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# Use of ornamental rock waste as a partial substitute for binder in the production of structural concrete

### Uso do resíduo do beneficiamento de rochas ornamentais como substituto parcial ao aglomerante na produção de concretos estruturais

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#### Abstract

Due to the population increase, to the improvement of life conditions, to the elevation of levels of consumption and the growing industrialization of developing countries, it is estimated that the production of concrete will present significant increase in the next decades. The process of producing cement is responsible for approximately 5 % of the emissions of  $CO_2$ , an expressive environment pollutant. In this context, this work presents an evaluation of the influence of partial substitution of cement by ornamental rock waste (ORW) on the physical and mechanical properties of concrete. ORW from a local marble and granite processing company was used. The waste was mineralogically characterized through X-ray diffraction (XRD) essays, energy-dispersive X-ray (EDX) microanalysis, and physically characterized through laser granulometry and specific mass. For this, was adopted a concrete trace as reference, produced with CP V-ARI cement (similar to ASTM Type III), from IPT dosage. Waste was used in proportions of 5 %, 7.5 %, 10 % and 12.5 % of substitution of cement by ORW, creating a concrete with proper resistance to Brazilian regulations regarding the classification as structural.

Keywords: waste management, ornamental rock waste, structural concrete, environmental impacts.

#### Resumo

Em decorrência do aumento populacional, da melhoria das condições de vida, da elevação dos níveis de consumo e da crescente industrialização dos países em desenvolvimento, estima-se que a produção de concreto apresentará expressivo crescimento ao longo das próximas décadas. O processo de produção do cimento é responsável por aproximadamente 5 % das emissões mundiais de CO<sub>2</sub>, poluente que causa danos expressivos no meio ambiente. Neste contexto, este trabalho apresenta uma avaliação da influência da substituição parcial de cimento Portland por resíduo do beneficiamento de rochas ornamentais (RBRO) em propriedades físicas e mecânicas do concreto. Utilizou-se o RBRO proveniente de uma empresa beneficiadora de mármores e granitos localizada na região sul do Rio Grande do Sul. A caracterização mineralógica do resíduo foi realizada por meio de ensaios de difração de raios X (DRX), microanálise de raios X (EDX), enquanto suas características físicas foram analisadas por ensaios de granulometria a laser e de massa específica. Adotou-se um traço de concreto de referência produzido com cimento CP V-ARI, utilizando o método de dosagem do IPT/EPUSP. A substituição do cimento Portland pelo RBRO foi realizada nos teores de 5 %, 7,5 %, 10 % e 12,5 % em relação à massa de cimento. Os resultados foram tratados por ANOVA e comparação múltipla de médias, indicando a possibilidade da substituição de té 7,5 % do cimento pelo RBRO, gerando um concreto com resistência adequada às normas brasileiras e com potencial uso estrutural.

Palavras-chave: aproveitamento de resíduos, resíduo do beneficiamento de rochas ornamentais, concreto estrutural, impactos ambientais.

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#### 1. Introduction

Concrete is the most used construction material in the world, with an annual production estimated in 23 billion tons, which corresponds to average consumption of 10 kg of concrete per person per day [1]. The manufacture of cement in particular has considerable impact over the environment, due to the high energy consumption and the emission of greenhouse effect gases, mainly carbon dioxide [2-3]. In 2016, cement industries were responsible for 5.6 % of the world's entire CO<sub>2</sub> emission. Compared to 2013, emissions due to burning of fossil fuels for cement production have increased 1.2% [4]. The CO<sub>2</sub> emission in the production of common Portland cement is composed of two sources: the is decarbonising of calcium carbonate (CaCO<sub>2</sub>), that releases approximately 528 kg of CO<sub>2</sub> per ton of cement, and the burning of fossil fuels, which releases approximately 367 kg of CO<sub>2</sub> per ton of cement, making a total of 895 kg of CO<sub>2</sub> per ton of cement [5].

The large environmental impacts generated by the cement industry foster the search for alternatives that make the production of concrete less aggressive to nature. Several studies have shown positive results in evaluating the incorporation of industrial waste into concrete, as this waste can increase the mechanical strength and durability-related properties of concrete.

Facing all that, the incorporation of industrial waste in concrete comes up as an alternative to minimize environmental impacts of cement production. Among such residuals, there is ornamental rock waste (ORW), a byproduct of the production of ornamental rocks, that has been showing potential to be used as a partial substitute for binder.

According to the Brazilian Association of Ornamental Rocks Industry (ABIROCHAS) [6], in 2017, the production of stones in Brazil was of about 9.24 Mt, of which 1.046 Mt were exported. It must be highlighted that 41 % of the blocks, in volume, were transformed into waste, producing 3.36 Mt of materials rejected during the processing of ornamental rocks. Thus, it is evident that the process of beneficiation of ornamental rocks is archaic, almost artisanal, and

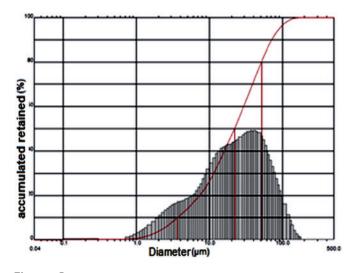


Figure 1 Granulometric distribution curves of ORW

that it has few investments in order to reduce the exorbitant waste generation during the process of rock processing.

In this context, the environmental issues exposed demonstrate the it is necessary to intensify studies on the ORW, with the objective of producing materials the incorporate it and, thus, mitigate its environmental impact.

Several researchers, like Ashish [7], Singh et al. [8], Rana et al. [9], Bacarji et al. [10] and Ergun [11], have demonstrated the viability of ORW as partial substitute for binder in the production of concrete. Most of these studies, however, focus only on mechanical properties. Considering that concrete is a hydrophilic material, studies that evaluate properties related to concrete durability, such as water absorption, are essential in the investigation of the feasibility of using ORW as a replacement for Portland cement in the production of structural concretes.

Thus, this work adopted a mixture of concrete produced with CP V-ARI cement (similar to ASTM Type III), as it contains the highest clinker content among the cements available in the regional market (southern Brazil) and, thus, consist of a "purer" cement in terms of mineral admixtures, and the feasibility of using the ORW as a partial replacement of cement was investigated. The waste was used in replacement of cement at the levels of 5 %, 7,5 %, 10 %, and 12.5 %.

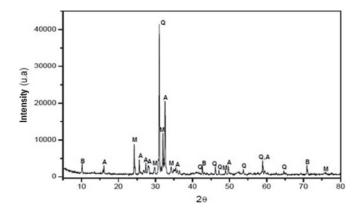
#### 2. Methods and materials

Following are presented the characteristics of the materials used to make the concretes that were studied, and the methods used to conduct the tests.

#### 2.1 Ornamental rock waste

ORW, collected as mud, was generated by a local company that processes ornamental rocks. The collection was made according to NBR 10007 [12], directly from the company's decantation tank, and all the material used in the work was collected the same day. After collection, the material went through a homogenization and quartering process, to obtain a representative sample. Then, the ORW mud was put into an oven, where it remained for 48 h, under a temperature of 100 °C. Next, waste was passed in a 1.18 mm sieve, to remove possible impurities and harrowing, eliminating the need to grind it. After that, it was passed in a 300 µm mesh sieve and stored in sacks, ready to be used. The granulometry of ORW was determined with a Cilas laser granulometer, model 1064. Figure 1 presents the data obtained in the test. The curve analysis shows that the average diameter of the ORW particle is 30,95µm.

To identify the presence of crystalline elements in the ORW composition, X-ray diffraction (XRD) analysis was carried out using a Shimadzu diffractometer, model XRD 6000, operating under radiation of CuK $\alpha$  (=1.5418 Å) and graphite monochromator, operating under a 40 kV tension and current of 30 mA, in a scanning range from 5 to 80° and angular velocity of 2°/min. Figure 2 presents the X-ray diffractogram of the waste. Analysing the results obtained from the XRD, it is verified the the ORW used is mostly composed of quartz (Q) and albite (A). It is also possible to observe less



#### Figure 2

X-ray diffractogram obtained for the ORW sample

intense peaks of microcline (M) and biotite (B). Through this essay, the non-pozolanicity of ORW was found, as a result of its quite defined crystalline peak regarding quartz ( $SIO_2$ ) and the absence of amorphous halo. So, ORW acts mainly as a nucleation agent within the microstructure.

To determinate ORW's chemical composition, an X-ray fluorescence spectrometer for energy dispersion, model Shimadzu EDX-720, was used. Table 1 presents the results of this test. According to NBR 12653 [13], the sum of oxides  $SiO_2$ ,  $Al_2O_3$  and  $Fe_2O_3$  must be higher than 70 % for the analysed material to be considered pozzolanic. The sum of them was 62.42 %, so, according to NBR 12653 [13], the material is not considered pozzolanic, presenting only physical effect, which corroborates what was previously discussed.

Table 2 shows results of the essays of ORW specific real mass, through NBR NM 23 [14], and unitary mass, according to NBR NM 45 [15].

## Table 1Chemical composition of ORW

Element	Quantitative (%)
SiO <sub>2</sub>	34.085
K2O	20.287
Al <sub>2</sub> O <sub>3</sub>	18.77
Fe <sub>2</sub> O <sub>3</sub>	12.57
CaO	12.181
TiO <sub>2</sub>	1.444
MnO	0.178
ZnO	0.122
ZrO <sub>2</sub>	0.118
SrO	0.103
CuO	0.101
Rb <sub>2</sub> O	0.024
Y_2O_3	0.006

#### 2.2 Cement

The cement used was CP V-ARI (similar to ASTMType III), as it possesses additions without reactivity, according to NBR 5733 [17], making it easy to understand the action of ORW and avoiding combined effects. That makes it possible to clearly visualize the effects of the substitution of cement by ORW in concrete.

#### 2.3 Aggregates

A natural medium-sized quartz sand, fit into the usable zone of NBR 7211 [16], was used and oven-dried until mass constancy. Granite gravel was classified as gravel 1 according to NBR 7211 [16]. Results of physical characterization obtained in natural aggregates are presented in Table 2.

#### Table 2

Aggregate and ORW characterization

Test method		Fine aggregate	Coarse aggregate	ORW
Granulometric composition/	Maximum Ø (mm)	4.75	19	_
NBR NM 248 (ABNT, 2003)	Fineness modulus	2.84	4.69	_
	/ NBR NM 52 (ABNT, 2009) 53 (ABNT, 2009)	2.62	2.6	2.64
	mass (g/cm³) / 5 (ABNT, 2006)	1.55	1.41	1.16

#### Table 3

Unitary mixtures used in this study

Mix	Cement (kg)	ORW (kg)	Sand (kg)	Gravel (kg)	w/c ratio
Reference	1	0	2.4	3.28	0.60
5 %	0.95	0.05	2.4	3.28	0.60
7.50 %	0.925	0.075	2.4	3.28	0.60
10 %	0.9	0.1	2.4	3.28	0.60
12.50 %	0.875	0.125	2.4	3.28	0.60

#### Table 4

Methodology of the essays conducted

Properties		Details	Methodology	
Mechanic	Compressive strength	5 samples by mixture at ages: 3, 7 and 28 days	NBR 5739 (2007)	
	Water absorption by immersion	3 samples by mixture at age: 28 days	NBR 9778 (2009)	
Physical	Sorptivity	3 samples by mixture at age: 28 days	NBR 9779 (2009)	

#### Table 5

Analysis of variance results - p value - of compressive strength

ANOVA table					
Factor	DF	Sum of squares	Average square	F-value	P-value
Substitution level	4	78.88236	19.72059	6.928441737	0.000118253
Age	2	1241.111976	620.555988	218.0201509	3.01E-28
Substitution level x Age	8	20.215224	2.526903	0.887777709	0.532279889
Residuals	60	170.77944	2.846324	—	—

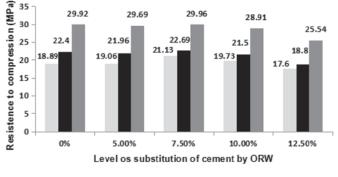
#### 2.4 Concrete production and assessment

For concrete dosage, was used the dosing methodology by IPT/ EPUSP [17]. Through an experimental procedure, the ideal percentage of dry mortar was set as 51% ( $\alpha$  = 0,51) and the amount of water necessary to obtain the depletion of the marsh cone as 70 ± 10 mm.

Through dosage equations and the pre-established value of the relation water/cement of 0.60, we determined the concrete's reference trace, according to Table 3. The relation of water/cement of 0.60 was defined because it is the maximum value considered for a structural concrete located in urban environment (Class II of aggressiveness), according to NBR 6118 [18].

From the reference mixture, partial substitution of cement by ORW was carried out, in levels of 5 %, 7.5 %, 10 % and 12.5 %. The choice of these values was based in the studies by Singh et al. [8], Rana et al. [9] Bacarji et al. [10] and Ergun [11].

In Table 4, there are details about all properties studied in this work, as well as the number of samples and the regulations used



🗏 3 days 🔳 7 days 🔳 28 days



in procedures. All specimens were moulded and cured according to the recommendations of NBR 5738 [19].

Finally, to understand the results obtained in terms of physical and mechanical properties of concrete, and if the independent variables analysed (age and replacement levels) are important in their alteration, they were submitted to statistical treatment by analysis of variance (ANOVA) and multiple comparison of means (Tukey's Test). The ANOVA, when indicates significance, makes sure that there are at least a couple of different averages, but not knowing how many and which ones. Thus, the Tukey's Test is necessary, where, in order to determine whether the pairs of means are different from each other, the comparison of means from two by two was made.

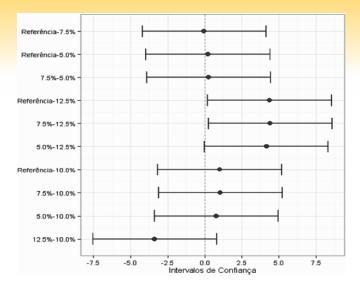
#### 3. Results and discussion

#### 3.1 Compressive strength

The means of the results of compressive strength along the ages are presented in Figure 3. At the first ages, except with value of 12.5 %, the mixes with replacemennt of cement by ORW provided greater resistance than the reference one, which demonstrates the physical action of the waste, once in this moment the greater part of the pozzolanic reactions did not occur. ORW's physical effect accelerates cement hydration when it acts as a point of nucleation for calcium hydroxide, as it possesses extremely fine particles, accelerating reactions and forming less crystals of calcium hydroxide. Through ANOVA, statistical significance of the related variables, which are replacement value and age in compressive strength was found. Table 5 presents the analysis of influence of these factors and possible interactions.

In Table 5, it is verified that the level of replacement and the age, when analysed separately, have significant effect over compressive strength.

However, there is no significant influence between the level of substitution and the age. The inexistence of interaction between



#### Figure 4

Tukey's Teat for concretes' compressive strength at 28 days

substitution level and age shows that substitution does not lead to significant increase throughout time. This behaviour was already expected, as the waste does not have pozzolanic activity.

Analysing the data in Table 5, it is possible to observe that the p value is lower than the expected level of significance ( $\alpha = 0,05$ ), so, the null hypothesis is rejected. This way, Tukey's Test was applied and the results obtained are shown in Figure 4.

Analysing the data obtained through Tukey's Test, Figure 4, it is possible to observe that, at 28 days, only the concrete with 12.5 % of replacement of cement by ORW can be considered statistically different from the reference concrete. At this level of substitution, the cementing effect has shown as preponderant regarding filler effect, causing a decrease of resistance, compared to the reference trace. Although the concrete with 10 % of substitution presents reduction in compressive strengthcompared to the reference concrete, this decrease in resistance cannot be considered significant, with a reliability level of 95 %.

The result obtained in the study corroborates with the results obtained by Ashish [7], Ergun [11], Agarwal; Gulati [20],Kockal [21], Ramos et al. [22]; Aliabdo et al. [23] and Munir et al. [24], who obtained increased compressive strength by replacing cement with ORW in the levels of 5 % and 7.5 %.

#### 3.2 Absorption by capillarity

The results obtained in the test of sorptivity are presented in Figure 5. The essay demonstrated a reduction of the absorption tax in the substitution levels of 5 % and 7.5 %, with the content of 7.5 % presenting the best performance (reduction of 10 %).On the other hand, thereplacement of 12.5 % presented the worst results, with an increase of 22.70 % in the sorptivity tax. This result of sorptivity aids to explain the decrease in compressive strength

#### Table 6

Analysis of variance results - p value - of absorption by capillarity

ANOVA table					
Factor	DF	Sum of squares	Average square	F-value	P-value
Substitution levels	4	0.244210037	0.061052509	22.57819744	5.42E-05
Residuals	10	0.027040471	0.002704047	_	_

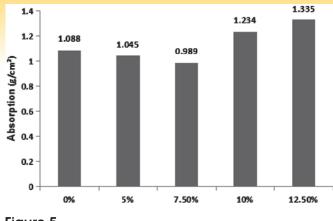


Figure 5 Water absorption by capillarity

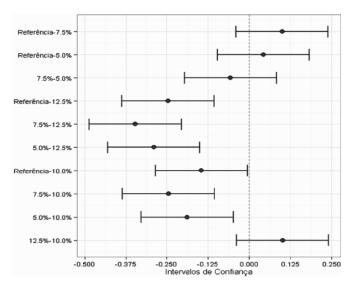
in substitution levels above 7.5 %. In this substitution value, the cementing effect has shown as preponderant regarding the filler effect, causing an increase of the sorptivity tax.

Through ANOVA, statistical significance of the variable substitution level in sorptivity was found. Table 6 presents the analysis of influence of this factor.

In Table 6, it is verified that the level of substitution has significant effect over water absorption through capillarity.

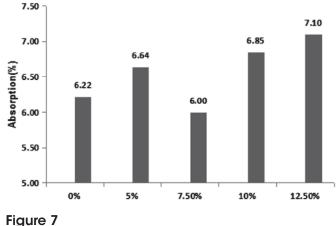
It is possible to conclude, based on the analysis of variance, that the hypothesis of the means being equal was rejected and that the effect of the relation of the substitution level is significant, with reliability of 95 %.

Through Tukey's Test, Figure 6, it was found that concrete with 10 % and 12.5 % of replacement of cement by ORW can be considered statistically different from the reference one.



#### Figure 6

Tukey's test for absorption by capillarity



Water absorption by immersion

#### 3.3 Absorption by immersion

Results of the measurement of water absorption by immersion are shown in Figure 7. The trace with 7.5 % substitution of cement by ORW presented better performance, with a decrease of absorption of 3.66 % compared to the reference mixture. All the other substitution levels promoted absorption increase, and the trace with 12.5 % substitution presented the worst performance, with increase of 14.14 % in the absorption tax, compared to the reference trace.

Through ANOVA, statistical significance of the variable replacement level in immersion absorption was found. Table 7 presents the analysis of influence of this factor.

In Table 7, it is verified that the level of replacement has significant effect over water absorption by immersion, indicated by the p-factor of 0.3956%, below 5 %.

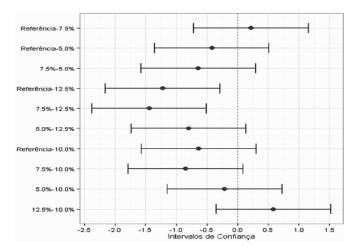
It is possible to conclude, based on the analysis of variance, that the hypothesis of the means being equal was rejected and that the effect of the relation of the substitution level is significant, with reliability of 95 %.

Through Tukey's Test, Figure 8, it was found that concrete with 12.5% of substitution of cement by ORW can be considered statistically different from the reference one.

#### 4. Conclusions

In this work, we aimed to verify the influence of partial substitution of cement by ORW over the physical and mechanical properties of concretes. With the results obtained, it was possible to conclude that:

a) Although reduction of the mechanical properties of concretes with ORW in substitution for cement was observed in 28 days,



#### Figure 8

Tukey's Test for absorption by immersion

there was no statistically significant difference regarding compressive strength in substitution values until 10 %;

- b) In the sorptivity test, mixtures with 10 % and 12.5 % presented statistically significant differences. These levels promoted increase of the sorptivity tax, in comparison to the reference mix;
- c) In the of absorption by immersion test, only the mix with 12.5 % replacement presented statistically significant difference in water absorption. This value promoted increase of the absorption tax, in comparison to the reference mixture;
- d) Concretes with 5 % and 7.5 % substitution did not present statistically significant difference in the tests of compressive strength, sorptivity and water absorption by immersion, compared to the reference mixture;
- e) The results show that the higher the waste content, the lower the compressive strength and the higher the water absorption rate. However, although smaller than the results obtained for the reference concrete, the compressive strength values indicate the possibility of use as a structural concrete. The results of the study demonstrate the partial replacement of the binder by OWR is satisfactorily feasible.

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#### Table 7

Analysis of variance results - p value - of absorption by immersion

ANOVA table					
	Factor	DF	Sum of squares	Average square	F-value
Substitution Levels	4	3.8088	0.9522	7.841339555	0.003956086
Residuals	10	1.214333333	0.121433333	—	—

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