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INVESTIGATION THE MECHANICAL PROPERTIES OF EPOXY POLYMER BY ADDING NATURAL MATERIALS

A. J. AL-OBAIDI[1,*], S. J. AHMED[2], A. T. ABBAS[1]

[1]Engineering College, University of Wasit, Kut, Wassit, Iraq
[2]Materials Engineering Department, University of Technology, Baghdad, Iraq
*Corresponding Author: aalobaidi@uowasit.edu.iq

Abstract

During the last few years, natural materials have been widely used in engineering applications due to their characteristic such as low cost, less dense, environment-friendly and high strength. Due to the suffering polymer composites from several shortcomings such as mechanical properties, the natural materials can be used as fillers in polymer composites to improve their properties. The present research investigates experimentally the effect of using natural pistachio shell particles on the mechanical properties of epoxy matrix composites. Three contents of the pistachio shell particles were used: 5, 10 and 15 weight %, whereas the size of particles was $120<d<180$ μm, $63<d<120$ μm, and $d<63$ μm. The produced samples at different particle contents and sizes were tested through different mechanical tests, such as tensile, flexural, Izod impact and hardness tests and the results compared with the pure epoxy polymer. The findings showed that the impact strength, tensile strength and flexural strength of epoxy composites improved around 75%, 56%, and 87.7% respectively at the content of pistachio shell 5 weight % and particle size $d<63$ μm. While the hardness of epoxy composite improved by about 28% when the content of pistachio shell in the produced samples was 15 weight% and particle size $63 <d <120$ μm.

Keywords: Composite polymer, Epoxy, Mechanical properties, Natural materials Pistachio shell, Reinforcement materials.

1. Introduction

During the last few years, the use of natural materials has been increased due to it characterize, such as low cost, less dense, environmental-friendly and high strength. There are a lot of examples of natural materials that use as a substitute for synthetic materials, such as peanut and pistachio shells, olive stone and sugarcane. These examples of natural materials have been generally applied in polymer matrices composites as toughened fillers to enhance the properties of composites besides lowering production cost [1, 2]. Gharbi et al. [3] investigated the mechanical properties of polyester matrix composites when olive nuts flour had used as a filler. The produced samples were prepared with the olive nuts flour contents of a range between 10 and 60 weight percent. The findings observed that the properties of composite such as modulus, flexural strength were significantly enhanced. Another natural material was used as reinforcement to polyester matrix composites for the production of low-cost composites and it was the sugarcane bagasse waste. Monterio et al. [4] found that the composites of sugarcane bagasse waste/ polyester resin have mechanical properties close to the ones normally associated with wooden agglomerates.

One example of a very tough natural material is pistachio shell (*PS*), where the chemical composition of pistachio shell is constituted of (42%) cellulose, (13.5%) lignin, (3.11%) cellulose lignin, (1.26%) ash, and (0.18%) extractable. Pistachio is one of the famous nuts (or fruit) of the world [5]. The particles of the pistachio shell have good strength and modulus properties [6]. Hence, pistachio shell (*PS*) can be used as a filler in polymer matrix composites and this usage is significant from an economical and environmental view. Besides the natural materials, epoxy resins have been extensively used in various engineering and structural applications because of their good chemical and mechanical properties, high bonding strength and solvent resistance. The matrix composite of polymer resin and pistachio shells particles is suitable for various applications of engineering fields [7].

Mohamad et al. [7] studied the influence of the microscale content of the pistachio shells (*PS*) on the mechanical properties of polyester matrix composites. The author found that the mechanical properties such as tensile and flexural strength of *PS*/polyester composites were enhanced at *PS* particle content of 10 weight %. Whereas the thermal and burning properties of *PS* were studied by Metin et al. [8], the author used fly ash as a flame retarder on the polymer composite particleboard- filled shell particles. The findings showed that the particles of *PS* improved the flexural and fire-retardant properties and therefore it can be securely and economically used in construction applications. The properties of rubber matrix filled with the particles of *PS* was studied by Bağdagül [9], The results showed the large particles of pistachio producing weak points at fracture test, additionally, an increase in the elongation occurred. The properties of pistachio shells (*PS*) were studied by Piness [10]. The results showed that the structure of pistachio shells contains a combination of amorphous and crystalline polymers and it’s a very fibrous structure. Hence, these quantifying the physical structure will help the mechanical testing by providing the foundation of explanation. Nayak et al. [11] investigated the possibility of using the pistachio shell flakes as a filler in polyester composites. The hand lay-up technique was adopted to produce the composite samples with pistachio shell flakes between 1 to 3 % by weight. The author found that the addition of pistachio shell flakes led to reduce the tensile strength, whereas the flexural and impact properties improved. Ghazanfari et al. [12] produced biodegradable plates that fabricated from high-density polyethylene, date pits, and pistachio shells. The findings showed that the composite plates made from pistachio shells had a greater stiffness than the composites with date pits.

In the previous studies, the mechanical properties of composites filled with natural materials have studied through many researchers. However, the tensile, flexural, hardness and impact properties of composite materials made from pistachio shell (*PS*) and epoxy polymer still uninvestigated. Moreover, the effect of particle size and the content percentage of filler on the mechanical properties of PS/ epoxy composites remain unstudied. Therefore, this research will produce composite natural materials from PS/epoxy polymer and evaluate the products by various mechanical tests, such as tensile, flexural, hardness and impact tests. The mechanical properties of produced samples will evaluate at various particle sizes of *PS* (120<*d*<180 μm, 63<*d*<120 μm, and *d*<63 μm), and different weight content (5%, 10% and 15 %).

2. Materials and Procedures

The technique was used in the current study is hand lay-up. The hand lay-up technique considers the simplest and least expensive method for moulding where the mixture of composite materials (particles and epoxy) is placed inside the mould by applying a brush or roller. This process has many benefits such as lower cost, typical for lower volumes and flexibility in design [13-15].

The production process of composite specimens includes many steps. At the first, immersion of the pistachio shells in water is necessary for two weeks to remove salt and any other suspended materials. Secondly, grinding pistachio shells into powder form is the next step using a jaw crusher mill (type: JF SD-100 PULVERIZED). Then, the powder that collected was sieved to obtained different particle size (120<*d*<180, 63<*d*<120, and *d*<63 μm), as presented in Fig. 1.

(a) **(b)**

Fig. 1. (a) Pistachio shells and powders of *PS*. (b) Sieving device.

The epoxy resin used for the present paper is a trademark (Euxit 50KI), while its hardener is Euxit 50KII. The epoxy resin and its hardener were supplied by the Egyptian Swiss Chemical Industry Co, as shown in Fig. 2. The epoxy resin and its hardener were added at an approximate ratio of (3:1) according to the supplier standard. The particles of pistachio shells were added to the resin, and then mixed at a constant speed (1000 rpm) by stirring for 20 min to ensure homogenous distribution of particles and avoid the air bubbles. Finally, and before the mechanical testing, the mixture was introduced into a mold and then samples were cured for one week at room temperature [7, 16]. The samples for tensile, flexural, impact and hardness tests have the same thickness and it was 5 mm. Table 1 shows the mechanical properties of epoxy resins used in the present work.

Table 1. Epoxy resin's properties.

Properties	Value
Glass transition temperature (T_g)	120 – 130 °C
Density	1.05 g/cm^3
Tensile strength	25 N/mm^2
Tensile modulus	1005 N/mm^2
Elongation at break	<6 %
Flexural strength	53 N/mm^2
Compressive strength	70 N/mm^2

(a) **(b)**

Fig. 2. (a) Epoxy resin. (b) Hardener.

ASTM D638 was adopted as a standard for preparing and testing the samples for the tensile test [17]. The universal testing machine (Type LARYEE) was used for a tensile test at across head speed of 5 mm/min and a gauge length of 50 mm, tensile samples as shown in Fig. 3.

ASTM D790 was adopted as a standard for the flexural test (3-point bending) using a universal test machine [18]. The testing machine gives us the curve of the relationship between the force and deflection at the midpoint of the composite specimens, flexural samples as illustrated in Fig. 4. The flexural and modulus strength are measured by using the following equations [18]:

$$\text{Flexural strength} = \frac{3PL}{2bd^2} \quad (1)$$

$$\text{Modulus strength} = \frac{L^3 m}{4bd^3} \quad (2)$$

where, *P*, Applied force (N), *L*, Effective length of the beam (mm), *b*, Width of the beam (mm), *d*, Thickness of the beam (mm), *m*, Slope of the initial straight line of the stress-strain curve.

The impact strength was obtained from the impact test by Unnotched Izod method which involved breaking the composite samples and these samples were prepared based on (ISO-180 standard) [19]. This test carried out at (impact device) with the energy of pendulum reach to (5.5 J), impact specimens as shown in Fig. 5.

Hardness test for composite specimens was performed by using (Shore-D device); the samples were prepared based on ASTM D2240 standard, hardness specimens as illustrated in Fig. 6 [20].

There are three different sizes of particles and each particle size has three different content of powder by weight and therefore the total number of experimental runs is nine, as shown in Table 2. Each mechanical test was repeated three times for calculating the average value and to verify the results. Hence, the total samples which are produced for each test are 27 samples.

Table 2. The total number of experimental runs.

Particle Size	Content of Particle
(d<63 µm)	5wt%PS
	10wt%PS
	15wt%PS
(63<d<120 µm)	5wt%PS
	10wt%PS
	15wt%PS
(120<d<180 µm)	5wt%PS
	10wt%PS
	15wt%PS

Fig. 3. Tensile specimens before the test.

Fig. 4. Flexural specimens before the test.

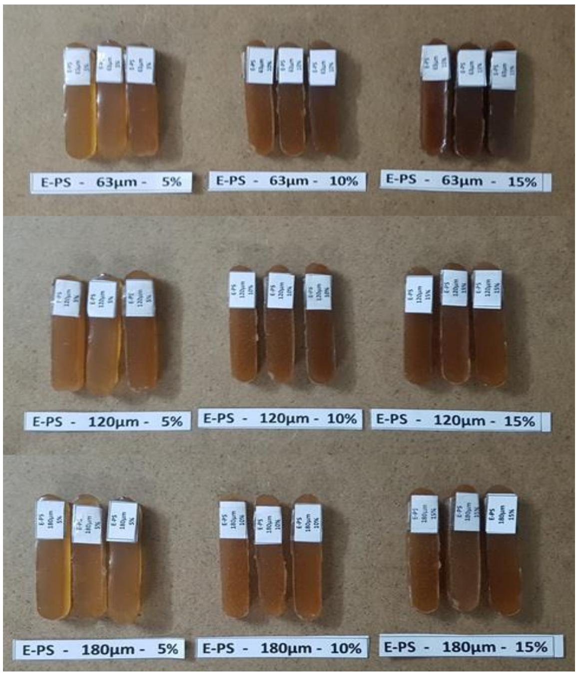

Fig. 5. Impact samples before the test.

Fig. 6. Hardness samples before the test.

3. Results and Discussion

3.1. Tensile strength properties

Figure 7 shows the stress-strain curves of PS-Epoxy composite samples. It is clearly seen that the properties of the epoxy matrix are significantly influenced by adding the particles of *PS.* The elastic limit of PS/epoxy composite is increased by around 100% compared with the pure epoxy when the content of pistachio powder was 5% by weight. However, the excessive increase of the powder content led to decrease in the elastic limit of composite materials. Moreover, the elastic limit increases when the particle size of pistachio shell powder decreased for all weight fractions. This is because the large particle size produced stress concentration points and eventually weak regions. This behaviour is similar to the previous study [8, 21] which used the pistachio shell with polyester.

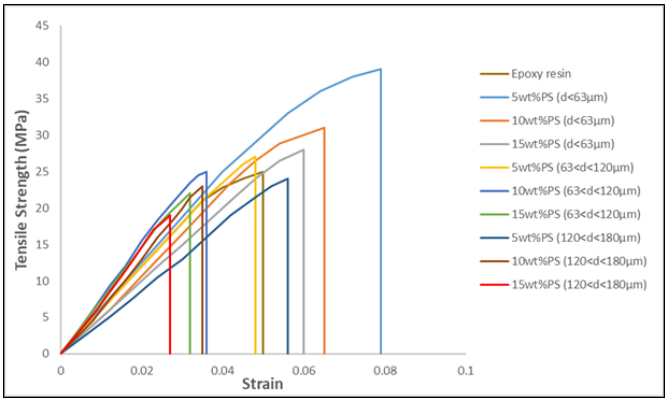

Fig. 7. Stress-strain curve of the PS-epoxy samples.

In Fig. 8, the tensile device and the samples after the test are shown. It can be clearly seen that all samples failed without necking. It means that the samples failed in a brittle type during the tensile test. Additionally, no effective permanent decreasing in the cross-sectional area was noticed. The same fracture type happened in the previous research [7] during producing of composite samples between natural material and polyester polymer.

Fig. 8. Tensile device and the failed samples after tensile test of PS/epoxy composite.

Table 3 shows the properties of the *PS*/epoxy composite during tensile, flexural, impact, and hardness tests for various filler contents and particle size.

Table 3. PS/epoxy composites' properties.

Type of composite specimens	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Absorbed energy (J)	Impact strength (KJ/m^2)	Hardness (Shore D)
Epoxy resin	25±0.93	1±0.16	53±2.9	2±0.02	0.4±0.07	8±0.6	60±2
5wt%PS (d<63 μm)	39±1.07	3±0.14	99.5±3	4.1±0.03	0.7±0.06	14±0.7	66±2
10wt%PS (d<63 μm)	31±0.99	2.7±0.2	71±2.6	5.7±0.02	0.65±.05	13±0.4	68±3
15wt%PS (d<63 μm)	28±0.65	2±0.13	69±1.8	6.6±0.05	0.5±0.08	10±0.6	75±3
5wt%PS (63<d<120 μm)	27±0.84	1.5±0.1	76.5±1	5.3±0.06	0.6±0.06	12±0.5	71±2
10wt%PS (63<d<120 μm)	25±0.55	1±0.16	65±2	7±0.02	0.5±0.03	10±0.2	74±4
15wt%PS (63<d<120 μm)	22±0.60	1±0.13	53±1.4	7.2±0.03	0.45±.02	9±0.5	76.8±5
5wt%PS (120<d<180 μm)	24±0.64	1±0.11	60±2	5.7±0.02	0.35±.02	7±0.3	68±2
10wt%PS (120<d<180 μm)	23±0.70	1±0.12	54±2.4	6.5±0.03	0.25±.01	5±0.1	71±1
15wt%PS (120<d<180 μm)	19±0.48	0.8±0.1	46±1.9	7.5±0.02	0.2±.01	4±0.09	71.7±3

Based on Fig. 9 and Table 3, the tensile strength of the epoxy matrix is enhanced around 56% through adding the particles of pistachio shell to reach its maximum value at the content of *PS* particle was 5 wt% (*d*<63 μm). Then, the strength of the PS/epoxy composite decreased when the content of the *PS* particle and size was excessive increased. While, the maximum percentage of increasing tensile modulus was 200% compared with the pure epoxy, which occurred at *PS* particle content of

5 wt % (d<63 µm). Whereas, the percentage of increasing the tensile strength was around 20% in the previous study [7].

The decrease in tensile strength during the excessive increase of the pistachio shell content particles might be occurred due to regions of ill-dispersed particles (agglomerated). This can be led to weak the proper bonding of ill-dispersed particles in the epoxy matrix and thus the stress concentrations can occur [22, 23]. Additionally, the stress concentration might occur when the individual particles of PS have an irregular shape and these stresses took place at the corners of the particle [24]. Furthermore, the increase of particle size results increasing the stress concentration at the interface between the particle and polymer matrix [25].

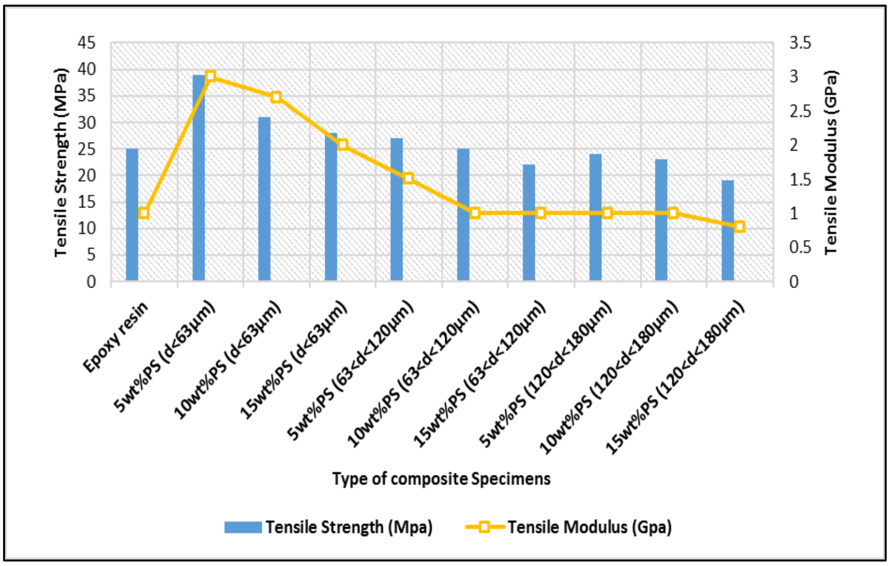

Fig. 9. Relationship between the type of composite samples versus tensile strength and tensile modulus of the PS/epoxy composites.

3.2. Flexural strength properties

The three-point bending test was adopted to obtain the flexural properties of PS/epoxy composite samples. The stress-strain curves of the composite samples are shown in Fig. 10, and it can be clearly seen that the behaviour of the curves was a practically linear up to failure stress (flexural strength) where an abrupt and that means the brittle failure occurred. The flexural device and the samples after the test are shown in Fig. 11.

Figure 12 shows the relationship between flexural strength and modulus with the type of composite samples. The flexural strength improved by adding the particles of *PS* as filler to reach its peak value and then the flexural strength decreased with increasing the *PS* content. This behaviour was also observed in the previous studies [3, 7] for pistachio shell/polyester and olive nut flour/ polyester respectively.

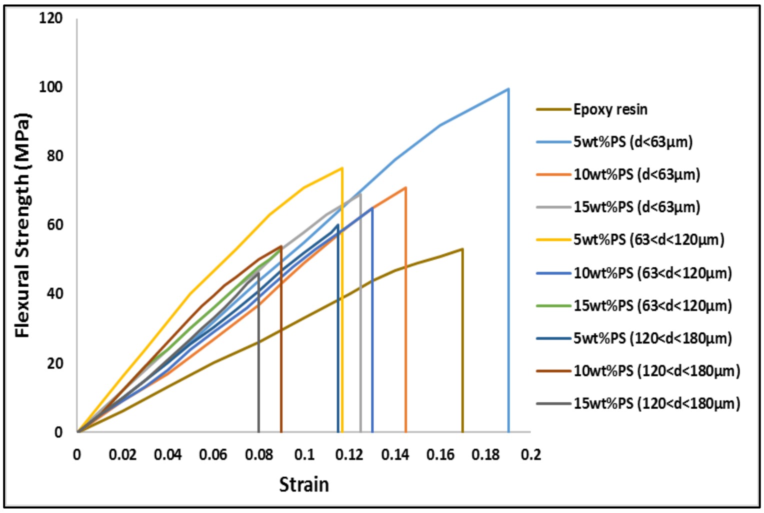

Fig. 10. Relationship between flexural strength and strain of the PS/epoxy composites.

Fig. 11. Flexural device and the failed samples.

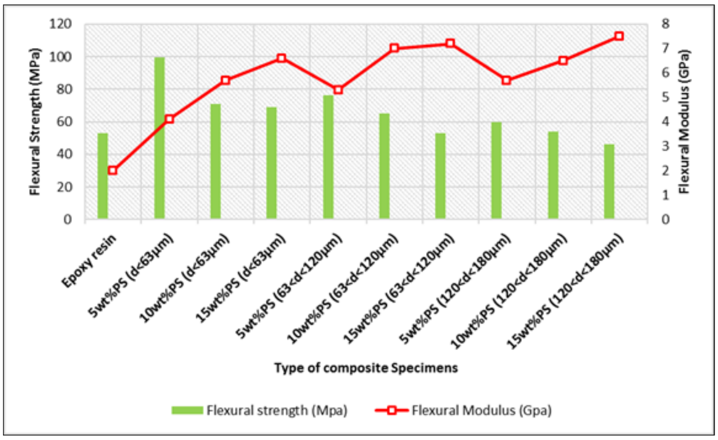

Fig. 12. Type of composite specimens versus flexural strength and flexural modulus.

The maximum flexural strength occurred at the content of 5 weight % *PS* (*d* <63 µm) corresponding to an increase of about 87.7%, but it decreased through the excessive increasing of *PS* content. While the increase of flexural strength was less than 10% in the previous study [7]. This increase in flexural strength occurred due to an increase in the surface area of filler particles because of a reduction in the particle size. Therefore, high surface energy results at the filler-matrix interface. Additionally, the content increase of *PS* particles leads to agglomeration and thus increases the stress concentration [26].

However, the agglomeration of *PS* particle does not influence the flexural modulus because it measured at stress levels under that causing crack propagation at agglomeration zones which leads to a decrease in the composite stiffness. Due to the incorporation of particles in the epoxy matrix, the flexural modulus was significantly improved because the stiffness of the rigid particles is much higher than the stiffness of the polymer matrix, and this behaviour was also observed by the previous research [7]. The flexural modulus was reached to its maximum value at the content of *PS* particle 15 *wt* % *PS* (120<*d*<180) corresponding to an increase of 200%. Whereas, the flexural modulus of the natural material/polyester composite increased by around 63% in the previous study [7].

3.3. Impact strength properties

The impact energy absorbed by the samples was measured using the Izod impact test. Figure 13 shows the impact of strength and absorbed energy of composite specimens. The addition of *PS* particles leads to an increase in the impact strength and absorbed energy to reach the peak value at content 5 *wt* % *PS* (*d*<63 µm). The percentage of increasing the impact strength at 5 *wt* % PS (*d*<63 µm) (14 KJ/m^2) is 75% compared with the pure epoxy (8 KJ/m^2). Hence, the reinforcement provides the required characteristics to the pure epoxy, thus the fracture resistance can be increased. The excessive increase of *PS* particle content and size leads to a decrease in the impact of energy and absorbed energy. The same trend observed by the previous research [7], when the impact strength of pure polyester improved by adding the powder of pistachio shell from 8.25 KJ/m^2 to 18 KJ/m^2. While the previous result [27] found the percentage of increasing the impact strength of polymer composite was about 49% when the peanut particle added to the polymer matrix.

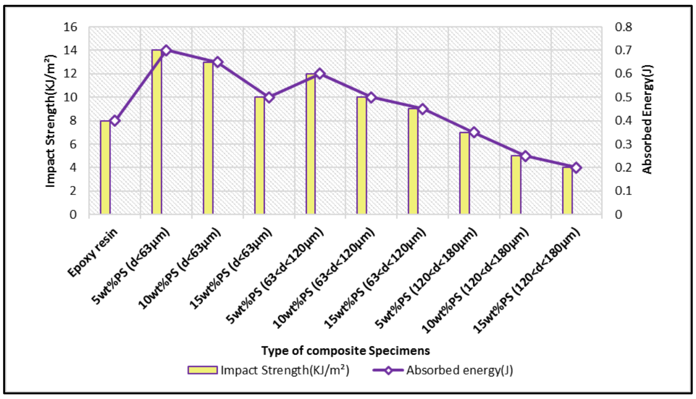

Fig. 13. Type of composite specimens versus impact strength and absorbed energy.

As explained previously, the excessive increase of *PS* particles produced the formation of *PS* particles agglomerates, which leads to deteriorating the impact strength, and absorbed energy by producing regions of stress concentration [7, 27]. Figure 14 shows the Izod impact device and the failed samples after the test.

Fig. 14. Impact device and the failed samples of the impact test.

3.4. Hardness properties

The hardness behaviour of PS/epoxy composites of various filler content is shown in Fig. 15. As shown in the Figure, the mean hardness increased when the pistachio shell filler content increases. Furthermore, the hardness was relatively improved by using large filler particles compared with small filler particles in the resin matrix. Based on Fig. 15, the findings show that the size of *PS* particles can positively affect the hardness of the composite. Therefore, the maximum enhancement of hardness was 28% which occurred at 15 wt % *PS* (63<*d*<120 μm) relative to the pure epoxy. Similar findings were also obtained for various natural filler filled polymer matrix [27-29] when the mean hardness increased by about 30% through using peanut filler in the polyester matrix [27]. The increase in hardness of the *PS*/epoxy composites can result because of the stronger interfacial bonding of the pistachio shell particles and epoxy matrix [27]. Figure 16 shows the hardness device (shore D).

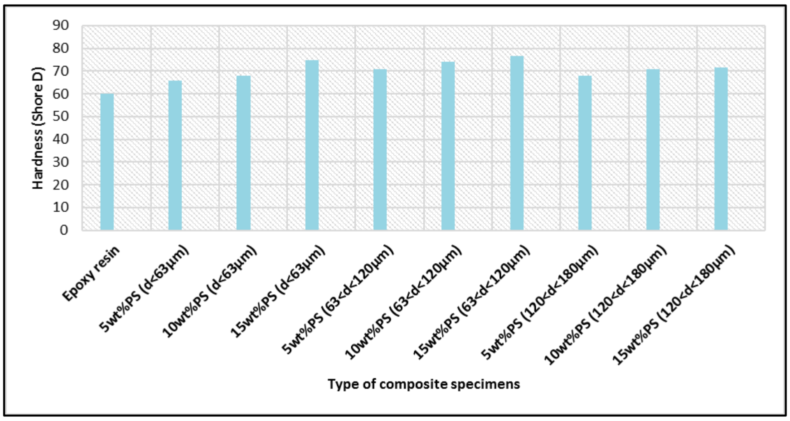

Fig. 15. Type of composite specimens versus hardness (shore D).

Fig. 16. Hardness device.

4. Conclusion

The present research was undertaken to produce composite materials made from pistachio shell (*PS*), epoxy polymer, and evaluate the effect of particle size and the content percentage of filler on the mechanical properties of *PS* / epoxy composites. Various mechanical tests, such as tensile, flexural, impact and hardness tests were carried out on the produced composite samples of *PS*/epoxy. These experiments have found the following:

- The research has found that generally the addition of pistachio shell powder importantly affects the mechanical properties of epoxy matrix composites. The results indicated that there was an increase or decrease in the mechanical properties of *PS*/epoxy composite compared with the pure epoxy and these changes depend on the quantity of the added powder or powder size.
- The tensile and modulus strength improved to reach their maximum values at the content of *PS* particles 5% by weight and the size of the *PS* particle was d <63 µm.
- The maximum flexural strength of composite samples was at *PS* content 5 weight% and particle size at d <63 µm whereas, at 15 weight % of the *PS* and particle size 120 <d <180 µm, the flexural modulus reached its highest value.
- The maximum impact strength and absorbed energy occurred in the content of *PS* 5 weight % and at d <63 µm, then they decreased with increasing *PS* (content and particle size) in the epoxy resin.
- The hardness of the PS/epoxy composite improved by about 28% when the content of the pistachio shell in the produced samples was 15 weight% and particle size 63 <d <120 µm.

The findings of this investigation complement those of earlier studies which showed the importance of using natural materials. Although this study focuses on improving the mechanical properties of *PS*/epoxy composite, the finding may well have a bearing on the environment by using a pistachio shell as particulate filler, which is low-cost and environmental-friendly.

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