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FOR SUSTAINABILITY AND A GREEN ENVIRONMENT: ARTIFICIAL MARBLE-GRANITE AS A VALUE- ADDED PRODUCT FROM MARBLE AND GRANITE WASTE RESIDUE

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ABSTRACT

This work aims to create artificial marble using marble and granite wastes by mixing them with polyester resin. The artificial marble slab is prepared by mixing the crushed fragments and powder with UPE in different ratios. It was compacted under different compression values using a vacuum vibratory compactor. The impact of resin ratio and compaction pressure on the characteristics of the artificial marble was investigated. The density, water absorption, porosity, compressive and flexural strengths, and micro-structure of the artificial marble produced were used for evaluation of the artificial marble. The results showed that the density of the artificial marble slabs decreased with increasing polyester resin content. The water absorption and porosity decreased with the increasing resin content due to the filling of the voids in the matrix. The maximum compressive strength (108.4 MPa) and flexural strength (13.8 MPa) were obtained with resin ratios of 25%. The porosity and the absorption of water decreased with increasing compaction pressure, while the density, compressive strength, and flexural strength increased with increasing compaction pressure up to 15 MPa. The results also showed that the artificial marble produced at a 25% resin ratio and under compression of 15 MPa meets the ASTM C503 and C615 standards for the raw materials of natural marble and granite.

KEYWORDS: Waste management, Waste treatment, Artificial marble, artificial granite, polyester resin, marble waste, granite waste, dimension Stone, Composite.

من أجل الإستدامة والبيئة الخضراء: إنتاج الرخام الإصطناعي كمنتج ذو قيمة مضافة من مخلفات الرخام والجرانيت

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الملخص

يهدف هذا البحث إلى تصنيع رخام صناعي بإستخدام مخلفات الرخام والجرانيت عن طريق خلطها مع البوليستر ، حيث تم تحضير ألواح من الرخام الاصطناعي بخلط مخلفات الرخام الناعم مع البوليستر بنسب مختلفة ، وضغطها بإستخدام ضاغط هزاز. وقد تم دراسة تأثير قوة الضغط ونسبة البوليستر على خصائص الرخام المنتج وتقييم المنتج بقياس الكثافة والقدرة على إمتصاص الماء والمسامية وتحمل الضغط والإنحناء والبنية المجهرية للرخام المنتج. وقد أظهرت النتائج إمكانية إستخدام مخلفات صناعة الرخام والجرانيت لإنتاج رخام صناعي يطابق المواصفات القياسية للرخام ، بخلطها مع البوليستر غير المشبع ، حيث أعطت نسبة خلط

75% من مخلفات الرخام والجرانيت و25% من البوليستر خواص فيزيائية وميكانيكية جيدة ، حيث بلغ تحمل الضغط حوالي (154.3 ميجا باسكال) ، ومقاومة الإنحناء (53.9 ميجا باسكال) ، والمسامية الظاهرية (0.06%) ، وإمتصاص الماء (0.013%) ، بكثافة (2.45 جم/سم3). كما أظهرت النتائج أن الرخام المنتج تحت ضغط 15 ميجا باسكال يفي بمعايير ASTM C503 و ASTM C615 للرخام والجرانيت الطبيعي.

الكلمات المفتاحية : إدارة المخلفات، معالجة المخلفات، الرخام الصناعي، الجرانيت الصناعي، البوليستر، مخلفات الرخام والجرانيت، أحجار الزينة.

1. INTRODUCTION

The marble and granite industry is developing consistently, so its creation could surpass one hundred million tons in 2025 [1, 2]. However, in 2019, the global gross creation of quarrying was around 317 million tons; around 53% of the item is viewed as waste from quarrying; generally speaking, between thirty and 35 percent of the garbage produced by marble processing operations is waste, and these leftovers require increasing regions for disposal every day [1]. Approximately 73% of garbage is produced worldwide [3]. From an environmental, economic, and social perspective, this poses serious issues. Carbonate stones, which include marble, limestone, and travertine, account for around 60% of the world's output of dimension stones; granites come in second with 38% [1, 2].

The marble and granite industry has expanded in Egypt over the past few years. The Shaq El-Thoaban neighborhood in Turah, Cairo, covers about 6.5 million square meters. It incorporates 3,500 processing plants and workshops for manufacturing and exporting marble [4]. According to the local estimation, Egypt is responsible for 2.24 million tons of waste created by this industry, which accounts for 70% of the total stone output of 3.2 million tons [5]. Twenty percent of the garbage is generated during the processing step, translating to 640,000 tons of waste overall. Ultra-fine sawdust sludge (448,000 tons annually) is produced in this area. Naturally, this is a huge amount that poses harm to the environment and those nearby and has long been seen as genuine waste. It is therefore incompatible with urgently needed eco-sustainability and environmental protection.

Unmanaged wastes are a significant environmental problem these days. Because of this, using these wastes to create new materials is a useful way to manage the waste that is produced and get around the problem of limited natural resources [6]. Numerous scholars have conducted research on the sustainable repurposing of these wastes in various industrial settings [7–15].

Much research and development is being done using epoxy resins, as these are widely used in all kinds of engineering applications. Due to the ever-increasing technical and aesthetic requirements in addition to the high prices of natural stones, it is necessary to utilize all the possible advantages of resins and to use them in the production of cheap artificial stones as an alternative to natural stones [16]. The mechanical and physical features of synthetic marble and granite are directly related to the resin content and the microstructure of the product [17]. During compaction, efficient vacuuming and vibrating may result in a more controlled formation of voids within the material, which reduces the product's porosity and water absorption [18, 19].

Artificial marble is well accepted in the market, and the factories that manufacture and offer it point to many benefits. The main feature that has been noted is its impermeability, which provides stain resistance. This is because the resin in its composition bonds the stone particles and fills in the typical porosity of the stone in its natural state. In this manner, liquids cannot penetrate through or percolate into the stone's interior [20–22].

Artificial stone slabs acquired go through conventional handling steps of crushing, cutting, and cleaning. Artificial marble slabs are superior to normal ones due to their low water absorption, porosity, and low flexural strength. As a result, they are perfect building materials for covering walls or paving floors. In light of the depletion of natural resources, recycling waste stone into building materials not only creates new goods but also provides a cost-effective and environmentally friendly alternative for waste management [23].

The demand for artificial stones has been rising in Egypt over the past several years due to their lovely, dazzling shape and affordable pricing; it has been predicted that this demand will rise by 30%. Thus, the artificial stone industry project is considered one of the most important industrial projects at present because of its high demand. It is considered a successful investment, as well as protecting the environment from the negative impact of these wastes.

The major objective of this study is to create an artificial stone from leftover marble and granite that has good physical and mechanical properties and can be utilized for flooring or tiles in the habitat business. As a result, it is suggested that this research make use of the characteristics of marble and granite wastes as well as unsaturated polyester resin (UPE).

2. Materials and methods

2.1. Raw materials

The granite waste fragments and slurry were collected from the Port-Said Factory for Granite & Marble, The Shaq El-Thoaban neighborhood in Turah, Cairo, Egypt. The waste fragments are chips as by-products of carving, chiseling, and finishing processes. A jaw crusher was used to crush them, and they were then ground in a ball mill. They were classified into three different fractions: aggregates (12.5 mm), coarse (4 mm), and washed fine (1 mm). The slurry wastes containing granite and marble dust were gathered from the settling tanks of the stone cutting. These slurries were air-dried for 4 days and then incubated in an oven for one hour at 100°C. The dried agglomerated slurry was ground in a ball mill to get a powder.

Unsaturated polyester resin was used as the binder in this study (UPE-2121) and hardener (HY 951, aliphatic amine) from Chemicals for Modern Building Company (CMB), Egypt. It has low viscosity, low exothermic temperature, mechanical and rheological properties, and is available at a reasonable price. At 25°C, the binder gels in around 35 minutes. The maximum sintering temperature is 130–155°C, and the shortest hardening period is 15 minutes.

2.2. Artificial marble manufacturing

The manufacturing process of artificial marble slabs was carried out by mixing the crushed fragments and powder with UPE in different ratios. The different solid fractions of 25% ultra-fine, 40% fine (1 mm), 25% coarse (4 mm), and 10% aggregate (12.5 mm) were pre-mixed. The latter was mixed with different resin ratios, as presented in Table 1. These ratios were selected to simulate the grains distribution of the natural marble.

The ultra-fine content of 25% of the total solids was used to avoid its negative effect on water absorption and porosity, and thus lower compressive and flexural strengths. After adding the accelerator and hardener to the polyester in a 2% ratio, thoroughly stir the mixture. The solid waste and resin were mixed mechanically at 500 rpm. Once well mixed, the mixture was poured into a mold of 250×250×10 mm dimensions; it was compacted under different compression values using a vacuum vibratory compactor. After that, the samples are cured in an oven at 95°C for 50 minutes, and then they are left for seven days at room temperature. Finally, the composite slabs are cut to the required dimensions for the tests to be carried out.

Table 1: Artificial marble mix design.

Resin, wt. %	Granite size fraction, wt.%				Total wt. %
	Ultra-fine	1 mm	4 mm	12.5 mm	
7	23.25	37.20	23.25	9.30	100
10	22.50	36.00	22.50	9.00	100
17	20.75	33.20	20.75	8.30	100
20	20.00	32.00	20.00	8.00	100
25	18.75	30.00	18.75	7.50	100

2.3. Physical and mechanical measurements

2.3.1. Density, apparent porosity, and water absorption

The ASTM C373-16 item 5.3 was used to calculate the artificial marble's density, apparent porosity, and water absorption. Six 50-mm cubes are tested for each mix design.

2.3.2. Compressive strength test

The compression strength of artificial marble was carried out according to EN1926:1999, as shown in Fig. 1. Six test samples with dimensions of 50 x 50 x 50 mm were used and oven-dried in accordance with standard procedures. To reach the last failure, a loading speed of 0.5 mm/min was used.

Fig.1. Compression Strength Test.

2.3.3. Flexural strength test

Flexural strength of composite marble under concentrated load was carried out according to EN 12372:1999, as shown in **Fig.2**. Ten specimens of dimensions 150×75×25 mm were tested.

Fig.2. Flexural Strength Test.

2.3.4. Micro-structural characterization

Using scanning electron microscopy (SEM), the micro-structure of the fracture of the test bodies subjected to flexural testing was assessed, allowing for an analysis of the particle adhesion by the epoxy resin. The tool utilized to carry out these investigations was the HITACHI TMT3030PLUS.

The mineral composition of the marble and granite residue was investigated using the German-made BRUKER X-Ray Diffractometer Model: AXS-D8 at 40 kV potential and 40 A with Cu-target (λ= 1.540 Å and n=1). At a scanning rate of 3°min−1, the diffraction data were obtained for 2θ values ranging from 3° to 70°.

3. RESULTS AND DISCUSSION

3.1. Mineral and chemical composition of the marble and granite residues

The XRD spectra of the marble and granite waste samples are shown in Fig. 4.1. The sharp peaks of both XRD spectra indicate the higher crystallinity of the samples. **Table 2** represents the mineral composition of marble and granite sludge obtained by XRD. The granite sludge sample is composed of 44.6% Albite, 15.2% Quartz, and 14.3% Dolomite, while the marble sludge is composed of 2.7% quartz, 2.5% albite, 2.6% illite, 64.3% calcite, 26.9% dolomite, and 1% bentonite.These results were confirmed by the complete chemical analysis, which was determined by XRF, (**Table 3).**

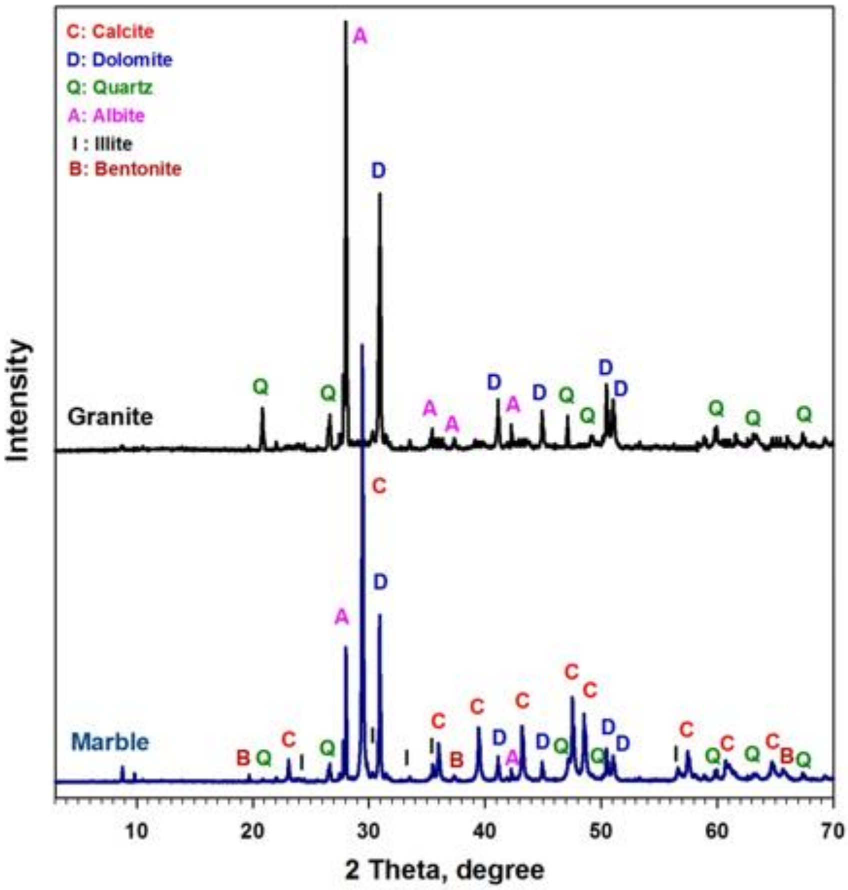

Fig.4.1. XRD patterns of the marble and granite dust samples.

Table 2: Mineral composition of the marble and granite sludge samples.

Waste sludge	Mineral	Formula	Weight %
Marble	Calcite	$CaCO_3$	64.3
	Dolomite	$CaMg(CO_3)_2$	26.9
	Quartz	SiO_2	2.7
	Albite	$NaAlSi_3O_8$	2.5
	Illite	$KAl_2Si_3AlO_{10}(OH)_2$	2.6
	Bentonite	$Na-Al-Si-O-OH-H_2O$	1.0
Granite	Albite	$NaAlSi_3O_8$	44.6
	Illite	$KAl_2Si_3AlO_{10}(OH)_2$	25.8
	Quartz	SiO_2	15.2
	Dolomite	$CaMg(CO_3)_2$	14.3

Table 3: Complete chemical analysis of the marble and granite sludge samples.

Waste residue	SiO_2	CaO	MgO	Al_2O_3	Fe_2O_3	P_2O_5	SO_3	Cl^-	TiO_2	Na_2O	K_2O	L.O.I
Marble	7.32	45.52	4.66	2.07	1.55	0.08	0.17	0.035	0.156	0.48	0.29	37.68
Granite	58.66	4.44	3.18	16.95	--	--	--	--	--	5.39	3.21	8.17

3.2. Evaluation of the artificial marble

3.2.1. Effects of resin ratio

The results of the effect of the polyester resin ratio at a compaction pressure of 4 MPa on the physical and mechanical properties of the artificial marble are presented in **Table 2**, which includes the average values of three tests for density, water absorption, apparent porosity, compressive strength, and flexural strength.

3.2.1.1. Density and water absorption

Table 3 shows the density of artificial marble slabs mixed at different polyester resin ratios. The density decreases with increasing the polyester resin ratio, which represents a satisfactory adherence between the particles and the resin. Since the specific gravity of resin is 1.38 g/cm^3, which is lower than that of marble and granite (2.6 g/cm^3) [24], along with the increase in the quantity of resin, a slight decrease was noticed in the densities of the resulting artificial marble.

Although the water absorption of fabricated marble products meets the specifications (< 0.5%) [24], Fig.3 shows that the water absorption and apparent porosity of artificial marble decrease along with the increase in the amount of resin. This may be due to the overfilling of the voids in the matrix of artificial marble with polymer resins. Therefore, the artificial marble is less porous with an increase in the amount of resin.

3.2.1.2. Compressive strength

Fig.4 illustrates how the ratio of polyester resin affects the compressive strength of artificial marble specimens. Evidently, 25% resin ratios produced the best compressive strength (108.4 MPa). It meets the standard specifications of marble and granite (ASTM C615), **Table 3**. From the standpoint of arranging particles, the polyester resin 25% mixing ratios appear to be the most effective.

It is important to remember that artificial marble is stronger mechanically than genuine marble. The poor mechanical strength performance is mostly caused by the porosity of the artificial marble matrix. It is commonly known that porosity reduces any brittle material's mechanical properties because the pores enhance stress, which aids in the initiation and spread of cracks [25]. The increased plastification of the polymeric phase as a result of the high thinner content added to the polyester might be another explanation for the low mechanical strength. Actually, adding thinner encourages a wider gap between the polymer chains, making it harder for them to reticulate and less rigid. Moreover, the thinner creates voids when it volatilizes and is non-reactive [26].

3.2.1.3. Flexural strength

The artificial marble's flexural strength specimens with different resin ratios were obtained using the three-point bending test (**Table 3**). **Fig.4** shows the variation in flexural strength. **Table 3** shows that the higher flexural strength (13.8 MPa) was obtained with a resin ratio of 25%. It meets the standard specifications of marble and granite [24].

Table 3: Characteristics of the artificial marble slabs at different polyester resin ratios.

Polymer %	Density g/cm^3	Water Absorption, %	Apparent Porosity, %	Compression Strength, MPa	Flexural Strength, MPa
7	2.36	0.97	1.24	62.7	5.10
10	2.32	0.46	0.97	73.1	7.90
17	2.24	0.37	0.91	87.9	9.40
20	2.21	0.29	0.83	96.4	11.60
25	2.15	0.23	0.78	108.4	13.80
Natural marble	2.59	0.20	0.50	52.0	7.00
Natural Granite	2.56	0.40	0.50	131	8.27

Note: kg/cm^2 = MPa / 0.0980665

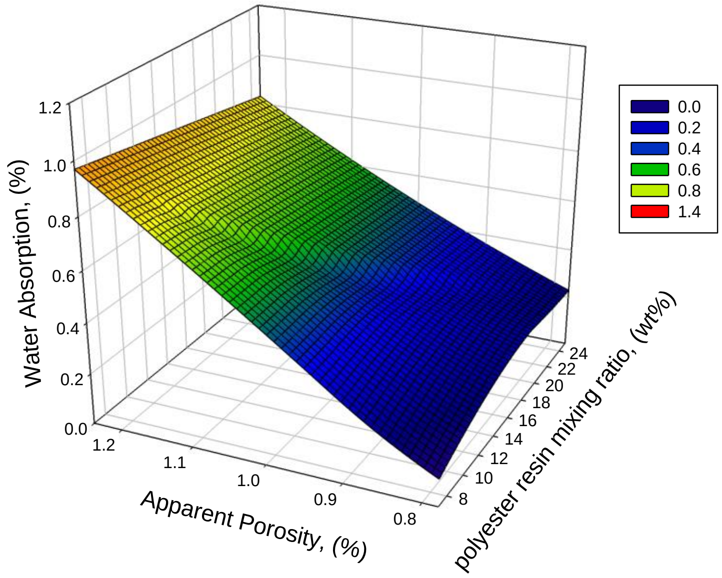

Fig.3. Effect of polyester resin mixing ratio on Water absorption and apparent porosity.

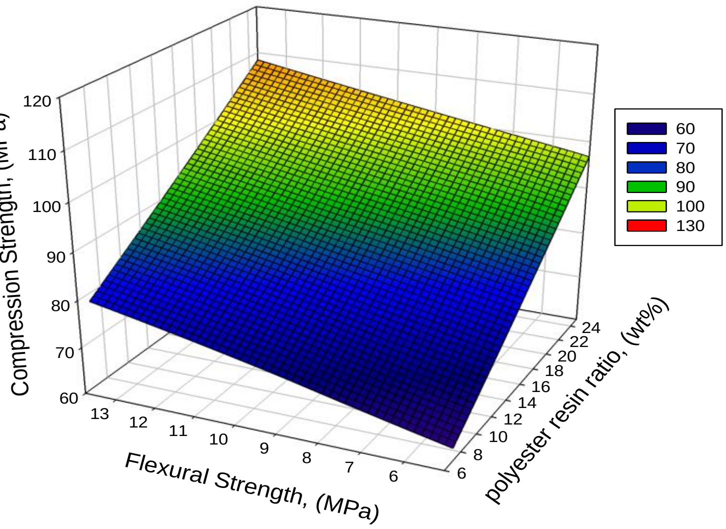

Fig.4. Effect of polyester resin mixing ratio on compressive strength and flexural strength.

3.2.2. Effects of compaction pressure

3.2.2.1. Density, apparent porosity, and water absorption

Six samples for each mix design were tested to measure water absorption, density, and apparent porosity. **Table 4** shows the average values for the outcomes obtained in the experimental tests for each mixture. The average value for the apparent density ranged between 2.15 - 2.40 g/cm^3, which is less than that of natural marble and granite (≈ 2.6 g/cm^3) [24]. The apparent porosity, as shown in **Fig. 5**, falls within the range of 0.06 - 0.78%, indicating a reasonable adhesion between the particles and the resin.

Demartini and Rodriguez [27] categorized the materials that were going to be used as building materials and found that materials with porosity values less than 0.5% were considered excellent grade. It was discovered that the artificial marble's water absorption ranged from 0.013% to 0.224 percent. The recommended value (ASTM C615) [24] is met by it.

3.2.2.2. Compressive strength

The compressive strength of the manufactured artificial slabs with 25% resin at various compaction pressures is displayed in Figure 6. As the compaction pressure rises, so does the compressive strength. A compaction pressure of 15 MPa achieved the maximum compressive strength of 154.3 MPa. However, the compaction pressure improved the compressive strength. This may be because the higher pressure compacts the aggregates and makes the structure more dense. Where the results showed that there is 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.

By using granitic particles, which possess higher mechanical strength than the marble particles used in this research, the compressive strength of artificial marble is higher than the compressive strength of natural marble, being almost double the minimum established value by the ASTM C503 Standard, which is 52 MPa [28].

3.2.2.3. Flexural strength

In **Table 4** and **Fig. 6,** the results obtained in the flexural strength test are described. Their values ranged between 13.8 to 53.9 MPa. This result indicates that the resin's polymeric chains are well-connected, which reduces slippage in the weak points of the artificial marble that often occurs when subjected to flexural pressures. This value complies with the criteria for marble and granite specified by ASTM C615 and C503 [24].

Table 4: Effect of compaction pressure on the produced artificial marble (25% resin).

Parameter	Compaction pressure, MPa				
	4	9	12	15	20
Density, g/cm^3	2.15	2.21	2.26	2.446	2.40
Apparent porosity, %	0.78	0.17	0.10	0.06	0.09
Water absorption, %	0.224	0.210	0.018	0.013	0.019
Compressive strength, MPa	108.4	113.5	135.2	154.3	126.3
Flexural strength, MPa	13.8	43.8	50.6	53.9	44.7

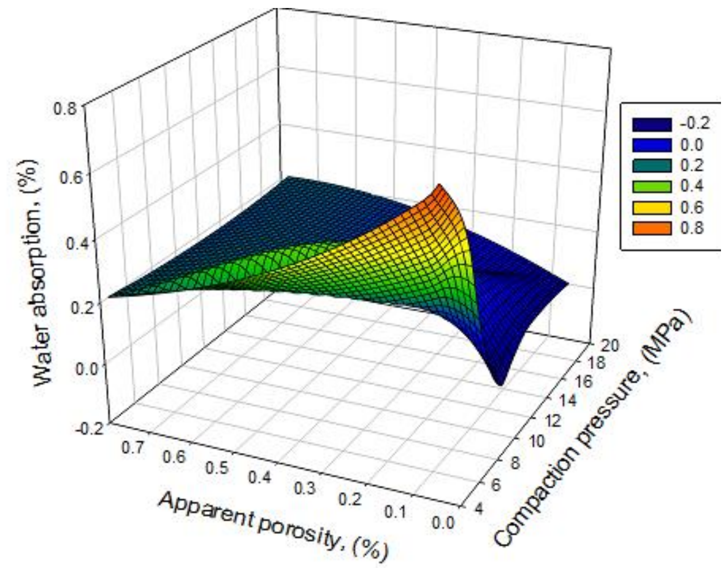

Fig.5. Effect of compaction pressure on water absorption and apparent porosity (25% resin).

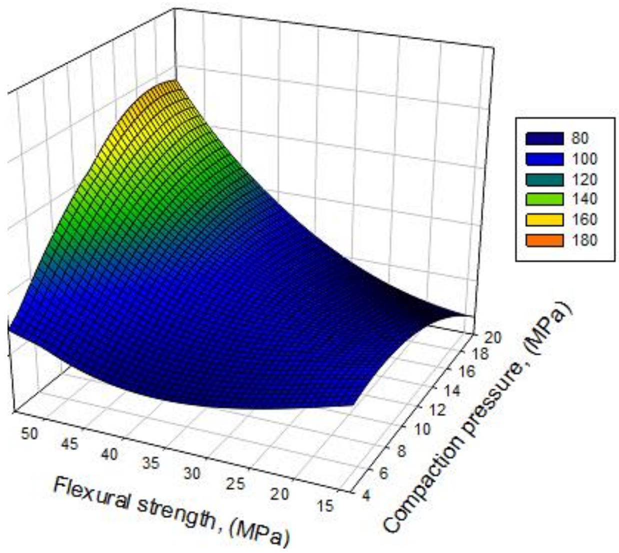

Fig.6. Effect of compaction pressure on compressive strength and flexural strength (25% resin).

Table 5 represents the comparison between the specifications of the obtained artificial marble and the Standard Specifications of Marble and Granite Dimension Stones according to ASTM C615 and ASTM C503 [24, 28]. The results showed that the artificial product meets the ASTM specifications for marble and granite.

Table 5: A comparison between the specifications of the obtained artificial marble and the Standard Specifications of Marble and Granite Dimension Stones according to ASTM C615 and ASTM C503 [24, 28].

Parameter	The artificial marble produced	Standard Specification of	
		Natural Marble	Natural Granite
Density, g/cm^3	2.446	2.59	2.56
Water absorption (%)	0.018	0.20	0.40
Porosity, (%)	0.100	0.50	0.50
Compressive strength, (MPa)	135.2	52	131
Flexural strength, (MPa)	50.6	7	8.27

3.2.3. Micro-structural characterization

The artificial marble with a low resin content is composed of open pores that allow for water absorption. The artificial marble porosity (< 1%), which is slightly higher than that of the natural marble, may indicate that the polymeric matrix contains the majority of the porosity. After the artificial composite was cured, some spaces and bubbles remained in the polyester phase. Although the fabricated marble has a slightly higher porosity compared to the natural marble, the water absorption and mechanical properties are better. This may be due to the strong bonding of resin.

Fig.7 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. Notably, a high number of pores are created when there is not enough resin (7% resin) to fill the voids. It displays spaces between the marble particles and the polyester matrix in addition to the pores. This might be related to ineffective load transfer between the marble particles and matrix due to poor interfacial strength. Therefore, utilizing 7% rupture at a much lower value (5.1 MPa) for the flexural strength results from both matrix porosity and the absence of adhesion between marble particles and polyester matrix. By adding more resin, the holes, voids, and pores were minimized. The mechanical tests and the water absorption measurement provide it.

By employing 25% resin, the smallest pores were achieved. This resulted in strong interfacial adhesion between the particles and the resin, which means that the resin adequately hydrated the particles. It was explained that a composite's mechanical characteristics might be improved by excellent interfacial contacts and that these interactions are directly correlated with the strength of the adhesive force generated by the interfacial areas' successful moistening. Furthermore, when the material is subjected to manufacturing operations such as polishing the surface and borders, the minimal pores increase its luster index [29, 30]. By reducing liquid penetration, these characteristics also increase the surface's mechanical qualities, which were found to be higher than those of the natural marble analyzed.

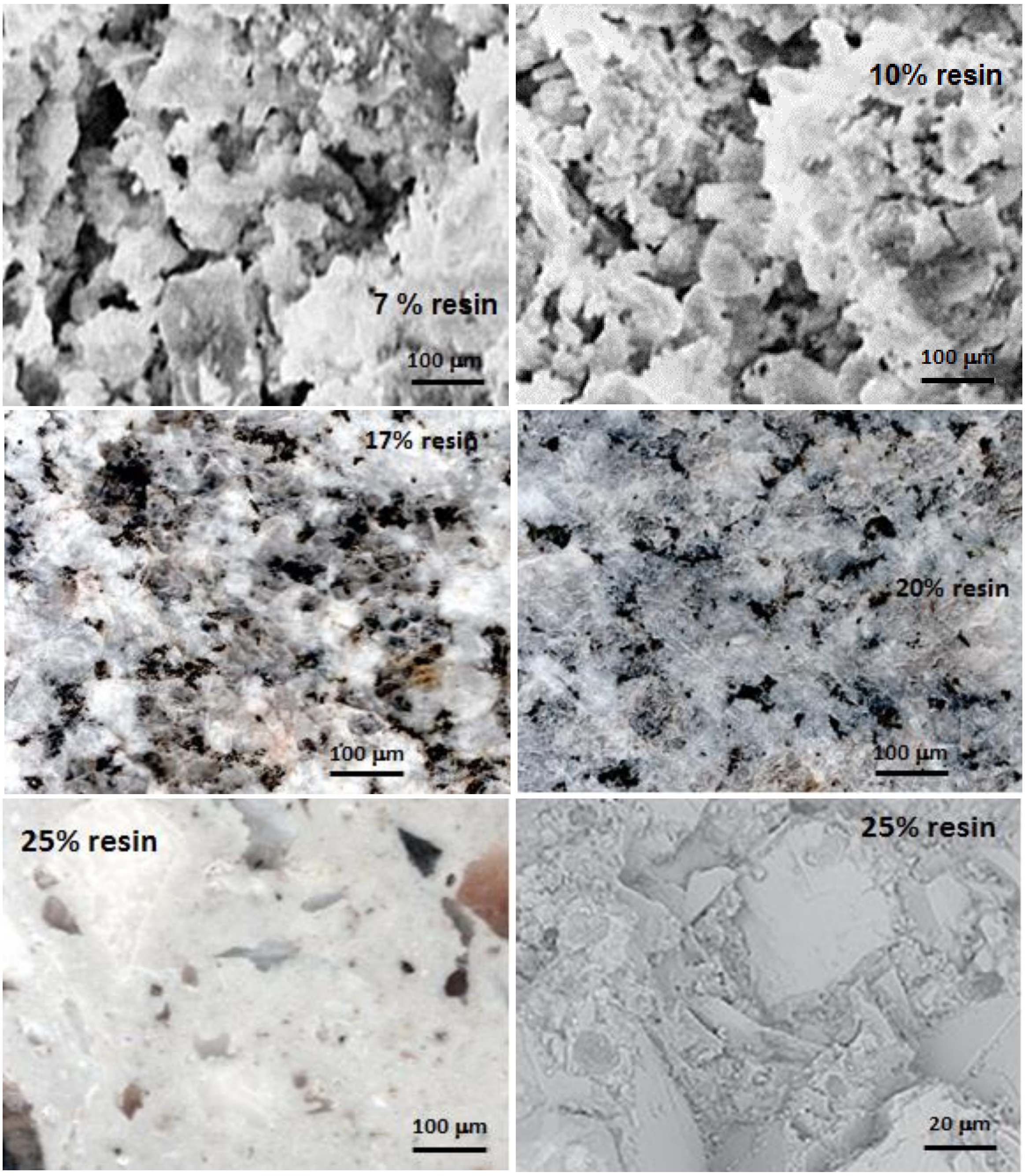

Fig.7. SEM micrographs of the fabricated artificial marble with different resin content.

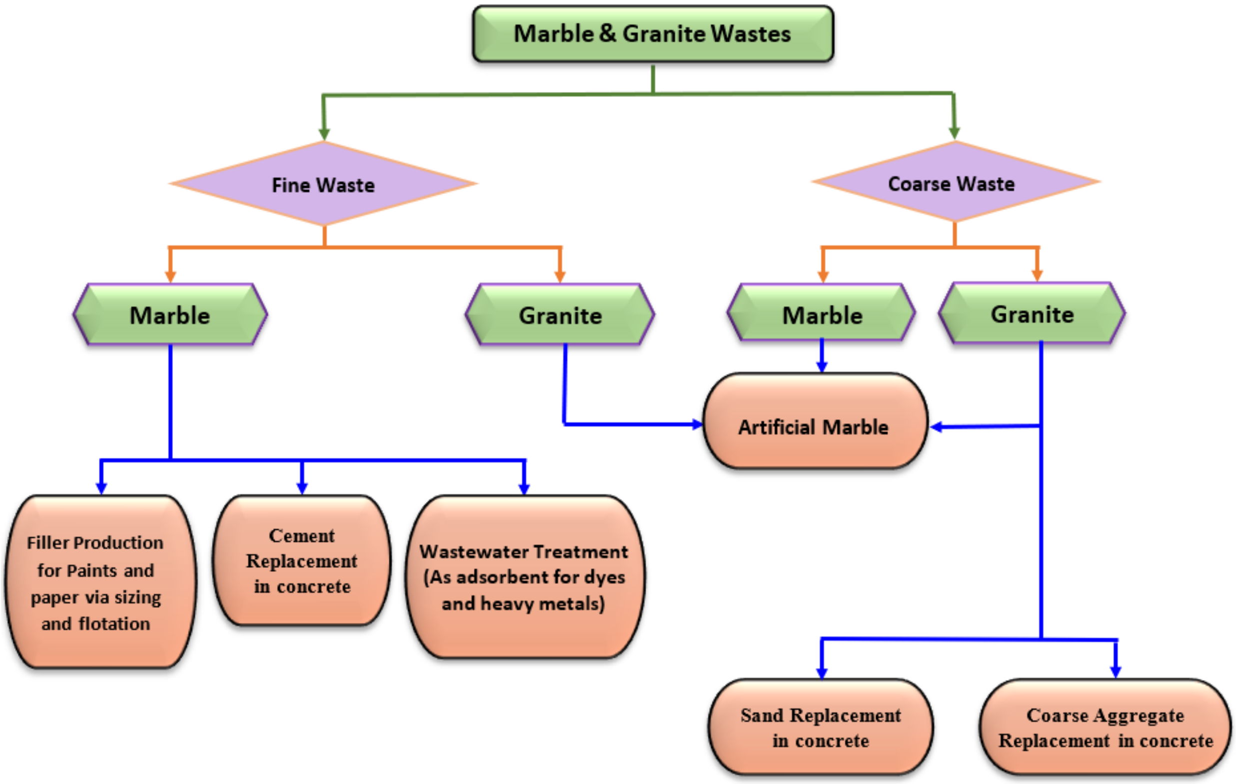

Fig.8. Flowchart for Expected Applications of Marble and Granite Wastes.

Conclusions

The granite and marble industry's solid waste was used to create artificial marble by mixing it with an unsaturated polyester resin. The outcomes indicated that the density of the artificial marble slabs decreases with increasing polyester resin content. The water absorption and porosity decreased with the increasing resin content due to the filling of the voids in the matrix. The maximum compressive strength (108.4 MPa) and flexural strength (13.8 MPa) were obtained with resin ratios of 25%. The porosity and the absorption of water decreased with increasing compaction pressure, while the density, compressive strength, and flexural strength increased with increasing compaction pressure up to 15 MPa. The artificial marble meets ASTM C615 and C503 for marble and granite.

Recommendations

- Studying the possibility of using waste from the marble and granite industry in more different industrial applications.
- An economic study should be conducted to reuse marble and granite waste in different applications.

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