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MECHANICAL AND THERMAL CONDUCTIVE PROPERTIES OF NATURAL AND SYNTHETIC CELLULOSE REINFORCED EPOXY COMPOSITES

OMRAN A. SHABEEB[1], DAWOOD S. MAHJOOB[2], HAMID M. MAHAN[2] AND MUAMMEL M. HANON[2,3]*

[1]Middle Technical University, Electrical Engineering Technical College, 10022 Baghdad, Iraq.
[2]Middle Technical University, Baqubah Technical Institute, 32001 Diyala, Iraq.
[3]Mechanical Engineering Doctoral School, Szent István Campus, MATE University, 2100 Gödöllő, Hungary.

**Corresponding author: Sharba.Muammel.M.Hanon@phd.uni-szie.hu*

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ABSTRACT: Natural and synthetic cellulose-based composites have been used widely as they have many advantages, the most significant of which are lightweight, durable, strong, flexible, and resistant to corrosion. Although several studies have reviewed the characteristics of these composites, only limited research has investigated combining both natural and synthetic cellulose together. In this study, the thermal and mechanical properties of epoxy resin reinforced with different additives (sawdust and industrial cellulose) will be explored. To do this, four samples of different materials were prepared at room temperature. The first sample consisted of 100% epoxy, whilst the next sample contained 95 wt.% epoxy and 5 wt.% industrial cellulose. The third sample contained 95 wt.% epoxy and 5 wt.% natural cellulose (sawdust), and the final sample contained 95 wt.% epoxy, 2.5 wt.% natural cellulose and 2.5 wt.% industrial cellulose. The findings indicated that mechanical properties and thermal insulation can be enhanced by adding natural cellulose to the mixture. Compared to the pristine epoxy, the improvement ratios for mechanical properties were as follows: tensile strength 25%, impact strength 16.6%, and hardness 6.9%, while the results were negative for bending resistance (3.9% less). In terms of thermal properties, the sawdust/epoxy composite showed better insulation (29% higher) than neat epoxy resin. These promising findings suggest the proposed composite can be a good alternative in numerous applications such as automotive parts and building construction that require superior mechanical characteristics and thermal insulation.

ABSTRAK: Komposit semula jadi dan sintetik berasaskan selulosa telah banyak digunakan secara meluas kerana ia mempunyai banyak faedah, dan yang paling penting ianya ringan, tahan lama, kuat, fleksibel dan tahan hakisan. Walaupun terdapat banyak kajian telah dilakukan pada ciri-ciri yang terdapat pada bahan komposit ini, terdapat kurang kajian dilakukan ke atas gabungan Bersama kedua-dua bahan semua jadi dan sintetik selulosa. Kajian ini adalah tentang suhu dan ciri-ciri mekanikal damar epoksi yang diperkukuhkan dengan pelbagai bahan tambahan seperti serbuk gergaji dan selulosa industri. Bagi menghasilkan bahan kajian ini, empat sampel dari bahan berbeza disediakan pada suhu bilik. Sampel pertama terdiri daripada epoksi 100%, sementara sampel berikutnya mengandungi epoksi 95 wt.% dan selulosa industri sebanyak 5 wt.%. Sampel ketiga mengandungi epoksi 95 wt.% dan 5 wt.% selulosa semula jadi (dari serbuk gergaji), dan sampel terakhir mengandungi epoksi 95 wt.%, dan selulosa semula jadi 2.5 wt.% dan selulosa industri 2.5 wt.%. Dapatan kajian menunjukkan ciri-ciri mekanikal dan penebat suhu boleh dipertingkatkan dengan menambah selulosa semula jadi dalam campuran. Jika dibandingkan dengan epoksi asal, nisbah penambah baikan bagi ciri-ciri mekanikal adalah seperti berikut: kekuatan tegangan 25%, kekuatan hentaman 16.6% dan kekerasan adalah sebanyak 6.9%, sementara dapatan kajian adalah negatif bagi rintangan ketika membengkok (iaitu berkurang sebanyak 3.9%). Dari segi ciri-ciri haba, komposit serbuk gergaji/epoksi menunjukkan sebagai penebat yang baik (meningkat 29%) berbanding damar epoksi bersih. Penemuan yang bagus ini menunjukkan, komposit yang di cadangkan ini dapat menjadi alternatif terbaik dalam pelbagai kegunaan seperti bahagian-bahagian automotif dan pembinaan bangunan yang memerlukan peningkatan ciri-ciri mekanikal dan penebat haba.

KEYWORDS: *natural composites; sawdust; mechanical properties; thermal conductivity; cellulose*

1. INTRODUCTION

Environmental awareness has led researchers to reconsider the design of engineering products for the construction, packaging, future, and vehicle industries. Polymer elements have become favorable parts for numerous applications due to their excellent characteristics [1]. In recent times, increasing attention has been paid to the use of a modified thermoset polymer [2]. Epoxy resins are polymeric or semi-polymeric materials that are part of the thermosets group and are widely used in composite materials [3]. Moreover, epoxy resin has become more popular than alternative products due to its beneficial properties, such as high chemical, thermal, electrical, and mechanical strength [4]. Multiple materials can be mixed to create a composite substance that has a unique combination of properties [5]. Epoxy resin modification is thus an innovative approach to polymer material production [6]. This material can be used to support other materials, which has enabled many workers to improve their properties in this field and to accomplish desired targets [7–9].

Recently, polymers have been reinforced with small amounts of strong fillers, as this can substantially enhance the mechanical and thermal properties [10–14]. It is thus sensible to seek other economically friendly raw materials (fillers) that could strengthen the properties of such products [15]. Modified epoxy resins are often used nowadays because they have great thermal, mechanical, and electrical properties when used to manufacture natural fiber-reinforced composites [16]. As they are environmentally safe and user-friendly, natural fibers are one of the most important components added to epoxy resin [17]. Unlike normal engineering fibers (such as glass, aramid, or carbon), natural fibers are widely usable, sustainable, recyclable, and cost-effective sources of fibers [18].

Natural materials, when compared to man-made materials, can provide a good combination of favorable properties, such as rigidity, strength, and low weight [19]. Natural fibers are also less harmful to the environment than synthetic fibers. Natural fibers are less erosive, which means that as they wear off, the devices' performance may not be affected, and they have a more hospitable manufacturing environment than synthetic fibers [20–22]. Studies exploring the topic demonstrate that sawdust can be added to polymer matrices from various sources, including trees, cellulosic powders, microcrystalline cellulose, natural fibers, and other waste materials produced during woodworking and food processing [23].

Composite materials are made up of two or more materials that have completely separate characteristics and do not dissolve or mix with one another [24]. By combining precise amounts of materials with inherited properties, new materials with modified and better properties can be created [25]. A chunk of wood is made up of long cellulose fibers that are held together by a chemical called lignin [26]. The overlapping materials are lightweight and provide excellent thermal and electrical insulation. Thus, in recent years, they have become increasingly more popular in various fields [27,28]. Natural fiber insulation is arguably the most practical way to produce environmentally friendly composites. Numerous studies have investigated natural fiber-reinforced composites. In recent years, polymer composites have been studied in great depth [29,30] and sawdust has been found to serve as an insulator, slowing heat flow and conduction. This ultimately enhances its capacity to isolate [31].

It has recently been discovered that, by adding a small quantity of strong fillers to polymers, the mechanical and thermal performance of the composites can be significantly improved [32]. Given their great insulation capacity, these composites can be applied for a variety of purposes. For instance, they can be used to fill the spaces between the interior and exterior sections of walls, or the floor, roof, ceiling, or insulation boards. Moreover, they can be used to manufacture aircraft and automobile parts and furniture. They are thus beneficial in many industries, including transport and even medicine [33].

Plenty of materials (natural and synthetic) have been investigated as reinforcements for polymer composites, as shown by the literature reviewed above. However, the additives utilized to form composites have not yet been fully covered, and there is still much to explore. Combining both natural and synthetic cellulose together in one composite was rarely inspected. Therefore, the present study aims to determine the effect of various combinations of additives, which are made from natural cellulose (sawdust) and industrial cellulose, on the mechanical properties (through tensile, flexural, impact, and hardness tests) and the thermal conductivity of composites, where epoxy is the matrix material. To the best of our knowledge, the combinations of additives utilized here have never been studied before.

2. EXPERIMENTAL WORK

2.1 Material Properties

Epoxy resin (ER) was used as a matrix material for the composites prepared. Epoxy is created by combining the liquid resin and an appropriate hardener. The ratio for resin to hardener is typically 2 to1, which enables the substance to cure perfectly. The epoxy resin used in this study was obtained from Sky Bahrain and contained the Sikadur-52 hardener. The specifications of the epoxy, as per the manufacturer, are presented in Table 1.

Table 1: Specifications of epoxy resin used in the study

Tensile strength (MPa)	Modulus of elasticity (MPa)	Percent elongation (%)	Density (g/cm^3)	Water absorption (%)	Shear strength (MPa)	Flexural strength (MPa)
37	1800	7	1.1	1.5	29.6	63

Industrial cellulose (IC), which is made up of carbon, oxygen, and hydrogen, is a very intricate carbohydrate. Properties of cellulose – $[(C_6H_{10}O_5)_n]$: Many properties are determined by the extent of polymerization, the chain length, and the number of glucose molecules that make up the polymer molecule. Cellulose is tasteless, odorless, and chiral, as well as being both insoluble and biodegradable, and appears in the form of white powder. The physical, mechanical, and thermal properties of the industrial cellulose are tabulated in Table 2.

Table 2: Specifications of industrial cellulose

Tensile strength (MPa)	Modulus of elasticity (MPa)	Percent elongation (%)	Density (g/cm^3)	Water absorption (%)	Flexural strength (MPa)	Thermal conductivity (W/m-k)	Specific heat capacity (j/g.ºC)
22.1 – 41.4	1005 – 1850	3.7 – 25	1.17 – 1.21	1.3 –1.8	29 – 55.8	0.17 – 0.33	1.26 – 1.67

The natural cellulose (NC) used is sawdust that is produced during the process of sawing wood (type jam wood). This sawdust was collected from the carpentry workshop of the Baquba Technical Institute, Middle Technical University, Iraq. It was then sifted using a sieve with apertures of 50 µm to obtain very fine particles (see Fig. 1a).

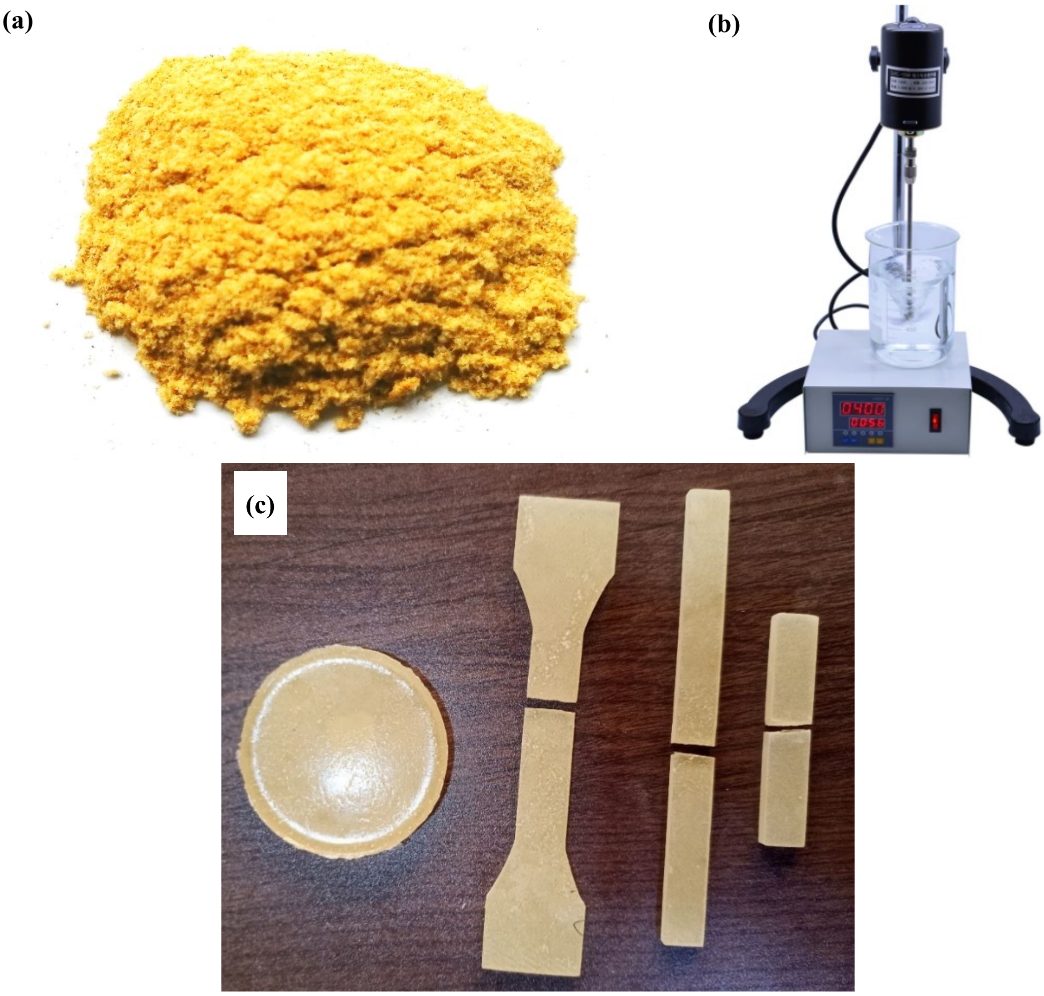

Fig. 1: (a) Physical appearance of natural cellulose (sawdust), (b) digital electric laboratory mixer, and (c) physical appearance of specimens (type ER+NC) after testing.

2.2 Preparing the Polymer Composite

Four square glass molds measuring (130 mm * 130 mm * 5 mm) were used in this experiment. To create casting samples, four polymeric compounds were prepared. These were made up of a base material epoxy resin mixed with a hardener at a mixing ratio (2:1). Once mixed, additives from industrial and natural cellulose were added to the composites for reinforcement (proportions presented in Table 3). The mixing process was carried out utilizing a digital electric laboratory mixer (see Fig. 1b) at room temperature and lasted 15 minutes. Subsequently, samples were left for a day and then placed in an oven for 5 hours at 60 °C. The primary reason for doing this was to eliminate internal stresses, tangle the chains in the mixture and harden and depressurize the sample [34].

Table 3: Designation and composition of composites

Sample No.	**Reinforcement %**		
	Cellulose (C)	**Epoxy (E)**	**Sawdust (N)**
ER	-	100	-
ER+IC	5	95	-
ER+NC	-	95	5
ER+NC+IC	2.5	95	2.5

2.3 Testing

Physical (thermal conductivity and hardness) and mechanical (tensile, bending, and impact) properties tests were performed on the polymeric compound samples. All experiments were carried out at room temperature, in accordance with American standards (ASTM). A CNC router machine was used to cut three samples of each polymeric compound to the required dimensions. The physical appearance of specimens (of one composite) is shown in Fig. 1c.

Regarding the physical properties' testing, Lee's Disc (type Griffin and George, England) was used to assess the thermal conductivity of the samples as per ASTM-D7340. The sample (disc) was sandwiched between the discs of the test setup. The heater was turned on, and the thermostat was adjusted to the desired temperature. In essence, thermal conductivity refers to a material's resistance to thermal transfer, which reflects the amount of heat that flows through it directly or indirectly. For each polymeric composition, three samples were tested for thermal conductivity. The sample's hardness was determined according to the ASTM D2240 using a durometer hardness tester (also known as the Shore D hardness tester) due to the low hardness of the polymeric compounds. When the indenter is forced into the testing material, the amount of resistance is measured and shown on a digital screen (scale). The key reason for performing the hardness test is to determine whether the material is suitable for use under specific conditions. For each polymeric compound, the hardness test was performed on three samples, during which the average readings for each sample were taken.

In terms of the testing of the mechanical characteristics, the sample tensile tests were carried out at room temperature using the universal testing equipment (see Fig. 2a) in accordance with ASTM D638-Type 1. Thus, a 5 mm/min crosshead speed was used. For each polymeric compound, the tensile test was carried out on three samples, and, during the process, the average results were recorded. During the tensile test, specimens were tightened adequately to avoid sliding. The flexural test of the samples was done in accordance with (ASTM-D790) and was measured using the universal hydraulic press, as the experimental setup seen in Fig. 2b. The two supports grip the specimen firmly, and the span length between them is 65 mm. The samples were tested with a loading rate of 2 mm/min. The flexural strength values are determined by the average of three bending tests. As illustrated in Fig. 2c, a Charpy impact test was performed in accordance with ASTM-E23 to evaluate the impact strength of the developed composites. A pendulum was used to break the unnotched specimen, with the test piece held tightly at each end. It is very important to perform a mechanical shock test to ensure that a material is safe. In this test, the extent to which a material is resistant to sudden shocks is evaluated. All specimens' dimensions used in the tests are shown in Fig. 3.

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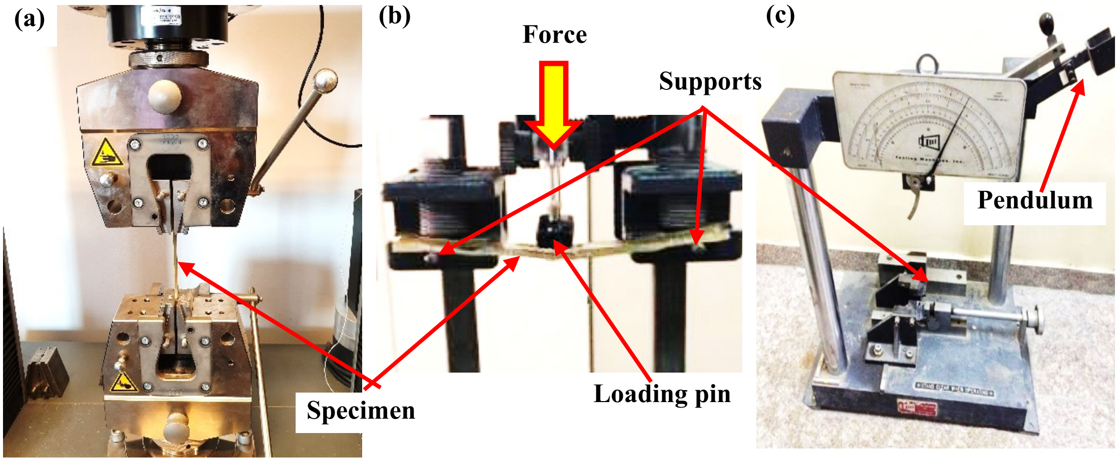

Fig. 2: Experimental setup of mechanical tests (a) tensile, (b) flexural, and (c) impact.

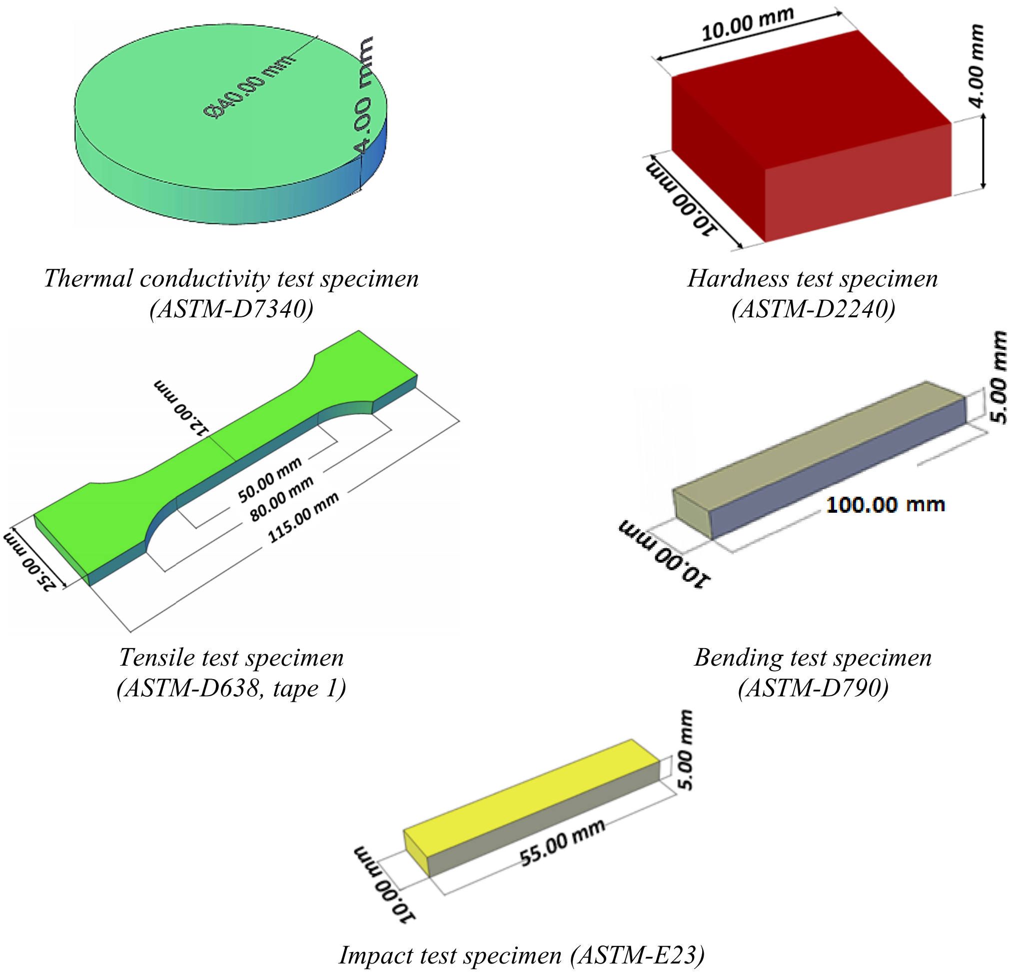

Fig. 3: Sample's dimensions of physical and mechanical properties tests.

3. RESULTS AND DISCUSSION

3.1 Physical Properties: Thermal Conductivity

The conductivity can be influenced by the existence of interfacial layers, free volume, and gaps or cracks within the inner structure. When fillers are loaded as particles during the sample's preparation, they might generate gaps because air voids (created while mixing) are unable to escape due to the particles. Therefore, all specimens containing particles were found to reduce the thermal conductivity of composites as compared to the pristine epoxy. As wood is a heat-insulating material, the sample with the polymeric compound (ER+NC) had the lowest thermal conduction efficiency (see Fig. 4) and hence provided the best thermal insulation. When compared to the epoxy resin specimen (the reference specimen), the improvement rate was 29%. This indicates that the presence of wood contributed to reducing the thermal conductivity of composites due to owning less thermal conductivity than epoxy as well as being in a form of particles within the matrix. The tendency of the results obtained agrees with the findings of a previous study [35]. They reported that adding 10 wt.% of wood dust to a composite reduces the composite's heat conductivity by around 67.1% compared to the neat epoxy resin.

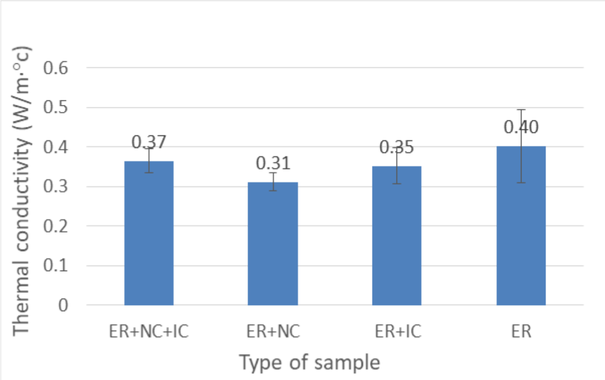

Fig. 4: Relationship between thermal conductivity and the proportions of additives to epoxy.

3.2 Physical Properties: Hardness (Shore D)

Figure 5 shows that the epoxy sample (ER+NC) reinforced with natural cellulose (sawdust) has the best hardness. This is because the natural cellulose particles in the composite are arranged in a uniform pattern. This strengthens the bonds in the polymeric chain. This is because adding natural cellulose to the compound impedes cracks formation during the preparation, which increases the bonding of the atoms, and thus increases the hardness value.

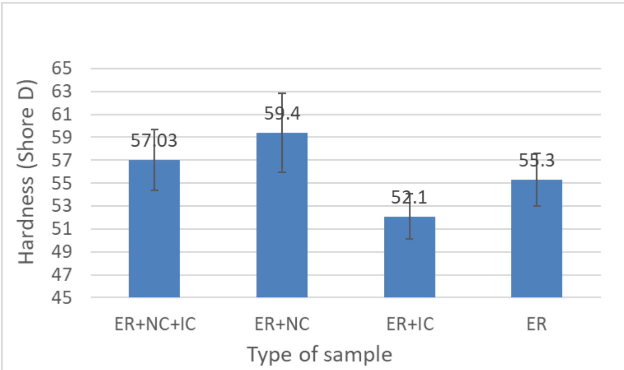

Fig. 5: Relationship between hardness and the proportions of additives to epoxy resin.

These findings suggest that indentation can be avoided through strong bonding. Thus, the improvement rate for this (ER+NC) sample was 6.9% higher than the reference sample. On the other hand, the hardness of the (ER+IC) was reduced by 6.1%. According to Homkhiew et al. [36], the Shore D hardness of composites (rubberwood sawdust reinforced thermoplastic natural rubber) increases as the amount of wood sawdust increased. They attributed this to the fact that wood sawdust has a far greater hardness than the matrix material (thermoplastic elastomer), and that adding wood particles reduces the elasticity or flexibility of polymer chains, which results in stiffer composites. This is in good agreement with the outcomes of the current study.

3.3 Mechanical Properties: Tensile Strength

The best tensile strength was identified (see Fig. 6) in the polymeric compound sample that was reinforced with natural cellulose (ER+NC) and this is due to the fact that the particles in sawdust join the fibers and strengthen the bonds between atoms. This is because sawdust, when loaded, serves as a barrier to interference movement inside the base material, decreasing the chance of plastic deformation [37]. As a result of this strong adhesive property, an improvement rate of 25% was achieved in comparison to the epoxy resin reference sample. However, the tensile strength of the (ER+IC) sample was found to be nearly 20% worse. According to Kumar et al. [38], wood dust (as filler) has good characteristics because it improves the mechanical properties of the polymeric resin. They stated that 10 wt.% fill provides the best tensile properties.

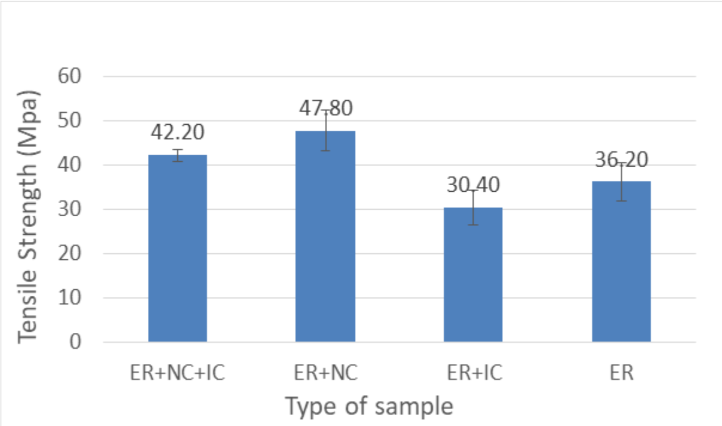

Fig. 6: Relationship between tensile strength and the proportions of additives to epoxy resin.

3.4 Mechanical Properties: Flexural Strength

The flexural strength of materials was measured in order to determine their capacity to withstand bending forces. The test results for the four samples revealed that the sample (ER+IC) prepared with industrial cellulose was the least resistant (36.4% lower) in comparison to the pure epoxy resin reference sample. However, as Fig. 7 shows, the sample (ER+NC) containing natural cellulose (sawdust) produced essentially identical results to the epoxy resin samples. This means that, even after the additive materials were added, there was no improvement in flexural strength. This can be ascribed to a lack of adhesion at the filler/matrix contact. Furthermore, as the particle-matrix interface is stressed, the number of microcracks increases, causing the crack to widen and fracture to occur. As a result, adding sawdust to the polymer matrix did not result in an improvement in flexural strength. Other researchers have disclosed similar outcomes [35].

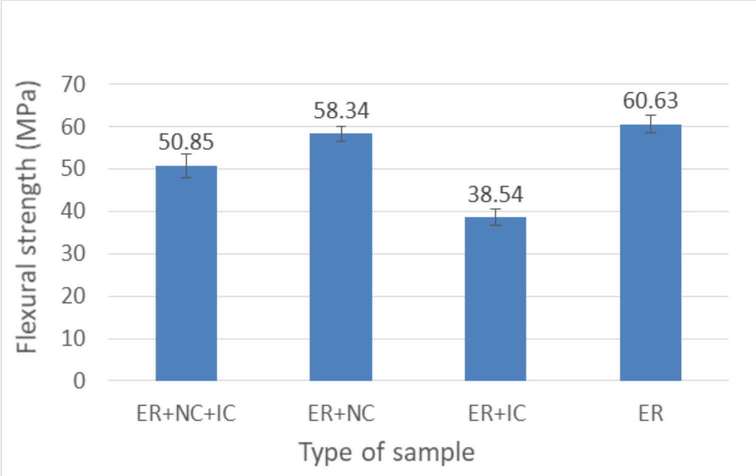

Fig. 7: Relationship between flexural strength and the proportions of additives to epoxy.

3.5 Mechanical Properties: Impact Strength

The findings are presented in Fig. 8. It is evident that the polymeric compound sample (ER+NC) reinforced with natural cellulose (sawdust) demonstrated the best impact strength. This is owing to the robust cohesion that exists between the sawdust and the epoxy. As a consequence, sawdust particles increase the amount of energy needed to break the sample. This energy is defined by the interface bonding's strength between the reinforcing materials' surfaces and the polymeric mix composite material's components [39]. Thus, a 16.6% improvement was achieved in this sample compared to the reference sample. On the other hand, the impact strength of the (ER+IC) sample was found to be reduced by 16.2%, whilst that of the (ER+NC+IC) sample was reduced by 2%.

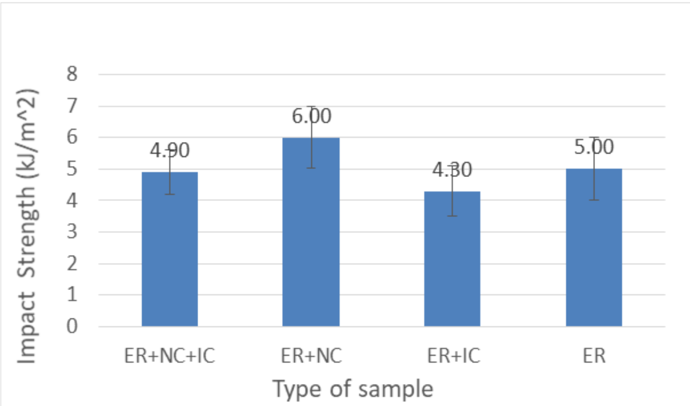

Fig. 8: Relationship between impact strength and the proportions of additives to epoxy.

4. CONCLUSIONS

Based on the experiments carried out in this study, the following conclusions have been made.

1- Physical properties:

- The thermal conductivity tests revealed that the polymeric sample with the best thermal insulation was the (ER+NC) sample containing 95% epoxy and 5% sawdust. This sample was found to be 29% more effective than the pure epoxy reference sample (ER).

- Sawdust was found to be most effective in the hardness tests. This was evident from the polymeric compound (ER+NC) sample which had the best hardness test results with an improvement rate of 69%.

2- Mechanical properties

- The findings of the tensile test also favored the (ER+NC) polymeric sample, as it showed the highest tensile strength with an improvement rate of 25%.
- The (ER+IC) sample was found to have the lowest bending strength in the bending test (which was 36.4% lower than the reference test), whereas the polymeric compound (ER+NC) sample seemed to have a bending resistance similar to that of the pure epoxy resin (ER) sample.
- The findings of the impact test also showed that the sample containing epoxy and sawdust had the highest impact strength and was 16.6% better than the reference sample.

Moreover, the findings of this work indicate that sawdust can be employed as a cost-effective and environmentally friendly reinforcer in polymeric compounds. This could be beneficial to many industries. Moreover, different types of wood and mixing ratios can be employed to achieve optimal mechanical and thermal properties.

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