2016; SADEK; EL-ATTAR; ALI, 2016; SINGH; NAGAR; AGRAWAL, 2016; MUNIR; KAZMI; WU, 2017; GHORBANI *et al*., 2018; KHODABAKHSHIAN *et al*., 2018; LI *et al*., 2019; SINGH; SRIVASTAVA; BHUNIA, 2019; ALMADA *et al*., 2020). These studies show that MGW does not, or has low, pozzolanic activity, but can act as a filler, improving some mechanical and durability properties of cementitious materials (BABOO *et al*., 2011; ALIABDO; ELMOATY; AUDA, 2014; ELYAMANY *et al*., 2014; VARDHAN *et al*., 2015; AREL, 2016; SADEK; EL-ATTAR; ALI, 2016; MUNIR; KAZMI; WU, 2017; BARBOSA; SANTOS, 2013; KHODABAKHSIAN *et al*., 2018; ALMADA *et al*., 2020).

According to Dill (2010), marble waste usually contains calcite and/or dolomite in their composition. These carbonate materials can participate in the cement hydration process. The small $CaCO_3$ particles react in the alkaline environment with the aluminates present in the cement to form hydrated carbonate products, such as monocarboaluminate ($Ca_4Al_2(CO_3)OH)_{12}\cdot 5H2O$) and calcium hemicarboaluminate ($(Ca_4Al_2(CO_3)_{0,5}OH)_{13}\cdot 5,5H2O$) (KAKALI *et al*., 2000; MATSCHEI; LOTHENBACH; GLASSER, 2007; DHANDAPANI *et al*., 2021; KECHAGIA *et al*., 2021). The growth of these products takes place competitively to monosulfoaluminate, a product of aluminate and sulfate interaction of cement. Thus, more sulfate ions remain in the solution and there is ettringite formation at more advanced ages (KECHAGIA *et al*., 2021). The formation of mono- and hemicarbonates provides a reduction in porosity over time, a gain in resistance to compression and carbonation (MATSCHEI; LOTHENBACH; GLASSER, 2007; BUCHER *et al*., 2017; DHANDAPANI *et al*., 2021).

It is also worth mentioning that the water-cement ratio (w/c) is one of the factors that has the greatest influence on the performance of these materials as it affects porosity and permeability, determining the microstructure of the paste, distribution and shape of the pores (KALLA *et al*., 2013). In addition, when at high values, it can facilitate the diffusion of chloride ions, impairing the durability of cementitious materials (KIM *et al*., 2014).

Singh *et al*. (2017) and Sardinha, Brito and Rodrigues (2016) studied concretes replacing cement with marble waste (MW) in different w/c ratios, using plasticizer and superplasticizer additives. In both studies, different levels of influence of the MW content were observed for each w/c ratio. Although an ideal w/c ratio for MW concretes was not specified, the best mechanical and durability results were obtained by the compositions with the lowest water content. However, studies were carried out using marble waste only and, as observed by Sadek, El-Attar and Ali (2016), the type and characteristics of the waste can have an important effect on the behaviour of concrete. Thus, it is important to evaluate the performance of cementitious materials introducing mixed MGW in different w/c ratios, which was the objective of this study. To do this, four compositions were made:

(a) two mortars without plasticizing additives;

(b) one with cement replacement by MGW;

(c) two with fixed plasticizer additives; and

(d) one with cement replacement by MGW.

Thus, it the influence of MGW can be evaluated on water demand and the impacts on mechanical and durability properties.

Materials and methods

The studied waste was collected in the form of abrasive sludge, from the wet processing of ornamental stone slabs in a marble factory located in the northern region of Belo Horizonte. Initially, the material was dried in an oven (105 ± 5 °C) until mass was constant. After drying, the sample was manually crushed, passed through a 75 µm sieve and stored in hermetically sealed bags. For the research, only a fraction of granulometry less than 75 µm was used, which corresponded to more than 75% of the material. The wastes were characterized in terms of granulometry, by laser diffraction, in a Sympatec Helos 12LA granulometer, with sodium hexametaphosphate as a dispersant.

The specific mass was determined by helium pycnometer in the Quantachrome SPY-3 pycnometer. The chemical composition was evaluated by X-ray fluorescence spectrometry (XFS) with sample melting in lithium tetraborate and loss on ignition by calcination up to 1000 °C, after sample drying. The mineralogical composition was obtained by X-ray diffractometer (XRD), in a Bruker D2 Phaser diffractometer, with CuKa copper radiation (1.54184 A) and a step size of 0.018°/s. Phase identification was performed using the X'Pert HighScore Plus 3.0 software and the Crystallography Open Database (COD 2020) database. Rietveld refinement was performed to quantify the phases, using an internal zincite standard (purity > 99%) and a GOF refinement parameter close to 1.0.

Portland cement CPI was used, as it does not contain additions, allowing a better assessment of the influence of MGW as a cement substitute. The cement was characterized according to its specific mass, using the helium gas pycnometry method, resulting in 3.221 g/cm . The chemical composition was determined by quantitative analysis by XFS.

As a fine aggregate, standardized quartz sand was used, supplied by the Institute for Technological Research, comprising four fractions (nº 16, nº 30, nº 50 and nº 100) equally proportioned. Water reducing additive of the multifunctional plasticizer type, normal setting, type 1 (RA1), Muraplast FK 97, from MC Bauchemie was used to fix the workability. According to the manufacturer, the product consists of a brown liquid, with a density of 1.18 g/cm and a recommended dosage of 0.2 to 1.0% on the cement mass.

Initially, a w/c ratio of 0.50 was defined, and two compositions were produced: one that was the reference (REF) without waste; and one with cement replacement by MGW at a content of 20% (MGW). Then, the consistency index obtained by the REF mortar (173 ± 10 mm) was fixed and the amount of water was dosed, using a plasticizer additive. The additive was applied at the maximum content allowed by the manufacturer (1% in relation to the binder mass), to obtain a greater reduction in the amount of water. Under these conditions, two mortars were produced, one of which was a reference with plasticizer (REF-P) and one with cement replacement by MGW at a content of 20% and plasticizer (MGW-P). The material consumption mortar production is shown in Table 1.

For specimen preparation, the NBR 7215 (ABNT, 2019) procedures were used, adapted so that the mixing time allowed the additive to activate. In the first 30 seconds, 60% of the water mass was added to the binding material and, in the next 30 seconds, when the mortar was at a low speed, the fine aggregate was added. At the end of the first minute, the rest of the water was added, with the diluted additive for the REF-P and MGW-P cases, allowing the mixture to proceed at a low speed for 1 minute. After that, during the following minute, scraping was performed in the first 30 seconds and, in the final 30 seconds, the mixture was left to rest, covered by a damp cloth. At the end, the mortar was turned on for another 1 minute at high speed.

In the moulding process, the moulds were greased with release oil and the densification was carried out on a vibrating table. The vibration was stopped when uniform exudation was observed on each layer. For the 5x10 cm cylindrical specimens, the consolidation was done in 3 layers and, for the 10x20 cm cylindrical specimens, in 5 layers. The specimens were demoulded within 48h ± 24h and identified. Then, curing was carried out in a humid chamber (Humidity ≥ 95%) for 28 days. The mortars were evaluated after 28 days of curing, according to the properties and standards described in Table 2.

Table 1 – **Material consumption per m³ of mortar**

ID	Cement (kg/m³)	Waste (kg/m³)	Sand (kg/m³)	Water (kg/m³)	Additive (kg/m³)	w/c ratio
REF	509.08	-	1527.25	254.54	-	0.500
REF - P	520.35	-	1561.05	233.64	5.20	0.449
MGW	404.63	101.16	1517.36	252.89	-	0.500
MGW- P	415.89	103.97	1559.58	227.70	5.20	0.438

Table 2 - **Properties evaluated in the mortars**

Property	Norm	Quantity and type of specimens	
Consistency index	NBR 13276 (ABNT, 2016)	3 flow measurements	
Water absorption by immersion	NBR 9778 (ABNT, 2009)	4 un	5x10
Water absorption by capillarity	NBR 9779 (ABNT, 2012b)	4 un	5x10
Dynamic modulus of elasticity	C215 (AMERICAN…, 2008) and Lee *et al*. (2015)	3 un	10x20
Electrical resistivity	NBR 9204 (ABNT, 2012a)	3 un	10x20
Compressive strength	NBR 5739 (ABNT, 2018)	4 un	5x10
Accelerated carbonation	ISO 1920-12 (INTERNATIONAL…, 2015).	3 un	4x4x16

In addition to the water absorption test, a visual analysis was performed to identify the shape and distribution of pores in the cement matrix. To do this, the specimens submitted to the immersion water absorption test were sectioned using a diamond saw. The sections were analysed using a digital microscope with a magnification of up to 1600x. From the scale of the images, measurements of the pores observed in the ImageJ software were carried out.

The dynamic modulus of elasticity, which expresses the mortar's stiffness or strength to elastic deformation, was determined by the forced resonant frequency method in longitudinal mode. For the test, the Erudite MKII Resonant Frequency Test System was used to measure the resonant frequency. Each of the three specimens evaluated was placed between two terminals (generator-receiver) so that the ultrasonic pulse passed through it. Then, the conduction frequency was varied until the response of the measured sample reached a maximum amplitude, which is considered the resonant frequency of the material. This measurement was performed five times per specimen. After having obtained the results of the experiment, the dynamic modulus of elasticity was obtained from Equation 1, developed by Lee *et al*. (2015).

$$E_d = 5.093\, x\, \frac{L}{D^2}\, x\, M\, x\, F^2 x\, 10^{-9} \qquad \text{Eq. 1}$$

Where:

E_d – dynamic modulus of elasticity (GPa);

L - length of the specimen (m);

D - diameter of the specimen (m);

F – frequency of the fundamental mode of longitudinal vibration (Hz); and

M – mass volumetric weight (kg).

For electrical resistivity, the specimens were completely immersed for 24 hours to become saturated and then an electrical circuit was set up with the sample inserted between two electrodes, located on the base and top surfaces of the specimens. As it is a series circuit, the current that passed through the specimen was the same that passed through the equipment. Direct electric current readings were performed, applied through the potential difference (8V) between the two electrodes, with the FG -8102 Digital Function Generator from Politerm. Thus, from the total circuit voltage (known), the measured current and the geometric parameters of the specimen, the electrical resistivity of the material was calculated, according to Ohm's Law (Equation 2).

$$\rho = R\, x\, \frac{A}{L} = \frac{V\, x\, A}{L\, x\, I} \qquad \text{Eq. 2}$$

Where:

ρ – electrical resistivity (Ωm);

R – electrical resistivity (Ω);

I – current (A);

V – potential difference (V);

L – Length of the specimen (m); and

A – cross-sectional area (m).

To determine the compressive strength, the specimens were capped with sulphur, to level the faces, and broken in an automated EMIC press, model DL2000, with a capacity of up to 200t and a speed of (0.45 ±0.15) MPa/s. Accelerated carbonation was evaluated in prismatic bodies (4x4x16) cm, after 60 days in a carbonation chamber (5% CO_2), using a phenolphthalein indicator solution (1%). Ten measurements of carbonation depth were performed in a specimen, perpendicular to the longest edge.

Results and discussion

MGW characterization

In the test to determine the specific mass, the result was 2.67 g/cm , within the values observed in previous studies (BARROS, 2008; DEGEN *et al*., 2013; SOARES, 2014; SADEK; EL-ATTAR; ALI, 2016). It is also observed that the waste is slightly lighter than the cement (3.22 g/cm).

Regarding the granulometric composition, shown in Figure 1, it can be observed that the waste has a granulometry close to that of cement, mainly in the finer fraction, with D50 = 12.0 µm greater than that of cement, with 9 µm. From 2 µm, the MGW granulometric distribution curve tends to move away, due to the greater amount of larger particles in the waste, reaching D90 = 41.7 µm (double that of cement – D90 = 22.55 µm). MGW particles in this granulometric range can contribute to better packing between the cement and sand granulometric range (< 0.15 mm), while the range below 2 µm contributes as nucleation points for cement hydration.

The results of the chemical composition, presented in Table 3, demonstrate that, unlike cement, the MGW mainly consists of SiO_2, with a lower amount of CaO. This shows that the collected MGW comes from processing a greater number of granite-type rocks, due to their more silicate nature, as observed Sadek, El-Attar and Ali (2016). This result differs from that found by Aliabdo, Elmoaty and Auda (2014), who studied marble waste with 83.22% CaO and 1.12% SiO_2, which shows the heterogeneity of waste, according to their lithological origin.

Using the equation proposed by Bogue (1929[1] *apud* COSTA *et al*., 2013), the contents of the anhydrous cement phases were estimated, according to Equations 3, 4, 5 and 6. Thus, it was found that the C_3A content is 4.01%.

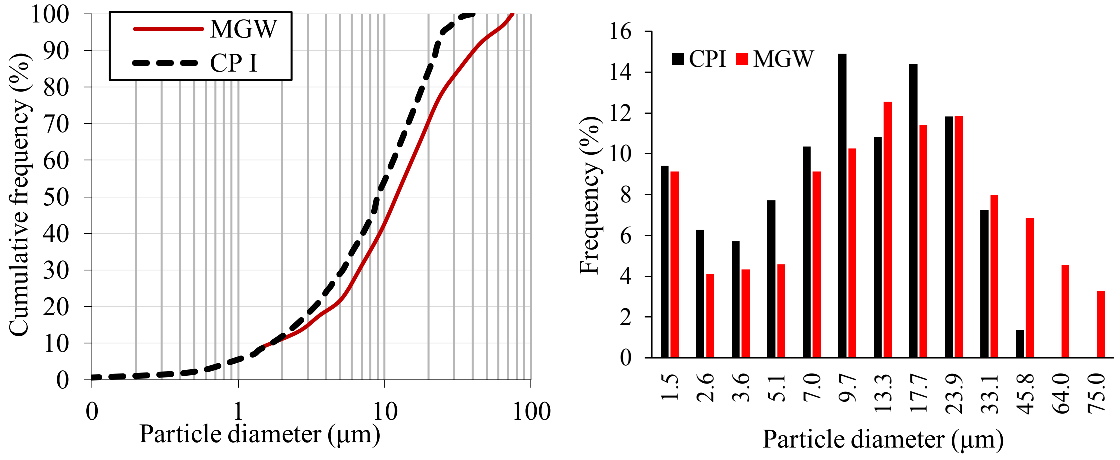
$$C3S = 4.07\,CaO - 7.60\,SiO_2 - 6.72\,Al_2O_3 - 1.43\,Fe_2O_3 \quad \text{Eq. 3}$$

$$C2S = 2.87\,SiO_2 - 0.754\,\%C_3S \quad \text{Eq. 4}$$

$$C3A = 2.65\,Al_2O_3 - 1.69\,Fe_2O_3 \quad \text{Eq. 5}$$

$$C4AF = 3.04\,Fe_2O_3 \quad \text{Eq. 6}$$

Figure 1 - **Granulometric distribution of MGW and cement**



[1]BOGUE, R. H. Calculation of the compounds in Portland cement. **Industrial and Engineering Chemistry**, v. 1, n. 4, p. 192, oct. 1929.

Table 3 - **Chemical composition of cement and MGW**

Material	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	TiO_2	P_2O_5	Na_2O	K_2O	MnO	Others	LOI
MGW	42.80	8.07	2.63	19.00	3.89	0.69	0.33	1.85	2.59	0.04	0.71	17.40
CP I	19.90	4.34	4.43	63.30	2.68	0.23	0.18	0.17	0.98	0.05	1.45	2.29

Note: *LOI = loss on ignition.

Considering that the loss on ignition of the MGW refers to carbonate compound decarbonation, such as calcite and dolomite, it can be observed that there is a possibility of calcium monocarboaluminate and calcium hemicarboaluminate formation in the matrix. However, this formation will be limited by the content of available aluminates, the solubility of calcite, which is lower compared to gypsum, as well as the granulometry of the $CaCO_3$ particles (KLEMM; ADAMS, 1990; DHANDAPANI *et al.*, 2021). According to Dhandapani *et al.* (2021), the reactivity of calcitic materials increases when they are inserted in association with aluminosilicates.

The calcite and dolomite decarbonation occur according to the chemical Equations 7 and 8:

$$CaCO_3 \overset{\Delta}{\rightarrow} CaO + CO_2 \qquad \text{Eq. 7}$$

$$CaMg(CO_3)_2 \overset{\Delta}{\rightarrow} CaO + 2CO_2 \qquad \text{Eq. 8}$$

By stoichiometry, the CO_2 content present in calcite and dolomite are 44% and 48%, respectively, and the CaO content present in calcite and dolomite are 56% and 30%, respectively. Considering the loss on ignition value and the amount of CaO in the MGW, obtained by XRF, the calcite (C) and dolomite (D) content in the material can be observed, according to the system described in Equation 9. Thus, the estimated contents of calcite and dolomite are 28.51% and 10.15% respectively.

$$\begin{cases} PF = 0.44\,C + 0.48\,D \\ \%CaO(FRX) = 0.56\,C + 0.30\,D \end{cases} \qquad \text{Eq. 9}$$

The MGW diffractogram is shown in Figure 2 and the amount of minerals identified in the waste is shown in Table 4. In general, the predominant phases in the MGW are oligoclase, quartz, dolomite and calcite. The first two are associated with granites and the others with marble (DILL, 2010). It can be observed that the contents estimated by stoichiometry with the XFS results were similar, but lower than those obtained by refinement. It is noteworthy that the zincite peaks refer to the internal standard used for Rietveld refinement. In addition, an amorphous material content of less than 5% was identified. According to the XRD results, the waste presents well-defined peaks and no amorphous halo was clearly identified, indicating the high crystallinity of the material. Thus, phases that are chemically reactive do not contribute or contribute little to any pozzolanic reaction of the material. Thus, MGW can be considered an inert material, corroborating the literature (SADEK; EL-ATTAR; ALI, 2016; KHODABAKHSIAN *et al.*, 2018; ALMADA *et al.*, 2020).

Consistency index

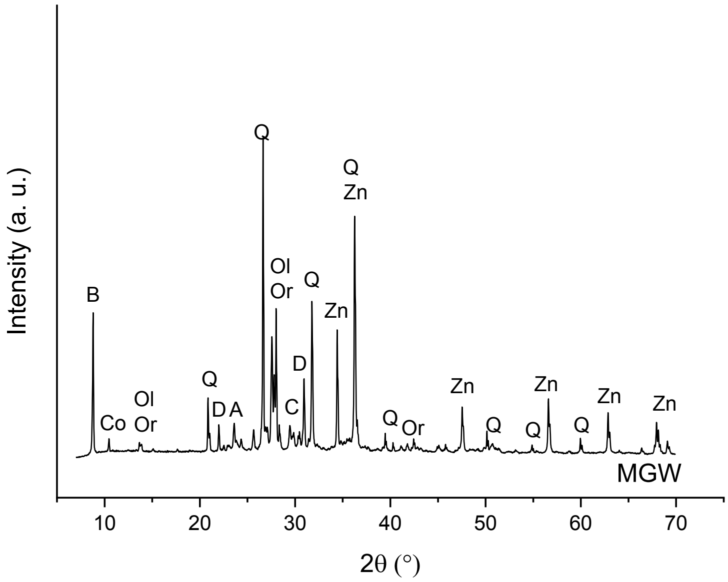
The results of the consistency index test are shown in Figure 3, as well as the water/cement ratios obtained for each mortar.

The consistency index of all dosages was kept within the spreading range determined from the reference mortar without the plasticizer (REF), which has a variation of only 5.78%. It can be observed that introducing the plasticizer allowed a reduction in the water/cement factor of 10.20% for the mixture without waste (REF-P) and 12.20% for the mixture with waste (MGW-P). Comparing the REF and MGW mortars, it can be observed that introducing MGW does not influence the workability, as already seen by Aliabdo, Elmoaty and Auda (2014), Almada *et al.* (2020) and Munir, Kazmi and Wu (2017), showing a scattering reduction of only 1.73%. In the samples containing additive (REF-P and MGW-P), the waste showed that it maintained workability with a low w/c factor.

Water absorption

Figure 4 presents the results of capillary absorption (Ca) and the absorption variation in relation to the REF mortar (Δ), calculated by the difference between the results at each time. The lowest final capillary absorption was obtained for the reference mortar. The mortars containing the additive resulted in an increase in the final absorption of 22.28% for the REF-P sample and 12.15% for the MGW-P sample, while for the MGW sample, this increase was 53.16 %. These results corroborate those found in the literature (ALMADA *et al.*, 2020; SARDINHA; BRITO; RODRIGUES, 2016).

Figure 2 -**XRD standard of MGW**



Note: Q - Quartz; D - Dolomite; C - Calcite; A - Albite; Co - Cordierite; B - Biotite; Ol - Oligoclase; Or - Orthoclase; and Z - Zincite.

Table 4 - **Amount of minerals in the sample**

Mineral		COD	Content (%)
Oligoclase	$NaAlSi_3O_8$	9011423	30.9
Dolomite	$CaMg(CO_3)_2$	9003518	17.2
Quartz	SiO_2	1526860	14.6
Calcite	$CaCO_3$	4502443	13.2
Orthoclase	$KAlSi_3O_8$	9000161	8.4
Albite	$NaAlSi_3O_8$	2310574	5.2
Biotite	$AlFHKMg_3Si_3O_{11}$	9000025	3.9
Cordierite	$Al_4Mg_2Si_5O_{18}$	9005806	1.6
Amorphous	-	-	5.1

Figure 3 - **Result of the mortar consistency index**

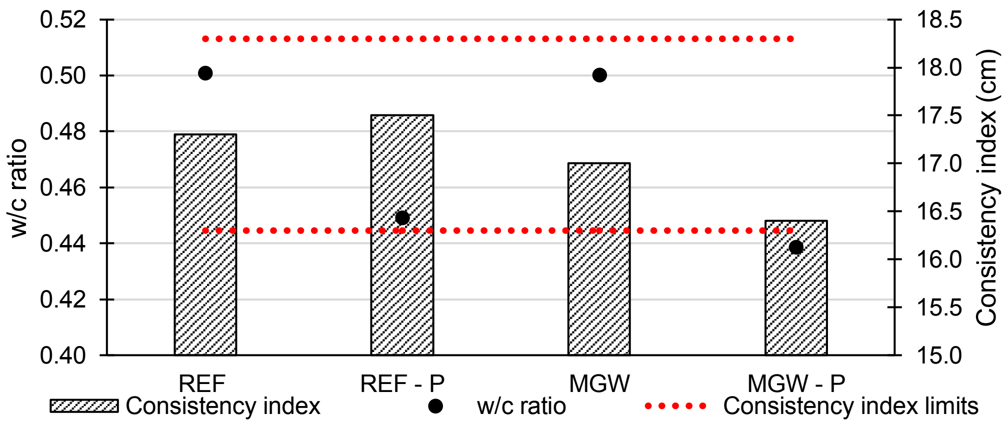
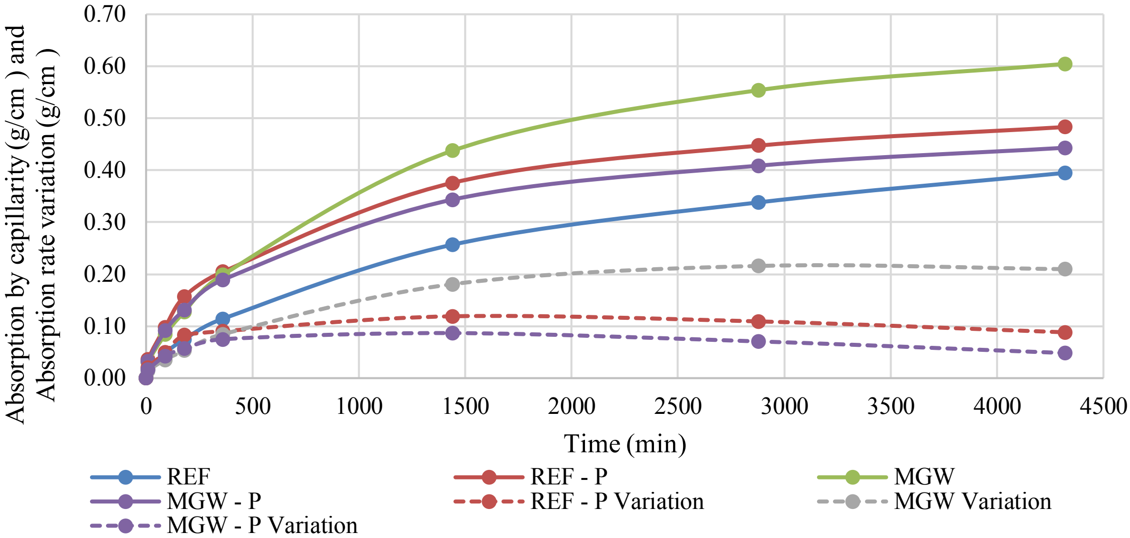


Figure 4 - **Result of absorption and variation of water absorption by capillarity of mortars**



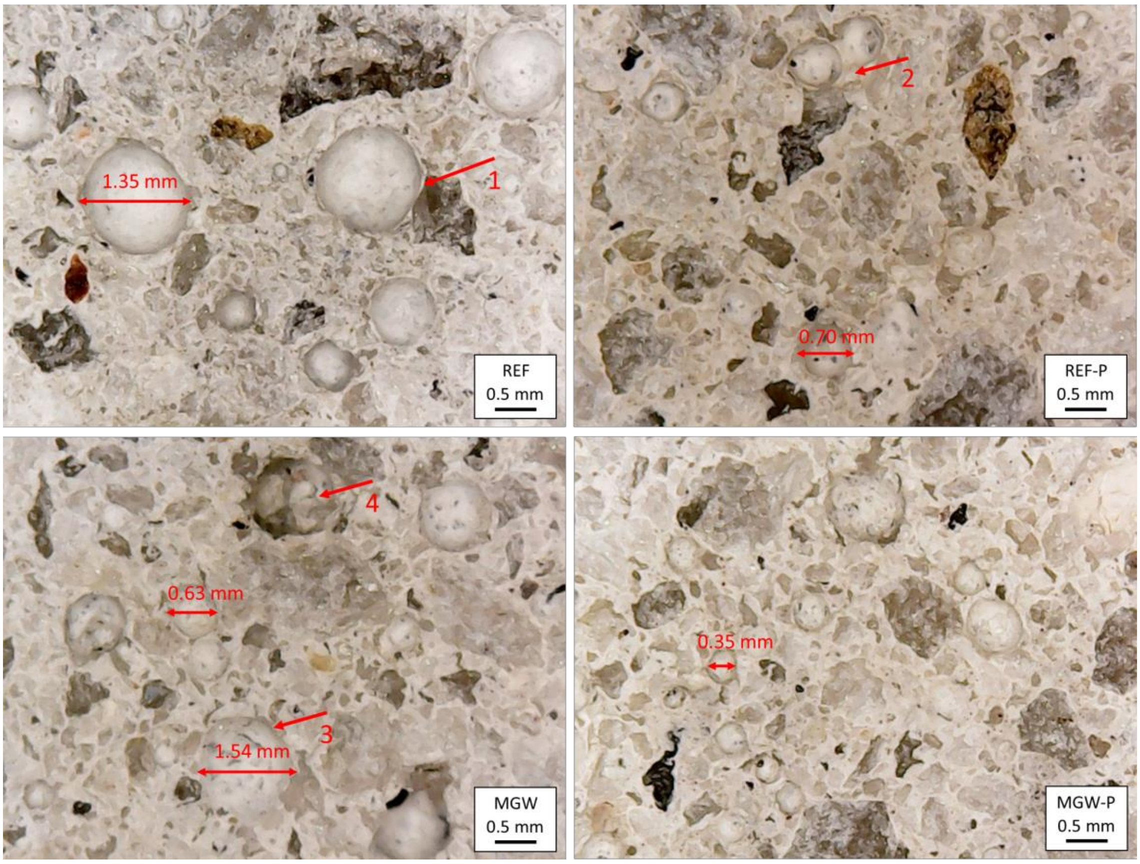
It can be observed that, in the initial minutes, the absorption variation of the mortars with the plasticizer, compared to the mortar REF, is higher than that of the MGW mortar. This result suggests that the additive provided a matrix with a more refined porous structure, which facilitates water absorption by capillarity. In addition, using the additive reduces the amount of water in the composition, which consequently reduces porosity. However, after 360 minutes, there is an inversion and the variation is greater for the MGW mortar, indicating a greater number of interconnected pores in this mortar. Comparing the REF-P and MGW-P samples, it can be observed that the cement replacement by the waste resulted in a lower final water absorption, which is due to the better packaging and the lower w/c ratio. By the slope of the curves in the initial stretch of absorption, it can be seen that the absorption speed is lower for the REF mortar. Therefore, both the waste and the plasticizer were responsible for the porous structure refinement. Furthermore, the results indicate that there are differences in pore size and connectivity.

To support the justifications, images were taken of the internal porosity of the specimens submitted to the absorption test, with a magnifying glass of 1000x, as shown in Figure 5.

In general, it can be observed that, for the REF mortar, there are large pores, with dimensions of up to 1.35 mm and more distant from each other. At point 1, it can be seen that the pore is formed near the paste-aggregate transition zone, indicating a weakening region that can affect the compressive strength of the material. Comparing the mortars containing waste (MGW and MGW-P), the reduction in the amount of water and the better packing of particles of the MGW-P mortar resulted in a more compact material, with smaller pores (0.35 mm) and more well distributed. In the same way, in the REF-P sample, there is a greater amount of smaller pores, approximately 0.70 mm, confirming the refinement caused by the use of the plasticizer and the reduction in the amount of water. However, it was still possible to observe interconnected pores, as indicated in point 2. The introduction of waste also reduced the pore size in the MGW mortar to dimensions of approximately 0.60 mm. However, larger and deeper pores were identified (points 3 and 4), which corroborates the higher porosity resulting from the sample.

Other properties such as porosity, capillarity coefficient and immersion absorption are shown in Figure 6. The results indicate that the lowest values of immersion absorption and porosities were obtained by mortars containing plasticizer additive, due to the lower w/c ratio, while the lowest capillarity coefficient was obtained by the mortar REF. Comparing the mortars with the same w/c factor (REF and MGW), an increase of 19.18% in the water absorption of the MGW sample is observed, although its total porosity has increased by only 1.13% (not representative). This increase in permeability is also represented by the higher capillarity coefficient of this sample. Between REF-P and MGW-P mortars there is a difference of 6.86% in total porosity, lower in MGW-P, associated with better particle packing (HADAD *et al.*, 2020), promoted by cement replacement by waste.

Figure 5 - **magnifying glass (1000x) images of the internal section of the specimens**



Singh *et al*. (2017) point to a higher density of concrete containing marble waste in the w/c factor of 0.40, when compared to concretes with w/c of 0.45 and 0.35, which indicates the existence of an optimal range (from the study in question) of water consumption to obtain more durable materials using MGW. In general, the results suggest that the REF sample presented a large volume of voids, but a smaller number of communicating capillary pores, while the MGW sample presented a more refined structure and interconnected voids, with a void volume similar to the previous sample. The REF-P and MGW-P samples presented a smaller amount of voids and smaller pores, compared to the other samples.

Dynamic modulus of elasticity

The results of the modulus of elasticity test are shown in Figure 7(a) and the relationship between this property and porosity is illustrated in Figure 7(b).

According to Apolinário (2014), the propagation speed of an ultrasonic wave in a solid material depends on several factors from the physical characteristics of the materials, degree of hydration, to the type of densification. In addition, the less porous the material, the shorter the propagation time and the higher the modulus of elasticity. In this study, a good relationship between porosity and modulus of elasticity was not observed, which suggests that not only the number of pores in the structure affects this property, but also their distribution in the matrix.

Pacheco *et al*. (2014) also state that the experimental determination of the modulus of elasticity of concrete is influenced by the test method, yield stresses, geometry of the specimens, degree of concrete saturation and others, in such a way that the dispersion of the results is still considerable. This dispersion was observed in the results, reaching 11% of variation in the same specimen. Thus, the resulting dynamic modulus of elasticity of all mortars was statistically equal.

Figure 6 - **Result of water absorption by immersion (Ai), capillarity coefficient (Cc) and open (open P) and total (total P) porosity**

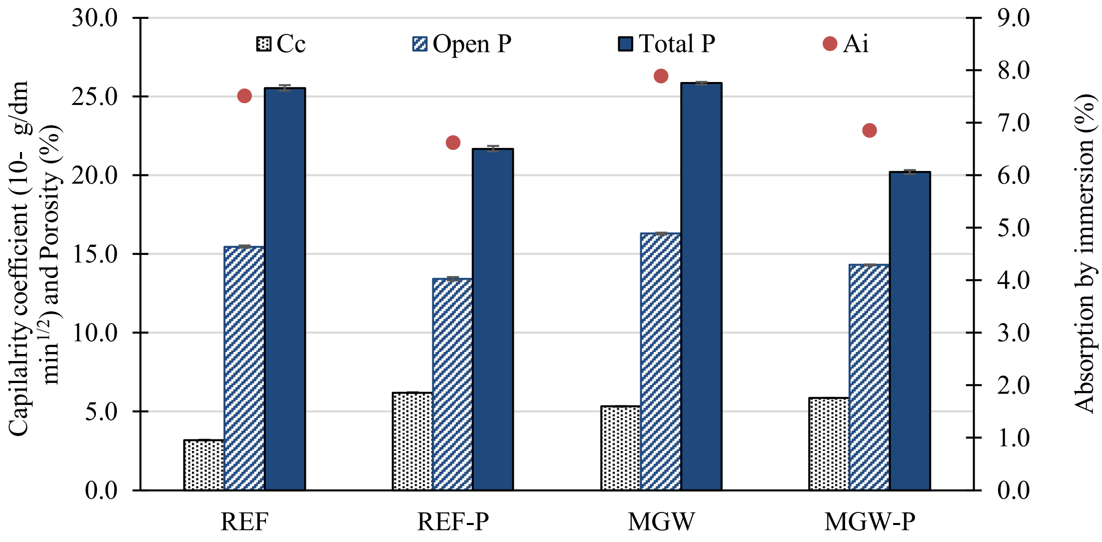
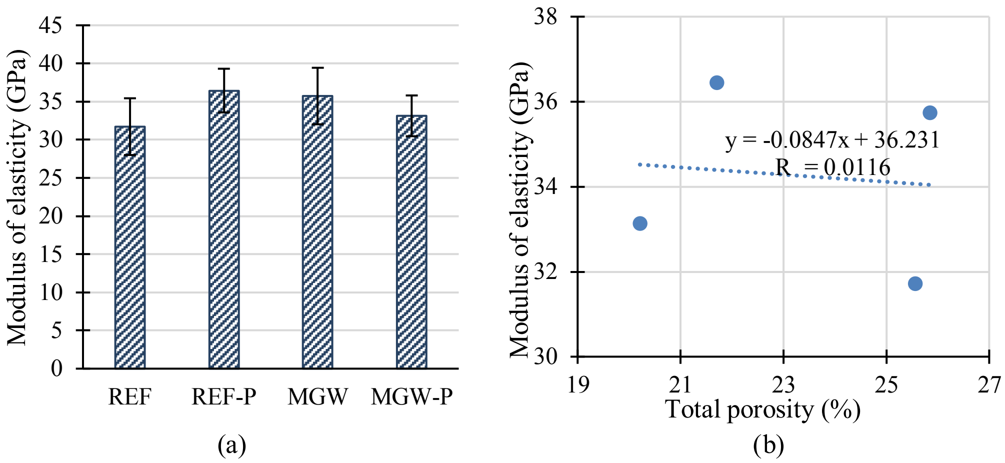


Figure 7 - **(a) Result of the dynamic modulus of elasticity (b) relationship between modulus of elasticity and porosity**



Considering the average value of the results, it can be observed that the use of the plasticizer additive and the cement replacement by MGW causes an increase in the modulus of elasticity from 31.7 (REF) to up to 36.4 GPa (REF-P). According to the study by Marques *et al.* (2020), cement mortars have a modulus of elasticity between 5 and 23 GPa. However, the values obtained in this study were closer to those found in the literature for concrete with marble waste (UYSAL; YILMAZ, 2011; BACARJI *et al.*, 2013; TENNICH; KALLEL; OUEZDOU, 2015) and structural mortars (ALMADA *et al.*, 2020). According to Uysal and Yilmaz (2011) and Almada *et al.* (2020), cement replacement by waste caused an increase in property, while for Bacarji *et al.* (2013) and Tennich, Kallel and Ouezdou (2015), the module was reduced. It is noteworthy that the methods for evaluating this property were different in each study.

Regarding the REF mortar, the REF-P and MGW-P mortars showed an increase in property of 14.88% and 4.44%, respectively, which is mainly due to the reduction in porosity caused by the lower w/c. The MGW mortar, despite having a porosity close to the REF, showed an increase of 12.64% in the modulus of elasticity. Therefore, it can be inferred that the waste led to a more homogeneous porous structure, resulting in a more intact mortar, with less propensity to internal failures.

Electrical resistivity

Cementitious material durability can be evaluated by measuring the electrical resistivity, which indicates the corrosion potential of reinforcement (SILVA; FERREIRA; FIGUEIRAS, 2011). The results are shown in Figure 8.

In general, electrical resistivity is dependent on the pore structure. However, the chemical composition of the solution and the presence of ions in the pores, as well as their mobility are also parameters that influence this property, although they do not significantly affect the compressive strength (SILVA; FERREIRA; FIGUEIRAS, 2011; VIJAYALAKSHMI *et al.*, 2013; KHODABAKHSHIAN *et al.*, 2018). Based on the results, it can be observed that the electrical resistivity followed the inverse behaviour of porosity and the samples with plasticizer, REF-P and MGW-P, when compared to REF, and showed higher electrical resistivity, 49.67% and 20.16%, respectively. This is due to the w/c factor, which is inversely proportional to resistivity, as reported by Sardinha, Brito and Rodrigues (2016). Furthermore, cement replacement by waste reduces the $Ca(OH)_2$ content in the mixture (dilution effect), decreasing the amount of OH-ions in the pore solution, which, according to Ramezanianpour *et al.* (2011), has great conductance. Thus, the electrical resistivity of compounds containing MGW should be greater than the reference. However, comparing the REF and MGW samples, it can be observed that there was no significant influence of the waste on this property, which resulted in a reduction of only 3.06%. This indicates that permeability is a more determining factor for resistivity than the chemical composition of the solution in the pores.

Whiting and Nagi (2003) present the corrosion potential ranges for concrete according to electrical resistivity, which is very high for values below 50 Ωm, high for values between 50 and 100 Ωm, moderate to low for 100 to 200 Ωm and low for values above 200 Ωm. However, as reported by Hou *et al.* (2017), this property is greatly influenced by coarse aggregates, which can present very high resistivity and, generally, of greater magnitude than that of cement paste, in addition to representing obstacles to the passage of current. The authors obtained a resistivity of 33 Ωm for mortar of composition 1:1 and w/c 0.40, a value close to those obtained in this research, and an increase of almost 50% when inserting coarse aggregate.

Compressive strength

Figure 9 presents the results of the compressive strength of the mortars. It can be observed that the highest strengths were achieved by the mortars containing the additive, as expected, due to the reduction of the w/c factor. The gain in compressive strength, in relation to the REF mortar, was 11.19% and 10.18% for REF-P and MGW-P, respectively. Despite having a lower w/c ratio and lower porosity, promoted by the filler effect of the waste, the MGW-P mortar did not obtain a better result than the REF-P. This is due to the reduction of the components responsible for the compressive strength of the mortar, when replacing the cement with the waste. It is likely that the chemical effect of the calcitic fraction of MGW to form hydrated carbonate products was not sufficient to maintain this property. This behaviour can be observed for MGW mortar, which had a small reduction (5.09%), as well as for cementitious materials in several studies (ELMOATY, 2013; ALIABDO; ELMOATY; AUDA, 2014; SINGH *et al.*, 2017; KHODABAKHSHIAN *et al.*, 2018). These authors point to an optimal replacement content between 5 and 15%, observing gains in compressive strength due to the filler effect.

Comparing the mortars containing waste (MGW and MGW-P), the reduction in the amount of water and the better packing of particles in the MGW-P mortar resulted in a more compact cementitious material, with a better porous structure, leading to 16.08% strength gain, Based on the results, it can be stated that the REF and MGW, REF-P and MGW-P mortars are similar, two by two, indicating that the w/c factor is mostly responsible for maintaining strength.

Accelerated carbonation

Figure 10 shows the carbonation results and Figure 11 shows the photos of the specimens after spraying the phenolphthalein solution.

The reference samples (REF and REF-P) did not show significant carbonation after 60 days in the chamber, although there was a difference of 15.08% between the total porosities. Thus, the use of plasticizer additive and the reduction of the w/c ratio were not influencing factors for the sample without waste. In the samples with waste (MGW and MGW-P), which showed a difference of 21.81% in total porosity, a great carbonation depth was identified for both samples. However, the MGW-P sample presented a more homogeneous carbonation front. This result can be attributed to the dilution effect of the waste (ALMADA *et al.*, 2020). Since the cement was replaced by a material that does not have pozzolanicity, the volume of hydrated products is smaller. Despite the reduction in porosity caused by the better packing of particles generated by MGW, the reduction in the alkalinity of the mortar was more decisive for this property.

Figure 8 - **Electrical resistivity result**

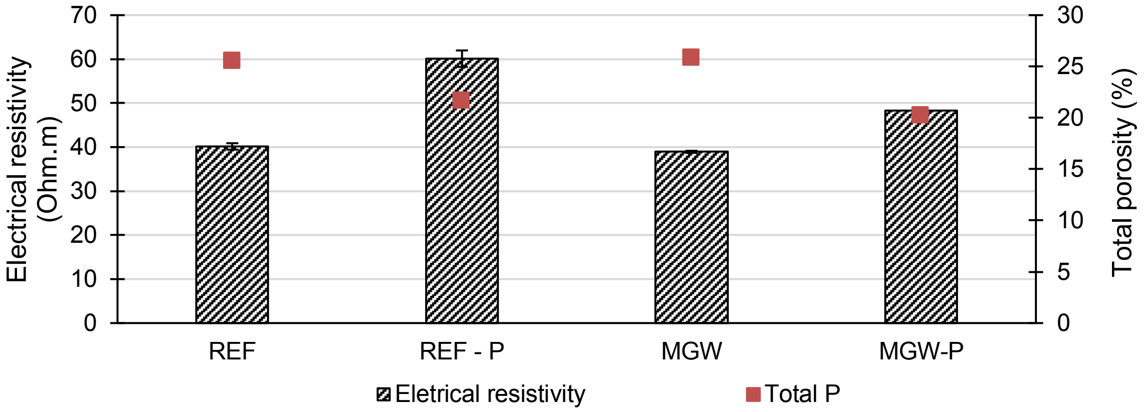


Figure 9 - **Result of compressive mortar strength (f_c)**

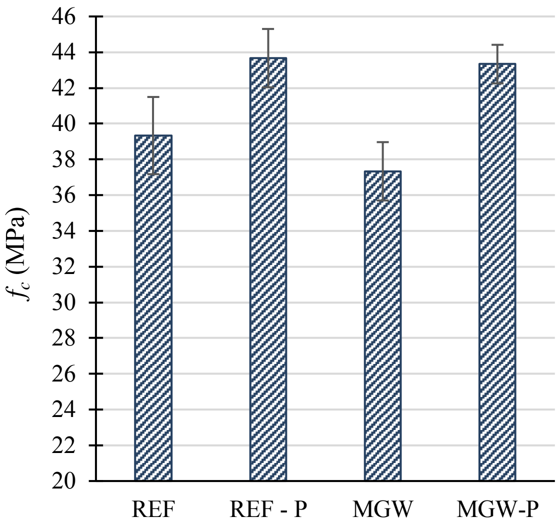


Figure 10 - **Result of the carbonation depth of the mortars**

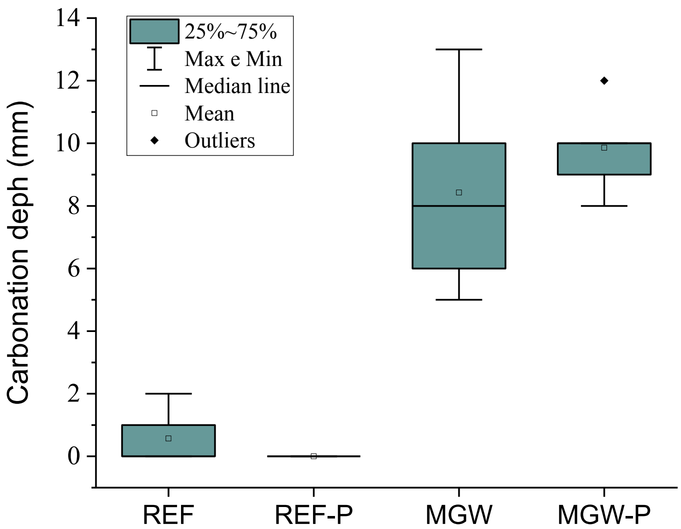
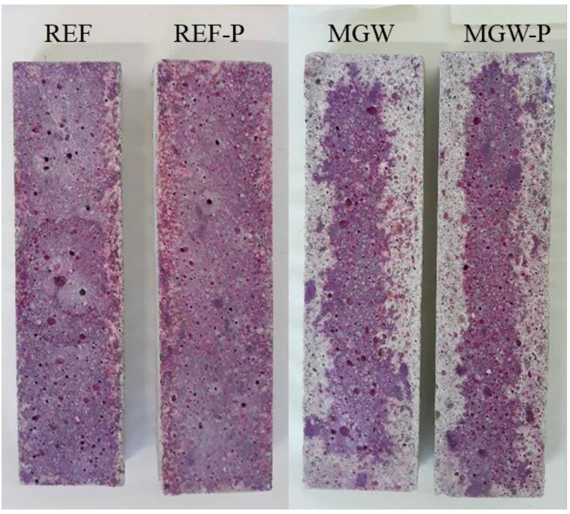


Figure 11 - **Photo of carbonated specimens**



Regarding the possible chemical effect of the carbonate components of the waste, the interaction of hemicarboaluminates, from the reaction of calcium carbonate with the aluminates of cement, with CO_2 can lead to a decarbonation process in the conversion of hemi to monocarboaluminate (DHANDAPANI *et al*., 2021). However, in this case, the dilution effect affected carbonation resistance more strongly.

Conclusion

After the experimental evaluations, it can be concluded that the waste mainly consists of silica, with a specific mass lower than that of cement and granulometry close to, but higher than that of cement. The insertion of the waste and the plasticizer additive allowed the maintenance of the workability with a greater reduction in the amount of water, promoting a refinement of the pores and an increase in the capillarity coefficient. There was no significant influence on the modulus of elasticity of mortars containing waste, with and without plasticizer, due to the high sensitivity of the test. However, by the average results, it is concluded that the waste improved the porous structure of the mortar, making it more homogeneous. There was an improvement in the electrical resistivity of the MGW-P mortar, compared to the reference mortar, due to the reduction of the w/c factor, lower pore connectivity and lower amount of OH-ions in the pore solution.

However, concerning the possibility of reinforcement corrosion, there was no difference between all samples. In the compressive strength, cement replacement by waste led to a small reduction in the property in the highest w/c factor. However, the variation of water content in the compositions was more relevant for the difference in compressive strength between the samples. Thus, the plasticizer and the waste generated a compact mortar, with a dispersed distribution of pores, allowing a compensation for the reduction of the cement content. This compensation did not occur in the accelerated carbonation test as the reduction of alkalinity with cement replacement by an inert material led to greater carbonation depths, regardless of the w/c ratio. In general, it can be observed that the waste is more efficient at lower water/cement ratios and its use as a filler promotes better particle packing, contributing positively to most properties, in addition to helping search for sustainable materials.

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