Tensile and Liquid Absorption Properties of Polyester-based Composites as Alternative Marble Materials

Lamees Khalaf[1*], Israa. Ghazi[2]

Abstract

Three polyester-based composite materials were prepared with different volume fractions of three types of fillers (i.e. cement, gypsum and limestone) with the aim of improving the tensile and liquid absorption properties of the former for the synthetic marble industry. The tensile strength, modulus of elasticity, toughness, ductility and liquid absorption percentage of the composites were characterized. Results revealed an increase in the modulus of elasticity, tensile strength and toughness of all prepared composites, as well as a decrease in their ductility, with increasing filler amount. The liquid absorption values of all composites increased with increasing filler content. The composites were able to absorb water extensively but absorbed benzene, kerosene and gasoil minimally.

Key Words: Tensile Properties, Liquid Adsorption, Polyester Composite Materials, Synthetic Marble.

DOI Number: 10.14704/nq.2021.19.12.NQ21197 **NeuroQuantology 2021; 19(12):55-68**

Introduction

Since hundreds of years ago, composite materials have appeared and are widely used in a variety of applications that increase life quality. Composite materials have a variety of features that make them useful for a variety of industrial applications because they combine the properties of two or more substances to overcome the shortcomings of each. They can also modify their properties by altering the kind and proportions of its constituent materials, as well as their design and production techniques. As a result, designers and engineers are concentrating on the usefulness of engineering materials that have made their way into numerous industrial domains. They were made through a series of processes in order to produce the required design and construction structures that fit the functional performance as well as analyze the failure in this performance. In response to the needs of development and renaissance, which is targeted at increasing the product's design and production performance. There are endeavors in structural engineering to construct structures that are strong, durable, and reliable in terms of their durability and corrosion resistance.

Polymeric materials have been increasingly popular in building construction, the automobile sector, and aerospace technology over the previous few decades. In most industrial fields, polymeric materials can virtually compete with conventional materials. Polymers have made significant contributions to the production of better, cheaper, and more functional products in the manufacturing, appliance, and amusement and leisure industries.

Corresponding author: Lamees Khalaf

Address: [1*]College of Engineering, Al-Muthanna University, Iraq; [2]Department of Materials Engineering, College of Engineering, University of Al-Qadisiyah, Iraq.

[1*]E-mail: drenlameesmat@mu.edu.iq

[2]E-mail: israa.ghazi@qu.edu.iq

Relevant conflicts of interest/financial disclosures: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Received: 18 October 2021 **Accepted:** 24 November 2021

Incorporation of stiff and hard fillers into weak polymers may result in improved polymeric composite materials with suitable mechanical and physical properties on account of synergistic effects between the filler and matrix (Termonia, 1990). Moreover, the resulting composite may gain new chemical and physical characteristics resulting from the combination of these materials (Constantinides, 2013).

Marble is a tough crystalline rock made from limestone or dolomite; calcite minerals make up the majority of pure white marble. Marble is often used for the interior design and exterior decor of houses and building, including walls, kitchens, stairwells and toilets; it may also be a source of raw materials for crafts and sculpture ((Machmud, Yudha, & Anhar, 2019). Although natural marble is a hard rock, it reacts with acidic substances and suffers from discoloration. They are also quite heavy and require high maintenance costs.

Artificial marble is a synthetic material composed of small marble debris, stone powder, quartz, sand, colophony, plastic, cement and acrylic glue mixed at a predetermined ratio. Compared with natural marble, artificial marbles is lighter and, thus, more portable; it is also easier to install and more cost effective.

Artificial marble is made of natural marble or granite crushed stone for the filler, with cement, gypsum and unsaturated polyester resin as a binder, after molding, grinding and polishing made. A number of researchers have attempted to produce synthetic granite for lightweight and low-cost seats, tables and benches with sufficient mechanical strength. Ismail used polyester resin as an in expensive polymer for fabricating polymer/composites reinforced with cement mortar and clay to produce high-performance composites for general purposes (Ismail, Ali, El-Milligy, Afifi, & chemistry, 1998). Ribeiro investigated the synthesis of synthetic marble by using a polyester matrix with varying amounts of calcified marble residue and then assessed the density, water absorption, mechanical resistance and stains and scratches of the resultant products. A composite with good strength was finally obtained (Ribeiro & Rodriguez, 2015). Mahayatra et al. determined how different particle sizes affect the mechanical properties of marble composites. The authors demonstrated that particles of 140 mesh provide maximum flexural strength and hardness via their even distribution and bond formation in the matrix (Mahayatra, Supriadi, & Savetlana, 2013).

Costa developed a polyester resin/marble powder hybrid made mainly from ornamental stone cuttings. The composite provided a suitable alternative to natural marble or granite for the production of structures with low strength requirements, such as shelves, tables, benches and bathroom pottery (Costa, 2017; Souza, Silva, & Souza, 2020). Luiz produced four composites of polyester and cement and then studied their tensile and flexural strength, impact capacity, water absorption percentage, and density, thermal and curing properties. In contrast to their resin strength, which increased, the mechanical properties of the composites generally decreased with increasing proportion of cement powder(Souza et al., 2020).

Also, the use of waste creates from mineral fillers like marble waste particulates in the manufacturing of polymer composites are usually cost-effective fillers, therefore Abenojar et al. developed a composite material for use in the habitation industry as a floor or wall in buildings, built of a natural raw material of marble (waste powder from quarries or plate manufacturing) with appropriate mechanical and fire resistant capabilities. Polyester matrix composites containing 50 wt.% of marble and 3 wt.% of glass fiber (short fiber or mesh) were created to attain this goal. Results show that marble improves the mechanical properties of polyester and the effect of the glass fiber depends on its morphology (fiber or mesh). The high percentage of ceramic material added to a polymer matrix composite makes the importance of this study.

The purpose of this study is to develop in expensive, tough and lightweight marble materials with improved properties compared with traditional materials, such as, natural marble and granite. To this end, the mechanical properties of three different resin composites with varying filler contents were studied. The minimum filler volume fraction that could endow the composites with sufficient strength was also determined. As a result, this research focused on creating composites with polyester resin as the matrix and (Cement, Gypsum, and Limestone) powder as fillers, which appears to be a scientifically recent idea.

Experimental

1. Matrix Material

Unsaturated polyester (UP) resin is the most commonly used matrix in polymeric composites because of its many advantages, which include low cost, ease of processing, cold and fast curing, good appearance, easy pigmentation and usability with clear and readily available moulds(Annamalai & Ramasubbu, 2018; Bodur, Englund, & Bakkal, 2017). The UP resins used in this work were supplied by Saudi (SIR) Company and mixed with a hardener (i.e. methyl ethyl ketone peroxide). The properties of the UP are listed in Table 1.

Table 1. Tensile properties of UP

Tensile Strength (MPa)	Modulus of Elasticity (MPa)	Ductility (%)	Toughness (J/mm^3)	Density (kg/m^3)
27.2	2750	1	0.135	1200

2. Fillers Materials

- *Cement*

Portland cement accounts for over 60% of all cement produced worldwide. This type of cement is carefully prepared from calcium, silica, aluminum and iron oxides at specific proportions (Barros et al., 2020).
The ordinary Portland cement used in this work was produced in Iraq by the Al-Muthanna Cement Factory (fineness, 3220 cm^2/gm).

- *Gypsum*

Gypsum is a very soft powder made up of calcium sulphate dihydrate; it exhibits high fire resistance, excellent workability and pleasing aesthetic properties ((Pervyshin et al., 2017)).
The ordinary gypsum used in this study was obtained from Al-Anbar Province (fineness, 8%) remained on sieve No.16 Not less than 8%.

- *Limestone*

Limestone with a fineness of 9800 cm2/gm was passed through a sieve to obtain particles of a uniform size (i.e., 0.125 mm).

Samples Preparation

Samples were prepared by the hand lay-up technique (Figure 1.a). The masses of the reinforcement materials (i.e. cement, gypsum and limestone) were weighed according to the required volume fractions and mixed with calculated masses of the UP resins and hardener.
The mixture was poured into the designed moulds, left to solidify for 24 h at room temperature, and then dried in an oven for 1 h at 55°C. This last step was necessary to achieve complete polymerization, optimal coherency and low residual stress (Qhazi, 2017).

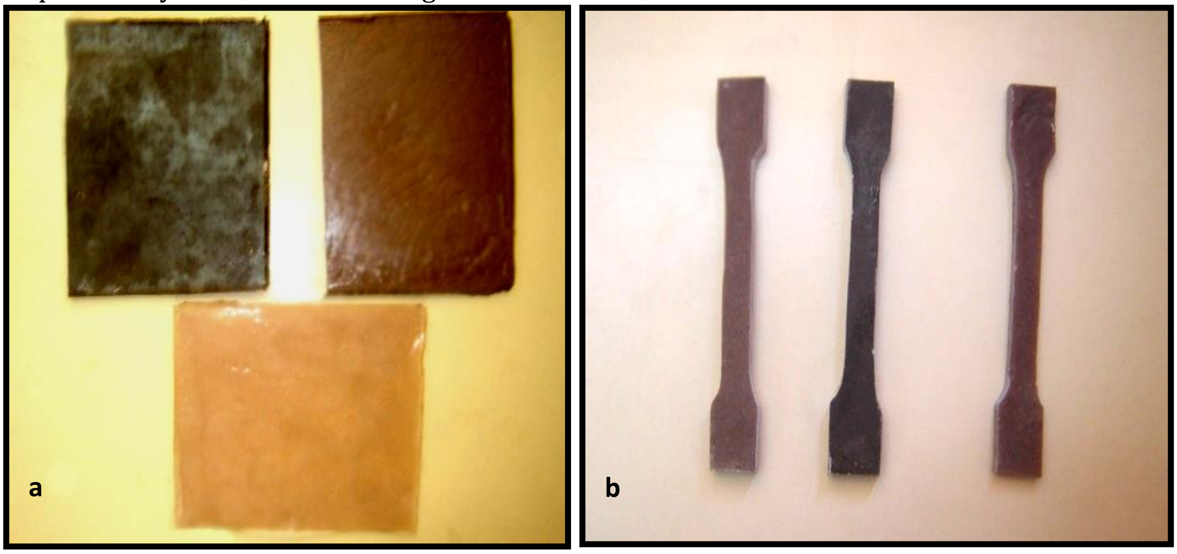

Figure 1a). Samples hand lay-up b). Tensile sample prepared according to ASTM D638

1. Tensile Test Samples

Tensile tests (Figure 1.b) were conducted on all of the composite materials (Table 2) according to ASTM D638M at room temperature.

2. Liquid Absorption Test

Liquid absorption tests were conducted on all composite materials according to ASTM 570 by immersion in distilled water, benzene, kerosene and gas oil at room temperature, as detailed in Table 5. At specific time intervals, the samples were collected, blotted dry and then weighed using a precise balance machine to determine the amount of liquid they absorbed. The specimens were weighed regularly after 24, 48, 72, 96, 120, 144, 168, 192, 216, 240, 264, 288, 312, 336, 360, 384, 408, 432, 456 and 480 h. Liquid absorption was calculated from the weight difference of the samples. The percentage weight gain of the samples was measured at different time intervals by using the following equation (Mohammed & Journal, 2016).

$$\text{Water absorption } (\%) = \frac{(W2-W1)}{W2} \times 100 \qquad (1)$$

where W_1 and W_2 are the weights of the dry and wet samples, respectively.

Table 2. Details of the test samples

Number of samples	Volume fraction of fillers	Gypsum	Lime	Cement
0	UP (pure)	-	-	-
1	5%	-	-	5%
2	10%	-	-	10%
3	15%	-	-	15%
4	5%	-	5%	-
5	10%	-	10%	-
6	15%	-	15%	-
7	5%	5%	-	-
8	10%	10%	-	-
9	15%	15%	-	-
10	15%	-	7.5%	7.5%
11	15%	7.5%	-	7.5%
12	15%	7.5%	7.5%	-

Results and Discussion

1. Tensile Properties

The mechanical properties of the composites obtained from the tensile tests are listed in Table.

Table 3. Tensile properties of the composite samples as per ASTM D638

Number of samples	Volume fraction of fillers	Tensile Strength (MPa)	Modulus of Elasticity (MPa)	Toughness (J/mm³)	Ductility (%)
0	UP (pure)	27.2	2750	0.135	1
1	5%	66.9	2800	0.77	2.2
2	10%	71.15	3800	0.68	1.8
3	15%	72.1	3000	0.64	1.9
4	5%	67.56	3050	0.8	2.3
5	10%	79.2	3960	0.75	1.9
6	15%	85.6	3490	0.68	2
7	5%	70.1	2995	0.96	2.3
8	10%	86.47	4100	0.9	2.2
9	15%	88.1	4150	1.02	2.5
10	15%	100.1	3150	1.35	2
11	15%	80.5	4306.8	1.27	1.5
12	15%	75.2	3707.4	1.05	1.8

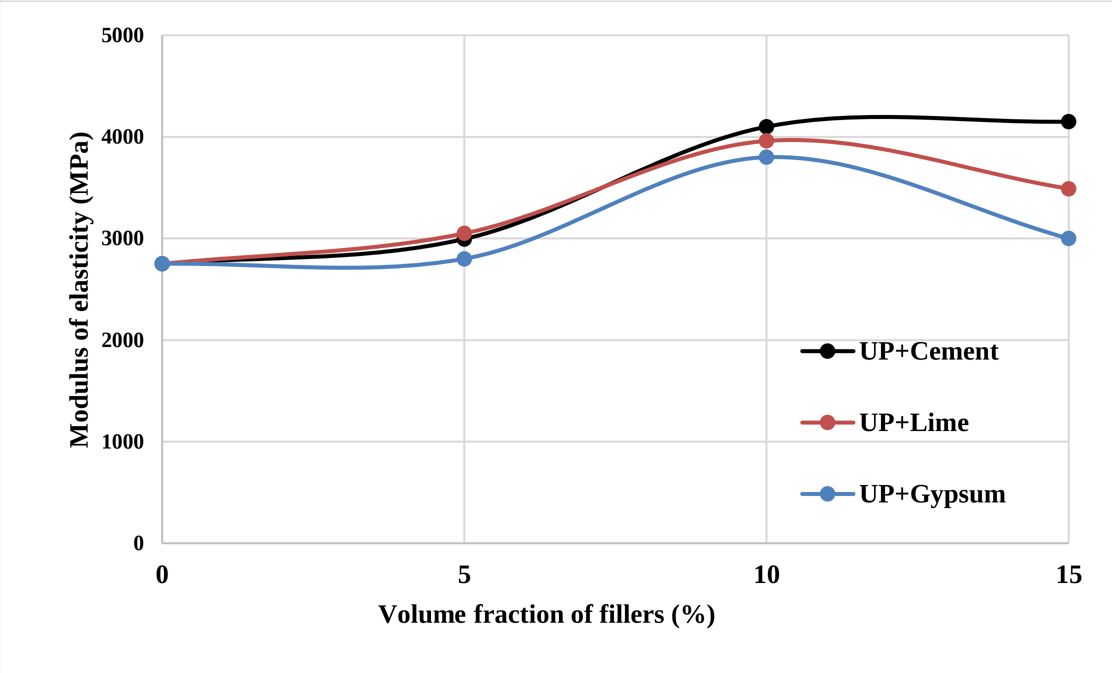

Figure 2. Modulus of elasticity of samples 0, 1, 2, 3, 4, 5, 6, 7, 8, and 9

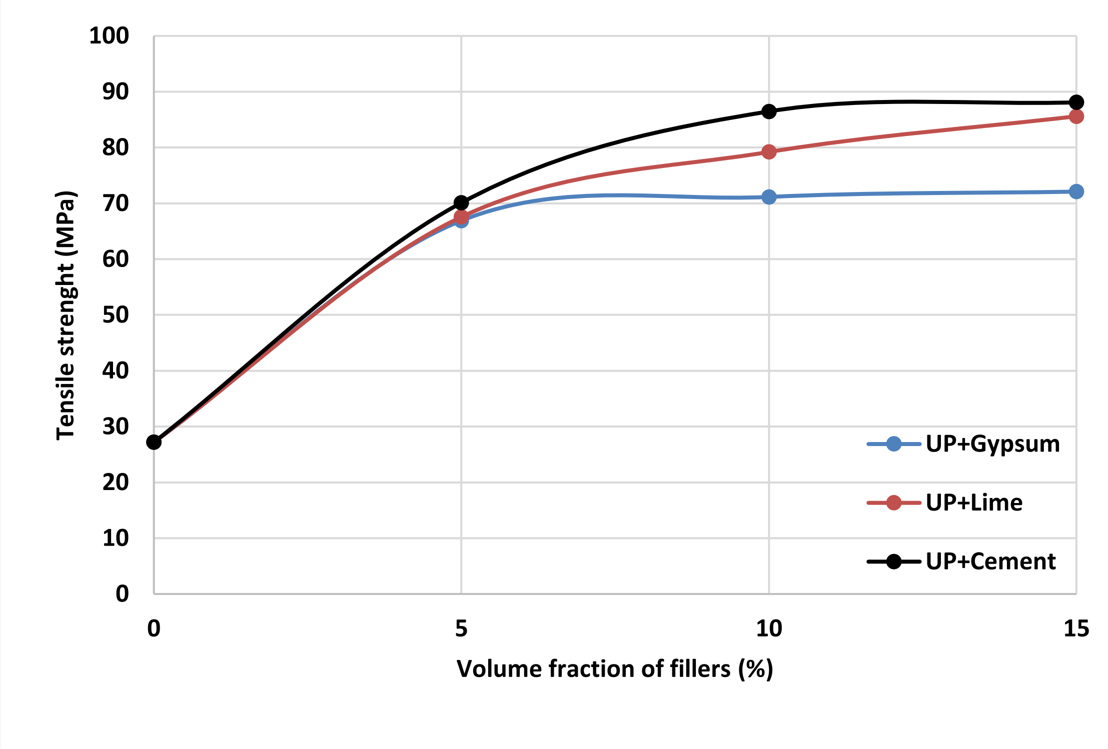

Figure 3. Tensile strength of samples 0, 1, 2, 3, 4, 5, 6, 7, 8, and 9

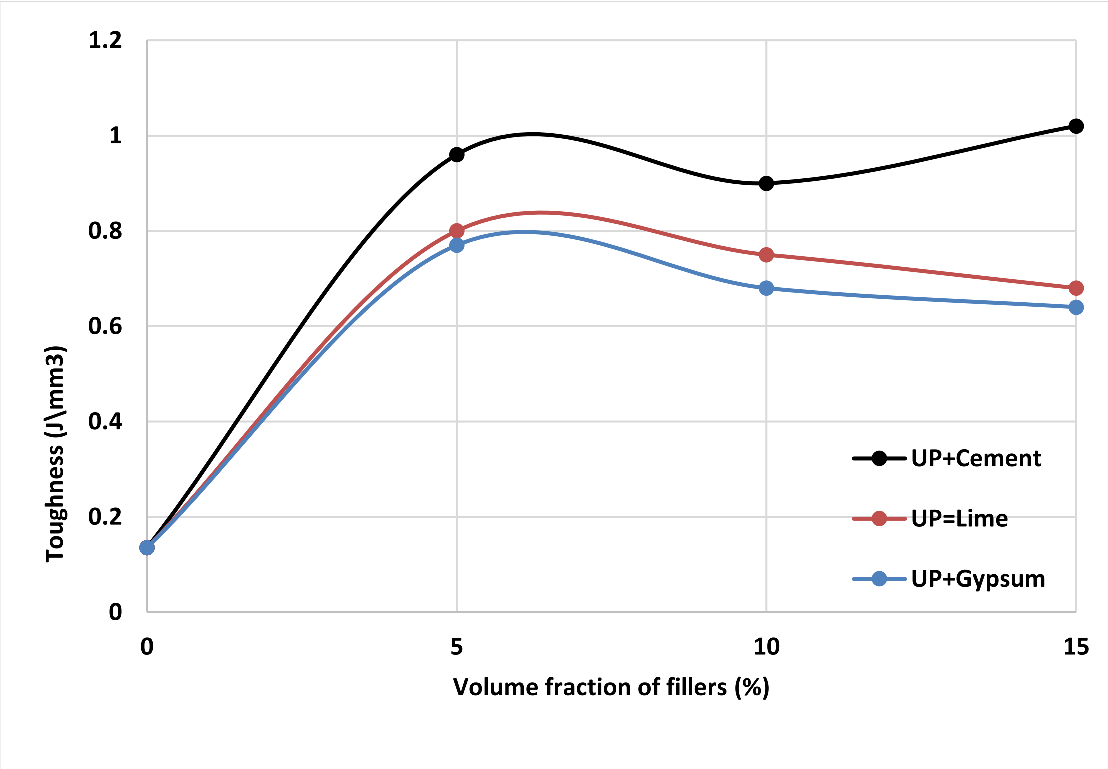

Figure 4. Toughness of samples 0, 1, 2, 3, 4, 5, 6, 7, 8, and 9

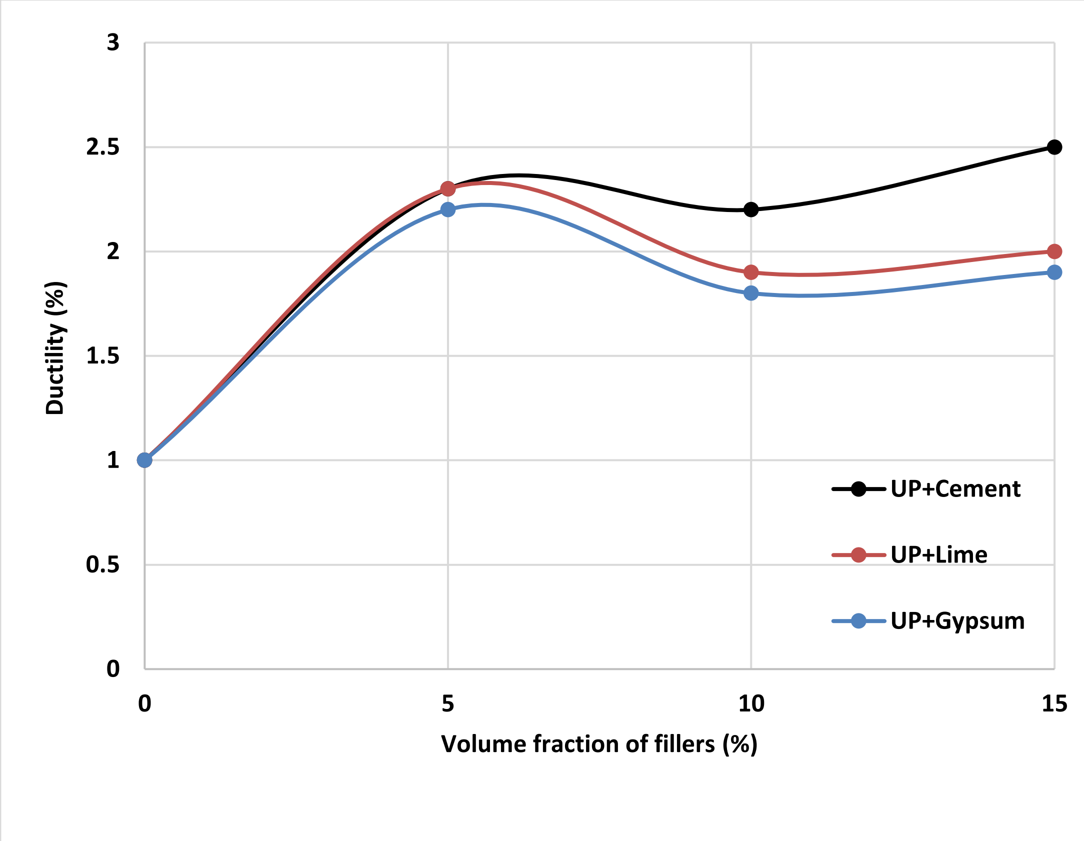

Figure 5. Ductility of samples 0, 1, 2, 3, 4, 5, 6, 7, 8, and 9

Figures 2, 3 and 4 respectively reveal increases in the modulus of elasticity, tensile strength and toughness of all samples containing cement, gypsum or limestone added to the UP. The decrease in ductility of the composites, which is shown in Figure 5, may be attributed to the distribution of the fillers throughout the matrix in a random manner, next to the heterogeneity in the length of polymer chain will create due to polymer interactions by fillers. Polymer absorption may result in entanglement interactions and polymer bridging between the fillers.

First, and in agreement with the Payne effect mentioned in E. Delebecq et al ((Delebecq, Hermeline, Flers, Ganachaud, & interfaces, 2012), which create a model composed of a filler network formed either explicitly between the fillers or through polymer domains that been used to describe it qualitatively. Moreover, because the polymer adsorbed on the filler surface is usually glassy, the modulus varies with the distance from the filler surface.

Many of the contributions, including chemical knots and physical interactions, must be considered. The latter, though weaker, plays an important role in improving the tensile properties of the composites. The filler increases the orientation and extension of the chains, resulting in an increase in stress and material modulus.

As stress is applied to the sagging chain, the latter may be adsorbed onto the filler surface. Thus, the chain bears the load by slipping along the filler surface. The fillers tend to restrain matrix phase movement in their vicinity, whilst the matrix transfers the applied load to the filler and a bear fraction of it. Thus, the filler particles improve the tensile properties of the UP.

It is important to be noted that the as-prepared polyester composite material has good values in all modulus, tensile and toughness strength as compared with other composite used in the same application.

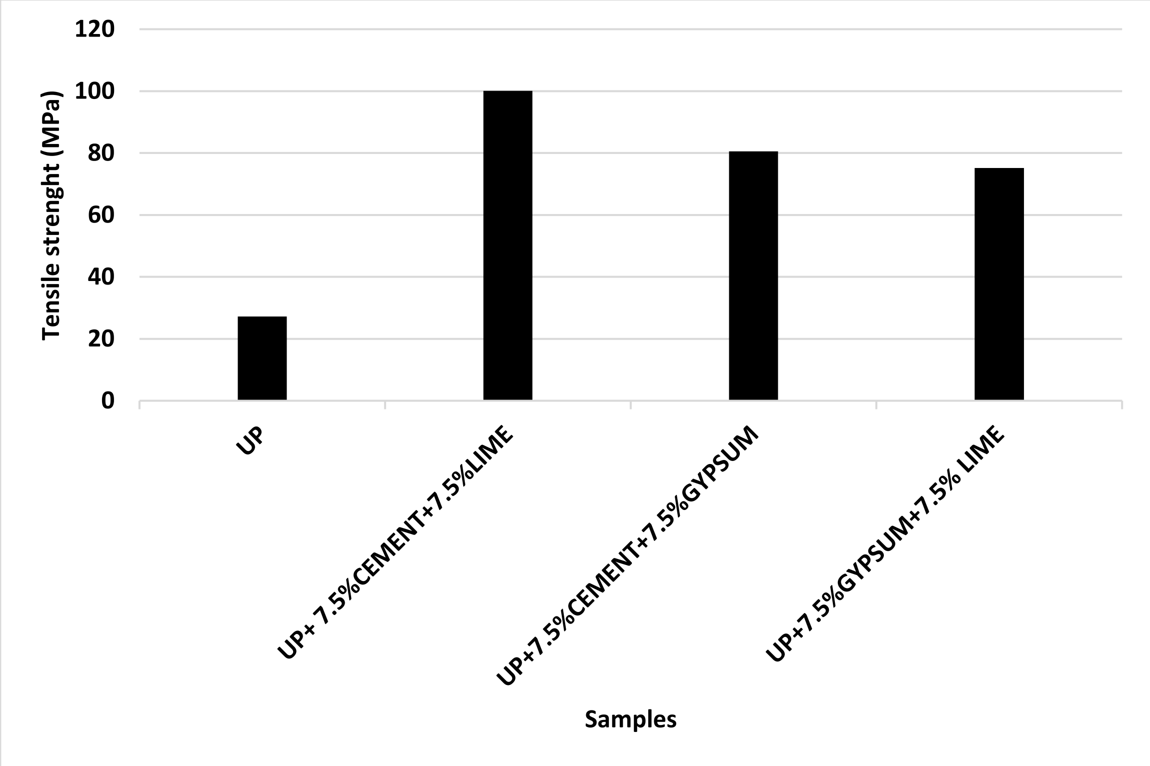

Figure 6. Tensile strength of samples 0, 10, 11 and 12

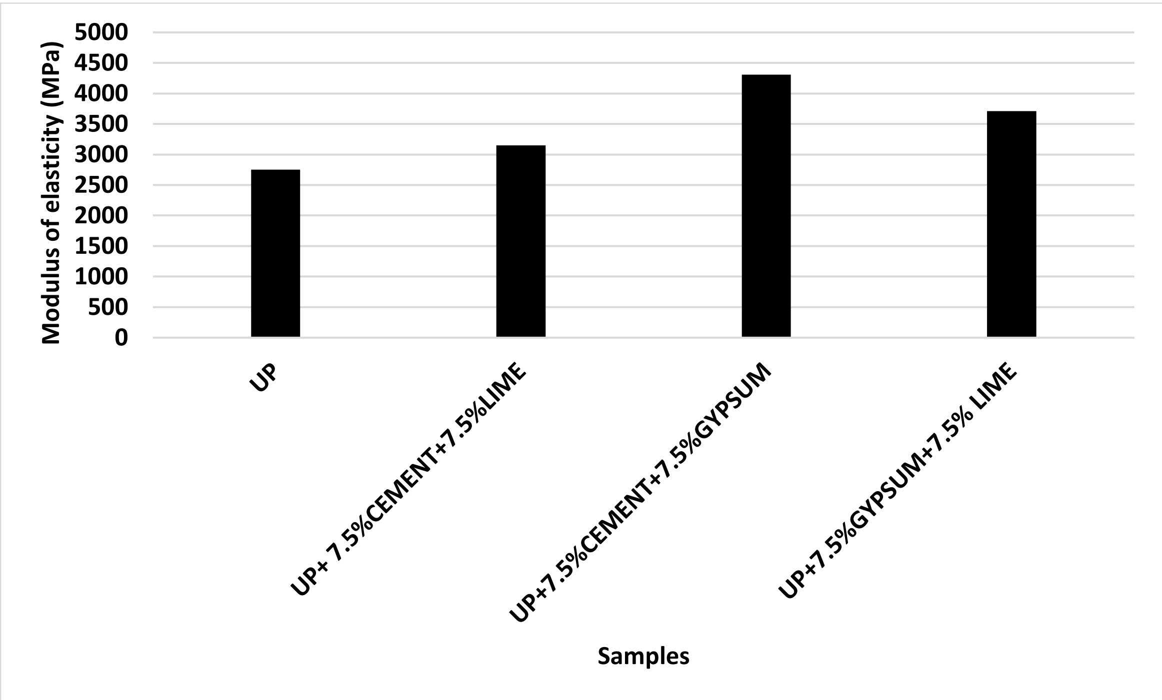

Figure 7. Modulus of elasticity of samples 0, 10, 11 and 12

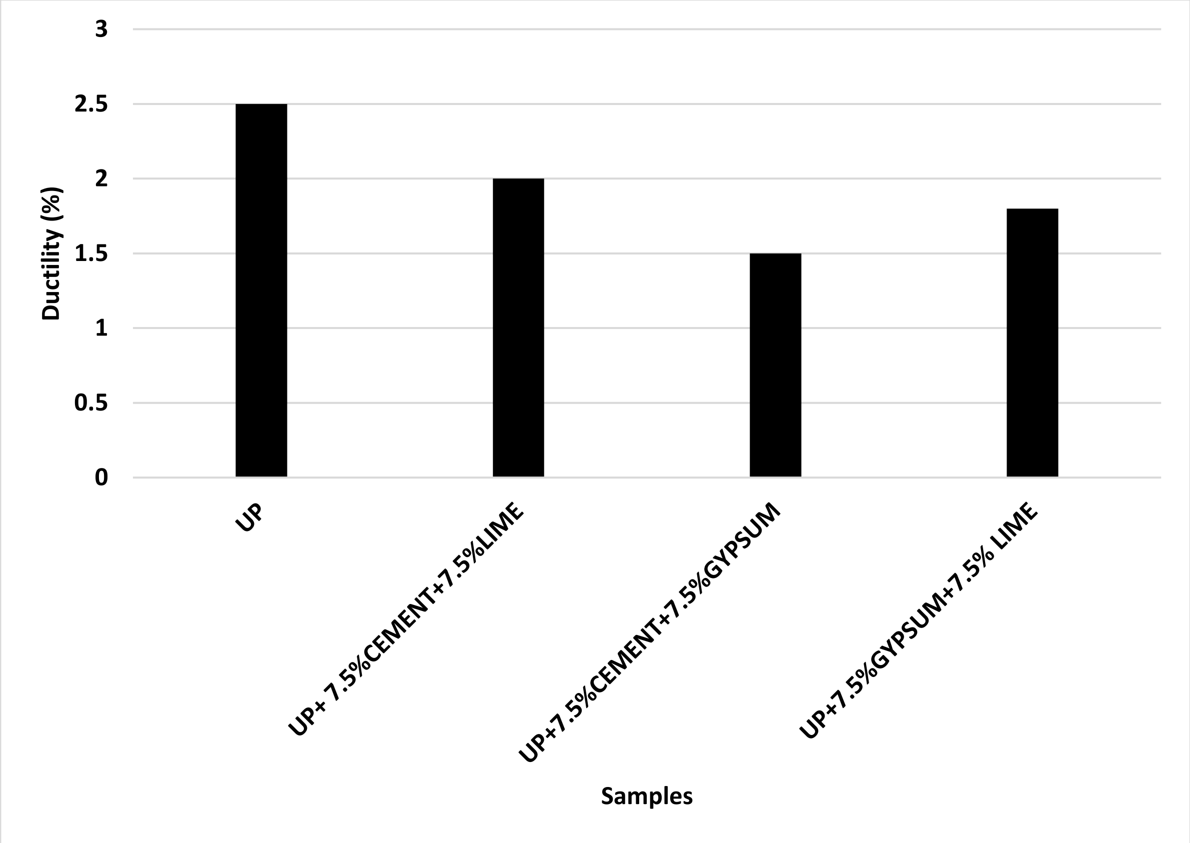

Figure 8. Ductility of samples 0, 10, 11 and 12

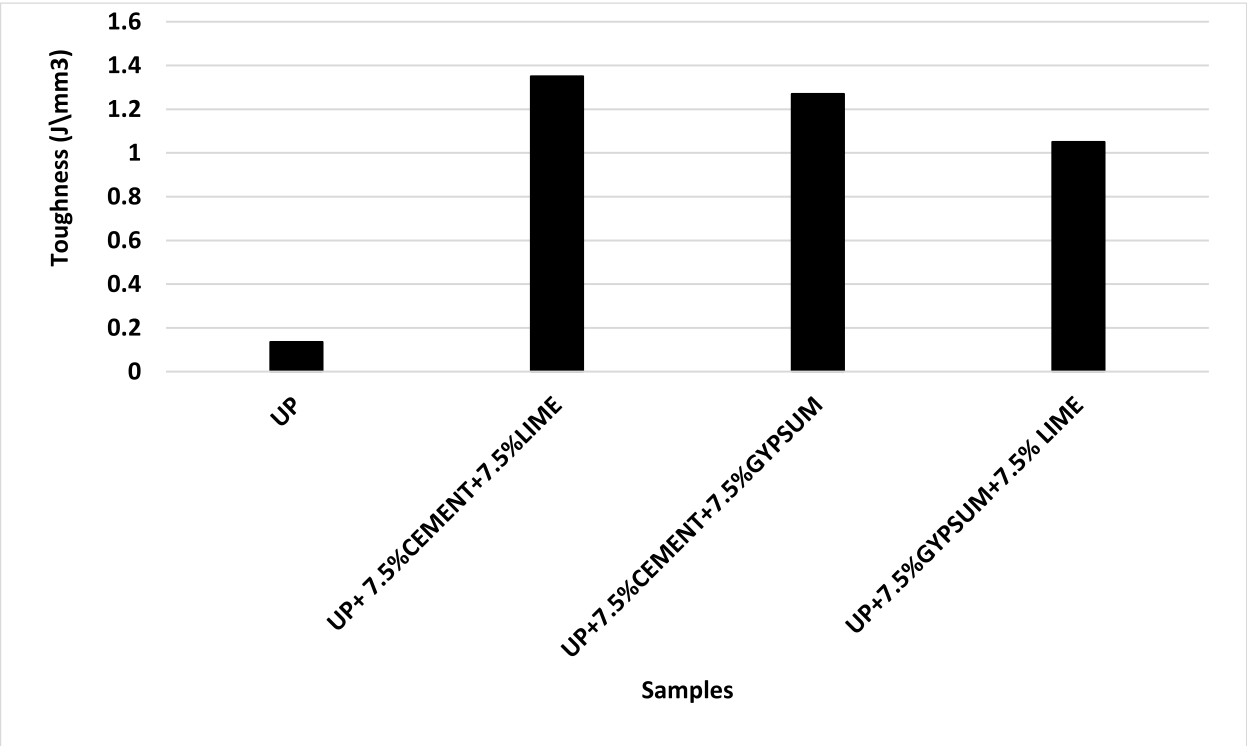

Figure 9. Toughness of samples 0, 10, 11 and 12

Figures 6, 7, 8 and 9 compare the effects of filler type on the tensile properties of the composites. Cement and limestone can remarkably improve the tensile properties of the composites whereas gypsum cannot because the former are more highly dispersed than the latter in the matrix. The fine

particles of cement and limestone provide a large surface area and, thus, a higher probability of forming physical bonds between the polymer and the filler. Indeed, improvements in modulus and ultimate properties occupy a hydrodynamic result to arise from the addition of stiff particles and a raise in the cross-linking density formed by polymer–filler bonding. By contrast, the large particles of gypsum (particle size ≤ 1.19 mm) may restrain the movement of the matrix phase, whereas the matrix removes some of the applied load to the particles and hold the fraction. Dispersion of cement and limestone, which have particle sizes of ≤0.0027 and 0.125 mm, respectively, tends to impede the slipping of polymer chains and requires high stress to bow them in narrow gap amongst particles compared with large particles of gypsum and the matrix hold the main fraction of the applied load that agrees with M. Sh. Abed (Abed, Oleiwi, & Hamza, 2016; Oleiwi, Hamza, & Abed, 2010).
Also, the attractive mechanical properties of the synthetic marble developed in this study can be explained by the reasonable interfacial adhesion between the fillers particles and the polyester matrix. This really is related to an effective load passage from the matrix to the filler particles, and it causes the high strength levels and also the greater modulus of the prepared composites.

2. Liquid Absorption Test

The absorption results of all composite samples immersed in distilled water, benzene, kerosene and gas oil at room temperature as listed in Table 4.

Table 4. Absorption of the composites according to ASTM 570

Number of samples	Volume fraction of fillers	Absorption%			
		Gasoil	Kerosene	Benzene	Distilled water
0	UP (pure)	0.51	0.75	0.95	1.1
1	5%	1.4	1.6	1.8	1.9
2	10%	1.8	2	2.3	2.4
3	15%	2	2.5	3	3.1
4	5%	1.2	1.3	1.5	1.6
5	10%	1.5	1.9	2	2.1
6	15%	1.9	2.2	2.4	2.7
7	5%	1	1.2	1.3	1.5
8	10%	1.1	1.3	1.5	1.7
9	15%	1.5	1.7	1.9	2
10	15%	3	3.8	4	4.9
11	15%	2.5	3	3.2	3.6
12	15%	1.9	2.1	2.5	2.9

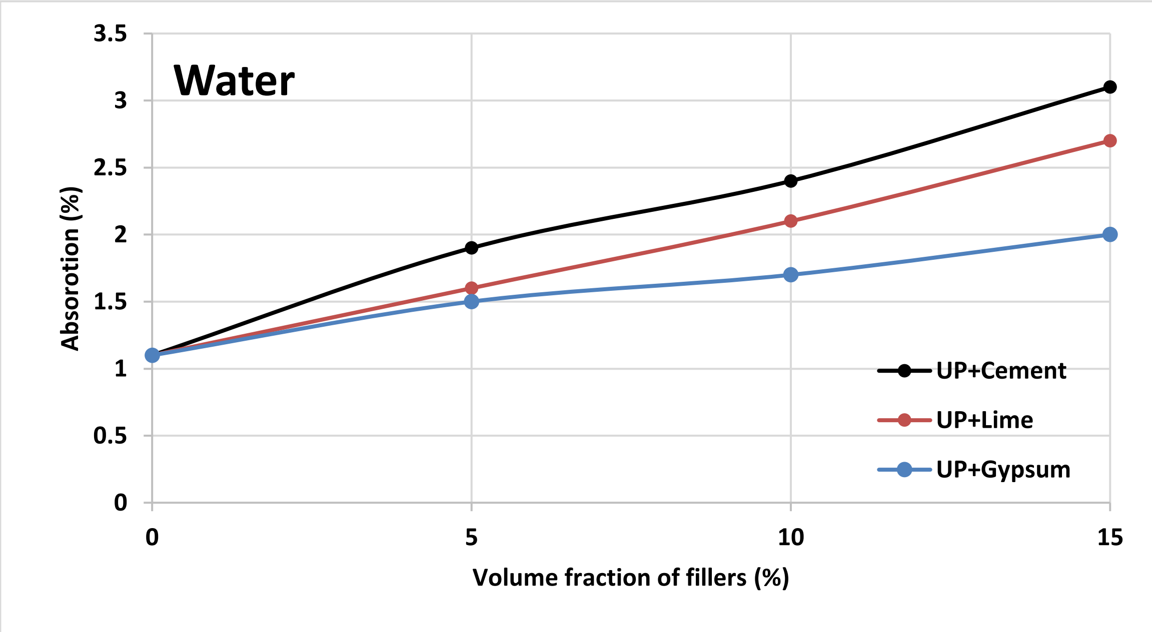

Figure 10. Water absorption (%) of samples 0, 1, 2, 3, 4, 5, 6, 7, 8 and 9

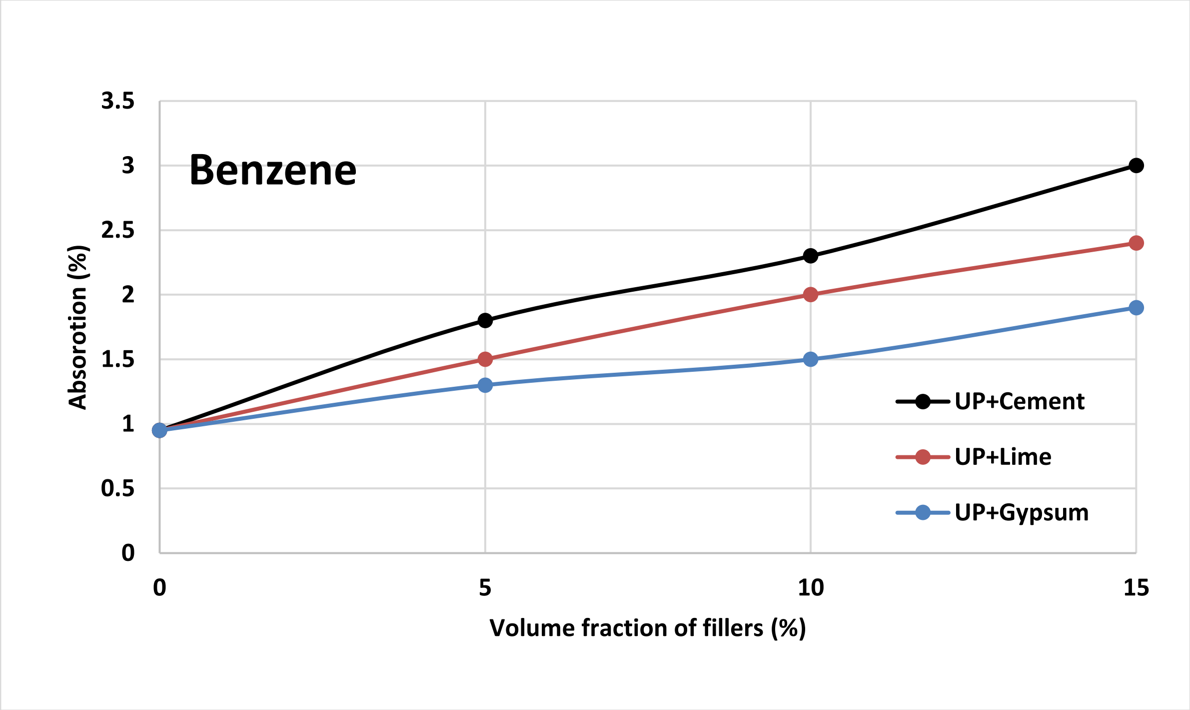

Figure 11. Benzene absorption (%) of samples 0, 1, 2, 3, 4, 5, 6, 7, 8 and 9

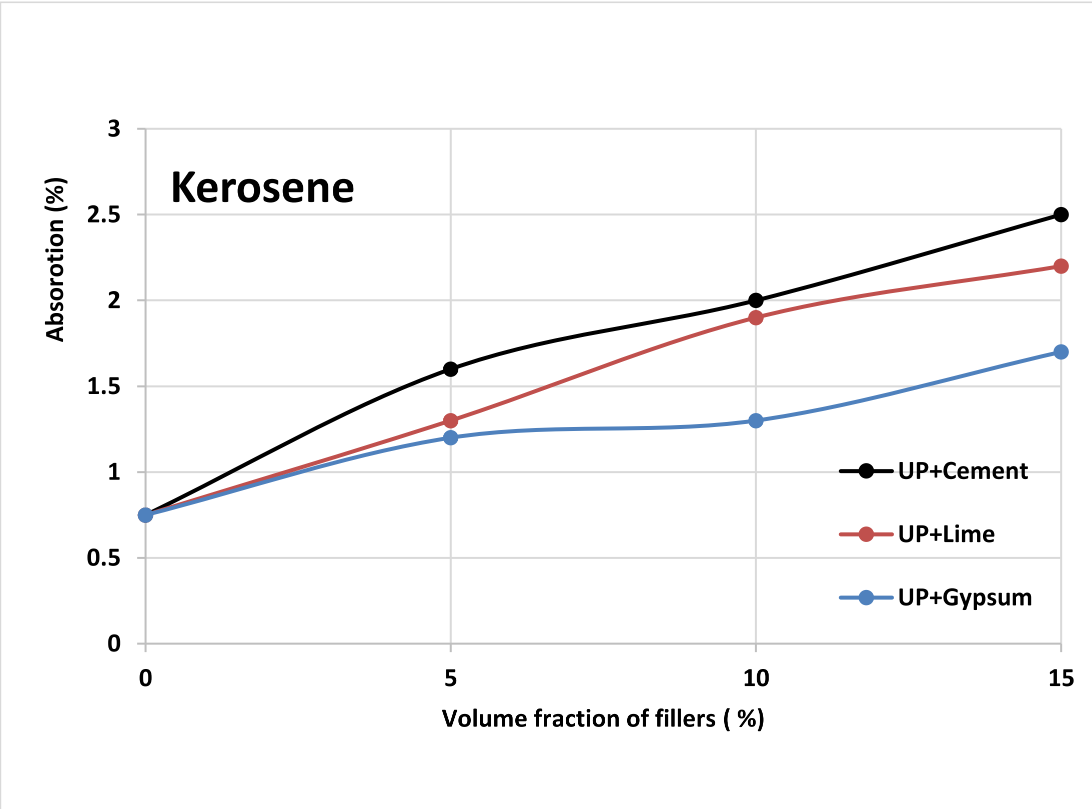

Figure 12. Kerosene absorption (%) of samples 0, 1, 2, 3, 4, 5, 6, 7, 8 and 9

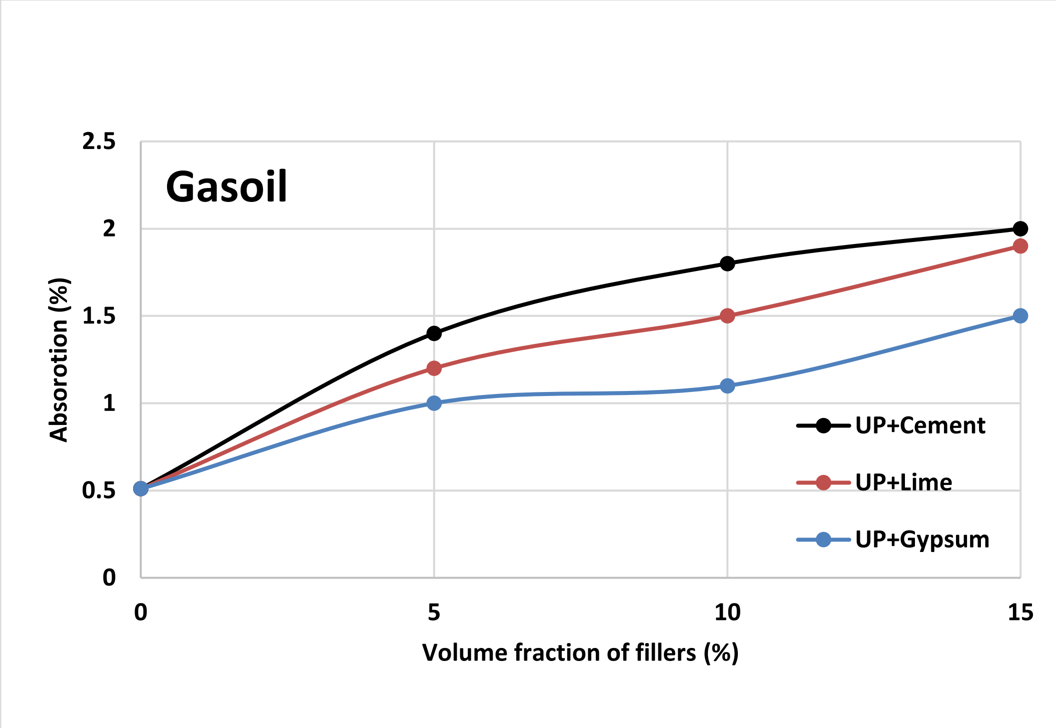

Figure 13. Gas oil absorption (%) of samples 0, 1, 2, 3, 4, 5, 6, 7, 8 and 9

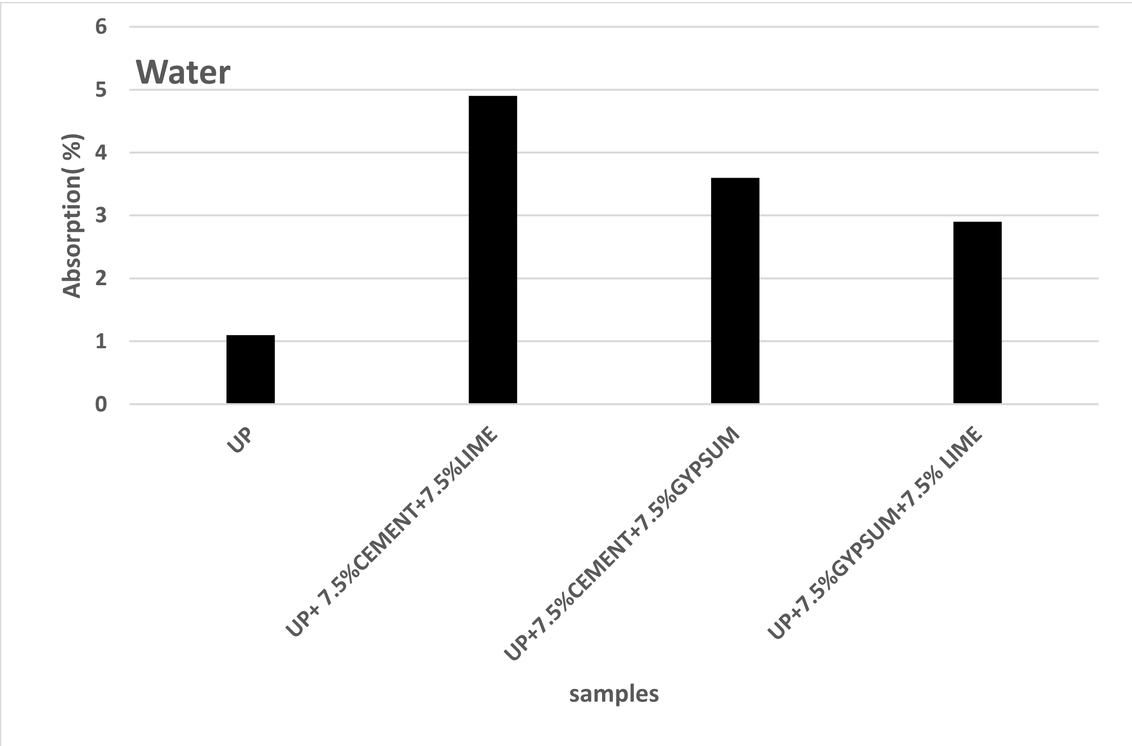

Figure 14. Water absorption (%) of samples 0, 10, 11 and 12

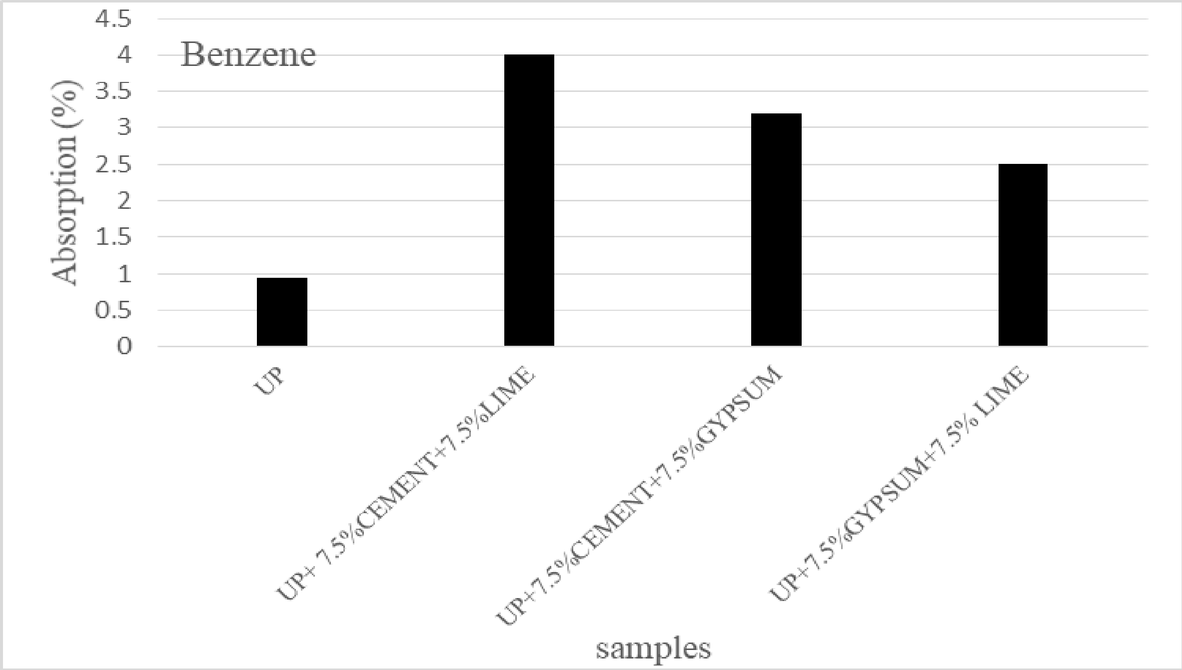

Figure 15. Benzene absorption (%) of samples 0, 10, 11 and 12

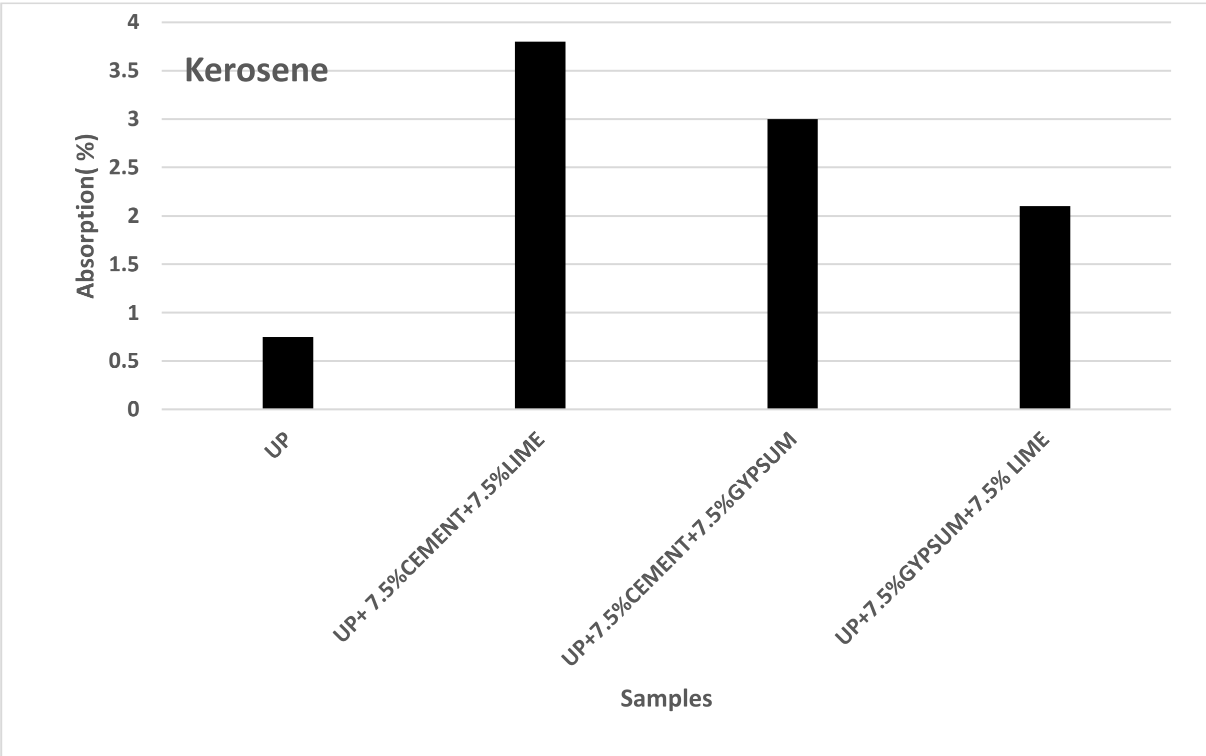

Figure 16. Kerosene absorption (%) of samples 0, 10, 11 and 12

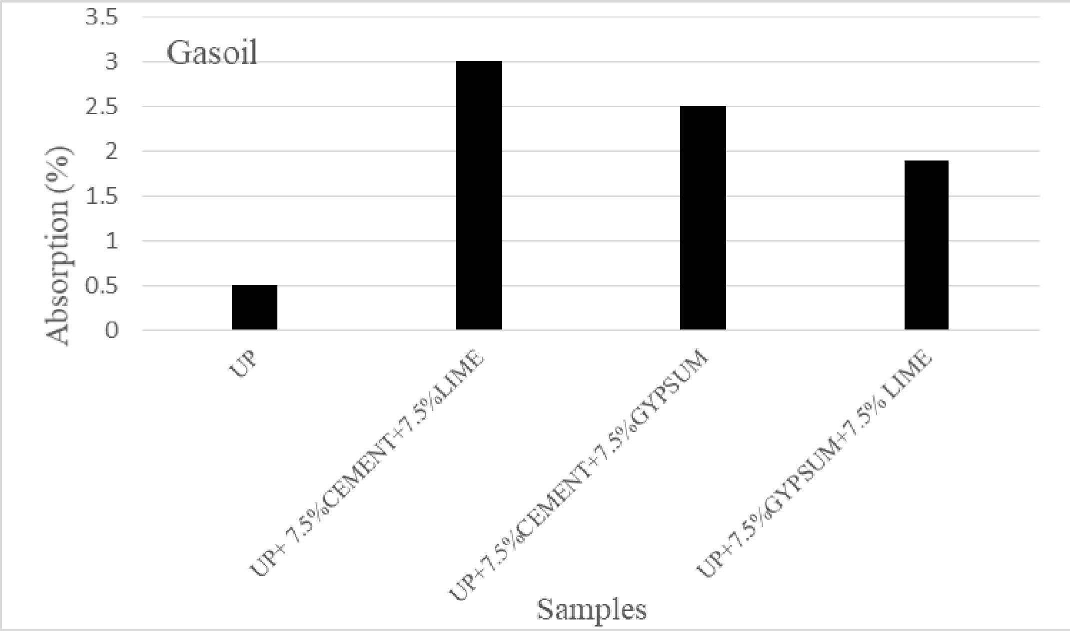

Figure 17. Gas oil absorption (%) of samples 0, 10, 11 and 12

Figures 10–17 show the effect of the volume fraction of the fillers on the liquid absorption properties of the prepared composites. The liquid absorption values of the prepared resin composites ranged from 0.51 mg/mm3 to 4.9 mg/mm^3. None of the tested specimens demonstrated maximum liquid absorption values exceeding those described in previous research (Das & Biswas, 2016; Ghazi, Oleiwi, Salih, & Mutar, 2021; Widiastuti, Pratiwi, & Cahyo, 2020). The liquid amount a polymeric composite can absorb depends on its chemical composition, particularly the hydrophilicity of the polymeric matrix and the nature of the nanofiller materials ((Ferracane, 2006).

The liquid absorption values may also vary according to the filler content. Increased absorption and diffusion were noted with increasing weight fraction of the fillers because the latter may have higher liquid penetration and a higher liquid absorption ratio than the matrix material (George & Thomas, 2001).

The porous structure of the filler materials also increased with increasing filler content in the composites. An increase in porosity could increase the free volume inside the UP resin and form tortuous paths through which solvent molecules can permeate matrix molecules, leading to an increase in liquid absorption (Mutar, Ghazi, & Mahdi, 2020).

Liquid absorption increased with increasing immersion time because of the capillary property. As the liquid solution spreads through the polymer–filler interfacial gaps produced by poor bonding between the filler particles and matrix, the composite structure is filled with the liquid, leading to increased absorption. Amongst the solvents tested, water was absorbed by the composites most extensively; by comparison, benzene, kerosene and gasoil were absorbed by the composites least extensively. This finding may be attributed to the high viscosity, which could lead to the inhibition of the capillary property of the composites. The results clearly show that liquid absorption depends on the particle size of the fillers. Composites filled with gypsum (particle size, 1.19 mm) exhibited an increase in absorption resistance compared with composites filled with cement and lime (particle sizes, 0.0027 and 0.125, respectively). In general, the rate of liquid absorption is affected by the density of the composites, the void content of the matrix and the particle size of the fillers(Mutar et al., 2020).

Also worthy to note is that in the field of composite materials, one of the most vital areas of research is the development of leak resistant forms.

Conclusions

1. Addition of reinforcement particles improves the tensile properties of the polyester composites.
2. The maximum increase in tensile strength was observed following the addition of 15% reinforcement fillers to the composites. However, the maximum modulus of elasticity was observed after the addition of 10wt% fillers.

3. High proportions of reinforcement fillers in the composites decrease the toughness and ductility of the polyester composites.
4. All polymeric composites absorb liquid when immersed in the latter. The amount of liquid absorbed by the resin composite showed variations according to the hydrophilicity of the polymers and filler content.
5. The percentage of weight gain of the prepared composites increases with increasing weight content of the fillers. Addition of 15wt% fillers results in the highest liquid absorption values.
6. These composite's innovations can have far implications, not just to expand the use of polyester resin composites in civil purposes, but also in order to try and make better use of a substantial industrial byproduct.

Acknowledgements

We are grateful to Department of Materials Engineering /University of Technology-Iraq for facilitating this research and we are very extremely grateful to Dr. hussein jabbar Hussein and (Aqeel Selman lifth) for their support to us.

References

Abed MS, Oleiwi JK, Hamza MS. *Improving the Properties of the Tire Tread by Adding* SiO_2 *and* Al_2O_3: LAP LAMBERT Academic Publishing 2016.

Annamalai M, Ramasubbu RJMT. Optimizing the formulation of e-glass fiber and cotton shell particles hybrid composites for their mechanical behavior by mixture design analysis 2018; 52(2): 207-214.

Barros MM, De Oliveira MFL, Da Conceicao Ribeiro RC, Bastos DC, De Oliveira MG. Ecological bricks from dimension stone waste and polyester resin. *Construction and Building Materials* 2020; 232.

Bodur MS, Englund K, Bakkal M. Water absorption behavior and kinetics of glass fiber/waste cotton fabric hybrid composites. *Journal of Applied Polymer Science* 2017; 134(47).

Constantinides G. Nanoscience and nanoengineering of cement-based materials. *In Nanotechnology in Eco-Efficient Construction* 2013, 9-37a.

Costa L. *Obtaining and studying a composite of polyester matrix and charge of marble residues.* Dissertação de mestrado. Universidade Federal do Rio Grande do Norte, Natal 2017.

Das G, Biswas S. *Physical, mechanical and water absorption behaviour of coir fiber reinforced epoxy composites filled with* Al_2O_3 *particulates.* Paper presented at the IOP conference series: materials science and engineering 2016.

Delebecq E, Hermeline N, Flers A, Ganachaud F. Looking over liquid silicone rubbers:(2) mechanical properties vs network topology. *ACS applied materials & interfaces* 2012; 4(7): 3353-3363.

Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. *Dental Materials* 2006; 22(3): 211-222.

George SC, Thomas S. Transport phenomena through polymeric systems. *Progress in Polymer science* 2001; 26(6): 985-1017.

Ghazi IF, Oleiwi JK, Salih SI, Mutar MA. Water Sorption and Solubility of Light-Cured Dental Composites Prepared from Two Different Types of Matrix Monomers. *In IOP Conference Series: Materials Science and Engineering* 2021; 1094(1).

Ismail MR, Ali MA, El-Milligy AA, Afifi MS. Physico-chemical studies of gamma-irradiated polyester-impregnated cement mortar composite. *Journal of radioanalytical and nuclear chemistry* 1998; 238(1): 111-117.

Machmud MN, Yudha BS, Anhar MP. Mechanical Properties Characterization of Marble and Resin Composite Materials. *In IOP Conference Series: Materials Science and Engineering* 2019, 506(1).

Mahayatra IG, Supriadi H, Savetlana S. Effect of Variation in Statutory Marble Particle Size on Mechanical Properties of Statuary Marble Particle Composites. *Scientific Journal of Mechanical Engineering* 2013; 1(4).

Mohammed RA. Effect of Al2O3 powder on some mechanical and physical properties for unsaturated polyester resin hybrid composites materials reinforced by carbon and glass fibers. *Engineering and Technology Journal* 2016; 34(12 Part A): 2371-2379.

Mutar MA, Ghazi IF, Mahdi MS. Preparation and characterization of Novel Bis-GMA Dental Nanocomposite and their application as dental material: Mechanical Properties and Water Sorption/volumetric shrinkage. *In IOP Conference Series: Materials Science and Engineering* 2020, 870(1).

Oleiwi JK, Hamza MS, Abed MS. Improving the properties of the tire tread by adding SiO2 and Al2O3 to SBR rubber. *International Journal of Applied Engineering Research* 2010; 5(9): 1637-1652.

Pervyshin GN, Yakovlev GI, Gordina AF, Keriene J, Polyanskikh IS, Fischer HB, Buryanov AF. Water-resistant gypsum compositions with man-made modifiers. *Procedia Engineering* 2017; 172: 867-874.

Qhazi IF. Study the Effect of the Particle Size on Mechanical Properties of Particulate Natural Composite Materials. *Al-Qadisiyah Journal for Engineering Sciences* 2017; 10(2): 120-132.

Ribeiro CEG, Rodriguez RJS. Influence of compaction pressure and particle content on thermal and mechanical behavior of artificial marbles with marble waste and unsaturated polyester. *Materials Research* 2015; 18: 283-290.

Souza LGMD, Silva EJD, Souza LGVMDJMR. Obtaining and Characterizing a Polyester Resin and Cement Powder Composites 2020.

Termonia Y. Tensile strength of discontinuous fibre-reinforced composites. *Journal of Materials Science* 1990; 25(11): 4644-4653.

Widiastuti I, Pratiwi YR, Cahyo DN. A Study on Water Absorption and Mechanical Properties in Epoxy-Bamboo Laminate Composite with Varying Immersion Temperatures. *Open Engineering* 2020; 10(1): 814-819.

Aziz SA, Ali RS, Abd AN. Characterization studies of nickel oxide nanostructure films prepared by electrolysis method for photo detectors applications. *NeuroQuantology* 2020; 18(2): 45-49.