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Thermal Conductivity Enhancement of Hybrid Epoxy Composites Using Copper Oxide Nanoparticles and Carbon-Nanotubes

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ABSTRACT

In this current experimental research, the amount of improvement in the thermal conductivity of HEC hybrid epoxy resins was studied by adding copper oxide nanoparticles CuONp and carbon nanotubes (CNTs) as hybrid additives in different proportions to select the sample with the highest thermal conductivity value to include it in the design of the Flat Plate Solar Collector FPSC as Thermal Interface Material TIM reduces thermal resistance between the absorber plate and the tube. Four groups of samples were prepared using a mass balance with a sensitivity of 0.01g and a magnetic mixing device, then poured into cubic plastic molds to take the shape of the sample. The first group consists of one sample of pure epoxy to calibrate the thermal properties testing device through it. The second group consists of five samples of epoxy loaded with CNTs by weight (1, 3, 5, 7.5, 10) %. The third group consists of five samples of epoxy loaded with CuONp with weight percentages of (1, 3, 5, 7.5, 10) %. The fourth group consists of five samples of epoxy loaded with CuONp and CNTs combined in weight percentages of (1, 3, 5, 7.5, 10) %. The thermal conductivity of the samples was measured experimentally using the hot disk analyzer technique to measure thermal specifications. After comparing the thermal conductivity values of the samples, the highest value was 1.57 W/mK for the HEC sample loaded with 10% CNTs, which represents 9.23 times higher than pure epoxy.

1. Introduction

Polymers are frequently covered materials due to chain chemical holding and arrangement. They have a few advantageous properties, such as tall attachment and great mechanical properties. They are less costly than other thermosets and have superior chemical resistance. However, due to their cross-linked structure, the higher thermodynamic properties are unimportant to the original copy and have higher electrical and thermal resistances.
The utilization of composite materials with an epoxy matrix has been prevalent in most cutting-

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edge plans due to its prevalence of thermal properties over other materials. There are many applications in which the need has developed for materials whose thermal conductivity can be changed (increase or decrease) depending on the applicable application. For example, but not limited to, researchers in the field of air conditioning homes and workplaces are working to find materials with thermal insulation properties to reduce heat loss. In contrast, researchers work in the field of heat dissipation from thermal applications to finding materials with high thermal conductivity.

Nanotechnology is an advancing field of science due to the broad examination of nanomaterials, particularly polymer composites. Polymer composites include two major components: lattice and fortification. The strengthening materials, which are, for the most part, nanomaterials, improve the properties of the framework. Thermosetting polymers like epoxy are utilized as network materials since they have excellent temperature resistance and crawl properties.

To achieve significant strides in thermal properties, size partitioning, and filler dispersion will be more demanding than the choice of polymeric materials. The thermal conductivity of the composite varies with temperature and the polymeric material content of the fillers. In many studies worldwide, researchers have summarized the field of changing the properties of thermoplastic polymeric materials by adding nanomaterials with high thermal conductivity, and they have reached better results. Among these studies are:

Noor Sabeeh Majeed et al. [1] experimentally studied the thermal behavior of the composite material of epoxy / TiO2 titanium dioxide in proportions (0, 2, 4, 6, 8, 10, 12) % and found that the thermal conductivity of the compound increases with increasing room temperature, reaching a limit of 1.9 W/m.K at the highest load ratio or 12%ratio epoxy / TiO2 titanium dioxide. Maher N. Abdullah et al. [2] presented a study on the use of epoxy resin with a hardener in a ratio of 1:2 at a temperature of 25 degrees Celsius, which was prepared using nanoparticles (nanomagnesium, nanozinc, nanoboron) with weight ratios of 1% and 2%. The results showed that the best thermal conductivity value at 2%wt reached 1.7 W/m.K . Kareem A. Jasim and Rihab Nassr Fadhil [3] presented a study on preparing epoxy compounds and copper powder. They used epoxy as a matrix with copper powder of medium diameter (240.91 nm). They prepared a circular mold with different weight percentages of the additive (0, 5, 15, 25, 35, 45%) to manufacturing models with specific measurements (diameter 30 mm) to study their thermal conductivity after conducting laboratory tests on them, the thermal conductivity value of the new material they installed increased as the percentage of the loaded material increased. The highest thermal conductivity value they observed was (1.502775 W/mK), representing 123.73% of the thermal conductivity of epoxy not loaded with the material. Yuexia Li et al. [4] presented a study on filling alumina nanowires (Al2O3-NWs) in epoxy composites with different shapes to understand the thermal behavior of the new composite material. They mixed Al2O3-NWs with lengths of 100 nm and diameters of about 5 nm and mixed them with epoxy to form a new composite material. They studied the effect of the percentage of the additive on the thermal properties of the new composite material. They observed that the compound with a 10% filler content increased its thermal conductivity by 134% higher than pure epoxy. Bassiouni et al. [5] experimentally studied mixing three different weight ratios of three different nanomaterials in a carbon fiber/epoxy (Al_2O_3, RGO, and MWCNTs) composite (1%, 0.3%, and 0.5%), (respectively) They noticed an increase in thermal conductivity of (0.52 W/mK) at ratio 1% of fillers after its value was (0.12 W/mK) before adding nanomaterials. Swapneel Danayat et al. [6] experimentally studied the thermal behavior of two samples, the first consisting of epoxy/graphene nanocomposites with weight percentages (3%, 5%, 7%) of both samples made of dimethyl formamide (DMF). They compared them to the second one made of acetone and found that the dimethyl formamide (DMF) compounds showed a 44% higher thermal conductivity than those made with acetone. Jivtesh Garg et al. [7] experimentally studied two models of epoxy and graphene composites containing graphene with a filler ratio of 7% by weight and found that dimethyl formamide (DMF) improved thermal conductivity by 44% compared to acetone. Po-Tuan Chen et al. [8] presented a practical study, part of which dealt with the thermal performance of compounds of epoxy, reduced graphene oxide rGO, graphene oxide, BaSO4 preparation, and Coating B in different proportions. Their study showed that the compound consisting of (epoxy, rGO reduced Graphene Oxide, and coating B) at a ratio of 5% by weight with the lowest percentage of BaSO4 preparation gave the best results. The thermal conductivity value reached around 160 W/m. K. Ismail Salih Mohammed et al. [9] presented a study on the effect of mixing micro- and nanoparticles of zinc oxide with an average size of (45 micrometers) and (50 nanometers) separately with epoxy on the thermal conductivity properties of the composite material. They prepared micro- and nanocomposites using the open-template method with different weight ratios of micro and nano (0.1, 0.3, 0.5, 0.7 wt), respectively. They examined the thermal performance of compounds using a lithium disk, and the relationship was inversely proportional between thermal conductivity and the increase in weight percentage of micro- and nano-additives, where thermal conductivity showed the lowest value, which is (0.62 W/m.C), at the highest loading percentage compared to the lowest loading percentage. R. Kochetov et al. [10] presented a study of the effect of adding nanoparticles (Sio2, Al2o3, aluminum nitride) in different weight ratios to epoxy to study the thermal performance of the composite material. Their study showed that the highest conductivity value reached 0.188 W/m K for the ER-AlN nanocomposite with a weight ratio of 5%. Junwen Ren et al. [11] presented a study on a nanocomposite material manufactured by blending fluorinated graphene (F-graphene) with epoxy resin. After examination, the F-graphene/epoxy composite showed a high thermal conductivity of (0.3304 W/m.K) at a low filler loading of 1.0 wt%, which is 67.63% higher than pure epoxy. Jianying Li et al. [12] presented a study on the preparation of epoxy compounds like Micro/nano-BN and a study of their thermal conductivity. Their study showed enhanced thermal conductivity in the compound with a loading concentration of 20% by weight BN with a micro/nano ratio of 95/5. Sriharsha Sudhindra et al. [13] studied theoretically the best technology findings in the field of thermal interface materials, and among the results of the study was the amount of improvement in the thermal conductivity of the epoxy compound loaded with nano-graphene, as the value of the thermal conductivity of the compound reached about (5 W/m.K) at a weight loading rate of 40%. Diyar J. Hassan. [14] They conducted an experimental study on mixing epoxy, palm oil, and zinc oxide at 1, 2, 3, and 5% concentrations. They reached the highest thermal conductivity value of 0.5 7 W/m.K at the highest loading percentage.

In this study, we seek to obtain a thermal conductivity value of more than 1.5 W/mK by mixing different proportions of the mentioned nanomaterials with epoxy. Research trends in this field are trying to reach an ideal value for the thermal conductivity of these materials for use in engineering applications related to thermal conductivity. In our current study, we seek to improve the thermal specifications of the nanocomposite material through practical and analytical research.

2. Experiential Part

2.1 Preparation of test samples

The composites were prepared to study their thermal behavior by mixing nanomaterials (CNT, CuONp) in different percentage ratios (1, 3, 5, 7.5, 10 %) with 2:1 wt/wt of epoxy resin and hardener. Plastic molds with internal dimensions of (2 * 2 * 1) cm was prepared, which are the dimensions required for the samples. As shown in Table (1), four sample groups were ready. The first group, G1, consists of epoxy plus a hardener. The second group, G2, consists of five epoxy resin samples loaded with CNTs in different weight percentages (1%, 3%, 5%, 7.5%, 10 %). The third group, G3, consists of five samples of epoxy resin loaded with CuONp in different weight percentages (1%, 3%, 5%, 7.5%, 10%). The fourth group, G4, consists of six epoxy resin samples loaded with (CNT\ CuONp) in different weight percentages (1%, 3%, 5%, 7.5%, 10%). Each Sample was mixed mechanically for 15 minutes at room temperature and then poured into the designated plastic mold until it completely solidified after 35 hours.

Table 1: Experimental Results

Group	Filler	Wt. %	Thermal conductivity (W/m. K)	Density Kg/m^3	Specific heat kJ/m^3. K	Diffusivity mm^2/s
1	–	0	0.170	1187	2763	0.061
	CNTs	1	1.028	1250	7612	0.147
	CNTs	3	1.059	1170	7889	0.145
	CNTs	5	1.16	1075	8120	0.142
	CNTs	7.5	1.16	1370	560	21.02
	CNTs	10	1.57	1110	15650	0.1
3	NCO	1	0.44	1150	6160	0.72
	NCO	3	0.47	1100	2800	1.64
	NCO	5	0.49	1075	3500	0.14
	NCO	7.5	0.406	1355	628	0.64
	NCO	10	0.69	1392	168	4.1
4	CNTs + NCO	1	0.83	1220	1330	6.87
	CNTs + NCO	3	0.88	1170	1230	7.78
	CNTs + NCO	5	1.34	1120	1101	0.12
	CNTs + NCO	7.5	0.819	1296	178	4.6
	CNTs + NCO	10	0.66	1312	6021	0.1

Figure 1 : Sample from G1

Figure 2 : Sample from G2

Figure 3 : Sample from G3

Figure 4 : Sample from G4

It should be noted that the density was calculated for each sample by determining the amount of mass and dividing it by the sample volume, relation (1).

$$\text{Density} = \frac{\text{mass}}{\text{volume}} \qquad (1)$$

To convert the volumetric specific heat capacity from J/m^{3}K to J/kgK, relationship No. 2 must be used.

Cp = Cv/density (2), where cp and cv have specific heat capacity in J/kgK and volumetric heat capacity in kJ/m3K, respectively.

2.2 Characterization

Hot Disk TPS Figure 5 is a fast and feasible way to measure (the thermal conductivity, thermal diffusivity, and volumetric heat capacity) of a material in the exact measurement based on the relationship between these thermal properties. The hot disk thermal constants analyzer relies on the transient level source (TPS) method. It is a transient heating level sensor (a thin spiral layer of nickel or other) with a known thermal resistance applied between two fragile layers of insulating material. To be able to measure, we place the sensor between the two samples whose thermal properties are to be measured so that they are identical in geometric shape, size, and surface flatness until complete contact and sealing are achieved between them and the sensor.

The tester was calibrated by measuring the thermal conductivity of the pure epoxy sample, which was 100% identical to the standard value of pure epoxy. This indicates that the results issued by the testing device are accurate and reliable.

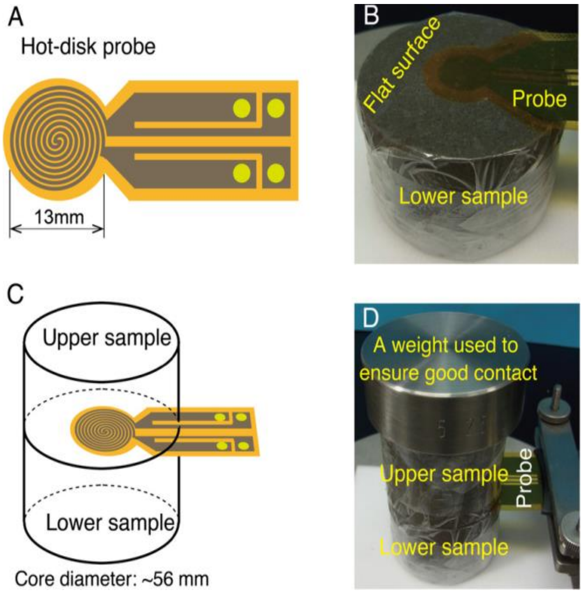

Figure 5. The hot disk probe with a bifilar spiral element. [15].

3. Results and Discussion

In Table No. 1, experimental results were obtained that included (thermal conductivity, heat capacity, diffusivity, and thermal density) for fifteen samples of epoxy compounds loaded with additives in different proportions, in addition to pure epoxy samples, where we note the variation in the values of the thermal properties of each sample. Regarding the thermal conductivity results of the samples, we notice an apparent increase in all samples when we compare them to the pure epoxy sample. The reason is that all additives have thermal conductivity compared to pure epoxy. We will go into deeper explanations when discussing the conductivity results in Figure 6. It shows the thermal conductivity of fifteen samples with different mixing ratios of additives, in addition to a sample of pure epoxy.

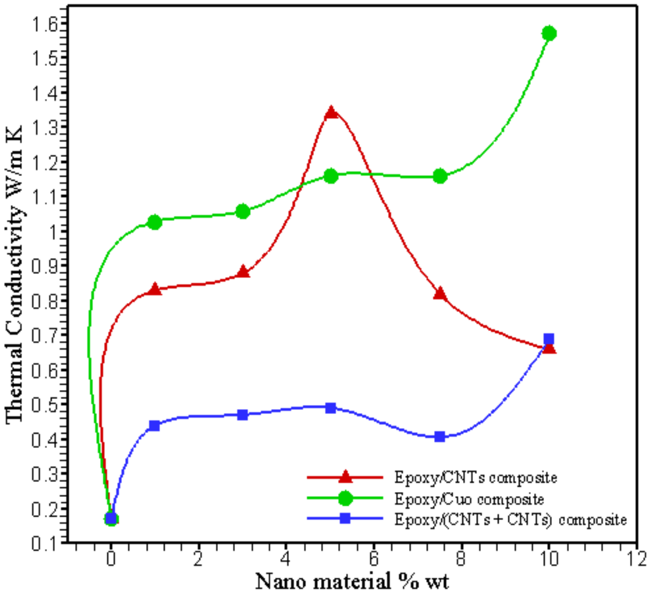

Figure 6. Nano additives ratio versus thermal conductivity

Regarding the samples' specific heat capacity results, we notice the difference in the results when we compare them to pure epoxy, and we will discuss deeper explanations when discussing the results of the specific heat capacity for each mixing ratio in Figure 7.

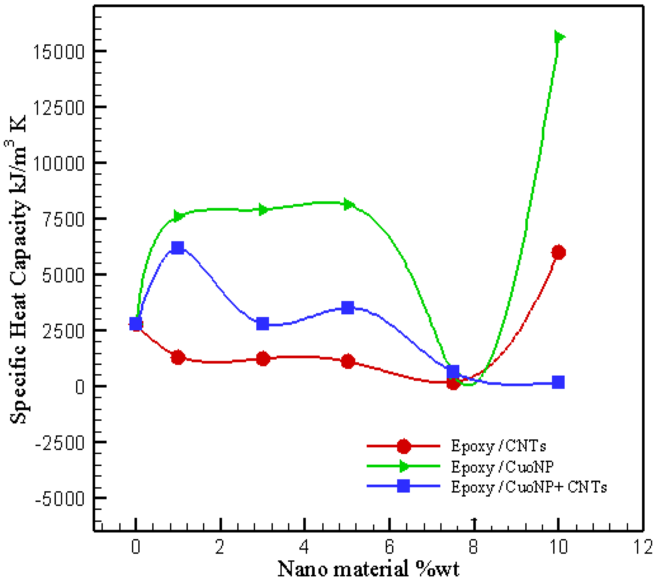

Figure 7. Nano material ratio versus thermal diffusivity.

Regarding the results of the thermal diffusivity of the samples, we notice an increase in all samples when we compare them to pure epoxy because the thermal diffusivity of the additives is higher than that of epoxy. We will discuss deeper explanations when discussing the thermal diffusivity results for each mixing ratio in Figure 8.

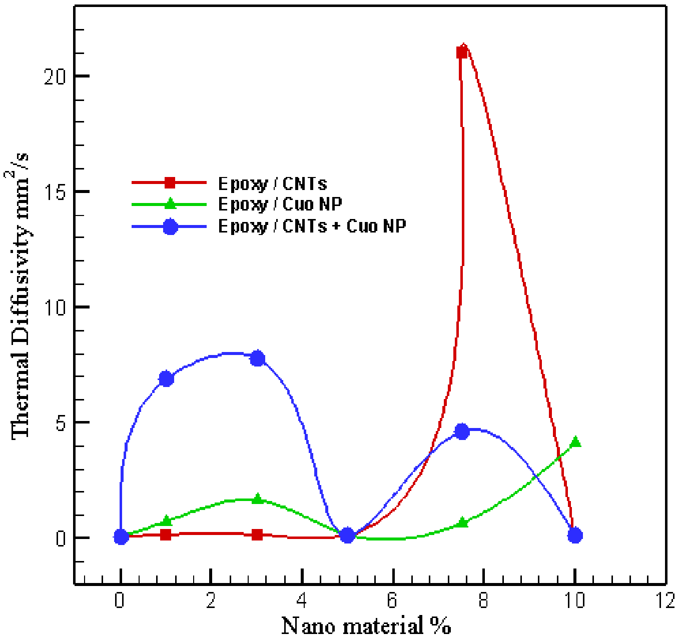

Figure 8. Nanomaterial ratio versus volumetric-specific heat

The starting point represents pure epoxy without additives; its thermal conductivity was 0.17 W/mK. At a concentration of (1, 3, 5)% by weight for each of (CNTs + CuONp, CNTs, CuONp), we notice that the value increases with increasing concentration, as the highest value at these concentrations was for compounds containing CNTs due to the higher thermal conductivity of this substance compared to the rest of the additives. The lowest value for the same concentration was for the compound containing CuONp due to the lower thermal conductivity of this substance compared to the rest of the additives, while the value was in the middle at the same concentration for the compound containing CNTs + CuONp. The reason is that the thermal conductivity reached the middle of the two values when mixed in the same compound. At a concentration of 7.5% by weight for each of (CNTs + CuONp, CNTs, CuONp), we note that the value at this concentration for the compound containing CNTs did not change compared to the previous concentration. Still, the value remained the highest among the rest of the compounds at the same concentration. In contrast, the value decreased significantly for the compounds containing (CNTs + CuONp, CuONp) from the lower concentration value. The reason is that we believe that the behavior becomes the opposite at this concentration.

At a concentration of 10% by weight for each of (CNTs + CuONp, CNTs, CuONp), we notice that the value returned to increase for the compound containing CNTs due to the higher thermal conductivity of this substance compared to the rest of the additives with increasing concentration, as it had the highest value among all compounds at all concentrations and It reached 1.57 W/mK.

Fig 7 shows the specific heat capacity for fifteen samples with different additive mixing ratios and a sample of pure epoxy. The starting point represents pure epoxy without any additives; its value was 2730 kJ/m3 K.

At concentrations 1, 3 , and 5 % by weight for each of (CNTs, CuONp), we notice that the value increases with increasing concentration. In contrast, the values decrease for the compound containing CNTs + CuONp compared to the value for pure epoxy.

At a concentration of 7.5, 10 % by weight for each of (CNTs + CuONp, CNTs, CuONp) we notice that the value returned to increase significantly for the compounds containing (CNTs + CuONp, CNTs) due to the higher thermal conductivity of this substance compared to the rest of the additives with increasing concentration as it had the value was the highest among all compounds at all concentrations and amounted to 15650 kJ/m3K, while the value continued to decrease for the CuONp compound .

Fig 8 shows the thermal diffusivity for fifteen samples with different additive mixing ratios and a sample of pure epoxy. The starting point represents pure epoxy without any additives; its value was 0.061 mm^2/s.

At concentrations of 1 and 3% by weight for each of (CNTs + CuONp, CuONp), we notice that the value increases with increasing concentration. In contrast, the values for the compound containing

CNTs did not change compared to the value for pure epoxy. This behavior at these ratios appears to be due to the type of these additives.

At concentrations of 5, 7.5, and 10 % by weight for each of (CNTs + CuONp, CNTs, CuONp), we notice that the values decrease but converge greatly at this percentage. The values increased again for all compounds at a rate of 7.5%.

4 .Conclusions

After manufacturing new epoxy composites loaded with different nanomaterials in weight percentages (1%, 3%, 5%, 7.5%, 10%), using carbon nanotubes and CuONp as additive fillers, testing their thermal properties. The thermal conductivity of epoxy/CNTs composites showed the highest value of about (1.57 W/mK) at a mixing ratio of (10%) of (CNTs), which represents an increase of (9.23) times the thermal conductivity of pure epoxy. Thus, we obtained nanomaterials with an essential value in thermal conductivity that can be used in engineering applications that depend on thermal conductivity to increase their thermal efficiency when used.

Founding

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Conflicts of Interest

None

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