Effect of Particle Size and Composition of Marble Quarry Waste on the Properties of Artificial Marble

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Abstract. The purpose of this study was to characterize and investigate the performance of local marble quarry waste to assess its use as filler in producing artificial marble. Marble waste was obtained from marble quarry located in Simpang Pulai, Ipoh and sieved into two different sizes; < 2mm as coarse particle and < 250 µm as fine particle. The artificial marble was prepared under vacuum condition and the composition of fine and coarse particles were modified between 40-70% (wt.) and 0-30% (wt.), respectively. The artificial marble was evaluated their performance by water absorption, flexural strength, compression strength, and Barcol hardness properties. It was found that artificial marble prepared with 60% (wt.) of fine particle and 10% (wt.) of coarse particle showed the best overall properties leading to lowest water absorption and good flexural and compression strength. However, modification of marble waste content in the composition of artificial marble showed insignificant influence on Barcol hardness properties.

1. Introduction

Globally, mining and quarrying sector is perceived as one of the important economic activities which contributing to the development of the country. However, the potential impact of mining and quarrying industries on the environment and human health continues to pose as the most challenging problems to the society. A significant increase in the extraction of natural stone such as granite and marble is observed due to the demand of these materials in building and construction industries. In 2019, there are 28 numbers of dimensional stone’s producer reported in Malaysia with Perak is still the main contributor and it includes marble slabs and marble furniture [1]. Nearly 70-75% of the whole extracted marble are lost as waste during mining, marble processing and polishing [2]. This huge amount of non-biodegradable waste generated are kept increasing year by year. In the current studies, it is seen that researchers have extensively conducted studies on utilizing many types of marble waste for different applications. For instances are; as partial replacement of aggregates in concrete, asphalt pavements and as well as particulate-filled polymer based composites materials [3,4,5]. The utilization of marble waste to produce artificial marble is also one of the alternative way to reduce the environmental impacts of inadequate waste disposal. Artificial marble also known as synthetic marble has been prepared in the market and demonstrated high demand in replacing natural marble. Generally, it consists of high content of mineral aggregates including marble, granite, quartz, and others and mixed with polymer resin and catalyst. There are many studies have towards the research specifically on different types of aggregate waste and resin. [6] investigated the properties of
an artificial stone made of granite residue in epoxy resin; [7] evaluated the mechanical properties of artificial stone produced using SiO₂ and quartz sand while study by [8] have developed a different kind of marbles using epoxy resin with marble particles. Thus, this study will recycle and reusing an abundance of local marble waste to produce artificial marble using the matrix of polyester resin. The influence of different size particle of marble waste and their composition on the physical and mechanical properties of artificial marble will be investigated.

2. Materials & Methods

2.1. Marble waste
Marble waste with particle size distribution between 45 and 8000 µm was collected from a marble processing plant in Simpang Pulai, Ipoh. The origin sample was dried directly under the sun to remove the moisture and ground into powder form using planetary ball milling (Retsch PM 400, German) to be used for characterization purpose. Marble waste have been characterized using X-ray fluorescence (XRF) (Shimadzu XRF-1700) and X-ray diffractometer (XRD) (D8 Advanced, Bruker, German). Figure 1 represents the marble waste used in this study to be acted as a main filler in the composition of artificial marble.

Table 1 shows the chemical composition of marble waste determined using XRF. The marble waste mainly composed of calcium oxide (CaO) with the content is 54.41 % and followed by low impurities levels of MgO, SiO₂, Al₂O₃ and etc.

| Oxides (%) | LOI |
|-----------|-----|
| SiO₂      | 0.77 |
| Al₂O₃     | 0.13 |
| CaO       | 54.41|
| MgO       | 1.09 |
| Fe₂O₃     | 0.10 |
| K₂O       | 0.02 |
| Na₂O      | 0.05 |
| TiO₂      | 0.04 |
| MnO       | 0.02 |
| Cr₂O₃     | 0.04 |

The XRD analysis as shown in Figure 2 revealed the presence of calcite (CaCO₃) as the main phase mineral and dolomite (CaMg(CO₃)₂) as traces mineral. This result is in agreement with XRF analysis which the chemical composition revealed small amounts of MgO hence exhibit low intensity diffraction peaks in XRD result.
2. Binder and hardener

The type of binder used in this study is unsaturated polyester resin with the appearance of hazy and pinkish solution. This resin has a low viscosity with the range between 450 and 600 cps that can be easily blended with fillers. While, the hardener or also known as catalyst is added into the mixture to initiate the polymerization reaction to transform the liquid to solid state. The most common hardener used is methyl ethyl ketone peroxide (MEKP) and in this work, the amount of 1\% by weight of resin content have been used.

2.3. Preparation of artificial marble

For artificial marble preparation, marble waste was sieved to obtain two different grain sizes; coarse (< 2 mm) and fine (< 250 µm). The polyester resin and fillers were mixed using an overhead stirrer for about 10 to 15 minutes and stirred at the constant speed of 200 rpm until the mixture well mixed homogenously. During mixing, the hardener is added into the mixture and then was cast into the mold and left in the vacuum at 700 mbar for 5 minutes to remove an air bubbles. For this study, a total of four samples has been prepared from the base ratio of filler to matrix at 70:30 (in weight \%). The mix proportions used in this study are shown in Table 2.

![Figure 2. XRD pattern of marble waste.](image)

**Table 2. Mix proportions of artificial marble.**

| No | Sample | Resin (wt %) | Fine particle < 250 µm (wt %) | Coarse particle < 2 mm (wt %) |
|----|--------|-------------|-------------------------------|-------------------------------|
| 1  | 70/0   | 30          | 70                            | 0                             |
| 2  | 60/10  | 30          | 60                            | 10                            |
| 3  | 50/20  | 30          | 50                            | 20                            |
| 4  | 40/30  | 30          | 40                            | 30                            |
2.4. Characterization of artificial marble

2.4.1. Water absorption test
The water absorption of the artificial marble specimen was determined as per ASTM D785. A total of five specimens with dimension of 12.7 mm x 12.7 mm x 12.7 mm was immersed in water bath at room temperature for 24 hours. The origin weight of the specimen before immersing is measured and denoted as W1. After immersing specimen in distilled water, each specimen was wiped off using a tissue to remove excess water from specimen’s surfaces and immediately weighed and denoted as W2. The rate of water absorption is determined using the following equation:

\[
\text{Water absorption (\%)} = \frac{(W_2 - W_1)}{W_1} \times 100
\]

(1)

2.4.2. Flexural test
Flexural strength of the artificial marble specimen was carried out according to ASTM D790. Five specimens of each composition with dimension 25.4 mm x 12.7 mm x 8 mm were prepared and tested under three-point bending load using the Universal Testing Machine (Model 3366, Instron) with a cross-head speed of 0.2 mm/min and the support span length is 20 mm.

2.4.3. Compression test
Compression test was performed according to ASTM D695. In order to determine the flexural strength, five specimens with dimension 12.7 mm x 12.7 mm x 12.7 mm were prepared. Compression tests specimens were also performed using the Universal Testing Machine (Model 3366, Instron) with a cross head speed of 0.2 mm/min.

2.4.4. Barcol hardness
The purpose of this test is to determine the hardness of artificial marble specimen by indentation of the portable Barcol Hardness Impressor indenter through the depth of penetration into the specimen. The marble specimen with dimension of 50 x 50 x 8 mm was prepared according to ASTM D258. A 10 readings of measurement were indicated and the average value was calculated.

3. Results and discussion
3.1. Water absorption
Figure 3 shows the effect of marble waste particle size and content on the water absorption properties of artificial marble. As observed water absorption values were varied between 0.32 to 0.39%. From this result, it can be seen that the rate of water absorption of sample incorporated with coarse particles at 10% (wt.) was found to be lower than the non-incorporated sample. This may be occurred when the fine particles fit easily within the pores between large particles, thereby decreasing the value of water absorption. However, when coarse particles increase by 20 and 30% (wt.), the rate of water absorption was observed slightly increases. As the higher content of coarse particle included in the mixture, the mixing process with the resin is more difficult due to the increase of the viscosity. The increasing of viscosity in the composition gives an effect to the flow capability and workability of the mixture, thus lead to entrap more air and produce a pores during the preparation of samples then water absorption increased simultaneously. However, the rate of water absorption found in this study is considered low which the commercial range of water absorption of artificial marble and as literature published reported from 0.09 to 0.40% [9]. Thus, it can be said that all the samples analyzed were in the range and the most optimum sample in this analysis is sample with composed of 60% (wt.) fine and 10 % (wt.) coarse particle (sample 60/10) with the value of water absorption is 0.32%.
The effect of marble waste particle size and composition on the flexural strength was analyzed as shown in Figure 4. As can be seen, the trend of the obtained result is not as expected, which there is generally slight decrease in strength with the addition of coarse marble waste particle from 10 to 30% (wt.). The decrease of flexural strength could be explained by the incorporation of larger size particle of marble waste which have an irregular shape as shown in Figure 1. During the mixing process with the fine particles, might be some of the mixed particles resulting an agglomeration in the sample thus increases the average particle size. The existed agglomeration in the sample could be acted as stress concentrations or weak point that caused a crack growth and finally breaks when poor stress transfer between the resin matrix and particulate is applied during testing. In spite of the incorporation of marble waste up to 10% (wt.) did not contribute to increase the flexural strength, the study conducted by Silva et al [10] has found that the flexural strength of artificial stone prepared by marble calcite waste and epoxy resin was between 30.5 to 37.4 MPa. While, Ribeiro et al [11] has obtained the value of artificial stone produced with marble residues, polyester resin and solvent was 21.5 MPa. Most probably the low value of the flexural strength is due to the use of the solvent. In present study, the average value of flexural strength was found between 51 to 60 MPa which much greater than literature reported and even higher than commercial value of solid surface in the market which recorded in between 28 to 54 MPa. Therefore, the results of this analysis have given an idea to the authors to enhance the aesthetic value of the artificial marble by modifying at least 10% (wt.) of coarse particle incorporated in the composition with no sacrifice of the technical properties.
Figure 4 presents the effect of marble waste particle size and composition on the compressive strength of artificial marble. The same trend as flexural strength was observed, where the compression strength was slightly decreased with the addition of 10 % (wt.) of coarse marble waste particles in the sample. Then, there are no significant changes in the value of sample incorporated of 20 and 30 % (wt.) coarse particle. According to Chiodi and Rodriguez [12], artificial stones with the value of compressive strength greater than 70 MPa is classified as high resistance materials. Hence, the obtained values of compressive strength in this study were recorded between 106 to 117 MPa, approximately 60% above the recommended value. It was verified that the artificial marble produced using a fine and coarse marble waste particle has a good mechanical performance. This is mainly due to the use application of vacuum during the preparation of the sample, which reduced the pores and also with the appropriate proportion of filler to the resin, which contribute a good wettability and adhesion between the marble particles.

3.3. Compression strength

Figure 5 presents the effect of marble waste particle size and composition on the compressive strength of artificial marble. The same trend as flexural strength was observed, where the compression strength was slightly decreased with the addition of 10 % (wt.) of coarse marble waste particles in the sample. Then, there are no significant changes in the value of sample incorporated of 20 and 30 % (wt.) coarse particle. According to Chiodi and Rodriguez [12], artificial stones with the value of compressive strength greater than 70 MPa is classified as high resistance materials. Hence, the obtained values of compressive strength in this study were recorded between 106 to 117 MPa, approximately 60% above the recommended value. It was verified that the artificial marble produced using a fine and coarse marble waste particle has a good mechanical performance. This is mainly due to the use application of vacuum during the preparation of the sample, which reduced the pores and also with the appropriate proportion of filler to the resin, which contribute a good wettability and adhesion between the marble particles.
3.4. Barcol hardness
From the Figure 6, it is found that the value of Barcol hardness has slightly decrease with addition of coarse particles at 10% (wt.) and the values are remains unchanged up to 30% (wt.). The values of Barcol hardness recorded in this study was between 25 to 27 HBa with sample composed of fine marble waste at 70% (wt.) shows the highest hardness value due to the compaction of fine particles which at this point, the fraction and distribution of filler are homogenous. Apparently that incorporation of coarse particle showed insignificant influence for the Barcol hardness properties.
4. Conclusion
In this work, artificial marbles were prepared with the composition of fine and coarse marble waste in the matrix of polyester resin was modified in order to assess its effect on physical and mechanical properties. It was found that incorporation of coarse particles in the composition did not showed any significant changes on the flexural and compression strength. However, for the effect of water absorption, artificial marble prepared with 60 % (wt.) of fine and 10 % (wt.) coarse particle (sample 60/10) showed the lowest value compared to non-incorporated. However, the mechanical strength of all the samples were in the range of commercial value of artificial marble properties and even higher from the specification reported in the literature review. As a conclusion, marble waste used in this study had showed a good potential to be utilized as particulate filler in producing artificial marble with enhanced properties with the suggested sample of 60/10 showed the optimum properties. Some recommendations for future work are also will be proposed as follows: 1) Modification of vacuum pressure to obtain the optimum parameter hence reduced the number of voids in the sample; 2) The use of different kinds of minerals and waste materials such as dolomite, silica sand and granite in the composition to improve their physical and mechanical properties and; 3) Modification of different particle size of fillers in the composition.

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