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Characterization of Artificial Stone Developed with the Incorporation of Granite and Mirror Wastes in an Epoxy Matrix

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The objective of this research was to produce artificial stone plates based on granite and mirror wastes and epoxy matrix, by vibration, compression, and vacuum and to characterize them. Plates were manufactured with 15%wt epoxy resin and 85% of aggregates in the proportion of 1/3 of granite waste from Ocre Itabira gray granite and 2/3 of mirror waste. The apparent density, water absorption, and apparent porosity values were 2.22 g/cm^3, 0.11%, and 0.25%, respectively, the flexural strength was 34.36 MPa, abrasive wear after a 1000m track was 2.28mm and the breaking height in the impact resistance test was 0.45m. In addition, the stone was resistant to several staining agents. Therefore, the technical viability of the material developed was verified, with results compatible with studies already carried out in the area, making it possible to apply it as coatings and countertops in civil construction.

Keywords: *Artificial stone; granite waste; mirror waste; epoxy resin.*

1. Introduction

Industrial activities frequently generate some types of waste that are hazardous to the environment, provoking concern about the correct disposal of these materials. On that account, studies aiming at the reuse of wastes to develop new materials have been rapidly increasing worldwide.

It is known that around 130 million tons of glass are globally produced per year. Nevertheless, the amount of recycled glass is low, around 27 million tons[1]. In addition, waste mirrors and flat glass require greater complexity for recycling[2] and therefore often end up in landfills or are wrongly disposed of. Consequently, a considerable part of this material needs novel applications for its reuse.

Likewise, in the ornamental stone industry, there is also a high amount of waste generation covering all stages of production[3,4]. In that scenario, Brazil is placed as one of the world's largest producers of ornamental stones and this market is rapidly expanding and enhancing its production volume[5], consequently generating more and more waste in different ways.

Considering the depletion of the natural resources supply, the recycling of stone fragments and glass waste when reused in the development of novel materials, not only creates new products but also offers an ecological and economical alternative for waste treatment[6]. Furthermore, research has shown that recycling waste from stone mining to develop artificial stones is technically and ecologically feasible[7,8].

For this reason, the authors have chosen to reuse waste from ornamental stone mining and waste glass from mirrors in developing artificial stones.

Artificial stones are polymer matrix composites that incorporate stony components, such as natural quartz and other minerals, usually in the range of 90 to 94% wt[9]. According to Lee et al.[6], artificial stones have superior properties when compared to natural stones, due to their low porosity, low water absorption, and high mechanical resistance. Besides, artificial stones are usually lighter than natural ones, which is attributed to its polymeric matrix, a low-density material, that turns artificial stone into an advantageous material for the execution of architectural and structural projects, diminishing costs related to the material's weight, such as transport logistics[7,10]. Hence, artificial stones are suitable materials for applications in wall coverings, floors, and countertops.

When analyzing the Brazilian ornamental stone market, ABIROCHAS[11] shows that artificial stone imports are twice greater than natural stone imports, pointing out its incipiency and highlighting the relevance of the research carried out in this area, for the development of novel materials for the Brazilian market.

Although artificial stone is a material whose composition comes mostly from mineral fillers, much research has been carried out with the incorporation of different types of wastes in polymeric matrices, with the aim of recycling and also of improving the physical, chemical, and mechanical properties. Among the aggregates commonly used are particles of glass,

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marble, granite, quartz, steel waste, crushed stone's powder, brick waste, iron ore waste, among others, agglomerated by matrices like epoxy polyester, polyurethanes, and natural source resins[6-10,12-21].

Epoxy matrix composites have been heavily researched in recent years[22]. Allied with this, the epoxy resin has good mechanical, thermal, and chemical properties as well as resistance to corrosion, low viscosity, and good adhesion to other materials[23]. Not to mention epoxy resin has slightly higher strength and stiffness than polyester, and also has comparatively lower density and better ductility[8], which motivated the use of epoxy resin as a binder in this research.

The vibration, compression, and vacuum (VCV) process is today one of the most used for artificial stone manufacturing, due to its efficiency in producing parts with excellent physical and mechanical properties and diverse aesthetic standards. According to Rubio et al.[24] the silestone, a type of artificial stone, is manufactured with the process of vibration, vacuum, and compression. First, the raw materials were mixed with polyester resin, and after homogenization, the mixture was placed in molds of varied dimensions, which were then fixed to a vibrating table and underwent vacuum and compression. Then, the material is placed in an oven at approximately 85°C to harden. This process results in a hard artificial stone plate resistant to scratches, stains, and burns and suitable to be used in kitchens, bathrooms, and floor and wall coverings.

Ribeiro et al.[25]'s work aimed to investigate the development of a synthetic marble processed using VCV with a matrix of 15%wt polyester resin and 85%wt marble waste. The composites showed properties within the expected range for an artificial stone. However, Ribeiro et al.[26] produced another artificial stone, with the same composition as the previous study but with another manufacturing methodology called resin transfer molding (RTM), a process that does not use vibration and vacuum. They compared both, and the artificial stone produced with RTM presented higher porosity and water absorption and lower mechanical resistance, even in comparison with commercial artificial stones, emphasizing the efficiency of VCV to manufacture artificial stones.

The research carried out by Lee et al.[6] aimed to study the effect of processing conditions variations in VCV (compaction pressure, vacuum, and vibration frequency) on the physical and mechanical properties of artificial stone plates manufactured with polyester resin and waste glass and granite and glass waste. The authors concluded that an increase in the compaction pressure, up to a certain limit, associated with the use of vacuum and vibration, reduced the porosity and water absorption while improving the mechanical properties. An excessive pressure increase, however, could crush the aggregates generating cracks that diminish the mechanical strength.

Under these circumstances, this research's main objective was to produce and characterize artificial stone plates through the process of vibration, compression, and vacuum, with the incorporation of granite and mirror wastes in an epoxy resin matrix, aiming at the recycling of wastes as well as the development of novel material with suitable physical and mechanical properties for application in civil construction projects.

2. Materials and Methods

2.1. Materials

For the development of the artificial stone plates (ASGM), the authors used wastes from the mining of a type of granite commercially known as gray granite Ocre Itabira mixed with mirror glass waste, arising from breaks during its processing in glassworking.

Epoxy resin, MC130, bisphenol A diglycidyl ether (DGEBA) type, was used as polymeric matrix, with 1.15 g/cm^3 density, hardened with Triethylenetetramine (TETA), FD 129, both supplied by the company Epoxyfiber.

2.2. Methods

The mirror wastes were subjected to a jaw crusher, brand ASTEMA, model BREAKER-FIX-50, with an eccentric shaft and jaws with an upper opening of 88mm and a lower opening of 11mm for crushing coarse particles. Then, the particles were put into the Marconi ceramic disk mill, model MA700/TR, with an opening of 0.85 mm, to produce medium and fine particles. Granite waste had already been collected in the form of powder, with fine grains. Both mirror and granite wastes (Figure 1) were sieved, according to ABNT/NBR 7181/2016[27], and then classified into coarse, medium, and fine grain sizes, according to Table 1.

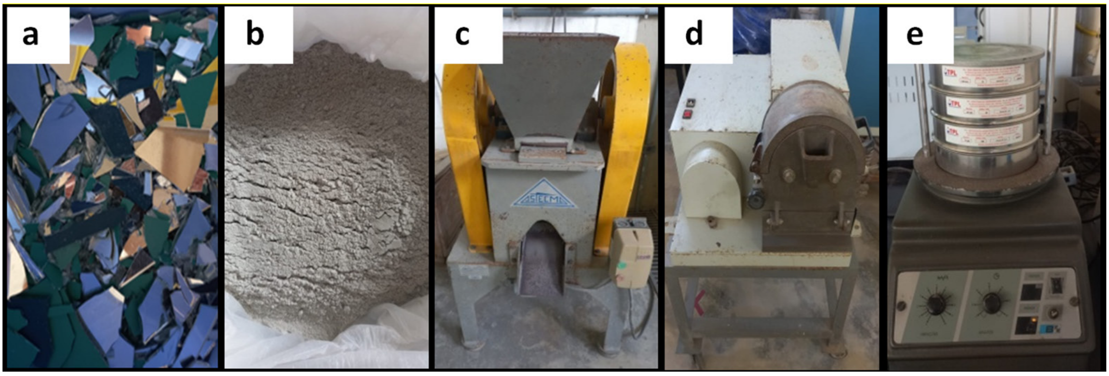

Figure 1. Mirror waste (a) and Ocre Itabira granite waste (b) without processing. Jaw crusher (c) disc mill (d) and sieve shaker (e).

Table 1. Granulometric ranges of the wastes.

Type	Granulometric size (ASTM)	Granulometric size (mm)	Residue
Coarse	10 – 40	2.000 – 0.425	Mirror
Medium	40 – 200	0.425 – 0.075	Mirror
Fine	< 200	< 0.075	Granite

To determine the highest packing granulometric composition among the mixtures of particles to be used for the development of ASGM, a "Simplex Centroid Model" ternary diagram was applied (Figure 2). Therefore, 10 mixtures with different proportions of the three granulometric ranges: coarse medium, and fine, were tested. The packing test was performed based on the Brazilian standard NBR 16843[28] - Determination of the minimum void ratio of non-cohesive soils.

For a better understanding of the data obtained after the packing test, statistical treatment was performed using analysis of variance (ANOVA) of the completely randomized design (CRC) ($p \leq 0.05$), aiming at validating the statistical significance between the treatments. Once the statistical difference was confirmed, a Tukey's average comparison test was performed ($p \leq 0.05$) to check the mixture with the best results. The calculations were performed using Excel from the Microsoft Office package.

To determine the minimum amount of resin (MAR), by weight, necessary to efficiently fill the volume of voids found between the particles, the Equation 1 (VV% = Void volume) and Equation 2 (MAR% = minimum amount of resin) were used. As a result, a resin percentage of 15%wt was obtained, being employed for the ASGM plates production.

$$VV\% = 1 - \left(\frac{particles\ apparent\ dry\ density}{particles\left(mirror\ glass + granite\ glass\right) density} \right) \times 100 \quad (1)$$

$$MAR\% = \frac{VV\% * \rho resin}{VV\% * \ \rho resin + \left(100 - VV\%\right) * \rho\left(particles\right)} \quad (2)$$

2.3. Artificial stone plates manufacturing

ASGM plates measuring 200x200x14mm were developed using the vibration, compression, and vacuum (VCV) method. The aggregates (granite waste + mirror waste) were dried in an oven at 100°C for 24 hours to reduce moisture and weighed in the appropriate proportions according to the result of the packaging test (composition 7), which was: 1/3 of fine particles (granite), 1/3 of medium particles (mirror) and 1/3 of coarse particles (mirror).

Wastes were then placed inside a planetary mixer, where the resin and the hardener, in the proportion of 15% by weight, were added to complete the mixture. The mixer was stirred until homogenization. The mixture was poured into a mold and taken to the hydraulic press, where it was vibrated under a vacuum while heat pressed at 90°C for 20 minutes under 12 tons of pressure. Then, the plates were sanded with a manual sander and cut to the dimensions specified for the characterization tests.

The production of ASGM plates (Figure 3) followed the steps shown in Figure 4.

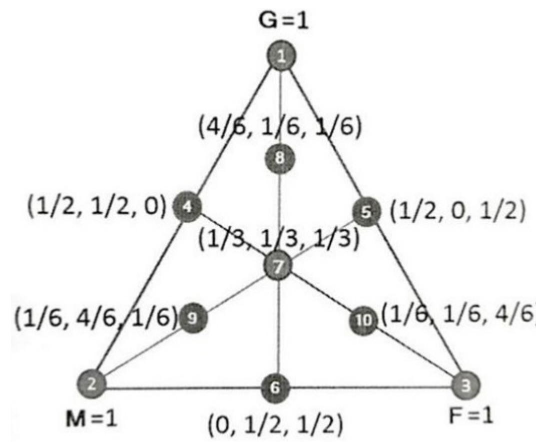

Figure 2. Simplex Centroid Model ternary diagram of compositions. Proportions of coarse (G), médium (M) and fine (F) particles[15].

Figure 3. ASGM plate.

2.4. Characterization tests

In the physical indices test, the apparent density, apparent porosity, and water absorption of the analyzed material were determined, using 10 specimens of 50x50x14mm, according to the procedure described in the Brazilian standard NBR 15845-6[29]. These samples were saturated in deionized water

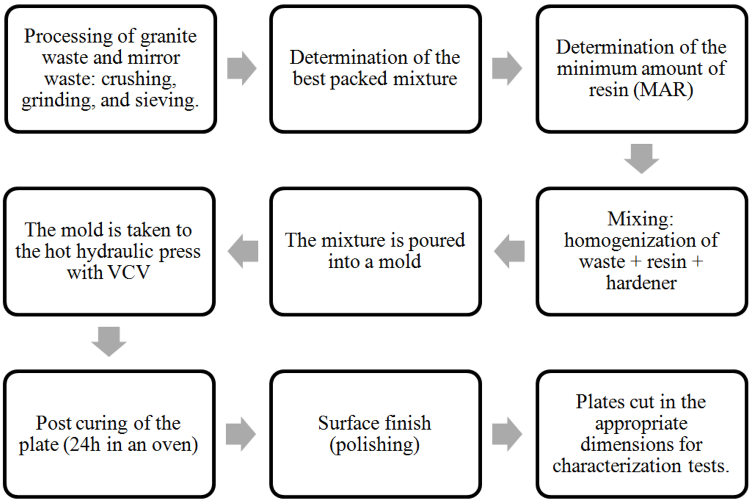

Figure 4. Stages of the ASGM plates manufacturing process.

following these steps: first, adding water up to 1/3 of the height of the samples, and after 4 hours, more water was added up to 2/3 of the height. After another 4 h, the samples were completely submersed for 40 hours. After that, the samples were weighed to determine the saturated mass (Msat) and also weighted submerged, to determine the submerged mass (Msub). To determine the dry mass (Msec) the samples were dried in an oven at 70°C until reaching a constant mass and then weighed. Apparent density, apparent porosity, and water absorption were calculated using the following equations:

a) Apparent density (g/cm³):

$$\rho a = \frac{Msec}{\left(Msat - Msub\right)} \times 1000 \tag{3}$$

b Apparent porosity (%):

$$na = \frac{\left(Msat - Msec\right)}{\left(Msat - Msub\right)} \times 100 \tag{4}$$

c) Water absorption (%):

$$\alpha a = \frac{\left(Msat - Msec\right)}{M\,sec} \times 100 \tag{5}$$

The 3-point bend strength test was carried out in 5 dried specimens of 14x25x100mm using an EMIC universal testing machine, model DL 10000, following NBR 15845-6[29], with a loading rate of less than 4450N/min.

The wear resistance test goal is to verify the thickness loss of the stone after an abrasive wear track of 500 and 100 meters. The test was performed in 2 samples of 70x70x30mm, in the Maqtest equipment, following the NBR 12.042[30] guidelines. The specimens had their thicknesses measured before and after each wear track using a dial indicator gauge.

The hard body impact resistance test was performed according to NBR 15845-8[29], using 5 specimens of 200x200x14mm. This test aims to determine the ASGM impact resistance by dropping a 1kg steel ball onto the specimen at increasing heights until the sample breaks.

Stain resistance test purpose is to verify the action of domestic daily and/or commercial staining agents that could jeopardize the ASGM aesthetics. The test was performed according to NBR 10545-14[31] with several staining agents such as penetrating staining agents (Cr_2O_3-green and Fe_2O_3-red), oxidizing agent (iodine), film-forming agent (olive oil) as well as everyday products like wine, coffee, ketchup, mustard, and lemon juice. Each staining agent was in contact with one sample for 24h. Then, the samples were submitted to the cleaning steps as described in the referred standard, to remove the stain. The material was classified according to the ease of stain removal.

SEM analysis was performed on the fractured region of 3 bend test specimens, to inspect the particles/matrix adhesion, as well as the presence of voids. A scanning electron microscope (SEM), model SuperScan SSX-550 by SHIMADZU, at 10 kV of secondary electrons, was used.

Some tests were also performed on other stones: natural gray granite Ocre Itabira (NG) and Galaxy White (CS), a commercial artificial stone, as a means to compare the properties of the developed artificial stone, ASGM.

3. Results and Discussions

3.1. Determination of the highest packing composition

Table 2 presents the results for the 10 mixtures of different particle sizes after carrying out the packing test.

As the vibrated density is an average of 3 densities found, the data were treated with analysis of variance considering a completely randomized design (CRD) with a 95% confidence level ($p \leq 0.05$), with a subsequent contrast Tukey test. Analyzing the results obtained in Table 3 , it is possible to verify that the studied treatments present statistical differences, meaning that among the 10 mixtures, at least two are differentiated.

They were differentiated with the Tukey test (Table 4), where it was possible to conclude that the mixtures with the highest densities are in 7 and 5. Therefore, mixture 7 was chosen for the development of ASGM.

3.2. Physical properties

Table 5 presents the physical properties of the ASGM, the artificial stone Galaxy White (CS), which contains mirror waste in its composition, among other particles not specified by the manufacturer, and the natural gray granite Ocre Itabira (NG), from which comes the granite waste used to develop ASGM.

As can be seen in Table 5, ASGM has a lower apparent density than CS and NG. Compared to the natural stone, NG, a low-density value of ASGM was expected, because ASGM is made of the polymeric matrix, a lighter material. The reduced density directly reflects the weight per square meter of the plates to be manufactured, which reduces logistical costs[7]. Furthermore, the ASGM density value is within the range of 2.03 to 2.45g/cm^3 found by Lee et al.[6], whose research produced stones with glass waste, granite, and polyester resin varying the compaction pressure, vibration frequency, and vacuum.

Analyzing the ASGM's apparent porosity and water absorption, it can be seen that the properties found are inferior when compared to CS and superior when compared to NG since ASGM has approximately 3 times less porosity and water absorption than NG. According to Rodrigues and Chiodi[32], coatings classified as high-quality materials must have less than 0.5% porosity. Therefore, as can be seen in Table 5, ASGM porosity of 0.25% is below that value, indicating that ASGM can be classified as a high-quality material for coating applications. Likewise, ABNT/NBR 15844[33] indicates that, for cladding applications, granite porosity must be less than 1% and granite water absorption must be less than 0.4%, and ASGM meets the parameters.

The ASGM's low porosity and water absorption indicate that there was satisfactory adhesion between the waste particles and the resin, highlighting the quality of the material, including for applications in humid environments, such as kitchens and bathrooms. In addition, the material's low porosity is directly reflected in the lower water absorption, since not all pores are interconnected, contributing to the reduction of water percolation in the material.

Other studies that used epoxy and different types of glass waste to produce artificial rocks found the respective values of 0.10% ± 0.01 and 0.21% ± 0.03 for water absorption and porosity[15] and 0.44% ± 0.06 and 0.83% ± 0.04[16]. Therefore, the physical properties of ASGM are in accordance with the literature, without significant changes in the results.

3.3. Three-point bend strength

According to Table 6 and Figure 5, ASGM presented superior mechanical properties when compared to natural granite, which can be attributed to its molecular interconnections between the matrix and the load. On the other hand, when compared to CS, the commercial rock, ASGM obtained inferior properties. The mechanical results found for ASGM, NG, and CS reflect their physical properties, as shown in Table 5, with emphasis on Ocre Itabira, which had lower bend resistance due to the higher porosity in its structure. According to Gomes et al.[10], low porosity can enhance the artificial stone's bend resistance once pores could act as stress concentrators, thus interfering with its resistance during requests for use.

According to Rodrigues and Chiodi[32], ornamental stones with bend strength above 20MPa are considered to be high-

Table 2. Vibrate density of the mixtures.

Mixture	Vibrate density average(g/cm^3)
1	1.55 ± 0.02
2	1.38 ± 0.01
3	1.49 ± 0.03
4	1.64 ± 0.01
5	1.85 ± 0.03
6	1.72 ± 0.01
7	1.88 ± 0.01
8	1.80 ± 0.00
9	1.62± 0.02
10	1.65± 0.00

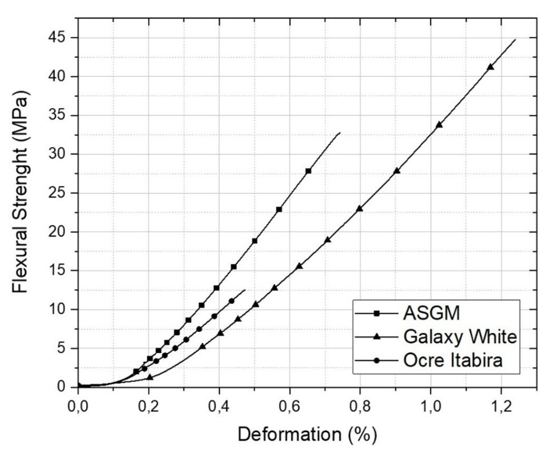

Figure 5. Bend stress x strain curves of ASGM, Galaxy White (CS) and Ocre Itabira (NG).

Table 3. ANOVA test results on the CRD of density (p≤0.05).

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Square	F Value
Treatment	9	0.6923	0.0769	265.5658
Waste	20	0.0058	0.0003	
Total	29	0.6980		

Conclusion: F calculated > F tabulated, there is a statistical difference. F tabulated = 2.39.

strength materials to be used as coatings in civil construction. Therefore, due to its 34.36 MPa bend strength, ASGM can be classified as a high-strength material and can therefore be applied in coatings, including worktops and countertops.

The flexural strength results of ASGM are in accordance with data found by other authors that developed similar materials. Carvalho et al.[8] developed an artificial stone with quarry wastes and 15% epoxy resin with 32 ± 2 MPa bend resistance, Agrizzi et al.[17] obtained 27.96 ± 1.86 MPa for their artificial stone based on quartzite waste and 13% of epoxy resin, Barreto et al.[15] manufactured a stone with glass waste, quartz and 15% of epoxy resin that obtained 33.54 ± 4.05 MPa and Gomes et al.[13] developed a stone with brick waste, quartz dust and 20% epoxy resin with 30 ± 3 MPa.

3.4. Abrasive wear resistance

Table 7 presents the ASGM, CS, and NG wear resistance after the Amsler wear test.

From the data in Table 7, it can be seen that the performance of the natural stone (NG) was superior to the commercial stone (CS) and the developed artificial stone (ASGM), presenting less thickness reduction. It was already expected due to artificial stone's polymeric matrix, which has lower hardness when compared to the constituent minerals of the natural stones, such as microcline, plagioclase, quartz, and biotite, with hardness ranging from 3 to 7, on the Mohs scale[34].

There are no standards that classify artificial stones in terms of abrasive wear. However, this test is important to define the developed stone's application in floor coverings, such as high, medium, or low-traffic environments.

Rodrigues and Chiodi[32] established parameters to classify artificial stones in terms of wear resistance, indicating that floors subjected to low traffic must have less than 6 mm thickness loss after a 1,000m track, as well as those subjected to medium traffic must have less than 3mm and the ones subjected to heavy traffic must be less than 1.5mm. Based on these parameters, ASGM can be classified as artificial stones to be applied on a floor with medium traffic, once its thickness reduction after the Amsler wear test was less than 3mm. In turn, CS and NG can be classified as high-traffic stones for flooring due to their thickness loss of less than 1.5mm.

The ASGM wear resistance does not differ from data found in the literature for similar materials. Barreto et al.[15] found 2.86 mm of wear (in 1000m) for their stone developed with quartz, bottle glass, and epoxy. Peixoto et al.[16] developed a stone based on epoxy and glass lamination waste that obtained 1.67 mm of wear and the artificial stone developed by Carvalho et al.[18] based on sinter waste and epoxy resin had 2.16 thickness loss. All those aforementioned artificial stones developed in these studies were manufactured under similar methods (vacuum, vibration, and compression) and with the same resin (epoxy resin) of ASGM, with similar results.

3.5. Hard body impact resistance

Table 8 presents the results found in the hard-body impact resistance test for the ASGM and CS.

The hard-body impact resistance test was performed to assess ASGM's cohesion and tenacity, as well as how much energy it dissipates before rupture, depending on the maximum drop height of an object dropped on its surface.

As observed in Table 8, CS's rupture energy was twice greater than ASGM's. It is worth noting the thickness difference between the specimens. ASGM plates were 14mm thick and CS plates were 30mm thick. The test was performed in accordance with ABNT/NBR 15845-8 (2015). Furthermore, as shown in Table 5 and Figure 7 (SEM), it was observed that ASGM's porosity was greater than CS's. Once pores act as stress concentrators, it may have influenced the impact resistance results.

Table 4. Tukey test for contrasting density averages ($p \leq 0.05$).

Treatment	Average	Tukey Test *
7	1.88	A
5	1.85	AB
8	1.8	C
6	1.72	D
4	1.65	E
10	1.65	EF
9	1.62	EF
1	1.55	G
3	1.49	H
2	1.48	HI

*The averages followed by the same letter in the Tukey test column are not different from themselves in a 5% of probability by Tukey Test.

Table 5. Physical Properties of ASGM, Galaxy White (CS) and Ocre Itabira (NG).

	ASGM	Artificial Stone "Galaxy White" (CS)	Natural Gray Granite "Ocre Itabira" (NG)
Apparent Density (g/cm³)	2.22 ± 0.01	2.38 ± 0.00	2.70 ± 0.01
Apparent Porosity (%)	0.25 ± 0.02	0.18 ± 0.01	0.92 ± 0.02
Water absorption (%)	0.11 ± 0.01	0.08 ± 0.00	0.34 ± 0.01

Table 6. Three-point bend strength resistance of ASGM, Galaxy White (CS) and Ocre Itabira (NG).

	ASGM	Artificial Stone "Galaxy White" (CS)	Natural Gray Granite "Ocre Itabira" (NG)
Three-point bend strength (MPa)	34.36 ± 2.01	44.73 ± 4.61	12.54 ± 3.68

Table 7. Thickness loss after Amsler wear test.

Materials	Thickness loss after Amsler wear test (mm)	
	500m	1,000m
ASGM	1.41	2.28
Artificial Stone "Galaxy White"(CS)	0.51	1.18
Natural Gray Granite "Ocre Itabira"(NG)	0.45	0.98

Table 8. Hard body impact resistance of ASGM and Galaxy White.

Material	Rupture Height (m)	Rupture Energy (J)
ASGM	0.45	4.41± 0.40
Artificial Stone "Galaxy White" (CS)	0.95	9.32 ± 0.98

Rodrigues and Chiodi[32] classified stones to be applied as cladding according to their resistance to impact in intervals of rupture height. According to them, very low-quality materials displays rupture heights lower than 0.30m; low-quality materials rupture heights range from 0.30m to 0.50m; medium-quality materials range from 0.50m to 0.70m; high-quality materials range from 0.70m to 0.95m and very high-quality materials must have rupture heights above 0.95m. Brazilian standard ABNT NBR 15844[33] establishes a minimum of 0.30 m of rupture height in hardbody impact resistance for a stone to be used as a coating.

Therefore, following Rodrigues and Chiodi[32] parameters and analyzing Table 8, it is possible to classify ASGM as low-quality material for cladding and CS as a high-quality one. In turn, taking into account ABNT NBR 15844[33] guidelines, both ASGM and CS meet the proposed parameter, as they have heights greater than 0.3m and can therefore be applied in coatings in civil construction.

Agrizzi et al.[17], Silva et al.[12], and Gomes et al.[10] used manufactured artificial stones with epoxy resin and different types of aggregates with 0.39m, 0.43m, and 0.4m of maximum rupture height, respectively. Therefore, the result obtained by ASGM is similar to those obtained by other authors.

3.6. Stain resistance

Figure 6 shows the result of the stain resistance test performed on ASGM, Galaxy White, and Ocre Itabira. In this test, the classification of resistance is related to the ease of removing stains after the cleaning steps described in the NBR 10545-14[31] standard, in which class 5 corresponds to greater ease of cleaning, meaning a greater stain resistance and class 1 corresponds to the impossibility of stain removal, meaning that the stone is not stain resistant to that specific product. This test is important to guide as to the proper application of the stone in coatings, kitchen countertops, bathrooms, or spaces that may be susceptible to contact of its surface with staining agents: Cr2O3-green, Fe2O3-red, iodine, olive oil, mustard, ketchup, wine, coffee, and lemon.

As seen in Figure 6, for the mustard, ketchup, wine, coffee, and lemon products, all the analyzed materials obtained an

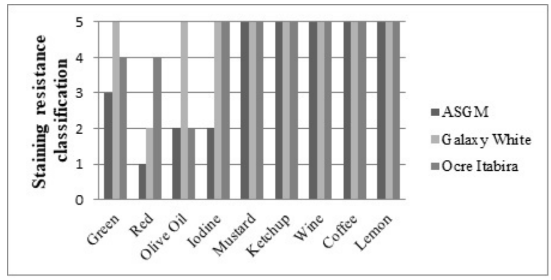

Figure 6. Classification in terms of stain resistance of ASGM, Galaxy White (CS) and Ocre Itabira (NG).

excellent performance, with high resistance, as they reached class 5. The stain removal was easy after cleaning with hot water. As for the other staining agents, each stone presented a different behavior.

ASGM was classified as class 2 for oil and iodine stains, once their stain was removed with solvents (hydrochloric acid, acetone, and potassium hydroxide), class 3 for the green agent, removed with abrasive paste, and class 1 for the red agent, the only stain that could not be removed after all the cleaning steps. It was possible to observe that the difficulty in removing the stain from some samples of ASGM was due to the penetration of the product into the pores of the material. Research on stains indicates that the pores on the surface are mainly responsible for the greater difficulty in removing stains[16].

Compared to ASGM, the CS obtained greater stain resistance for the green and red agents as well as the olive oil and iodine, whose results range from classes 2 to 5, as can be seen in Figure 6. This can be attributed to CS's lower porosity (Table 5) apart from the fact that this material's surface has a high-gloss treatment, meaning a greater closure of surface pores, therefore, lower liquid penetration and hence, fewer stains.

The NG was classified ranging from classes 2 to 5 for some staining agents such as green, red, olive oil, and iodine agents, as can be seen in Figure 6. Despite having greater porosity (Table 5), its porphyritic texture with heterogeneous coloring not to mention the high-gloss surface finish may have contributed to a good result in the stain resistance test.

Barreto et al.[15] and Peixoto et al.[16] developed artificial rocks using epoxy resin with the incorporation of waste glass from bottles and lamination waste glass, respectively, and evaluated the stain resistance of the materials with the same methodology and the same staining agents tested in ASGM. Peixoto et al.[16] artificial stone based on lamination waste glass had no stain remaining after the test was completed. Barreto et al.[15] artificial stone based on waste glass from bottles had low resistance to the iodine oxidizing, different from what happened with ASGM, which was resistant to iodine stain.

In the research developed by Borsellino et al.[35], the staining resistance of artificial marble based on marble powder and epoxy and polyester resins was also evaluated. Although, the authors used a different methodology and other staining agents. The authors concluded that the plate developed with a higher content of epoxy resin suffered fewer surface changes after the test, which was attributed to

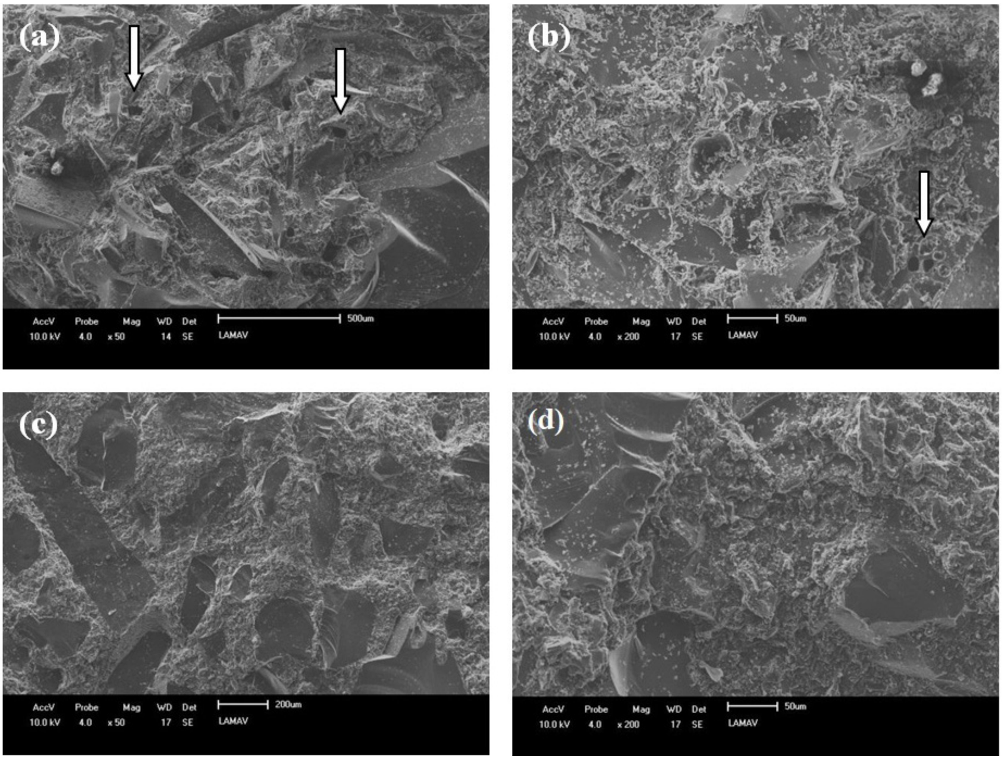

Figure 7. SEM micrographs of the fracture region at different magnifications ASGM (a) 50x and (b) 200x; Galaxy White (CS) (c) 50x and (d) 200x.

its greater stain resistance. Furthermore, there were greater changes in the samples submitted to Coca-Cola, wine, and lemon stains. Yet, all samples of ASGM, CS, and NG that were tested with wine and lemon were stain resistant to its materials, contrasting with Borsellino et al.[35] results.

3.7. Analysis of the microstructure

Figure 7 shows the SEM micrographs of ASGM and CS's fracture region after the three-point bend test.

From Figure 7a-7b, one can observe the ASGM'S good interaction between the granite and mirror particles and the epoxy resin, indicating a fine homogeneity in the mixture. Besides, it is possible to notice a low incidence of pores, which appear in isolation, which is a determining factor for low water absorption and apparent porosity (Table 5). The VCV production process contributed to the removal of air and, consequently, reduced the amount of empty space between particles. According to Debnath et al.[36], a good interfacial interaction is a consequence of an efficient interfacial wetting of the resin, which directly influences the improvement of the mechanical properties of the material.

From Figure 7c-7d the presence of pores or voids in the CS is almost imperceptible, highlighting the optimal interaction between the particles evidencing the physical (Table 5) and mechanical characteristics (Table 6) obtained in the tests performed.

Through the micrographs, it was possible to confirm the results mentioned above, in which ASGM has a greater porosity than CS. Therefore, microscopic analysis confirmed the physical indices and bending test results.

4. Conclusions

The research's main objective was to produce and characterize artificial stone plates by the process of vibration, compression, and vacuum, with the incorporation of gray granite Ocre Itabira wastes and waste glass from mirrors in an epoxy resin matrix. Characterization tests of this material were carried out, obtaining the following conclusions:

- ASGM is a material of low apparent density (2.22 g/cm^3), favoring the reduction of costs involved with transportation logistics. The material developed presented 0.25% apparent porosity and 0.11% water absorption, values that classified them as suitable for its application in humid environments, such as kitchens and bathrooms, due to its low water absorption, hence, lower liquid penetration.
- ASGM can be classified as a material of high mechanical strength, due to its 34.36 MPa bend strength, which exceeds the recommended value of at least 20 MPa for coating materials in civil construction.
- In the Amsler abrasive wear resistance test, ASGM showed 2.28mm thickness loss after a 1000m track, classifying it as a material that could be applied for floor covering submitted to medium traffic.
- In the hard-body impact resistance test, ASGM presented a 0.45m rupture height, meeting the parameter proposed by ABNT NBR 15844[33] of a rupture height higher than 0.3m for stones to be applied as coatings.

- ASGM demonstrated good stain resistance for mustard, ketchup, wine, coffee, lemon, iodine, olive oil, and green staining agent. However, the red staining agent caused a stain that was impossible to remove. Therefore, ASGM can be applied on coatings and countertops, as it is resistant to most staining agents tested.
- The SEM micrographs evidenced the ASGM's low presence of pores and excellent interfacial residue/resin adhesion, confirming the results found for the physical indices and bend strength.
- Some characterization tests were also carried out on CS and NG for comparison endorsing the material's quality, which was in general superior to NG and slightly inferior to CS.
- The results found for ASGM were mostly in accordance with similar materials proving its technical feasibility, with adequate properties for application in civil construction projects.

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6. References

1. Robert D, Baez E, Setunge S. A new technology of transforming recycled glass waste to construction components. Constr Build Mater. 2021;313:125539.
2. ABIVIDRO: Associação Brasileira das Indústrias de Vidro [Internet]. Why is glass the best option to recycle? São Paulo: ABIVIDRO; 2021 [cited 2022 May 20]. Available from: https://abividro.org.br/wp-content/uploads/2021/08/E-book_Porque-o-vidro-e-a-melhor-opcao-para-reciclar-1.pdf. [Portuguese].
3. Vidal FWH, Azevedo HCA, Castro NF. Ornamental stone technology: research, mining and processing. Rio de Janeiro: CETEM/MCTI; 2013. 700 p. [Portuguese].
4. Simão L, Souza MT, Ribeiro MJ, Montedo ORK, Hotza D, Novais RM et al. Assessment of the recycling potential of stone processing plant wastes based on physicochemical features and market opportunities. J Clean Prod. 2021;319:128678.
5. ABIROCHAS: Associação Brasileira da Indústria de Rochas Ornamentais [Internet]. The Brazilian performance of the ornamental stone sector. Brasília: ABIROCHAS; 2021 [cited 2022 Mar 31]. Available from: https://abirochas.com.br/wp-content/uploads/2022/01/Informe-02_2021-Desempenho-do-Setor-v3-1.pdf. [Portuguese].
6. Lee MY, Ko CH, Chang FC, Lo SL, Lin JD, Shan MY et al. Artificial stone slab production using waste glass, stone fragments, and vacuum vibratory compaction. Cement Concr Compos. 2008;30(7):583-7.
7. Demartini TJC, Rodríguez RJS, Silva FS. Physical and mechanical evaluation of artificial marble produced with dolomitic marble residue processed by diamond-plated bladed gang-saws. J Mater Res Technol. 2018;7(3):308-13.
8. Carvalho EAS, Vilela NF, Monteiro SN, Vieira CMF, Silva LC. Novel artificial ornamental stone developed with quarry waste in the epoxy composite. Mater Res. 2018;21(1):1-6.
9. Santos JPL, Rosa LG, Amaral PM. Temperature effects on the mechanical behavior of engineered stones. Constr Build Mater. 2011;25(1):171-4.
10. Gomes MLPM, Carvalho EAS, Demartini TJC, Carvalho EA, Colorado HA, Vieira CMF. Mechanical and physical investigation of an artificial stone produced with granite residue and epoxy resin. J Compos Mater. 2020;55(9):1247-54.
11. ABIROCHAS: Associação Brasileira da Indústria de Rochas Ornamentais [Internet]. Balance of Brazilian exports and imports of ornamental stones in the period January-November 2021. Brasília: ABIROCHAS; 2021 [cited 2022 Mar 31]. Available from: https://abirochas.com.br/wp-content/uploads/2022/01/Informe-08_2021-Exportacoes-novembro.pdf. [Portuguese].
12. Silva FS, Ribeiro CEG, Rodríguez RJS. Physical and mechanical characterization of artificial stone with marble calcite waste and epoxy resin. Mater Res. 2018;21(1):1-6.
13. Gomes MLPM, Carvalho EAS, Nascimento L So, Monteiro SN, Rodríguez RJS, Vieira CMF. Production and characterization of a novel artificial stone using brick residue and quarry dust in epoxy matrix. J Mater Res Technol. 2018;7(4):492-8.
14. Silva CB, Paiva PRP. Artificial stone production using iron ore tailing (IOT). Ceramica. 2020;66:164-71.
15. Barreto GNS, Carvalho EAS, Souza VS, Gomes MLPM, Azevedo ARG, Monteiro SN et al. Engineered stone produced with glass packaging waste, quartz powder, and epoxy resin. Sustainability. 2022;14:7227.
16. Peixoto J, Carvalho EAS, Gomes MLPM, Guimarães RS, Monteiro SN, Azevedo ARG et al. Incorporation of industrial glass waste into polymeric resin to develop artificial stones for civil construction. Arab J Sci Eng. 2021;47:4313-22.
17. Agrizzi CP, Carvalho EAS, Gadioli MCB, Barreto GNS, Azevedo ARG, Monteiro SN et al. Comparison between synthetic and biodegradable polymer matrices on the development of quartzite waste-based artificial stone. Sustainability. 2022;14:6388.
18. Carvalho EAS, Marques VR, Rodriguez RJS, Ribeiro CEG, Monteiro SN, Vieira CMF. Development of epoxy matrix artificial stone incorporated with intering residue from steel making industry. Mater Res. 2015;18(2):235-9.
19. Carvalho EAS, Souza VS, Barreto GNS, Monteiro SN, Rodriguez RJS, Vieira CMF. Incorporation of porcelain powder and mineral wastes in epoxy matrix for artificial stone purchase. In: Li J, Zhang M, Li B, Monteiro SN, Ikhmayies S, Eren Y et al., editors. Characterization of minerals, metals, and materials. Cham: Springer; 2021. p. 435-43.
20. Gomes MLPM, Carvalho EAS, Nascimento L So, Monteiro SN, Rodríguez RJS, Vieira CMF. Physical and mechanical properties of artificial stone produced with granite waste and vegetable polyurethane. In: Ikhmayies S, Li J, Vieira CMF, Margem JI, Braga FO, editors. Green materials engineering: an EPD symposium in honor of Sergio Monteiro. Cham: Springer; 2019. p. 23-9.
21. Cunha TP, Siqueira FB, Holanda JNF. Development of sustainable eggshell waste- polyester resin composite material for using as artificial rock. Mater Res. 2019;22(1):e20180865.
22. Kumar TN, Vikas B, Krishna MR, Jyothi Y, Imran SK. Development of composite slabs of marble powder embedded epoxy resin. Mater Today Proc. 2018;5:13031-5.
23. Adak NC, Chhetri S, Murmu NC, Samanta P, Kuila T. Analytical and experimental investigation on magnetorheological behavior of $CoFe_2O_4$-rGOincorporated epoxy fluid composites. Adv Compos Hybrid Mater. 2019;2:266-78.
24. Rubio MC, Moreno F, Belmonte A, Menèndez A. Reuse of waste material from decorative quartz solid surfacing in the manufacture of hot bituminous mixes. Constr Build Mater. 2010;24:610-8.
25. Ribeiro CEG, Rodríguez RJS, Vieira CMF, Carvalho EA, Candido VS, Monteiro SN. Production of synthetic ornamental marble as a marble waste added polyester composite. Mater Sci Forum. 2014;775-776:341-5.
26. Ribeiro CEG, Rodríguez RJS, Vieira CMF, Carvalho EAV, Cândido S, Monteiro SN. Fabrication of artificial stone from

marble residue by resin transfer molding. Mater Sci Forum. 2014;775-776:336-40.

27. ABNT: Associação Brasileira de Normas Técnicas. NBR 7181: soil - grain size analysis. Rio de Janeiro: ABNT; 2016. [Portuguese].
28. ABNT: Associação Brasileira de Normas Técnicas. NBR 16843: soil - determination of the minimum void ratio of non-cohesive soils. Rio de Janeiro: ABNT; 2020. [Portuguese].
29. ABNT: Associação Brasileira de Normas Técnicas. NBR 15845: rocks for cladding - test methods. Rio de Janeiro: ABNT; 2015. [Portuguese].
30. ABNT: Associação Brasileira de Normas Técnicas. NBR 12042: inorganic materials - determination of the resistance to abrasion. Rio de Janeiro: ABNT; 2012. [Portuguese].
31. ABNT: Associação Brasileira de Normas Técnicas. NBR 10545-14: ceramic tiles - part 14: determination of resistance to stains. Rio de Janeiro: ABNT; 2017. [Portuguese].
32. Rodrigues EP, Chiodi C Fo [Internet]. Guide application of stone coverings. Brasília: ABIROCHAS; 2020 [cited 2022 Dec 12]. 232 p. Available from: https://abirochas.com.br/ebooks/bula/#p=5. [Portuguese].
33. ABNT: Associação Brasileira de Normas Técnicas. NBR 15844: rocks for cladding -requirements for granites. Rio de Janeiro: ABNT; 2015. [Portuguese].
34. Sardou R Fo. Atlas of ornamental rocks in the state of Espírito Santo. Brasília: CPRM; 2013. [Portuguese].
35. Borsellino C, Calabrese L, Di Bella G. Effects of powder concentration and type of resin on the performance of marble composite structures. Constr Build Mater. 2009;23(5):1915-21.
36. Debnath S, Ranade R, Wunder S, McCool J, Boberick K, Baran G. Interface effects on mechanical properties of particle reinforced composites. Dent Mater. 2004;20:677-86.