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Original Article

Physical and mechanical evaluation of artificial marble produced with dolomitic marble residue processed by diamond-plated bladed gang-saws[☆]

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ABSTRACT

The production of artificial stone from the agglutination of the epoxy resin fine waste can be a viable alternative from technical, economic and ecological points of views. The residue derives from the dolomitic marble produced by diamond-plated bladed gang-saws and it is usually discarded. With this new application, there is value added to the residue, reducing the consequent environmental impact. The main objective of this research is to evaluate the physical and mechanical properties of an artificial marble produced through vacuum vibro-compression. With this purpose, stone plates were produced, using appropriate tools, characterized by a distribution of 85% of residual particle weight and 15% of epoxy system. The material was manufactured with the following conditions for 20 (twenty) minutes, compacting pressure of 0.5 MPa and temperature of 90 °C. Later on, the samples were characterized by density, water absorption, porosity, flexural strength, compressive strength and scanning electron microscopy. The result was an artificial marble produced applying vacuum vibro-compression with water absorption levels below 0.06%, flexural strength of 33.93 MPa and uniaxial compression of 96.49 MPa, thus indicating residual particles and epoxy resin grip at satisfactory levels.

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1. Introduction

Brazil is the fourth largest producer of ornamental stones in the world, with an estimated production of 9.5 million tons in 2015 [1].

During the production of ornamental stones, residues are generated mainly during the extraction and manufacturing of stone blocks into slabs, for instance, the waste generated by marble processing plants tends to be between 30 and 35% and these residues need larger areas for disposal daily [2].

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Since they cause damage to the environment, the stone residues, which are generally discarded, could be recycled and reused in a sustainable manner through the implementation of projects based on academic research.

The alternative of recycling the residues that come from manufacturing ornamental stones with diamond-coated tools proposed by this research is its agglutination with epoxy resin through vacuum vibro-compression, which would develop in return an artificial stone, in this case, a compact artificial marble.

The physical and mechanical properties of the artificial marble are the main influencing factors to its use in the construction industry. These properties are directly related to the resin content and the microstructure of the artificial marble which can be seen in the production process [3].

An advantage of the vacuum vibro-compression process is that the marble particles are bonded by the before molding. Vacuuming and vibrating during compaction in an efficient way may incur in more controlled void formation within the material due to its better efficiency in removing the air bubbles existing in the pre-cured mix [4,5].

Artificial marble is well accepted in the market and the companies that produce and offer this product mention various advantages. The main characteristic mentioned is its impermeability, which offers protections against stains, which is attributed to the resin in its composition that bonds the Stone particles and fill in the typical porosity of the stone in its natural state. This way there is no penetration or percolation of liquids in the stone's interior [6].

In Brazil and the rest of the world, throughout the last few years, the demand for artificial stones has constantly increased. Between 2011 and 2016 alone, it has been estimated that there would be a 36% increase in its demand [7].

The main purpose of this research is the production of an artificial marble from the residues of dolomitic marble, with good physical and mechanical properties tied to an efficient bonding of the particles by the epoxy resin to a microstructural level, and to compare this artificial marble produced with other artificial marbles manufactured by the industry.

When the recycling of the residues of ornamental stone production is proved as a viable alternative from both, technical and ecological viewpoints, it will be possible to aggregate value to something that, while adequate to environmental laws present, would be otherwise discarded in nature.

2. Materials and methods

2.1. Dolomitic marble waste

Two types of residues originated from manufacturing of natural dolomitic marble were used in this research; this marble is commercially called "Marmo-Onyx Blue", originated from the company Santa Clara, located in Cachoeiro de Itapemirim, Espírito Santo State, Brazil.

The first residue is semisolid and corresponds to the material collected directly at the company, in an abrasive clay aspersión system, called shower, from their diamond plated steel gang saws. Upon collection, this residue was dried on an oven to extract the humidity at 70 °C for 24 h. This

process of humidity removal was used to increase the efficiency of the curing process as well as the adhesion between the epoxy resin and the dolomitic marble particles [8]. The second residue is solid and corresponds to the leftover stone discarded from manufacturing of the marble at the same gang saw; the leftover stone was machine-crushed and then ground in a porcelain disk grinder, sieved by vibratory sieves with mesh between 10 and 200 in two grain sizes, coarse (2.000–0.425 mm) and medium (0.425–0.075 mm). The dry clay was also sieved and separated to a fine grain size (<0.075 mm).

Taking in consideration the three ranges of grains obtained, there were determined 10 mixtures with different percentage of coarse, medium and fine particles. A complete ternary diagram of cubic modeling was utilized, it was developed in the experimental numeric-modeling grid Simplex (Simplex-Lattice Design) [9]. The main objective was to determine which one of these mixtures presented the maximum apparent dry density. The mixture with highest apparent dry density represents the highest packing of the particles (4/6 of coarse particles, 1/6 of medium, and 1/6 of fine), which was chosen for the production of the slabs of artificial stone.

2.2. Epoxy resin

The marble residue particles were agglutinated by the epoxy resin of type diglycidyl ether of bisphenol A (DGEBA) hardened by a commercial amine (TEPAC) composed approximately of 60% Tetraethylenopentamine (TEPA) and 40% of Triethylenotetramine (TETA) with some impurities, medium viscosity, 1.97 g cm⁻³ density at 20 °C, and a gel time of 70–90 min at 20 °C.

2.3. Artificial marble production

A minimum resin rate was calculated to be used to fill the porosity within the particles, and thus the mixture utilized was 85% particles and 15% epoxy resin.

The stone plates were produced with dimensions 200 mm × 200 mm × 10 mm by the vacuum vibro-compression method. The mixture was put in vacuum while at the same time a pressure of compaction of 0.5 MPa at 90 °C was being applied for 20 min, resulting in its curing.

Later on, the plates were cut in the dimensions specified in the tests performed to characterize it.

2.4. Water absorption, density and apparent porosity

The values of density, water absorption and apparent porosity were obtained from the physical index tests ruled by the norm ASTM C373-16 item 5.3 [10].

There were 10 test bodies made with dimensions 30 mm × 30 mm × 30 mm.

2.5. Three-point flexural strength

To perform the test for three-point flexural strength a universal test machine EMIC model DL10000 was used. The test is standardized EN 14617-2 [11]. In total there were 6 test bodies used with dimensions 10 mm × 25 mm × 100 mm. At the end of

the test the maximum tensile stress and standard deviation were calculated.

2.6. Compression strength

The test for compression strength was based on the establishments of the norm Spanish UNE-EN 14617-15 [12] and was performed in a universal test machine EMIC model DL10000. In order to reach the desired length for the test bodies of the artificial stone produced, three layers were glued together with epoxy resin. There were used 5 test bodies oven-dried according to the norm with dimensions 30 mm × 30 mm × 30 mm, the speed of load application was 0.5 mm/min until the final failure was reached. The maximum tensile stress and standard deviation were calculated.

Besides the artificial marble produced, a commercial artificial marble Bianco Prime, provided by Emporio Stone, was tested in the mechanical tests (flexural strength and compression) for further comparison of results.

2.7. Micro-structure

The micro-structure of the fracture of the test bodies submitted to flexural testing was evaluated through an Electronic Scanning Microscope, enabling the particle adhesion by the epoxy resin to be analyzed. The equipment used to perform these analyses was TMT3030PLUS manufactured by HITACHI. The samples were prepared previously using an adhesive carbon tape enveloped by a gold surface.

3. Results and discussion

3.1. Water absorption, density and apparent porosity

The results to the physical indexes found for the artificial marble produced by the vacuum vibro-compression method, its respective averages, and its standard deviations are presented in Table 1.

For the apparent density it was found the average value of $2.10 \pm 0.06 \text{ g/cm}^3$, which is approximately 15% less than the values informed by the companies that produce artificial stones.

In his work on artificial stones produced under varying of compression pressure, vibration frequency, and vacuum

levels, Lee et al. [4] reached density values within $2.03\text{--}2.45 \text{ g/cm}^3$. This is indicative that the artificial marble produced in this research has density within the values found by the author.

The lowered density reflects directly on the weigh per square meter of the tiles to be fabricated from the artificial Stone produced, so the material with the same dimensions would be lighter, which lower transportation costs.

In regards to the apparent porosity, the value found was $0.13 \pm 0.02\%$, which represents a satisfactory adherence between the particles and the resin.

Chiodi and Rodriguez [13] classified the materials destined to covering in construction and determined that they have high quality when they have porosity values under 0.5%. The apparent porosity found for the artificial marble produced is approximately 4 times lower than the one determined by the authors.

The water absorption of the artificial marble produced was found to be $0.06 \pm 0.01\%$. The value found is below the recommended value for natural dolomitic marble, which is indicated to have a value less or equal 0.20% [14].

Chiodi and Rodriguez [13] established that a value below 0.1% represents that the Stone has a very high quality, and the value found in the test is 40% below this classification.

Carvalho et al. [15] found the value 0.17% of water absorption. The artificial marble produced in this research has water absorption approximately 33% below the minimum range of values informed by the industry and almost three times below the one found by Carvalho et al. [15]. With these comparisons, the low water absorption of the material is proved for its application in humid environments.

3.2. Three-point flexural strength

In Table 2 the values are found for 3-point flexural strength of the 6 test bodies produced from the artificial marble developed with 85% dolomitic marble residue and 15% of the resin epoxy.

The standard deviation of 0.49 MPa found shows a very reduced range of variation in the mechanical behavior of the material in this test, allowing to state that the artificial Stone plates produced and utilized in the test were very homogeneous in their composition, which can be a reflection of an efficient interlocking of the polymeric chains while curing of the resin and its good adhesion to the residue particles throughout the entire stone surface.

Borsellino et al. [16]. Used marble particles and agglutinated them with epoxy resin without the method of vacuum vibro-compression and obtained a flexural strength value

Table 1 – Results of physical indexes.

Test body	Density (g/cm^3)	Porosity (%)	Water absorption (%)
1	2.21	0.15	0.07
2	2.07	0.12	0.06
3	2.19	0.12	0.05
4	2.03	0.15	0.07
5	2.11	0.12	0.06
6	2.11	0.12	0.08
7	2.05	0.16	0.06
8	2.11	0.12	0.07
9	2.08	0.15	0.06
10	2.08	0.12	0.06
Average	2.10 ± 0.06	0.13 ± 0.02	0.06 ± 0.01

Table 2 – Results of 3-point flexural strength.

Test body	3-Point flexural strength (MPa)
1	34.05
2	33.64
3	33.95
4	33.42
5	34.83
6	33.72
Average	33.93 ± 0.49

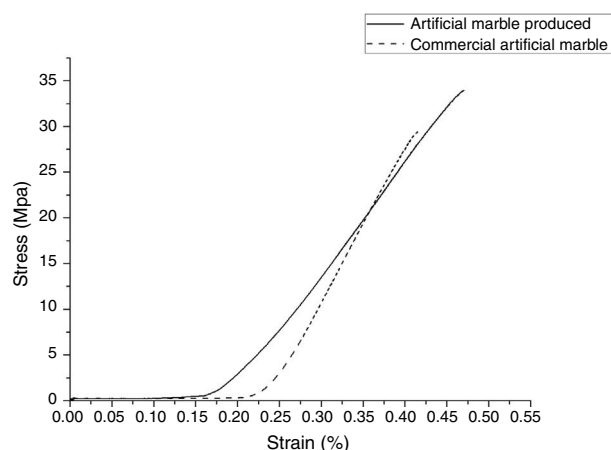


Fig. 1 – Comparison of mechanical behavior in the 3-point flexural strength.

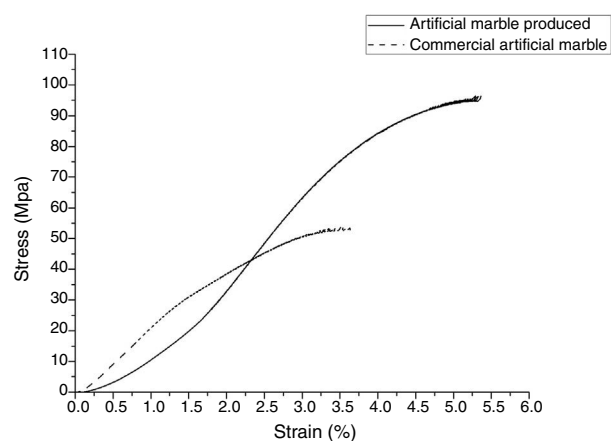


Fig. 2 – Comparison between the mechanical behavior in compression.

Table 3 – Results of uniaxial compression strength.

Test body	Uniaxial compression strength (MPa)
1	93.13
2	96.39
3	95.81
4	100.95
5	96.19
Average	96.49 ± 2.82

between 10.6 and 22.2 MPa. The strength value found for the artificial marble produced is 53% higher than the maximum value found by the author. This is because the use of the method of vacuum vibro-compression while producing the artificial marble in this research lowered the porosity levels, preventing the formation of tension concentration points.

The modulus of elasticity found was 7.10 ± 0.6 GPa, indicating that the artificial marble produced has high rigidity during bending. The good interfacial adhesion between the dolomitic marble particles and the homogenous dispersion in the epoxy resin, with good hydration, benefitted the increasing in the modulus of elasticity, being higher than the ones obtained for the epoxy system that defines the value for natural marble.

In Fig. 1 it is possible to establish a comparison between the mechanical behavior of the artificial marble produced and the commercial artificial marble. For the commercial artificial marble, the maximum tensile strength obtained was 30.42 ± 2.37 MPa, showing that the artificial marble produced has a 3-Point Flexural Strength almost 12% higher. This demonstrates that the material produced is in conformity with what is expected in terms of strength as a material made by the industry.

3.3. Compression strength

In Table 3 the results obtained in the uniaxial compression strength test are described.

A value of 96.49 ± 2.82 MPa was obtained in the compression strength test. This result shows that there was a good

interconnection in the polymeric chains of the resin, reducing slippage in the planes of weakness that normally happen in artificial marble when under compressive forces.

When this value is compared to the regulations defined by the ASTM C503 Standard for the raw material of natural dolomitic marble, it proves to be higher, being almost double the minimum established value by the norm, which is 52 MPa [14].

Lee et al. [4], using granitic and vitreous particles, which possess higher mechanical strength than the dolomitic particles used in this research, varied the parameters of artificial marble production and obtained values from 78.7 to 151.3 MPa.

For classification purposes, since there are no regulations in the artificial stone field, the values used were the ones defined by Chiodi e Rodriguez [13]. The authors define that, when used for structural purposes, the artificial stones should present compression strength values within 70 and 130 MPa for average resistance and this classification englobes the artificial marble produced in such category.

In the comparison between the commercial marble Bianco Prime and the artificial marble produced, the later obtained a result of almost double the resistance of the former, 96.49 ± 2.82 MPa against 56.46 ± 14.46 MPa, as can be seen in Fig. 2.

3.4. Microstructure

In Fig. 3(a–d) it is presented the microscopic analyses obtained by Electronic Scanning Microscope performed on the fracture sections obtained in the 3-point flexural strength test of the produced artificial marble.

There are few evident pores, because as proved by the physical indexes, the porosity is very low. Nevertheless, they may have contributed to the occurrence of the fracture, since it can be seen that there was good interfacial adhesion between the particles and the resin, meaning that the particles were well moistened by the resin.

Miller et al. [17] and Debnath et al. [18] described that good interfacial interactions in the composite could benefit its mechanical properties, and that this interaction is directly

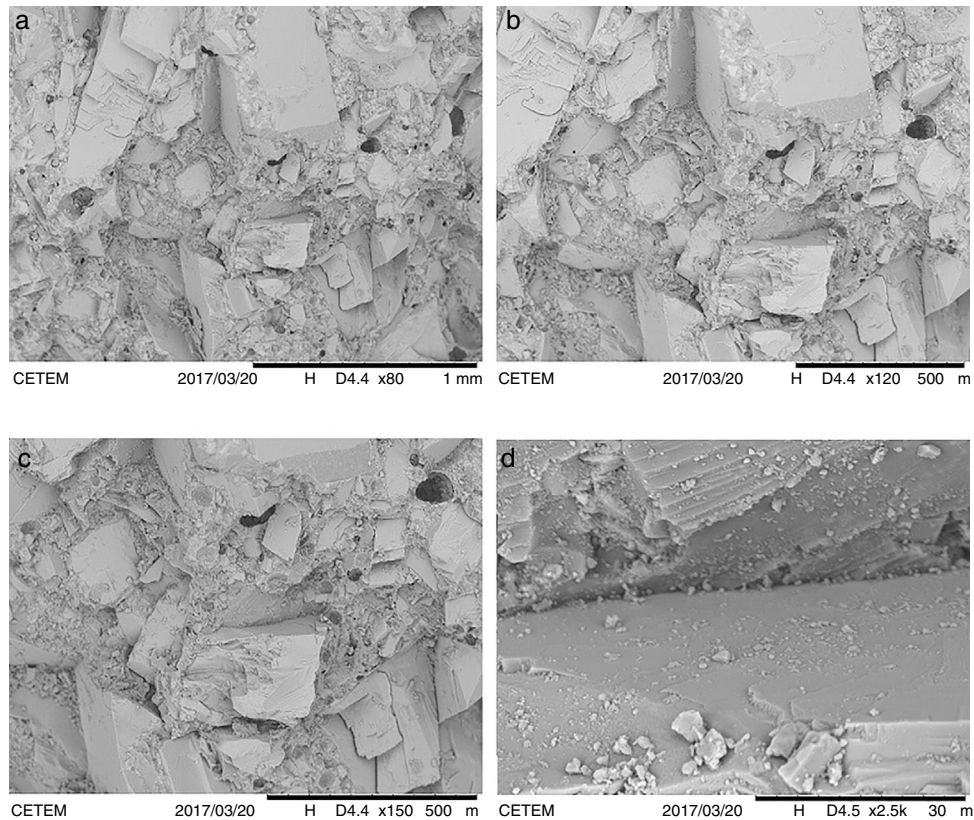


Fig. 3 – Electronic scanning microscopy of the fracture section.

related with the strength of the adhesive force provided by the successful moistening of the interfacial regions.

Karaca et al. [19] cites that Dolomite is a mineral with hexagonal crystalline structure, commonly presenting rhombohedral crystals with curved faces.

The intergranular fracture happened mainly in the cleavage planes of the dolomite, because it has rhombohedral shaped surfaces. This fracture happened due to the mechanical force applied on these weakness planes. In the interfacial region, the good adhesion allowed, as seen by the flexural and compression strength tests, a satisfactory result for these properties.

It is inferred that the compaction foreseen in the ternary diagram resulted in good adherence in the interfacial regions and few surface failures, resulting in the low porosity found; this increases its luster index when the material goes through manufacturing such as polishing of the surface and borders. These factors create a surface less penetrable by liquids, making it more difficult for stains to form, and they increase the mechanical properties as seen, which were higher than the properties of the artificial marble analyzed.

4. Conclusions

It was verified through the characterization of the artificial marble produced by the vacuum vibro-compression method, with compaction pressure of 0.5 MPa, at a 90 °C temperature, for a 20-min period and constituted by 85% in weight of

dolomitic marble residue from diamond-plated bladed gang saws and 15% of DGEBA/TEPac epoxy system, that:

- A compact artificial marble with excellent physical and mechanical properties was produced. It has only 0.13% and 0.06% of porosity and water absorption, respectively, emphasizing its high impermeability. The 3-Point Flexural Strength Test indicated that the material has maximum tensile strength of 33.93 MPa and 96.49 MPa in compression, higher than a similar commercial artificial marble, which shows its high mechanical resistance.
- In the Electronic Scanning Microscopy, the fracture regions showed that the dolomitic particles adhered well to the DGEBA/TEPac epoxy system, with few visible pores, which was confirmed in the mechanical tests, highlighting how important it was to define the best mixture of particle size distribution and to choose the highest packing factor by the ternary diagram.

In this way, the agglutination of particles of marble residue from diamond-plated bladed gang saws by the DGEBA/TEPac epoxy system allows for the development of a compact sustainable artificial marble with excellent physical and mechanical properties.

Conflicts of interest

The authors declare no conflicts of interest.

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