Article

Developments New Processing Of Marble Synthetic Based On Blend Polymers And Glass Sand As Reinforcement Filler

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Abstract

Artificial marble could be defined as a substitute product of natural marble. It is a new composite contain resin such as unsaturated polyester and reinforcement material as filler. Filler used in this study was glass sand. Both resin and filler are mixed mechanically or hand layout, then molded after cured to produce marble. Artificial marble is fabricated as the real marble by using pigments, chips etc. Onyx which is a kind of quartz that very transparency including aluminium trihydroxide, glass frits is used also as secondary fillers for decorative purposes. A detailed of resulting properties such as mechanical behavior are presented in this work.

The present study is directed to solving problems and provides artificial marble which exhibits an aesthetic pattern equivalent or superior to that of natural marble, has excellent mechanical strength and gloss, and does not contain impurities including bubbles therein.

Keywords: Marble, Unsaturated polyester, Glass Sand, Blend.

1. Introduction

Natural marble is an ideal material for the production of floor tiles, as it is not affected by environmental temperature changing. However, natural marble deposits are not adequate to satisfy the requirements of the structure and construction industry, and it is therefore necessary to investigate alternatives to natural marble [1].

Natural marble is generally heterogeneous with impurities and cracks. They are available in far off regions and require broad dealing with in molding and finishing. This increase the price for these types of marbles. To overcome these problems, artificial marble is produced [2].

Over the world, synthetic marble is quickly replacing natural marble. Its properties are almost better than those of the natural product. Besides, it displays high strength, lowshrinkage, minimal water absorption, high protection to corrosion by aggressive chemicals and to ageing and the effects of weather, easy maintenance and high abrasion resistance.

Artificial marble is prepared by mixing very strength resin, suitable filler in present of specific catalyst. Natural marble is present in faraway regions, it requires high treating in form and fibricating.This cause increasing in the cost of this industry. To solve these problems, artificial marble is produced. Artificial marble has high mechanical properties like weather resistance, flexural strength, surface hardness. There are a number of advantages provide by artificial marble above natural marble like low cost and amazing formation compared to natural marble. Artificial marble could be easily being formed for different shapes while natural marble is limited to flat surfaces. In spite of many best properties introduced by artificial marble, it also causes some flaws. For example, the marble surface could be exposed to scratch and so it must pay attention when put objects on it. So, a safety of marble could be enhanced by Pay attention to a number of important things such as molar ratio of resin, gel time,amount of catalysts,oven time, etc[3] .

Natural marble is a rock that forms through a natural process. It is characterized as does not change colours that stay a long time[4]. It is not effect by UV radiation , so it keeps the colour as the first day. Also it resist fire and heat and the marble surface is resistant to high temperatures. However, there are many disadvantages that natural marble has such as:

- Marble requires suitable care and keeping.
- Some marble is soft and suffers to break.
- Some marble has pores .
- Marble is very heavy, so it need more of labor for lifting it.

Aim of the work

The study aims to find a new method in the production of artificial marble by replacing the traditional fillers used with glass sand as an economically feasible method.

This is in addition to the use of polymeric blends to improve some of the mechanical properties of the composite, such as fracture when exposed to stresses or strong impact, by adding polymeric materials with the base resin to gain additional properties.

1.1. Marble's Composition

Artificial marble requires specific compounds as raw materials such as resin, filler, gel coat, pigment and catalyst. Fillers must be present at range of 80-85 % by weight [4]. Generally, the filler used could be calcium carbonate or silica. In this work, glass sand was used as filler. The resin behave as abinder and Subsequently increase the forcement of the marble. Resin content in artificial marble always about 15 to 20%. The resin used in this work was unsaturated polyester (UPE). Artificial marbles are manufactured by mixing the components, fillers, pigment and catalyst with the resin. Then, the mixture will be molding and curing in a mold. Pigment is used to get the required color. Marble could be pre-designed included shapes similar to the natural marble in color and veining . Decorative additives are used depending on fashion, design and should not exceed 2%byweight. Gel coats require for artificial marbles to

prevent susceptible to stress cracking and to give marble an attractive exterior appearance. Also, there are some other constituents included in marble preparation such as hardener, accelerator, coupling agent, curing agents, antioxidants, catalyst, coupling agent, etc. Coupling agents are chemical compounds that form a bonding between the mixture components to combine fibers or particles with the matrix polymer. These materials modify the functionality of surface filler effectively to strong bonding with the polymer chains. Initiators like methyl ethyl ketone peroxide MEKP provide free radicals that required for polymerization.

Fillers are mixed with the resin and filled in amould and followed by a curing process. Conventionally hand lay upmoulding had been used [5].

1.1.1. Unsaturated polyester Resin

Unsaturated polyester resin could be prepared by condensation polymerization reaction between unsaturated acid like phthalic acidand anhydride like maleic anhydride, and glycol like ethylene glycol, propylene glycol. [6]. In curing of unsaturated polyesters,which its convert from liquid state to solid by crosslinking that occurs between styrene monomer and the unsaturation bonds in the polyester. Maleic anhydride provides unsaturation to the polyester which assistto copolymerized with styrene in presence of initiator like MEKP. The density of crosslinking could be adjusted by controlled of the molar ratio of maleic anhydride [7]. The observability of the inside patterns and the designs of marble prepared by unsaturated polyester is very less because it have lesser transparency [8]. The ratio of glycol in the resin at least is more 30 mole % . Metallic salts of organic acids were used as cure accelerators, especially cobalt salts, for example, cobalt naphthenate and cobalt octoate.

Styrene is used as crosslinker in the copolymerization process.Copolymers composite at least one vinyl monomer.

1.1.2. Plasticizer

Plasticizer is chemical compound used to enhance the flexibility and elongation of polymer's that ease the workability. Addition of a plasticizer generally causes a reduction in the cohesive bonding forces along the polymer chains.After that the chains of polymers could be move more freely to one another, and the hardness of the polymer is reduced.An example of specific type of plasticizer is the phthalate plasticizer.

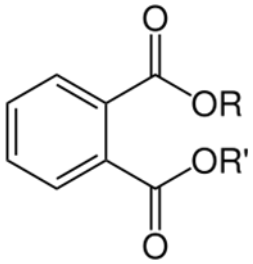


Figure 1.phthalate plasticizer

1.1.3. Filler

In this study, glass sand is the filler used as reinforcement with unsaturated polyester composites in artificial marble preparation which considers as an important materials in construction process with size of less than 4.75mm.It is also termed as fine aggregate. When it used as reinforcement mix, it must be cleaned from clay, salt and other organic compounds at specified limit" [9].

Figure 2 Glass Sand

2. Materials and Methods

2.1. The Matrix

The matrix used this study was unsaturated polyester resin(UPE), brought from (Sika Building Trust company), which is in a yellow liquid form at room temperature. It is a thermoset polymer, that converts from the liquid state to the solid at the addition of the initiator (MEKP), in a ratio of 0.2% wt. of UPE**.**

2.2. Reinforcing materials

Sand glass was used as reinforcement filler as powder after grinding it to obtain the granular size 75μm. Before grinded it must be washed and cleaned from impurities.

2.3. Silane Coupling Agent

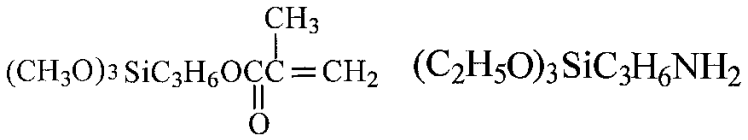
Coupling agent is an additive used with the mixture of composite by 0.2% wt of blend. It is a chemical which improves the adhesion between the components of composite material. It is a chemical compound has molecules contain functional sites that bond organic with inorganic compounds.

"Silanes" are compounds of binary silicon-hydrogen and compounds have four replacement on silicon, contaning organosilicon compounds. Examples

like **trichlorosilane ($SiHCl_3$), tetramethylsilane ($Si(CH_3)_4$), and tetraethoxysilane ($Si(OC_2H_5)_4$)**

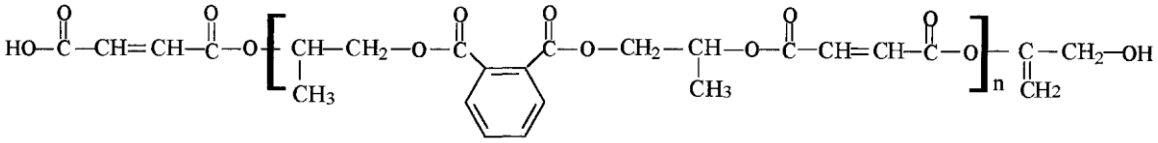
-methacryloxypropyl trimethoxy silane (MPS) and

-aminopropyltriethoxy silane (APS),



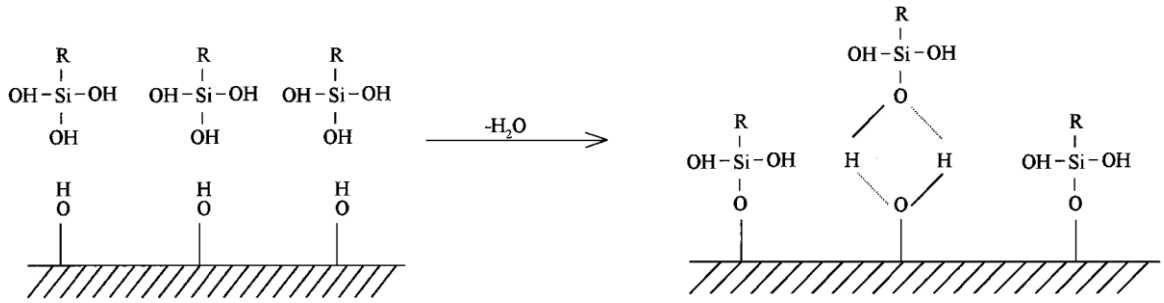
MPS **APS**

Figure 3. types of Silane Coupling Agent



UPE

Figure 4. Chemical structures of the UPE resin



Scheme 1. Structure of of silane surface -treated glass fiber

2.4. Manufacture of Artificial Marble

Specimens of composite with different ratios of filler ranging from 10 to 30% were prepared using hand lay-up method. First, the glass sand (**G.S)** and blend of UPE resin with plasticizer by 5% wt of resin were mixed until completing of **G.S** wetting, then 2% MEKP was added as initiator of free-radical polymerization. Mechanical mixers have been used for the composites mixing. The components are then mixed until the resin is distributed during the filler. After mixing of composite, a little amount of pigment is then added to the mixture. Finally, the mixture was poured into the mold coated with a thin layer of wax as a release agent [10].

Gel coat is applied on the mold. It used in general coating thixotropic applications. Gel coats are very important application of unsaturated polyester resins. It is the essential layer that is applied on the mold surface and it will form the visible side of the last product after demolding .

To make sure the preparation of high-quality products, the manufacturing process of artificial marble involves several steps as following[11]:

Step 1: Preparation – The crushed glass sand particles are cleaned and classified based on size. They are must dried to remove the moisture content that may effect on binding with resin.

Step 2: Mixing – The dried particles are combined with the unsaturated polyester resin, silane coupling agent, and pigments. The mixture is blended to achieve a compatable composition.

Step 3: Molding – The mixed material is poured into molds that are tailored for specific requirements like countertops, sinks, bathtubs, shower panels, and other architectural elements.

Step 4: Curing – The molds are placed in a warm place at which the mixture undergoes a curing process. This assist the resin to harden and bind the glass sand particles together, create a solid and durable structure.

Step 5: Finishing – After the process of curing, the artificial marble product was released from the mold. Then it was polished, trimmed, and refined to get the desired texture and appearance.

Figure 5 Samples of Synthesis Artificial Marbles

2.5 Improving of Mechanical and Surface Properties

When studying the mechanical properties of artificial marble as a polymeric composite we find that it gets cracks and fractures when subjected to high loads and stresses, so it is characterized brittle similar to the rest of the composites. To treated or avoid this problem that affects the properties of marble, it must submit to a variety of procedures.

One of these treatments by use blends polymer method. It is carried out by addition of plasticizer to UPE resins in specific ratio which been noticed caused an increase in the mechanical properties which develop resistance from cracking and coated marble pieces. Experimental with different ratios of plasticizer have shown that improvement in UPE properties which directly proportional to the amount loading. This is due to elasticity created in the UPE matrix which conferred flexibility to the cured polymer. Also tensile, elongation affected by blending idea. Therefore, all other mechanical properties must be taken into account when a new type is used in the modification[12].

Also, glass fiber reinforcing increased the resistance to cracks and bend resistance of the product. When fiber content increases than of 50 wt. % highly reduced the strength of the composite because of poor.

3. Results and Discussion

3.1. Mechanical Properties Polymer Blends

Economically**,** modification of polymers is more feasible than progress of latest monomers for the synthesis of new kinds of polymers. Much properties could be added to obtained composites either by specific polymerization methods or by blending method, and reinforcing polymer compounds. polymer blends are polymer modified by simple mechanical mixtures of two or more polymers created a new type of materials[13].

The target of blending is to get best properties of blend polymer from mix two or more polymers. As example,to solve brittle polymer problem ,flexible polymer used to decrease the rigidiy and prevent cracks

In this study, the work on blends of UPE with plasticizer to modify the mechanical properties of UPE resin in composites to make it suitable for artificial marble applications. Two main steps were conducted; the first was mixing the plasticizer with unsaturated polyester resin and studying its results. The second step was adding the filler and/or glass fibers with the plasticizer and studying the resulting mechanical properties to see the development of these characteristics [14]

3.2. Mechanical Preparation of (plasticizer / UPE) Blend

A plasticizer is a polymer additive that serves to increase the polymer's flexibility, elongation or ease of processing (workability). In more technical terms, the addition of a plasticizer generally causes a reduction in the cohesive intermolecular forces along the polymer chains. The chains can then move more freely relative to one another, and the stiffness of the polymer is reduced. Mixing of UPE resin and plasticizer get best

mechanical properties blend. Below are the results of those checks in order of mixing percentages.

It was observed that the mechanical properties of the polymer changed with the change in the percentage of plasticizer added. We noticed that values of some mechanical properties increase with increasing in the percentage of plasticizer, then they begin to decrease gradually, and these properties include compressive strength, fluxrural strength, and impact strength. While we noticed a decreasing in tensile strength and hardness with an increase in the ratio of plasticizer to polymer. As for elongation, we notice an increase in its value with increases in the percentage of plasticizer.

Among the percentages of plasticizers, the addition rate of 5% was the best in giving the mechanical properties of the mixture .This ratio was chosen to make a polymeric composite for the marble model. Following table 1 show the mechanical properties of (plasticizer / UPE) Blend.

Table 1 the mechanical properties of (plasticizer / UPE) Blend

Mechanical Test	**UPE 0%**	**plasticizer / UPE 2.5%**	**plasticizer / UPE 5%**	**plasticizer / UPE 7.5%**	**plasticizer / UPE 10%**	**plasticizer / UPE 12.5%**
Compression Test	110	115	111	95	84	71
Flexural Test	80	88	99	87	73	61
Impact Test	2.71	2.87	2.93	3.2	2.85	2.64
Tensile Strength	38	34	30	26	22	19
Elongation %	2	2.3	2.7	3	3.3	3.6
Hardness Test	41	38	34	29	25	20

3.3. [G.S / (Plasticizer/ UPE) 5%] Composites

The optimal proportion of the polymeric blend with good mechanical properties was chosen for the purpose of mixing it with G.S as filler to prepare the polymeric composite. Mechanical properties were investigated for the purpose of exploring its suitable industrial application. Following are the discussions of the results[15]

Table 2.The mechanical properties of [G.S / (Plasticizer/ UPE) 5%] Composites

Mechanical Test	G.S/ Blend 0%	G.S/ Blend 10%	G.S/ Blend 15%	G.S/ Blend 20%	G.S/ Blend 25%	G.S/ Blend 30%
Compression strength	111	116	112	107	102	96
Flexural strength	99	95	91	87	82	78
Impact strength	2.93	2.87	2.81	2.77	2.7	2.1
Tensile Strength	30	35	39	44	48	53
Elongation	2.7	2.3	2.0	1.7	1.2	0.93
Hardness	34	38	43	48	52	57

3.3.1. Compressive Strength

Figure 6 shows a clear increase in the compressive strength at the first addition, and then the strength begins to gradually decrease. The reason for the decrease is due to the penetration of the filler between the chains of the matrix (the polymeric blend), which impedes the full crosslinking of the polymerization of free radicals.

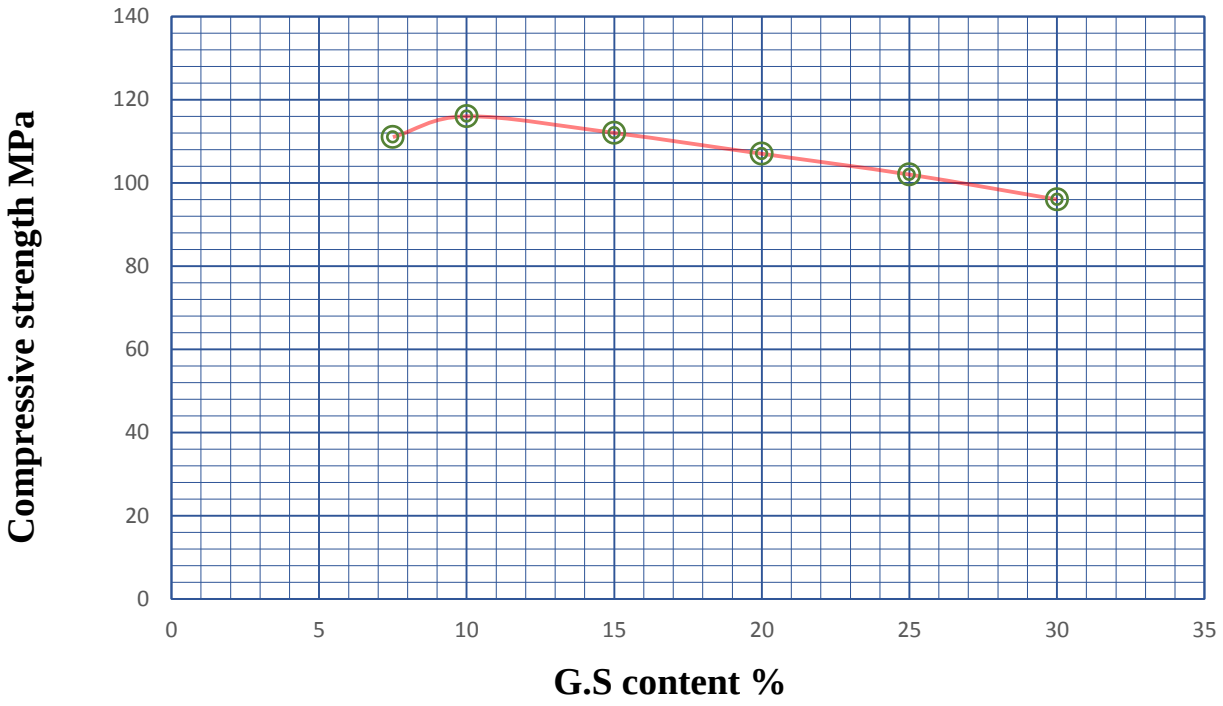


Figure 6 Compressive strength of [G.S/(Plasticizer/UPE) 5%] Composites

3.3.2. Flexural Strengt

Figure 7 shows a decrease in the flexural strength values with the increase in the ratio of the filler to the polymeric mixture. The reason for this is due to the decrease in the flexibility of the polymeric composite as a result of its increased brittleness. It may be attributed to the occurrence of cavities in the structure of the composite.

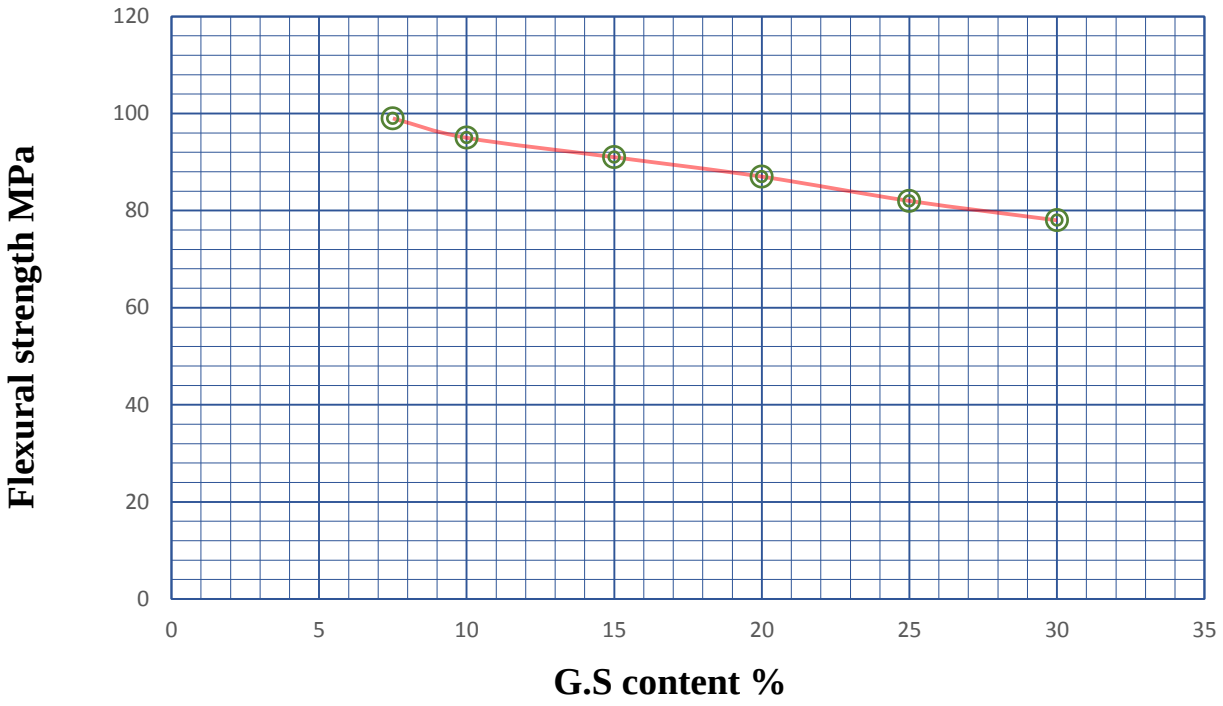


Figure 7 Flexural strength of [G.S/(Plasticizer/UPE) 5%] Composites

3.3.3. Hardness

Figure 8 shows increases in the hardness values with the increase in the ratio of the filler content in the polymeric blend. The reason for this is due to the ability of G.S reinforcement with low ductility.

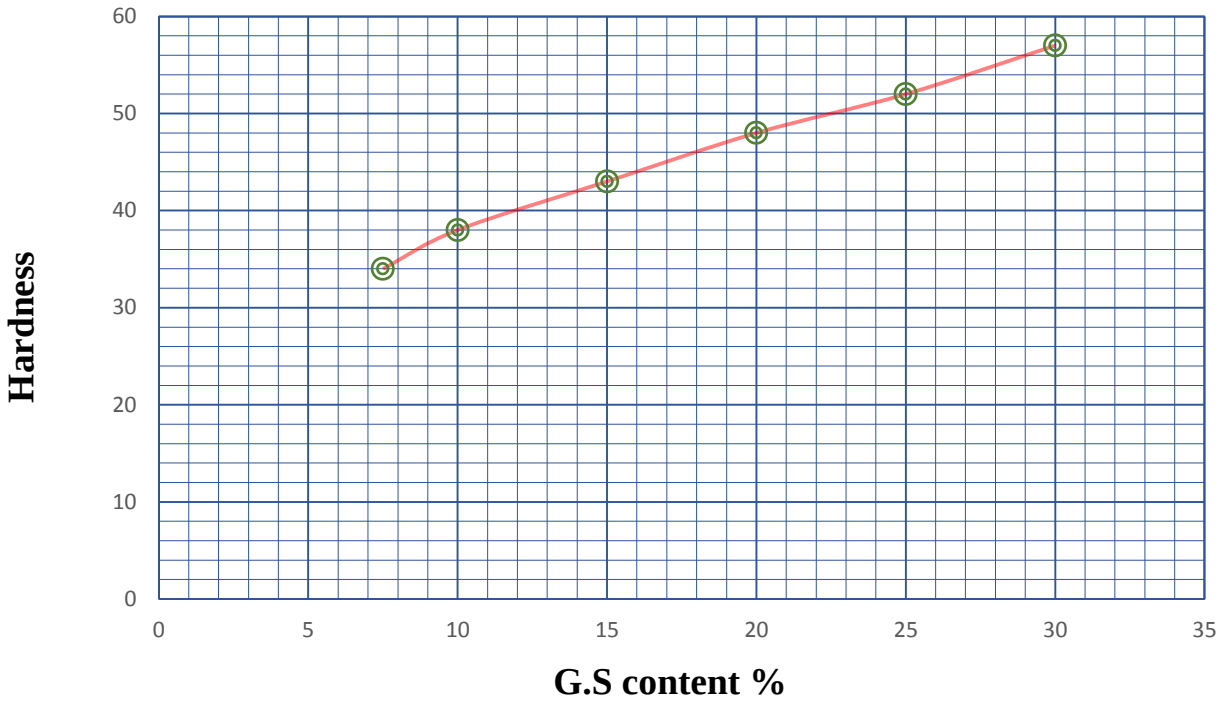


Figure 8 Hardness of [G.S/(Plasticizer/UPE) 5%] Composites

3.3.4.Tensile Strength

Figure 9 shows an increase in tensile strength gradually with filler content increases. The reason for the increase in tensile strength at first is due to the increase in the stiffness of the polymeric composite and the decrease in its flexibility.

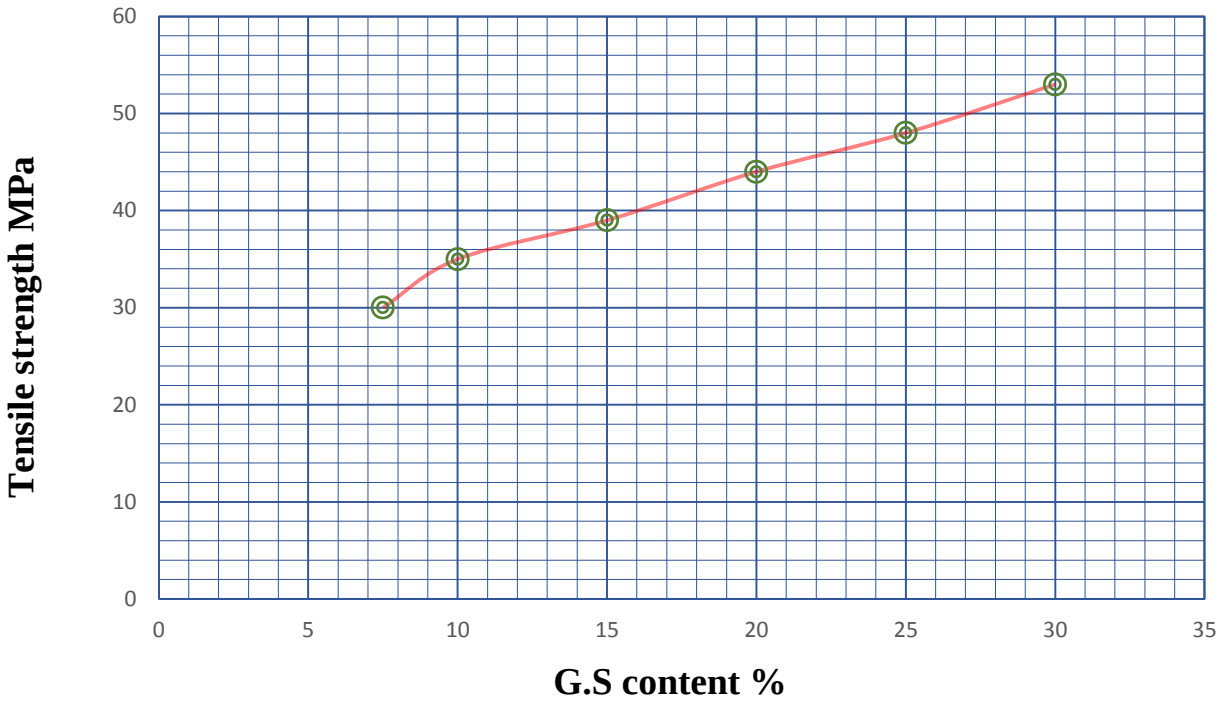


Figure 9 Tensile strength of [G.S/(Plasticizer/UPE) 5%] Composites

3.3.5. Elongation

Figure 10, showed that increasing of G.S content will decrease the crosslinking of the two polymer chains Thus, the elasticity of the mixture will decrease, which means that the elongation will decrease.

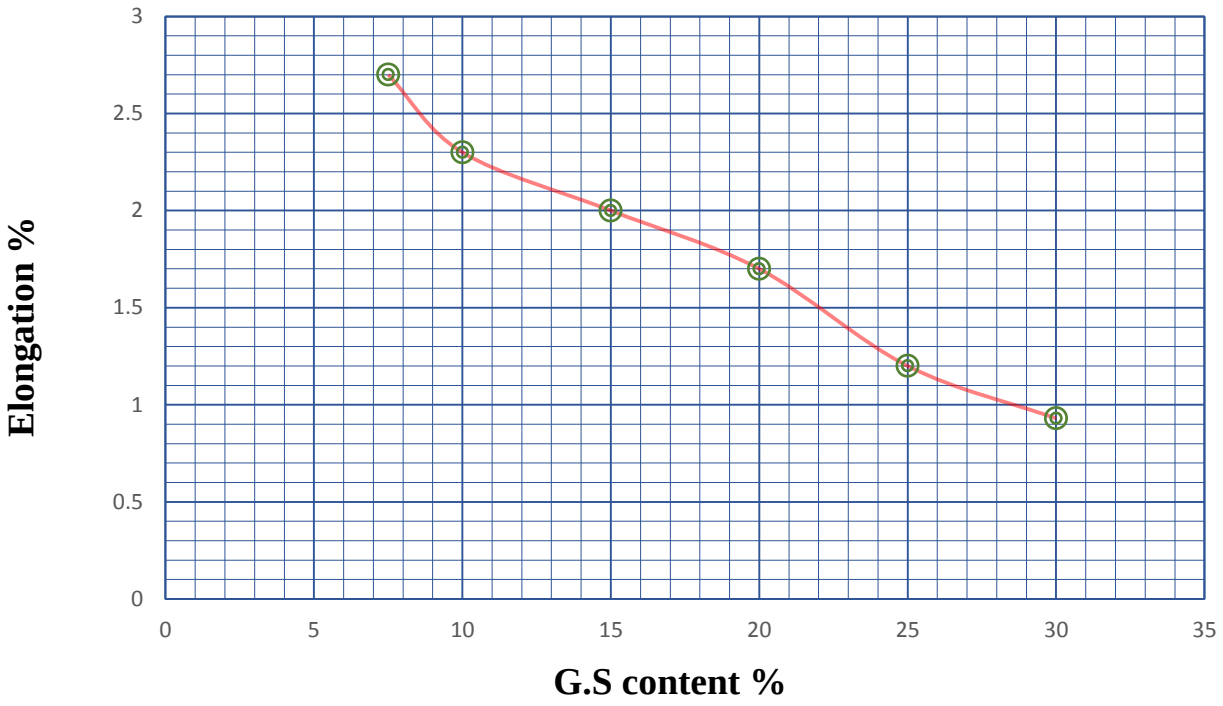


Figure 10 Elongation of [G.S/(Plasticizer/UPE) 5%] Composites

3.3.6. Impact Strength

As shown in Figure 11, the impact strength of the filler decreases when the G.S content increases. This indicated the decreases of the plasticity of the blend.

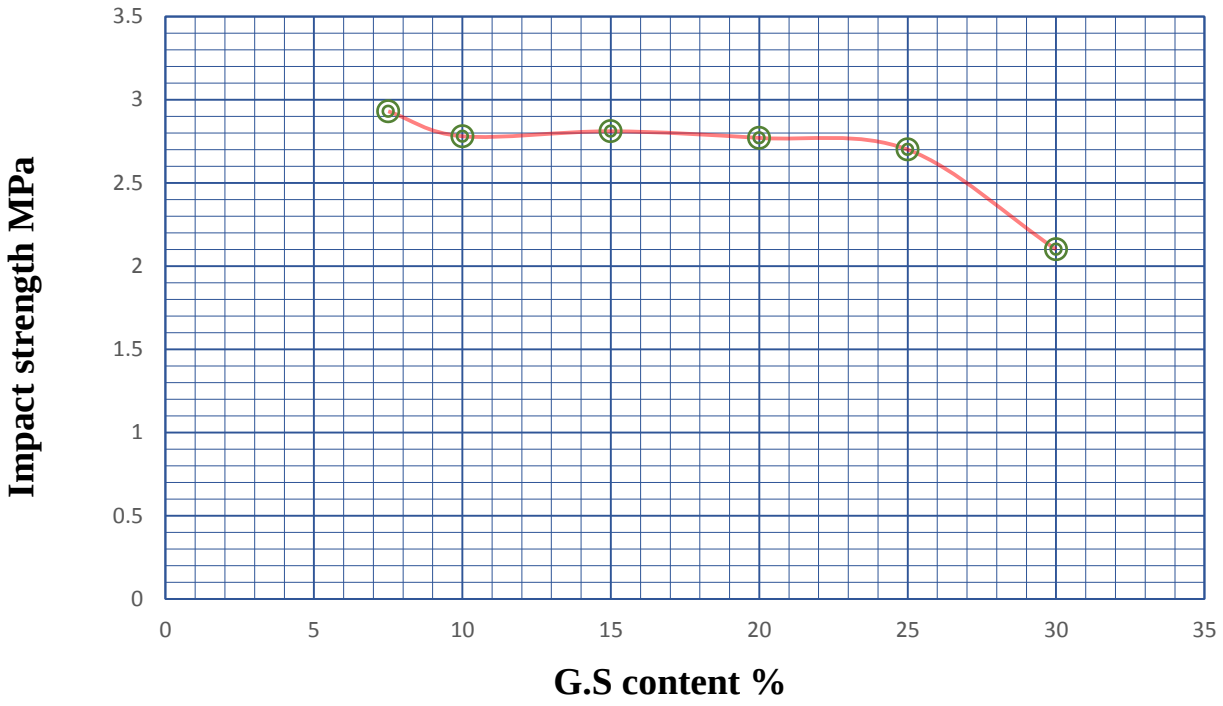


Figure 11 Impact strength of [G.S/(Plasticizer/UPE) 5%] Composites

3.4. Microstructure Tests

3.4.1.Microstructure of UPE-Plasticizer Blend

The SEM micrograph for the morphology analysis of the blend of UPE with Plasticizer that prepared is shown in Figure 12.The glassy fracture surface in the figure showed not flat ripples which cause the brittle fracture. It also showed that there was two-phase exist in the morphology of the sample where the resin is completely cured. SEM micrograph showed a well dispersion of UPE- Plasticizer blend which spherical particles with smaller size compared to other UPE -plasticizer sample blend which show that polymer is compatible with UPE in this concentration[16].

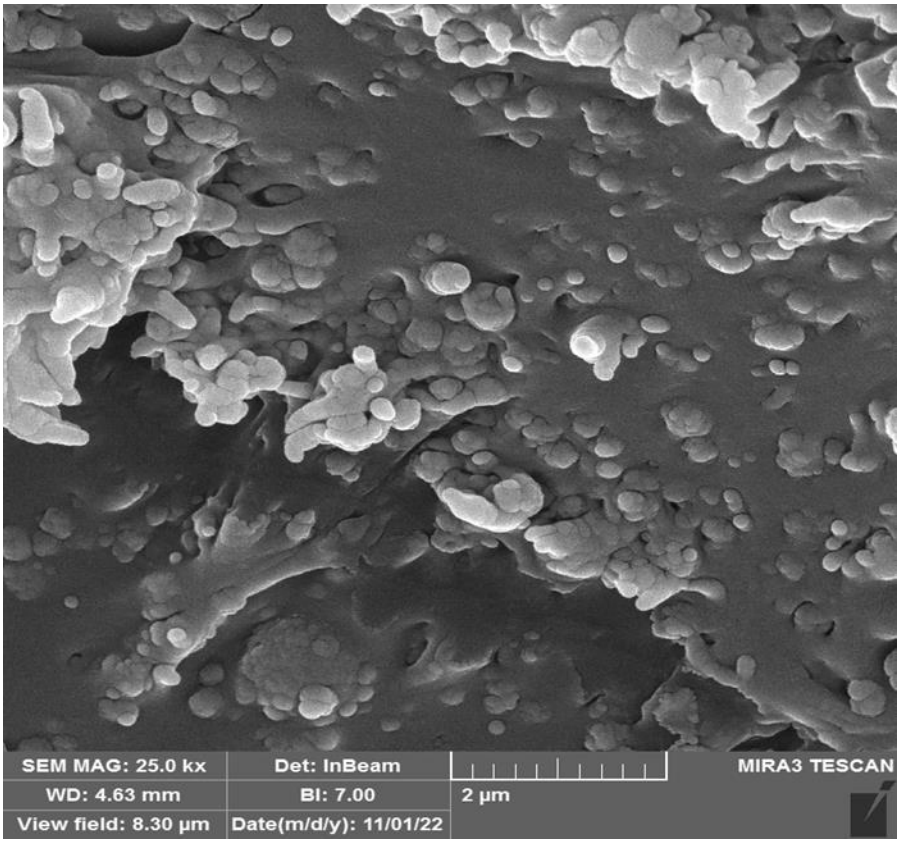


Figure 12 SEM micrographs of UPE-Plasticizer Blend Sample

3.4.2. Microstructure of UPE-Plasticizer Blend Composite

G.S particles showed **irregular** distribution in UPE resin matrix because of the mixing was carried out by hand layout and the **viscosity** of the UPE was rather high. These reasons made the G.S particles aggregate to form different shapes in some regions which take place in a whole matrix and occupy the spaces among the molecules(see figure 13) .

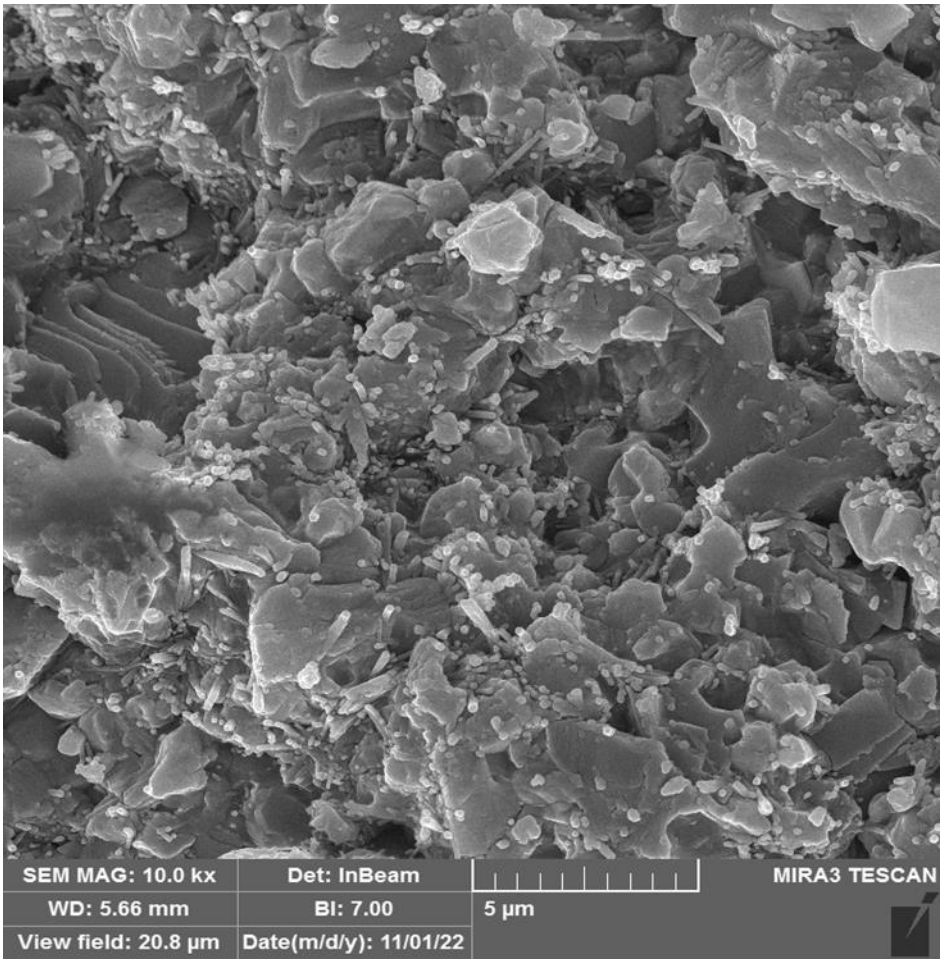


Figure 13 SEM micrographs of UPE-G.S Composite Sample

Conclusion

The high quality of artificial marble, light weight, beautiful shape, less expensive cultured marble has become a competitive alternative for natural marble. Traditionally, resins like unsaturated polyester are mixed with fillers, pigments and applied on molds followed by curing to yield artificial marble. The mechanical and aesthetic properties of artificial marble depend on the resin and filler composition together with other decorative substances like pigments, chips etc. Many works have been done to simulate the look of natural marble as closely as possible. Transparent chips have been used to give a transparent surface so that the inner veining is visible. Despite these several

advantages, it also has some drawbacks like lower hydro thermal shock resistance and proneness of surfaces to easily get scratched. Yet its grand look and cheaper price makes it very attractive. In general, the unsaturated polyester resin is suitable for mixing with suitable fillers and other additives in the production of artificial marble.

Properties of artificial marble depend on the composition and other decorative materials such glass fractures ,pigments, etc. Many things have been present to produce marble has the look of natural marble. Also transparent surface has been done to give visible inner veining. Artificial marble could be easily manufactured into shower rooms. Artificial marble has some disadvantages like lower impact strength and ability of surfaces to easily get scratched. However, it's very attractive because of its grand look and cheaper price.

In this study, the results obtained exhibit the feasibility of insert glass sand into the composite to manufactured artificial marble. In spite of the insert of sand resulted in the collaps of some mechanical properties of the composites, the manipulation concerning the ratios of the components of the resin when preparing and changing the proportions of mixing glass sand with the resin gives different results that can be used in choosing the optimal ratio for mixing.

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