Thermal and Natural Particles Addition Effects on the Mechanical and Physical Properties of Epoxy–Polyurethane Resin Blend Polymer

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***Abstract*—In this study, natural particles were added to blend polymer resin. Composite material prepared from pistachio shells was added to epoxy–polyurethane blend polymer. The weight ratio of additive for liquid polyurethane to epoxy was 12 wt.%. In addition, their mechanical and physical properties were studied depending on a range of variables, such as temperature (25 °C, 35 °C, 45 °C and 55 °C) and volume fraction (3%, 6%, 9% and 12%). Hardness, impact strength and thermal conductivity were studied. Results showed that the impact strength was increased and that the hardness and thermal conductivity values were reduced at high temperatures. Meanwhile, the impact strength and hardness increased and the thermal conductivity values decreased with increased volume fracture of pistachio shells particles.**

***Index Terms*—Pistachio Shells; Particles; Thermal; Volume Fraction; Blend Polymers; Impact; Hardness; Thermal Conductivity.**

I. Introduction

Industrial and technological development depends significantly on the progress and expansion of the field of engineering materials, especially polymeric materials. These materials are widely used in daily life because of their features and characteristics, which make them indispensable in aircraft, ship and car industries [1],[2]. Polymeric mixtures (polymer blends) are important and widely used engineering materials in many modern industrial and technological applications because they possess features that allow them to replace many other materials. They are used since ancient times because of their low cost, ease of formation for complex forms, recyclability, light weight and high resistance to corrosion. However, they lack hardness and durability, especially when exposed to external stresses. Moreover, they can withstand high temperatures [2]-[4]. With industrial development and technological progress, researchers are working on the development of polymers to improve their properties and expand their uses in complex applications. Polymers have desirable properties, especially when they are mixed together to form what is known as polymeric mixtures. However, the physical combination of two or more types of polymeric materials depends on the polymer quality and mixing method [4],[5].

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Al-Rawi et al. studied the mechanical properties of a polymer mixture of epoxy resin–polyurethane with different polyurethane concentrations (20%, 40%, 60% and 80%) and then cured it thermally at 75 °C for different durations (3, 6 and 9 h). Researchers found that high or low impact strength depends on polymeric blend concentrations and thermal curing of some mixtures [1]. Maki et al. studied the effect of reinforcement and temperature on the impact strength for a polymeric mixture of epoxy resin and 10% polyurethane reinforced with polyvinyl chloride and aluminium fibres in the form of a bidirectional mat with a volume ratio of 15%. They performed the reinforcement at different temperatures (20 °C, 40 °C and 60 °C). The authors found increased impact strength with the addition of reinforcing materials and increased temperature. However, no changes were observed in the mixture reinforced with PVC fibres as the impact resistance was decreased by increasing temperature [4]. Jawad studied the effect of compressive and electrical insulation of the strength of epoxy resin–polyurethane reinforced with glass fibre. Different concentrations of polyurethane were used (0%, 12.5%, 25%, 37.5% and 100%). The study showed that compressive strength increased and electrical insulation resistance decreased with increasing polyurethane ratios [6]. The present work aims to study the effect of temperature and natural filler volume fraction on the mechanical (impact strength and hardness) and physical properties (thermal conductivity) of a binary polymeric mixture consisting of epoxy resin with polyurethane.

II. Experimental

A. Materials

Epoxy resin (Quick mast 105, DCP Co., Jorden) was used as the base material in this research. It has a density of 1.04 g/cm^3 and a transparent liquid colour. It was converted into solid state by adding a hardener (metaphenylene diamine). It is also a transparent liquid, and the addition was 1:3 at room temperature. Flexible aliphatic polyurethane two component (PU 780; BVR Co., Turkey) was used as an additive to the base material with a density of 0.97 g/cm^3. Similar to the resin, it has a transparent colour at room temperature. It was turned into a solid state by adding a hardener material with resin-to-hardener mixing ratio of 3:1. The pistachio shells, obtained from a local market, was used as a filler material and soaked in 3% NaCl solution for 2 h. It was then washed in distilled water and dried in an electric oven for 24 h at 50 °C. Then, it was collected and crushed using an electrical mill to attain an appropriate size of less than 106 μm. The

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resulting particles were obtained by using a vibratory sieve shaker.

B. Sample preparation

A glass mould with a dimension of (20 cm × 20 cm × 1 cm) was used to form the samples in the research. Before mixing and casting, the mould was thoroughly cleaned and left to dry. Then, a thin film was used for the inside part to ensure easy sample removal by preventing the adhesion of resin material with the mould and obtain a smooth and regular surface. In terms of addition ratios, composite material specimens were synthetic with volume fractions of 3%, 6%, 9% and 12%, depending on (1), (2) and (3) [7]:

$$\varphi = \frac{1}{1+\frac{1-\psi}{\psi}\cdot\frac{\rho p}{\rho m}} \quad (1)$$

$$\psi = \frac{w_p}{w_c} \quad (2)$$

$$w_c = w_p + w_m \quad (3)$$

where ψ is the weight fracture of the pistachio shells for the composite material; w_p, w_m and w_c are the weight of the pistachio shells, matrix material and composite, respectively; ρ_m and ρ_p are the density of the pistachio shells and matrix material, respectively; and φ is the volume fraction of pistachio shells in the composite.

The epoxy resin was mixed with 12% polyurethane inside a glass container by using a mechanical mixer for 5 min to ensure full homogeneity between the base material. The pistachio shells were added to the base (matrix) material. Then, the component was remixed for 5 min to ensure homogeneity. Then, epoxy and polyurethane hardeners were added to the mixture and then mixed manually for 15 min. The final mixture was poured very slowly onto the glass mould to prevent bubble formation inside the cast and failure. Afterwards, the cast was left in the mould after being covered tightly with a glass cover for 48 h to dry and solidify. Then, the cast was extracted from the mould and cut according to the required dimensions for the tests, where the surfaces and edges of the samples were smoothened before the test to remove surface defects. The final dimensions were controlled using a polish device and a 2500-grit polishing paper.

C. Composite specimen tests

The results of the mechanical tests on the impact strength and hardness on blend epoxy/12% polyurethane (BEP) resin reinforced with pistachio shells at different temperatures (25 °C, 35 °C, 45 °C and 55 °C). Taking into consideration the volume fraction (3%–12%) of pistachio shells that was added, a special heating oven consisting of an air blower with temperature control system on inlet air was prepared for hardness and impact tests to retain the specimen temperature during examinations. Warm air was blown continuously by the system into the chamber until thermometer reading at a specific temperature.

1) Impact test samples

Unnotched impact samples were prepared with a standard dimension of 80 mm×10 mm×4 mm. These are standard samples according to international standards (ISO-179). Impact detection was performed with an HSM41–25J pendulum impact tester (P. A. Hilton Ltd.) by applying (4) to calculate impact strength.

$$a_{cU} = \frac{E_c}{h.b} \quad (4)$$

where a_{cU} is the impact strength (kJ/m^2); E_c is the energy absorbed by breaking the test specimen (J); and h and b are the thickness and width of the test specimen (m), respectively.

2) Hardness test samples

The samples used in the hardness test were in accordance with ASTM D2240. Five readings were obtained for each composite material specimen. The hardness test was conducted based on Shore D hardness by using Qualitest HPE.

3) Thermal conductivity

The thermal conductivity of the material was calculated using Lee's Disk method, in which heat is transferred from the heater to the following disc until it reaches the last disc. The specimen was placed between disks A and B, whilst the electric heater was placed between disks B and C. The temperature of the three discs, T_A, T_B and T_C, was measured using the thermometers inside them. Then, the thermal conductivity value (k) of a specimen in the form of a disk with a diameter of 40 mm and a thickness of 5 mm was extracted using (5) and (6) [8],[9]:

$$k = \frac{e[T_A + \left(d_A + \frac{d_S}{4}\right)\frac{2T_A}{r} + \frac{d_S T_B}{2r}]}{\frac{T_B - T_A}{d_S}} \quad (5)$$

where e represents the amount of thermal energy passing through the unit area of the disk per second (W/m·°C) and can be calculated using the following equation:

$$e = \frac{I \times V}{\pi r^2 (T_A + T_B) + 2\pi r[d_A T_A + d_S \frac{1}{2}(T_A + T_B) + d_B T_B + d_C T_C]} \quad (6)$$

where T_A, T_B and T_C are the disk temperatures of A, B and C (°C), respectively; d_A, d_B and d_C are disc thicknesses (mm); d_s is the specimen thickness (mm); r is the disk radius (mm); and I and V are electric currents (A) and applied voltages across the heater (V), respectively.

III. RESULTS AND DISCUSSION

A. Impact test samples

Fig. 1 shows the impact strength values of epoxy–polyurethane matrix composite at different temperatures. The figure shows that the impact strength of BEP increases with increasing volume fraction and reaches its maximum value by using 12% vol. at 55 °C. After reviewing the results, we noted that the energy needed to break the unreinforced blend matrix material is less than the reinforcing pistachio shells. The increase in the fracture energy of composite materials was due to the pistachio shells bearing a part of the impact stress. The pistachio shells acted as impediments to the progression of cracks. This impedance depended on the strength of the intermediate interfacial surface between the pistachio shells and the matrix material due to transfer of cracks through the interfacial surface (around the pistachio shells) in case that the pistachio shells is not ruptured or torn. This impedance also depended on the volume fraction ratio of the pistachio shells and the method of its distribution in the matrix material [10]. Fig. 1 shows the increase in impact strength with high temperature of the blend composite materials due to the loosening of bonds between the molecules of the material and its sliding motion, which enables them to absorb a part of the energy, thereby increasing the energy needed for the break.

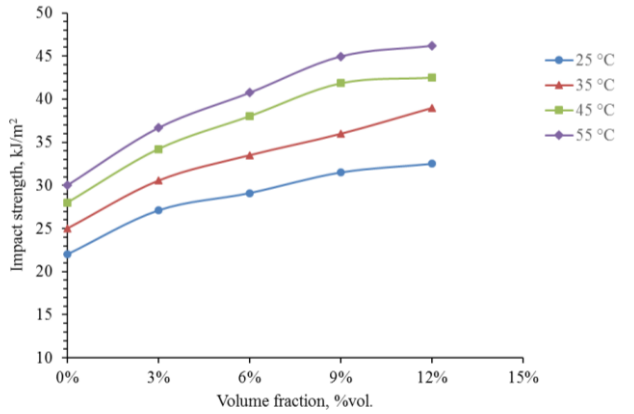

Fig. 1. Relationship between impact strength and temperatures at different pistachio shells volume fractions of the BEP materials used

B. *Hardness test*

Fig. 2 shows the values of Shore D hardness. The hardness values of the BEP were increased by the volume fraction of pistachio shells particles with a maximum value at 12% vol. The increase in hardness with increasing pistachio shells volume fraction might be related to the types of particle reinforcement and wettability of the matrix. The figure also shows the effect of temperature variations on the hardness values of the materials used. The hardness values of the models used were reduced by high temperature. The hardness values of BEP were reduced by high temperature and reached its lowest value at 3% vol and 55 °C. This result was due to the transformation of BEP polymer from fragile form to soft form because of the softness or loosening of bonds and the movement of molecules bound to it, thus weakening its resistance to scratching and sorting [11].

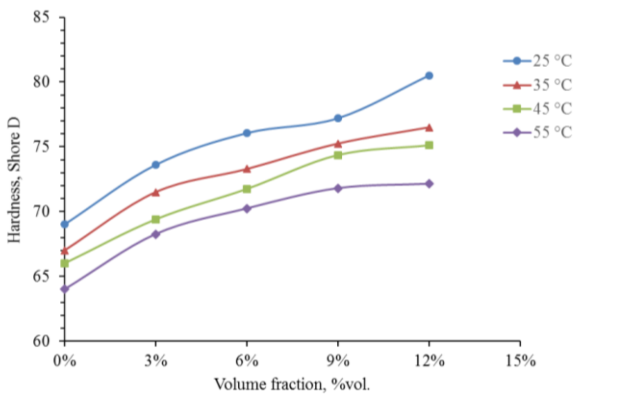

Fig. 2 Relationship between hardness and pistachio shells volume fraction at different temperatures

C. *Thermal conductivity*

Fig. 3 shows the relationship between thermal conductivity and volume fraction. The thermal conductivity of the BEP–pistachio shells composite increased with increasing temperature and decreased with increasing volume fraction. This finding shows that the specimens had higher heat resistance features than the base matrix materials. The decrease in thermal conductivity with pistachio shells particles addition might be related to interfacial bonding between pistachio shells filler particles and BEP matrix. The presence of interfaces plays an important role in the thermal connectivity of polymer materials. Generally, heat is transferred as flexible waves within the structure in polymeric and ceramic materials. Therefore, the presence of interfaces leads to the obstruction of the movement and passage of these waves, and the transmission of thermal energy as waveform remains a difficult and complicated process due to a discontinuity and alteration of the structure. This means that the wave loses part of its energy at the interfaces between the polymeric matrix material (BEP) and reinforcement materials (pistachio shells particles). In addition, another part of the energy is lost during wave transmission between the pistachio shells particles [12].

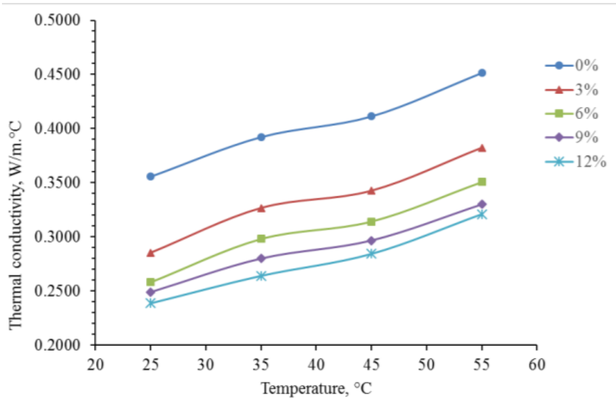

Fig. 3 Relationship between thermal conductivity and temperatures at different pistachio shells particles volume fractions

IV. CONCLUSION

The mechanical and physical properties (hardness, impact strength and thermal conductivity) of the blend polymer (epoxy resin/12 wt.% polyurethane) are improved by the addition of pistachio shells particles. Moreover, increasing the volume fraction of pistachio shells particles increases the hardness and impact strength values, and their maximum values are reached at 12% vol. Increasing the volume fraction of pistachio shells particles also leads to reduced thermal conductivity, and their minimum values are reached at 12% vol. Meanwhile, increasing temperature leads to the increase in thermal conductivity and impact strength values of the blend epoxy–polyurethane, and their maximum values are reached using 12% vol. at 55 °C. By contrast, rising temperature leads to a decrease in the Shore D hardness of the blend composite, and its minimum value is reached at 3% at 55 °C.

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