

Evaluation of Mechanical Properties of Epoxy Resin Nanocomposite Reinforced by CuO–GO Hybrid

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Abstract: In this study, an epoxy-based nanocomposite reinforced with copper oxide-graphene oxide hybrid was investigated. Initially, the hybrid powder of CuO–GO with a weight ratio of 9:1 was prepared. The hybrid filler with different weight percentages ranging from 0.1–0.5 was used to reinforce the epoxy resin. The prepared samples were analyzed using XRD, FTIR, FESEM, TEM, and tensile testing. According to the XRD results and SEM images, the hybrid powder was successfully prepared, and the mechanical testing results showed an improvement in tensile strength in the composite samples. The best composite sample in terms of tensile strength was the one containing 0.3 wt% of hybrid reinforcement, which exhibited a 71% and 161% increase in the tensile strength and elastic modulus, respectively, as compared to the neat resin sample.

Keywords: Nanocomposite, Epoxy, Copper oxide, Graphene.

1. INTRODUCTION

Epoxy resin was first introduced commercially in the late 1940s and is currently widely used in various industries [1]. Epoxy is a polymer composed of epoxy groups, also known as polyepoxide. Epoxy is produced by the reaction of epichlorohydrin and bisphenol A to yield bisphenol A diglycidyl ethers (DGEBA), which is a key component in the formation of epoxy resins. Epoxy is characterized by its three-membered ring structure. Epoxy resins have various types, such as trimethylpropane, tetrafunctional epoxy resin, novolac epoxy resin, cycloaliphatic epoxy resin, and bisphenol A diglycidyl ether. The properties of epoxy resin depend on the number of their monomers [2]. Among all available polymeric materials for use as a matrix, epoxy is one of the best choices due to its low cost, ease of manufacturing and processing, lightweight, and excellent chemical properties [1, 2]. In epoxy-based nanocomposites, the incorporation of nanofillers can lead to increased flexural and tensile strengths [3], enhanced toughness and impact resistance [4], improved thermal and electrical properties [5], and heightened corrosion resistance [6].

The properties of a nanocomposite are affected by various types of nanofillers. For instance, adding silica nanoparticles can increase corrosion resistance and mechanical properties [3], while

adding carbon nanofillers can enhance the composite's mechanical and interfacial properties [7]. The properties of the composite can also be impacted by the size and shape of the nanofillers. For example, incorporating graphene, transition metal dichalcogenides, MXene, and hexagonal boron nitride (h-BN) can enhance the composite's structural and flame-retardant properties [6]. Copper oxide nanoparticles (CuO) are widely used in producing heterogeneous catalysts due to their high activity and selectivity, as well as their accessibility and low production cost. Copper oxide nanoparticles possess excellent properties such as a direct bandgap of 1.3–2.1 eV, non-toxicity, thermal and chemical stability, high melting point, good semiconductivity, and electrochemical activity [8, 9]. Sunny et al. [10] achieved an upsurge in the hydrophilicity of the epoxy resin by the incorporation of nanosized copper oxide particles. The improvement in hydrophilicity of nanocomposite was attributed to the enrichment of the polymer surface with nanoparticles. The influence of copper oxide nanoparticle dispersion on the thermophysical properties of the epoxy nanocomposite was studied by Zabihi and Ghasemlou [11]. They found that the nanocomposite with 5% CuO nanoparticles possessed optimal performance in terms of thermal behavior. In another work, the mechanical and thermal properties of a composite made from different ratios of alkyd/epoxy resins

reinforced with CuO nanoparticles were evaluated [12]. They concluded that the tensile and flexural strength of the mixture samples decreased with increasing alkyd content. This loss was compensated by the incorporation of nano-CuO, so that their nanocomposite (alkyd/epoxy=30:70) showed an improvement in tensile and flexural strength of about 18 and 41%, respectively, compared to the purely mixed resins. Various carbon-based fillers have been used to reinforce polymer matrices. These fillers, mainly derived from graphite, include carbon black, fullerene, carbon nanotubes, graphene, etc. Carbon nanotubes were first reported in 1991, followed by a report on graphene in 2004. When added in small amounts to epoxy resin, carbonaceous nanofillers can enhance its mechanical properties [13]. Graphene and graphene oxide (GO) exhibit excellent electrical, mechanical, and thermal properties due to their unique structural and morphological characteristics. The elastic modulus and failure strength of pure, defect-free graphene has been reported to be 800–850 GPa and 100–150 GPa, respectively [13]. The presence of graphene in the epoxy resin increased the viscosity by 4.2% and the shrinkage from 3.2% to 5.7% [14]. Furthermore, the addition of graphene oxide to epoxy resin at an amount of 0.1 wt.% increased the tensile strength and tensile modulus of the composite by 40% and 31%, respectively [15]. Graphene oxide has great potential due to its excellent mechanical properties and high aspect ratio. The influence of graphene oxide on the mechanical properties of epoxy composites was studied, and it was found that the addition of graphene oxide can improve the mechanical properties of the composite [16]. Graphene-based nanostructures have been proven to be good nanofillers for epoxy resins, as the fabricated nanocomposites have demonstrated remarkable mechanical properties [17, 18].

Hybrid reinforcements have recently attracted more attention for use in polymer nanocomposites. To achieve higher thermal conductivity and lower electrical conductivity, a magnesium oxide-coated graphene nanostructure was employed, which may be a suitable candidate for use in electronic packaging and insulation applications [19]. In another research, ZnO-decorated graphene was synthesized and dispersed in an epoxy matrix to improve water

barrier properties and corrosion resistance [20]. Furthermore, the use of GO–TiO₂ nanohybrid in the epoxy resin resulted in an increase of up to 59% in tensile strength and 79% in flexural strength compared to unmodified epoxy resin [21]. In this study, for the first time, the effect of adding CuO–GO nanofiller to an epoxy-based matrix on the mechanical properties of the fabricated nanocomposite was investigated.

2. EXPERIMENTAL PROCEDURES

A 99% pure graphene oxide powder with a sheet thickness of 3.4–7 nm and 6–10 layers was purchased from Pishgaman Nanomavad Iranian Co. Cupric oxide nanoparticles with chemical formula of CuO and dimensions of 30–50 nm and a purity of 99% were purchased from Nanomvad Gostaran Pars Co. A Kumho epoxy resin (model EPON 828) with its corresponding hardener (Epikure F205) was purchased. To prepare a hybrid nanofiller, a weight ratio of CuO:GO of 9:1 was assumed. First, 0.45 g of copper oxide nanopowder was mixed with 0.05 g of graphene in 20 mL of dimethylformamide (DMF) solution. The mixture was subjected to ultrasonic treatment using a probe sonicator at a power of 750 W for half an hour. The solution was then centrifuged and the resulting precipitate was dried at 100°C. The powder samples were analyzed by X-ray diffraction powder (XRD), Fourier transform infrared (FTIR) spectroscopy, scanning electron microscopy (SEM), and transmission electron microscopy (TEM) techniques.

To prepare composite samples with a resin-to-hardener weight ratio of 2:1, the hybrid powder was dispersed in the hardener using an ultrasonic probe with a power of 400 W for 5 min. The resulting solution was then added to the resin and mechanically stirred to obtain a homogeneous mixture. Finally, the mixture was poured into a silicone mold and cured at 60°C for 8 h. Dumbbell-shaped specimens were prepared for the tensile testing according to the ASTM D638. Tensile tests were conducted using a SANTAM testing machine (STM-50) at room temperature. The strain rate for this test was determined to be 2 mm/min. Fig. 1 shows a diagram illustrating the procedure of composite fabrication. For comparison purposes, the neat specimen without nanofiller was also fabricated in the same way. Table 1 provides the specifications of different specimens.

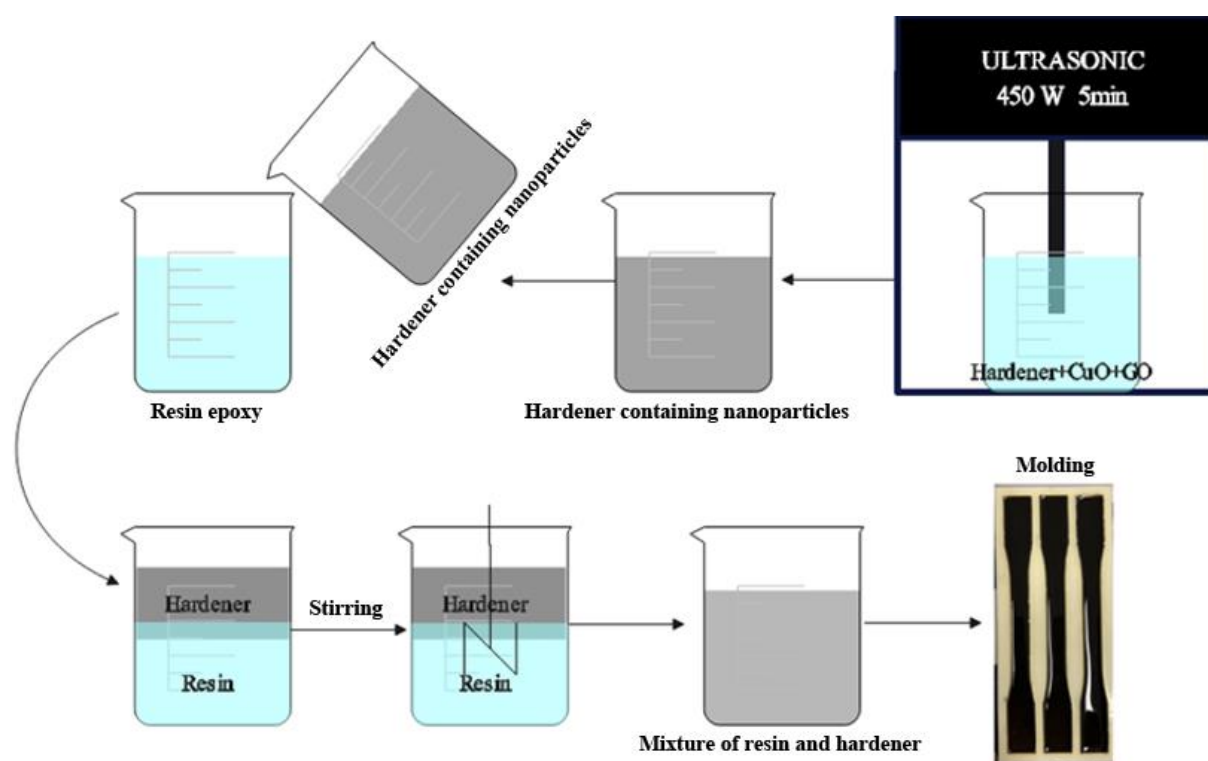


Fig. 1. Diagram illustrating the procedure of the composite fabrication.

Table 1. Specifications of different composite specimens

Specimen code	Specification
EC0	Epoxy + Hardener
EC1	Epoxy + Hardener + 0.1 wt.% CuO-GO
EC2	Epoxy + Hardener + 0.2 wt.% CuO-GO
EC3	Epoxy + Hardener + 0.3 wt.% CuO-GO
EC4	Epoxy + Hardener + 0.4 wt.% CuO-GO
EC5	Epoxy + Hardener + 0.5 wt.% CuO-GO
EC6	Epoxy + Hardener + 0.3 wt.% CuO

3. RESULTS AND DISCUSSION

Fig. 2 shows the X-ray diffraction spectrum of copper oxide, graphene oxide, and their hybrid. The copper oxide pattern shows peaks located at angles of 32.5°, 35.5°, 38.9°, 48.7°, 53.5°, 58.3°, 61.5°, 66.2°, 68.9°, 72.4°, and 75.2°, which are associated with the planes (110), ($\bar{1}$ 11), (200), ($\bar{2}$ 02), (020), (202), ($\bar{1}$ 13), ($\bar{3}$ 11), ($\bar{2}$ 21), (311), and ($\bar{2}$ 22), respectively, of the monoclinic crystal structure of CuO. The XRD pattern of GO exhibits a sharp peak at an angle of about 10°, which corresponds to the (002) plane of the honeycomb structure of graphene. In general, the characteristic carbon peak in pure graphite appears at an angle of approximately 26°, representing the (002) plane with an interlayer spacing of 0.34 nm [22, 23]. Following the

chemical oxidation of graphite, the 2 θ peak shifts to smaller angles of about 7–10°, which vary depending on the degree of carbon oxidation. This shift indicates the oxidation of graphite to GO with a d -spacing of ~0.86–0.93 nm. The increased d -spacing is a result of the formation of oxygenated functional groups (carbonyl, epoxy, hydroxyl, and carboxyl) between the carbon layers caused by the oxidation process [22–24]. For the XRD pattern of the hybrid powder, CuO peaks are observed without any shift. However, the GO (002) peak in the hybrid powder shifts toward smaller diffraction angles (~7°) with lower intensity, which is most likely due to the decrease of GO layers by the rigorous ultrasonic treatment.

FTIR spectra of three materials, copper oxide, graphene oxide, and copper oxide-graphene oxide

hybrid, are shown in Fig. 3. Based on this figure, it can be observed that the broad absorption peak at around 3417 cm^{-1} is attributed to the O–H bond. Two peaks are observed at around 2925 cm^{-1} and 2855 cm^{-1} , which correspond to asymmetric and symmetric C–H stretching vibrations, respectively. The peak at 2350 cm^{-1} is attributed to C=C stretching. The range between 1636 and 1618 cm^{-1} corresponds to the hydrogen bond vibrations of water.

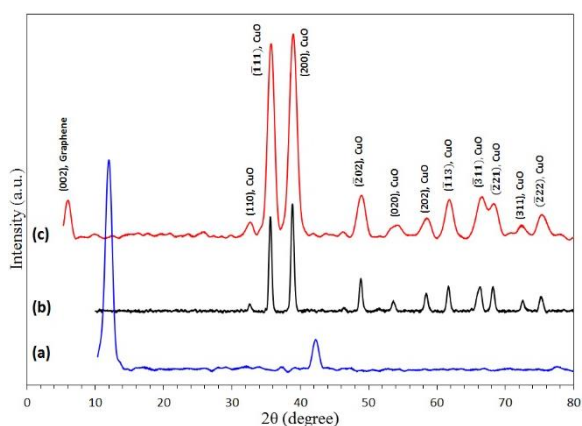


Fig. 2. X-ray diffraction patterns of (a) GO, (b) CuO, and (c) CuO–GO.

The peak at 1380 cm^{-1} is assigned to symmetric C–H bending vibrations. Generally, copper oxide has characteristic peaks in the range of $600\text{--}400\text{ cm}^{-1}$, and this study, showed an absorption peak at 512 cm^{-1} , which corresponds to the stretching bond between copper metal and oxygen. The OH stretching peak for graphene appears at 3402 cm^{-1} , and the C=O stretching peak appears at 1717 cm^{-1} . In the CuO–GO hybrid, all these peaks are also present, but their intensity is reduced due to

the low content of GO [25–28].

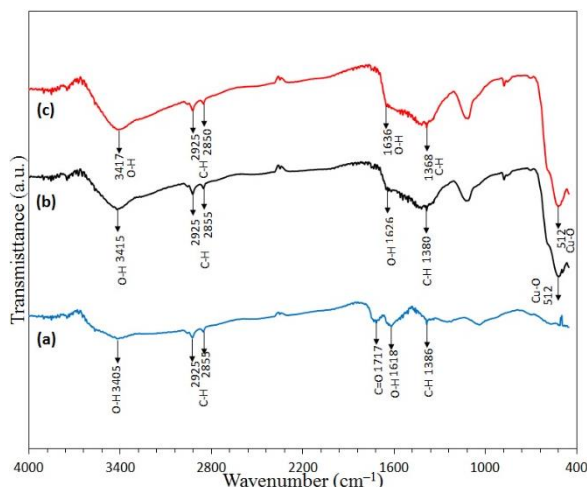


Fig. 3. FTIR spectra of (a) GO, (b) CuO, and (c) CuO–GO

Transmission electron microscopy (TEM) was used to study the morphology of nanoparticles. TEM images of copper oxide powder at different magnifications are shown in Fig. 4. CuO nanoparticles exhibit a spherical morphology with an average particle diameter of $20\text{--}40\text{ nm}$. Furthermore, scanning electron microscopy (SEM) was performed to determine the morphology of the decorated nanoparticles on the GO nanosheets and their distribution in the hybrid powder (Fig. 5). As can be seen, copper oxide nanoparticles successfully anchored on the GO nanosheets while maintaining their spherical morphology. Considering the ratio of copper oxide to graphene, it can be concluded that the surface of the graphene nanosheets is fully covered by copper oxide.

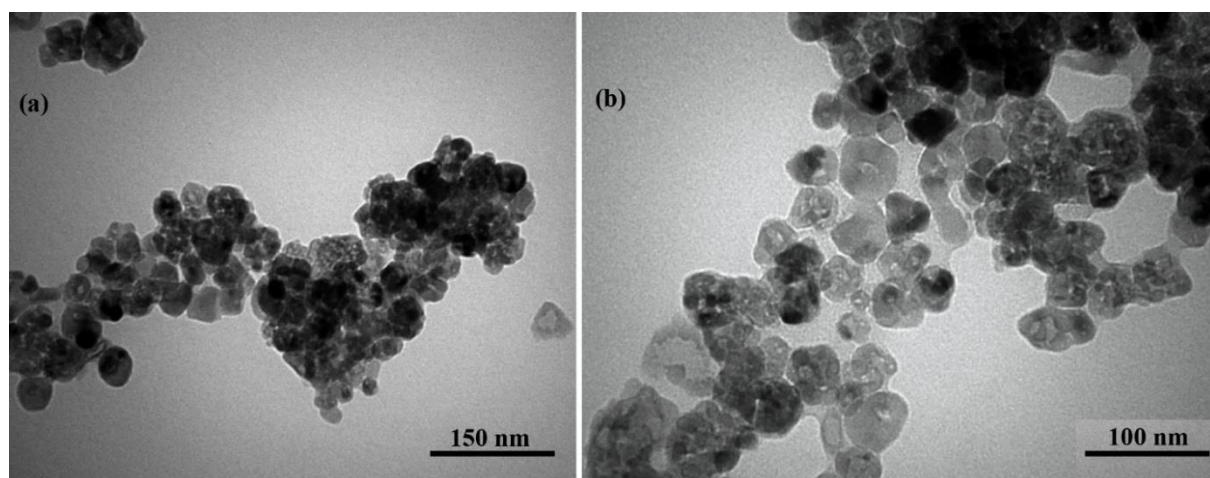


Fig. 4. TEM images of CuO nanoparticles at different magnifications.

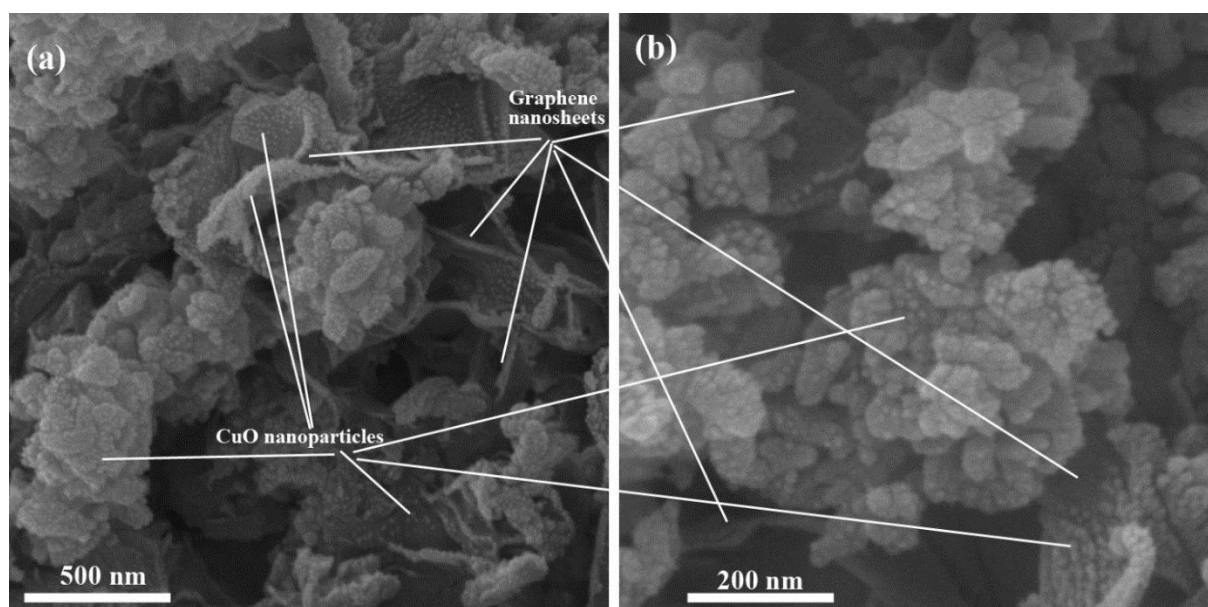


Fig. 5. SEM micrographs of CuO-GO nanocomposite at different magnifications.

The elemental distribution maps for carbon, oxygen, and copper in the hybrid powder are also presented in Fig. 6. Based on the distribution of copper and oxygen points in this map, it can be inferred that copper oxide nanoparticles are

uniformly distributed on the graphene layers. Additionally, it can be observed that the amount of carbon is low, which is attributed to the low ratio of GO to CuO. Overall, it can be concluded that the hybrid powder has been successfully synthesized.

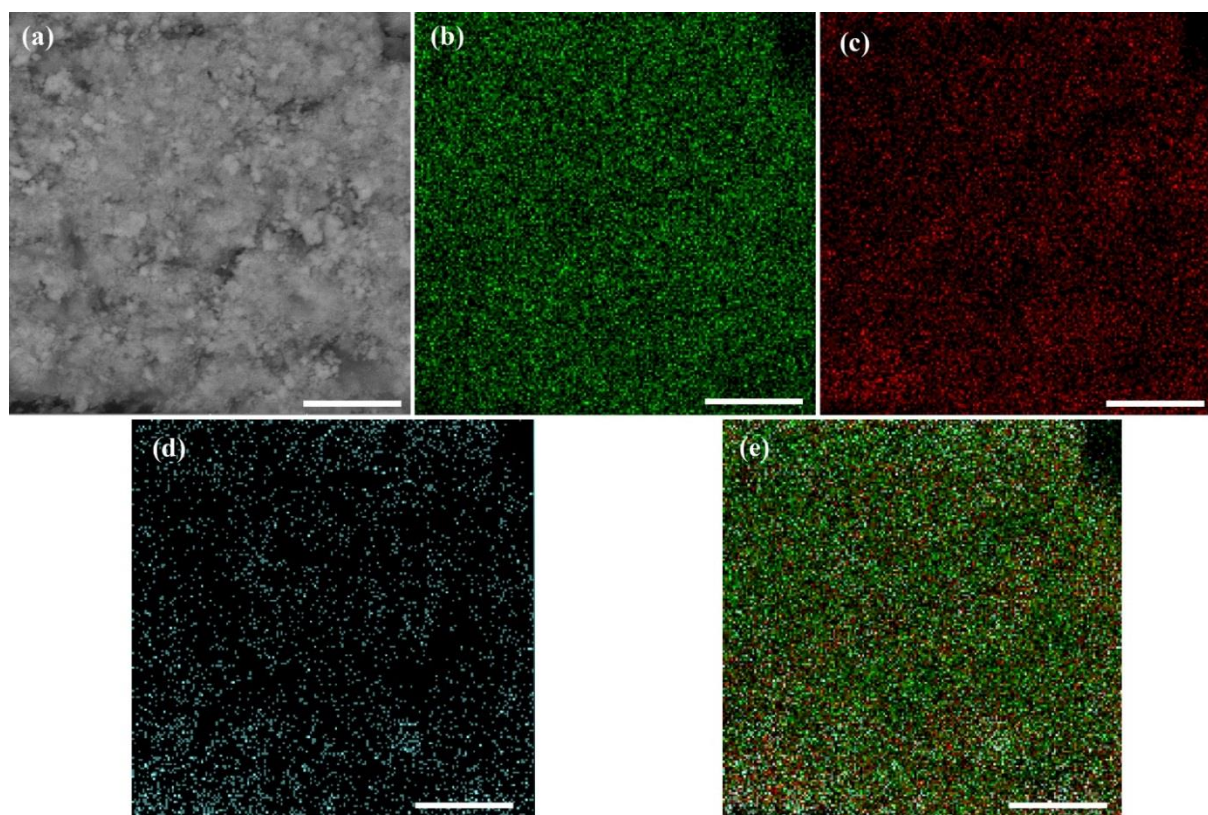


Fig. 6. SEM micrograph of the CuO-GO nanocomposite (a) and its elemental distribution maps for copper (b), oxygen (c), carbon (d), and their combination (e). Scale bars in the image are 10 µm.

Fig. 7 (a) shows the stress–strain curves of the different epoxy-based composites with 0–0.5 wt.% CuO–GO hybrid nanofiller. The influence of the weight fraction of the nanofiller on the tensile strength and elastic modulus of the nanocomposite is also presented in Fig. 8 (b). Neat epoxy resin (EC0) had the lowest tensile strength and elastic modulus. It can be also seen that both tensile strength and elastic modulus initially increased by increasing the amount of nanofiller up to 0.3 wt.% and decreased after the maximum point with a further increase in the reinforcement content (Fig. 8 (b)).

The sample EC3 (as the best sample) containing 0.3 wt.% CuO–GO hybrid nanofiller with a tensile strength of ~29 MPa and elastic modulus of ~2040 MPa exhibited an improvement of approximately 71 and 161%, respectively, compared to those of the neat sample (EC0: ~17 MPa and 782 MPa). A critical point in the relationship between strength and nanofiller content was observed, and the strength decreased at nanofiller concentrations above 0.3 wt.%. This can be attributed to the agglomeration and clustering of the nanofiller in the resin matrix at higher concentrations, disrupting their homogeneous distribution and effective

reinforcing function. For comparison purposes, the epoxy composite containing only 0.3 wt.% CuO nanoparticles (without GO) were also tested (EC6 sample). The EC6 nanocomposite with a tensile strength of 21 MPa and an elastic modulus of ~1060 MPa (23 and 92% increase compared to EC0, respectively) had lower mechanical properties than EC3, indicating the considerable effect of graphene nanosheets in preventing crack propagation. In addition, graphene nanosheets can improve the mechanical properties of the matrix because of their high mechanical properties. The better dispersion of nanoparticles leads to a lower stress concentration. It seems that graphene nanosheets can facilitate a better dispersion of nanoparticles within the matrix, which has also been reported previously [29].

Fig. 8 compares the fracture surface of different specimens after the tensile test: (1) the neat epoxy specimen (EC0), (2) the epoxy loaded with only 0.3 wt.% CuO nanofiller (EC6), and (3) the epoxy loaded with 0.3 wt.% CuO–GO nanofiller (EC3). For neat epoxy resin, a quite smooth fracture surface after the tensile test (Fig. 8 (a)) is observed, which is representative of the brittle failure.

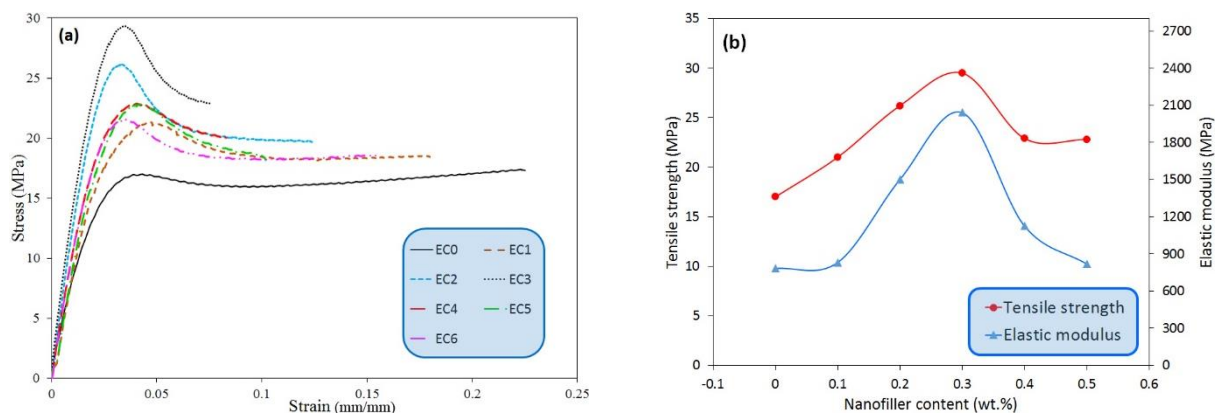


Fig. 7. (a) Stress-strain diagram for different specimens. (b) Variations of tensile strength and elastic modulus of the nanocomposite with the nanofiller content.

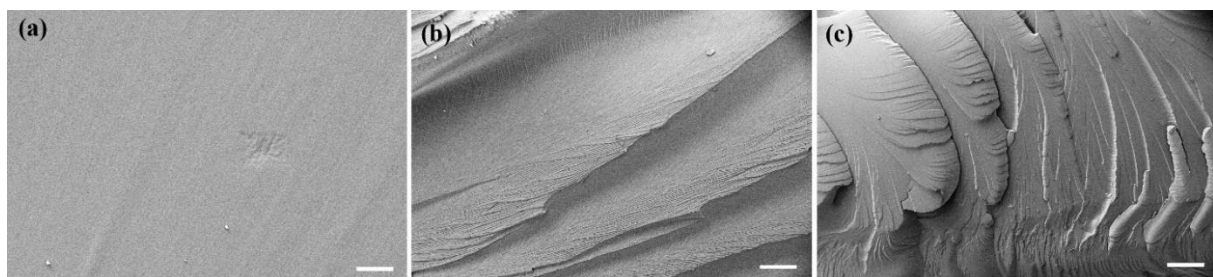


Fig. 8. Fracture surface of different specimens: EC0 (a), EC6 (b), and EC3. Scale bars in the image are 10 μm .

Its smooth surface suggests that the fracture occurred along a single direction, which is consistent with the direction of applied stress, implying that the neat epoxy resin had low resistance to crack propagation [30–32]. Loading nanofillers into the epoxy matrix resulted in roughened fracture surfaces as shown in Figs. 8 (b) and (c). When microcracks encounter nanostructured fillers, the cracks must bypass them and seek alternative paths, consequently creating ductile fracture zones, which consume more energy in the fracture process. Therefore, the nanocomposites failed in a more ductile manner and exhibited plastic deformation before fracture. Furthermore, a rougher surface with more crack deviation can be distinguished in the microstructure of EC3 than EC6. The more crack deviation and rougher surface are likely due to the presence of the hybrid nanofiller (CuO–GO) in the composite epoxy, which can affect the fracture behavior of the material, resulting in the epoxy-based nanocomposite having higher resistance to crack propagation compared to the neat epoxy [32, 33]. Therefore, the nanocomposite containing both graphene nanosheets and oxide nanoparticles (EC3) experienced more crack deviation, while the nanocomposite containing only CuO nanoparticles (EC6) experienced less crack deviation, demonstrating the significant influence of graphene nanosheets on the failure mechanism. This was also confirmed by the higher tensile strength of the EC3 specimen than the EC6 specimen, as mentioned above.

4. CONCLUSIONS

In this research, copper oxide–graphene oxide hybrid powder was prepared using the ultrasonic treatment. The effects of this hybrid on the mechanical properties of epoxy resin-based composites were investigated. The various characterizations of the hybrid powder and epoxy resin-based nanocomposites were conducted using different analytical instruments. The X-ray diffraction spectroscopy confirmed the formation of the copper oxide–graphene hybrid structure. The FTIR spectroscopy revealed the presence of the bond between copper and oxygen, while due to the surface-sensitive nature of this type of spectroscopy and the high amount of copper oxide compared to graphene, the presence of graphene in the hybrid sample spectrum was not

detectable. Therefore, we relied on scanning electron microscopy images and observed that the copper oxide particles were well seated on the graphene sheets. The mechanical tensile test proved the effect of the copper oxide–graphene hybrid on the mechanical properties of epoxy resin. A comparison between the mechanical properties obtained from the testing of the copper oxide–graphene hybrid composite sample and the copper oxide composite sample demonstrated the positive effect of graphene on the composite's mechanical properties due to its excellent mechanical properties.

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