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Research article

Improving the mechanical properties of fiber-reinforced polymer composites through nanocellulose-modified epoxy matrix

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Abstract. In this study, the potential use of nanocrystalline cellulose (CNC) modified epoxy nanocomposite as a matrix is investigated for both glass and carbon fiber-reinforced composites. Various amounts of CNCs (1, 2, 4, and 6 wt%) were added to bisphenol A diglycidyl ether-based epoxy resin (DGEBA), and the optimum CNC loading was determined as 4 wt% in terms of mechanical and thermal properties. Compared to the reference sample containing a neat epoxy matrix with the obtained carbon fiber/CNC-epoxy (CNC/epoxy/CF) and glass fiber/CNC-epoxy (CNC/epoxy/GF) hybrid nanocomposites, significant improvements have been determined in the in-plane shear modulus and strength, and flexural modulus, respectively. The mechanical properties improvements of CNC/epoxy/CF hybrid composites are approximately 0.9% higher than the CNC/epoxy/GF hybrid composites. Additionally, the distribution of CNC in hybrid nanocomposites is also investigated by scanning and transmission electron microscopies. It is noted that the homogenous dispersion of CNCs in the epoxy matrix and their diameters varied from 10 to 100 nm are detected at higher magnification.

Keywords: *bioadditive, carbon fiber, glass fiber, nanocellulose, hybrid composites*

1. Introduction

Recently, composite materials have been utilized in various applications because of their extraordinary properties that cannot be achieved by single materials, arising from the merge of the characteristics of different components [1]. Among the various composite materials, fiber-reinforced polymer (FRP) composites appear to be the most used materials. Basically, FRP composites consist of fibers (either synthetic or natural) reinforced in a polymer matrix [2]. Fibers of carbon, glass, basalt, aramid, sisal, and kenaf have been used as reinforcing elements in the polymer matrix structure of composite materials. Similarly, different polymeric resins such as poly(dicyclopentadiene), polyimide, polyester, and epoxy have been employed as matrices in FRPs. Predictably, the FRP composite properties depend on both the fiber and the matrix. Moreover, the fiber/matrix interface behavior can also play a key role in dictating the overall performance of composites [3]. Usually, this behavior has been improved by enriching the chemical interaction between the fiber and matrix or by increasing the interfacial area to facilitate the load transfer to the fiber. For instance, Wang *et al.* [4] functionalized carbon fiber surfaces with different functional groups, and Ren *et al.* [5] modified glass fiber surfaces to promote covalent bonding between fiber and matrix for composite materials with improved properties. Electrochemical oxidation or plasma treatment approaches have also been utilized for

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fiber surface treatment [6]. However, it is experimentally difficult to control structure parameters such as functionalization density, surface waviness, *etc*. Moreover, poor out-of-plane properties arising from the differences in orientation of fiber and matrix remain a drawback. Thus, new approaches to improve the composite properties appear as a challenge.

The recent progress in nanocomposites has led the way to hybrid composites with modified polymer matrices [7]. Thus, numerous nanoparticles were incorporated into the polymer matrices utilized in conventional FRP composites to elevate the mechanical, thermal, electrical, and barrier-related properties [8–10]. Such modifications elevate the polymer matrix properties, resulting in relative improvements in the structural properties of the composite. Moreover, nanoparticles [11–13] in the matrix also strengthen the interface interactions and, thus, more efficient load transfer between the epoxy matrix and fibers [14]. For instance, it has been shown that epoxy/clay nanocomposite (with 6 wt% of nanoclay) matrix reinforced with 10–12% glass fibers represents similar properties with 30% glass fibers reinforced neat epoxy matrix composite [15]. Another study showed that an important improvement of around 74% in the fatigue life of carbon fiber-reinforced polymer composites can be achieved by using 3 wt% nanoclay in the polymer matrix [16]. Gojny *et al.* [17] reported that the incorporation of only 0.1 wt% carbon nanotubes resulted in a 20% improvement in interlaminar shear strength for glass fiber/epoxy composites. Silica or rubbery nanoparticles and Cloisite 25A® have also been shown to be efficient nanofillers for various composites in different application-oriented systems. Although a great number of compounds have been tested, interestingly, there has not been much focus on the utilization of nanocellulose in epoxy matrix for fiber-reinforced polymer composites.

Cellulose, discovered and isolated by Payen in 1838, is the most richly found natural polymer on Earth [18]. It is one of the most crucial structural components in plants, forming the wall of the cell structure. Parallel to the increasing environmental awareness and demand for eco-friendly materials, cellulose-based materials received great interest. Cellulosic nanoparticles, also known as nanocellulose, extracted from native cellulose are particularly important [19]. Based on their inherent properties, including sustainability, biodegradability, strength, natural abundance, and low cost, nanocellulose-based materials have been the subject of various applications. In addition to bio-applications such as drug delivery, medical implants, and wound dressing, they also took place in sensor and packaging applications [20, 21]. There are several comprehensive reviews summarizing the nanocellulose-reinforced polymer nanocomposites and their applications [22]. However, as can be seen from those reviews, the general trend is the usage of nanocellulose with polymers to form cellulosic bio-nanocomposites or to utilize them for replacing conventional fibers, such as glass or carbon fibers in reinforced composites. Yet, there has not been any research for the improvement of fiber composites via cellulosic epoxy nanocomposites as matrix [23]. Herein, we report the utilization of CNC-modified epoxy nanocomposite as a matrix for both glass and carbon fiber-reinforced composites. The obtained hybrid composites possessing CNC nanoparticles represented improved mechanical properties compared with the reference composites containing neat epoxy matrix.

2. Experimental

2.1. Materials

Nanocrystalline cellulose (CNC) (10–20 nm in diameter and 300–900 nm in length, dry white powder at room temperature) was purchased from Nanografi Nanotechnology Co. Ltd. (Ankara, Turkey) and used as received. The epoxy resin consisted of two components, epoxy oligomer (A) based on diglycidyl ether bisphenol A based epoxy resin and amine-based curing agent (B), mixing ratio is A:B = 100:26 by weight and was supplied from Gurit, Isle of Wight, United Kingdom as a trademark of Prime™ 27. Its gelling and curing time at room temperature are 1 and 24 h, respectively. The multiaxial carbon and glass fabrics as non-crimp forms were supplied from Metyx Composites, Istanbul, Turkey as a gift. They have longitudinal (0°) and transverse (90°) fiber orientations and their areal density was 300 grams per square meter [gsm].

2.2. Optimization of CNC loading into the epoxy systems

A typical procedure for the preparation of CNC/epoxy nanocomposites was as follows. First, a series of epoxy resins containing various amounts of CNC (0.5, 1, 2, 4, and 6 wt%) were mixed on a magnetic stirrer (250 rpm) at 50 °C for 30 min. Subsequently, these mixtures were sonicated in an ultrasonic bath

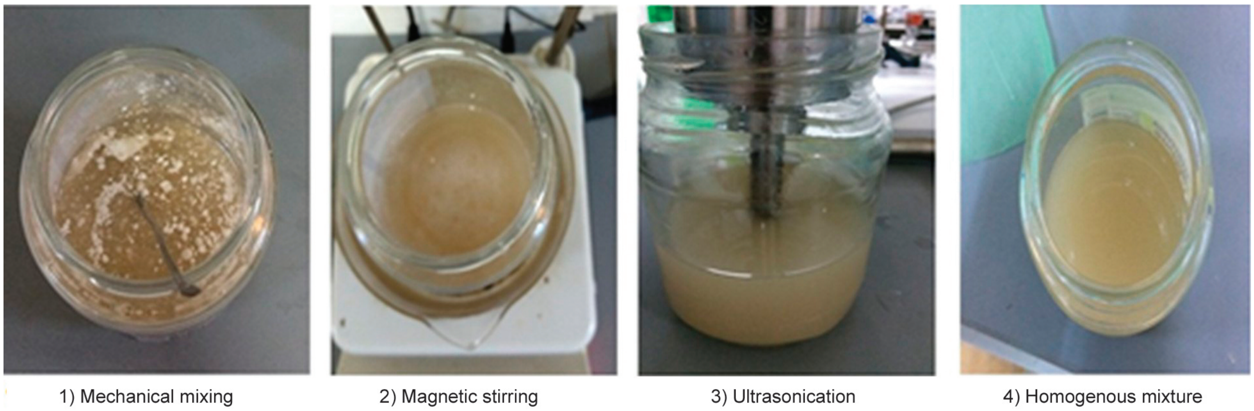

Figure 1. Dispersion of CNC in the epoxy resin.

for 1 min to promote homogenization (Figure 1). Then, the required quantity of the amine-based curing agent (B) was included in all mixtures that were degassed in a vacuum for a short time to remove the air bubbles. Finally, the mixtures were cast into silicon and left under vacuum for molds according to the ISO 178, 180, and 527-2 test standards for corresponding flexural, Izod impact, and tensile tests.

2.3. Preparation of carbon- and glass-fibers/CNC-epoxy hybrid composites

The carbon fiber/CNC-epoxy and glass fiber/CNC-epoxy hybrid nanocomposites were fabricated by vacuum bagging technique. In the first step, a release agent was applied to the surface of a flat polished steel plate to dismiss the bonding between the plate and resin material. Secondly, non-crimp multiaxial glass or carbon fabric layers were placed and epoxy resin containing 4 wt% of CNC was applied to each layer of the fibers using a roller and a scraper to facilitate the impregnation of the fibers with matrix as well as to compact the fabric layers together. After, a peel ply and a porous release film were placed over the surface of the impregnated fabrics before resin infusion. These peel ply and release films were not integrated into the composite laminates, they allowed for easy separation of the fiber-reinforced composite from the mold after curing of matrix. Subsequently, a non-woven breather fabric was laid, and it was used to distribute the vacuum pressure over the entire plate and to absorb excess matrix resin drawn out of the laminate via vacuum. Finally, the whole system was isolated with a vacuum bag and sealed with an elastomeric double-sided sealant tape. Then, a vacuum pump was connected to the vacuum bag through a vacuum port. The –760 mmHg pressure was applied to remove air bubbles and excess matrix resins, and a leak check was performed. After 24 h, the vacuum was turned off, and the composite test panels were removed from the mold. At the end of this process, a total of four composite test panels were produced (carbon fiber/CNC-epoxy, carbon fiber/epoxy, glass fiber/CNC-epoxy, and glass fiber/epoxy). The control samples (glass fiber and carbon fiber reinforced epoxy composites) were fabricated under identical conditions, replacing the matrix used from CNC-epoxy nanocomposite with neat epoxy.

2.4. Characterizations

The thermal and mechanical properties of neat epoxy, both CNC/epoxy nanocomposite and fiber-reinforced CNC/epoxy hybrid composite samples were investigated via appropriate characterization methods. Thus, in-plane shear, tensile, and flexural tests were performed by Zwick/Roell Z020 (ZwickRoell GmbH & Co, Ulm, Germany) electromechanical test machine, and Izod impact test was performed by INSTRON Ceast 9050 (Instron, Norwood, MA, USA) at ambient temperature for the mechanical properties. The differential scanning calorimetry (DSC) using Seiko Exstar DSC 7020 (Seiko, Tokyo, Japan) with a heating rate of 10 °C/min under nitrogen flow was performed to examine the thermal properties. About 3–5 mg samples were operated in DSC measurements by following three steps: heating from 30 to 180 °C and keeping for 5 min to delete the effect of thermal and mechanical histories, then cooling to 30 °C and holding for an additional 5 min. At the end, the samples were heated to 180 °C. In addition, the thermal stability of neat epoxy resin and epoxy/CNC nanocomposites were tested through thermogravimetric analysis (TGA) via Seiko Exstar TG/DTA 6300 (Seiko, Tokyo, Japan) with a heating mode from 30 to 800 °C at a heating rate of 10 °C/min under nitrogen atmosphere. The SEM imaging was carried out by Quanta FEG 250 (Thermo Fisher Scientific, Hillsboro, OR, USA) instrument operating at an acceleration voltage of 15 kV. The surfaces of the samples were coated with gold to make the surface conductive before the SEM imaging. The TEM analysis was performed with the instrument Joel JEM-2100 (JEOL GmbH, Eching, Germany) at 300 kV. Thin film samples were prepared by cutting the cross-linked composite using an ultra-microtome and then by putting it into a suitable grid.

3. Results and discussion

Epoxy resins have been widely used as matrix material in fiber-reinforced composites based on their good chemical/temperature resistance, low creep properties, and ease of processing. However, their average mechanical properties limit further utilization in high-tech engineering applications. These properties can be improved by the introduction of nanofillers. Yet, there exists an optimum loading capacity for the nanoparticles, as overloading results in increased resin viscosity and air bubbles during the mixing process. Thus, with our aim to utilize epoxy-nanocrystalline cellulose composites as a matrix, we first determined the optimum CNC loading ratio. Accordingly, different amounts (0.5, 1, 2, 4, and 6 wt%) of CNCs were added to bisphenol A diglycidyl ether-based epoxy resin (DGEBA). The obtained nanocomposites were then tested in terms of mechanical and thermal properties.

3.1. Optimization of CNC loading

The tensile, flexural, and Izod impact test results of the epoxy-CNC nanocomposites with various CNC content were compared with the control sample containing a neat epoxy matrix and summarized in Table 1. The tensile strength and modulus of Epoxy/CNC-0% as the reference sample were determined as 55.3 MPa and 2.40 GPa, respectively. The addition of CNC with 0.5, 1, 2, and 4 wt% in the epoxy matrix remarkably increased the tensile strength (up to 18.9%) and modulus (up to 5.4%) of the nanocomposites compared to the reference sample. However, the higher CNC loading (6 wt%) resulted in decreases in both tensile strength and modulus of the nanocomposite. The flexural test results of the epoxy/CNC nanocomposites were also compared with the reference sample in Table 1. A similar trend continued, and the maximum improvement for the flexural strength and modulus was observed at 4 wt% CNC loads with an increase of 19.4 and 30.0% compared to the control epoxy/CNC-0% sample. At 6 wt% CNC loading, decreases around 4.0 and 7.7% were detected in both flexural strength and modulus, respectively.

The effect of CNC content in DGEBA-based epoxy resin on the Izod impact strength (unnotched) was also shown in Table 1. This effect could be attributed to the strong secondary interactions and the possibility of chemical bonding. According to the Izod impact test, 5.2, 7.4, 42.3, 61.0, and 44.0% improvements with the respective loadings of 0.5, 1, 2, 4, and 6 wt% CNC where the highest value was reached at the 4 wt% CNC loads. Consequently, the 4 wt% appeared to be the ideal CNC loading value for improved mechanical properties between the functional groups on the CNC and epoxy matrix. However, in the case of CNC loading over 4%, the nanofiller-nanofiller interactions were more dominant than nanofiller-polymer matrix interactions and formed agglomerated structures as heterogeneous regions in the epoxy matrix. These macro-/micro-sized heterogenous CNC regions appeared as weak points and prevented the stress transfer [24].

Table 1. The effect of CNC on the tensile and flexural properties of the nanocomposites.

Sample name	Tensile strength[a] [MPa]	Tensile modulus [GPag	Flexural strength[b] [MPa]	Flexural modulus [GPa]	Izod impact strength [kJ/m^2]
Epoxy/CNC-0%	55.38±0.90	2.40±0.10	95.00±1.81	2.60±0.07	10.20±0.86
Epoxy/CNC-0.5%	58.75±1.83	2.42±0.05	100.27±3.86	2.90±0.12	10.73±0.84
Epoxy/CNC-1%	59.78±1.59	2.44±0.05	104.83±2.65	3.08±0.11	11.17±0.49
Epoxy/CNC-2%	63.32±1.86	2.52±0.15	106.45±2.31	3.19±0.06	12.51±0.32
Epoxy/CNC-4%	65.83±2.33	2.63±0.07	113.41±1.79	3.38±0.05	14.80±0.34
Epoxy/CNC-6%	58.97±1.38	2.50±0.08	108.92±2.88	3.12±0.21	13.19±0.45

[a]The tensile specimens were loaded with a constant speed of 5 mm/min until breaking.
[b]The flexural samples were loaded with a constant speed of 2 mm/min until breaking.

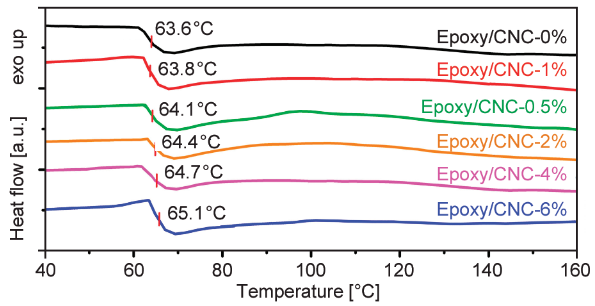

Figure 2. DSC curves of the neat epoxy resin and epoxy/CNC nanocomposites containing 0.5, 1, 2, 4, and 6 wt% CNC.

The influence of CNC loading on the glass transition temperature (T_g) of epoxy/CNC nanocomposite samples was investigated by DSC and compared with T_g of neat epoxy resin (Figure 2). By addition of 0.5 wt% amount of CNC nanoparticle, the T_g of neat epoxy resin was slightly improved from 63.6 to 63.8 °C. The inclusion of CNC could hinder the molecular motion of polymeric chains, which were responsible for an increased T_g of CNC/epoxy nanocomposites [25–28]. At higher CNC loading, the T_gs of epoxy/CNC-1%, epoxy/CNC-2%, epoxy/CNC-4%, and epoxy/CNC-6% samples were detected as 64.1, 64.4, 64.7, and 65.1 °C, respectively.

To further investigate the thermal stability of obtained epoxy/CNC nanocomposites, the thermogravimetric analysis was performed under an inert atmosphere with a heating rate of 10 °C/min from 30 to 800 °C and compared with the neat epoxy resin data. The TGA thermogram of neat epoxy resin displayed a single degradation step between 310 and 530 °C due to the oxidative degradation of carbon residue in the presence of oxygen (Figure 3). In addition, all epoxy/CNC nanocomposites displayed similar degradation behavior but with slightly higher char yields compared to the neat epoxy resin. This could be explained by the exchange of epoxy monomer with CNC that contains a more thermally stable crystalline [29].

Once the ideal CNC loading content was determined as 4 wt%, the epoxy-CNC nanocomposite with this content was adapted as a matrix to both glass and carbon fiber-reinforced composites. Mechanical properties of the composites, synthesized via vacuum-bagging technique as specified in the experimental part, were examined by in-plane shear, flexural, and Izod impact strength tests. Results were compared with the data of the control sample that did not contain CNC nanofiller.

3.2. In-plane shear test results

Comparative graphs representing the shear strength and modulus values of in-plane tests of carbon and glass fiber-reinforced hybrid composites are given in Table 2. According to the results, improvements of 13.07% in shear modulus and 10.81% in shear strength were achieved in the CNC/epoxy/CF hybrid composite compared to the control sample (Figure 4). While 24.27% in shear modulus and 10.36% in shear strength improvements were observed for CNC/epoxy/GF hybrid composite (Figure 4).

3.3. Flexural test results

The flexural strength and modulus results were collected by considering the fiber orientation (0°, 90° and ±45°) of the CNC/epoxy/CF and CNC/epoxy/GF hybrid composites are shown in Figure 5. Improved

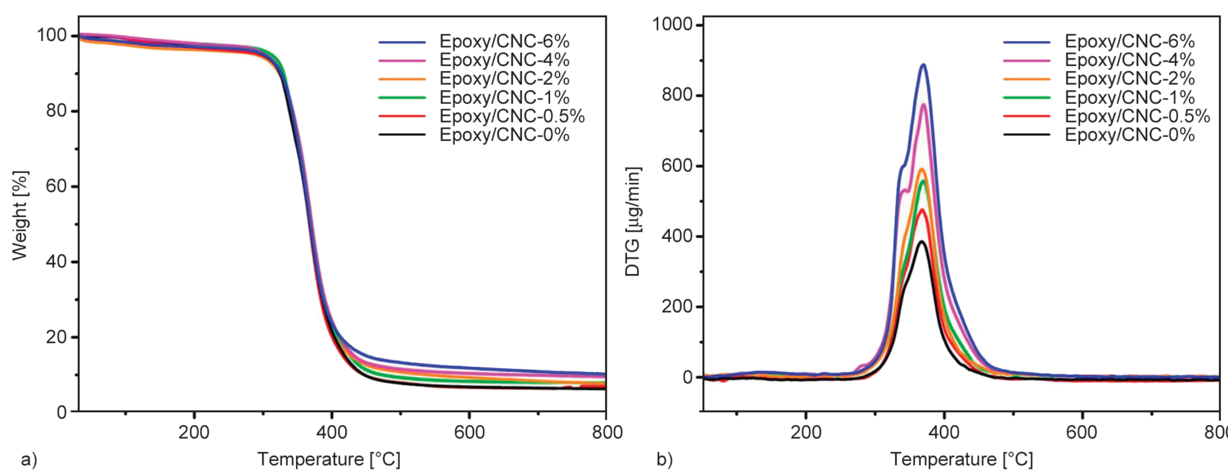

Figure 3. a) TGA and b) DTG curves of neat epoxy resin and epoxy/CNC nanocomposites containing 0.5, 1, 2, 4, and 6 wt% CNC.

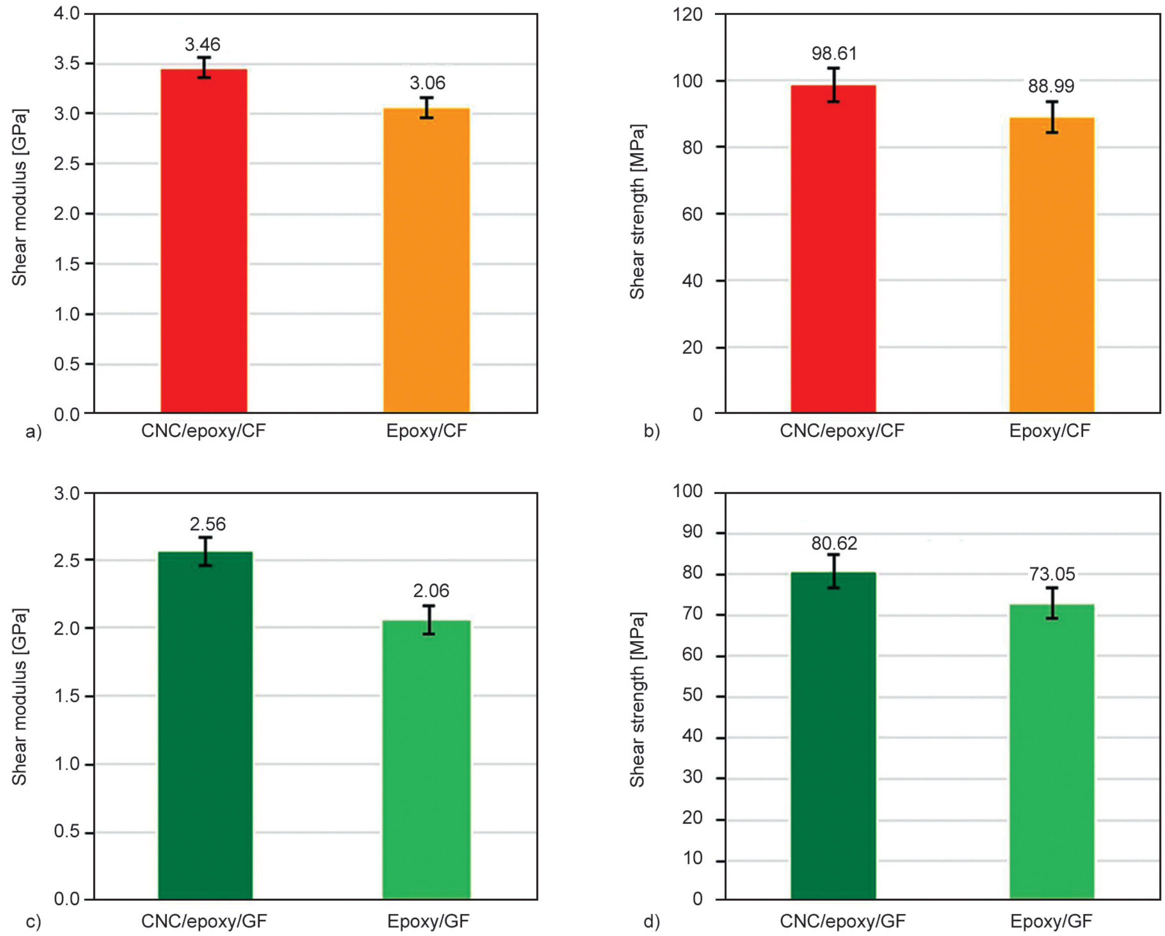

Figure 4. In-plane shear test results of the neat epoxy/CF and epoxy/GF composites with CNC/epoxy/CF (a and b) and CNC/epoxy/GF (c and d) hybrid composites, where the fiber orientation in the tests was ±45°.

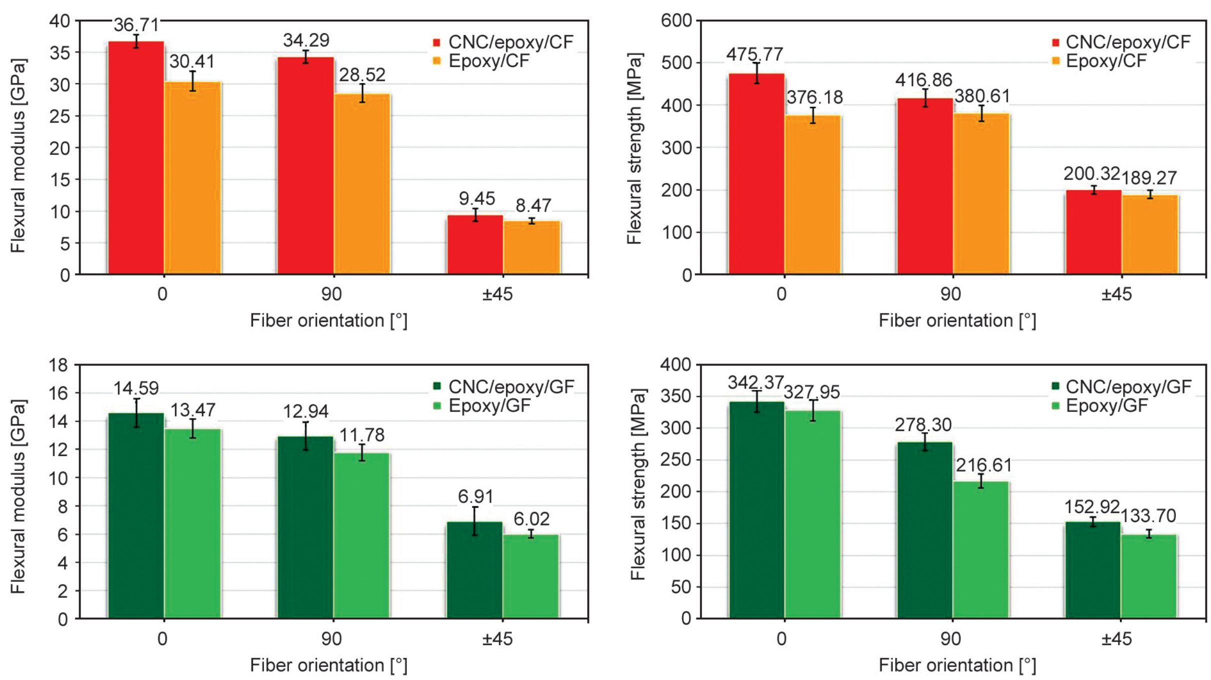

Figure 5. Flexural test results of the neat epoxy/CF and epoxy/GF composites with CNC/epoxy/CF (a and b) and CNC/epoxy/GF (c and d) hybrid composites.

flexural properties were observed in all three carbon fiber orientations. The positive effect of CNC nanofiller on the flexural modulus varied between 5.84 and 26.47%, while flexural strength was elevated between 11.51 and 20.71. As can be seen from Table 2, similar improved characteristics were observed also for glass fiber/CNC-epoxy hybrid composites. Enriched flexural properties were noticed for CNC/epoxy/GF hybrid composites in all glass fiber orientations.

Unnotched Izod impact strength tests were applied in two different directions (perpendicular or parallel) for both carbon and glass fiber reinforced, considering the fiber orientations as stated in the standard (ISO 180). Figure 6 summarizes the parallel Izod impact strength test results of the CNC/epoxy/GF hybrid composites with all three carbon glass fiber orientations. Obvious improvements were observed in all samples. As can be seen from Figure 6, developments have been attended also in perpendicular Izod impact strength tests in all fiber orientations. The CNC/epoxy/GF hybrid composites represented even better improvements compared to their carbon fiber analogs in parallel Izod impact strength tests. The positive effect of CNC nanofiller was varying from 8.53 to 23.08% (Table 2). While 4.80 and 12.65% increase have been monitored in perpendicular tests (Table 2).

In summary, all the achieved mechanical tests showed that the inclusion of CNC nanoparticles in the epoxy matrix improves all mechanical properties, including in-plane shear modulus and strength, flexural modulus and strength, and Izod impact strength of both glass and carbon fiber reinforced hybrid composites. In the cases of in-plane shear modulus, the glass and carbon fiber-reinforced hybrid composites increased 1.13 and 1.24 fold compared to neat composites. Similar mechanical results contributed to the improvement of in-plane shear strength, flexural modulus and strength, and Izod impact strength were also determined. The uniform dispersion of CNC nanoparticles in the epoxy matrix enabled the strengthening the fiber-matrix interface. These strong interactions provided more effective load transfer from the matrix to the fibers by acting as a bridge. Because of improved interfacial strength, characteristic deformations such as debonding and pull-out of fibers from the matrix, fiber fracture, and matrix breakage are able to withstand higher mechanical loads.

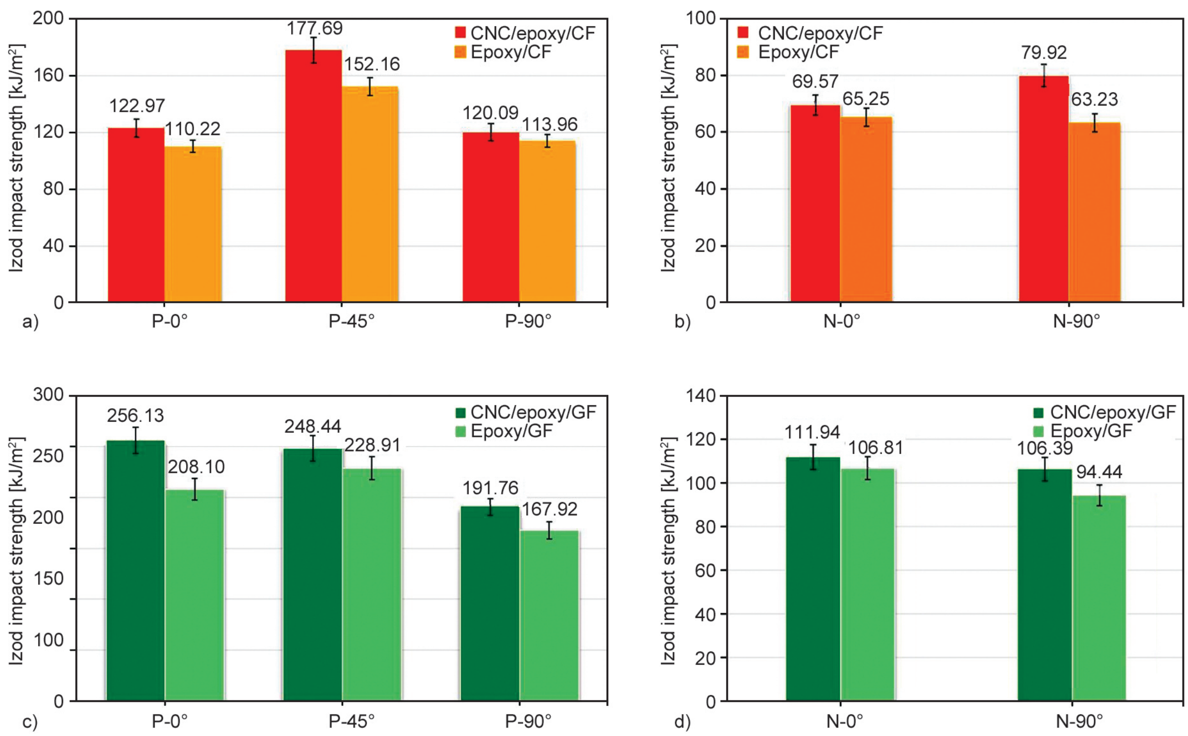

Figure 6. Izod impact test results of the neat epoxy/CF and epoxy/GF composites with CNC/epoxy/CF (a and b) and CNC/epoxy/GF (c and d) hybrid composites (P: direction of blow parallel to the plane of reinforcement, N: direction of blow normal to the plane of reinforcement).

Table 2. Effect of CNC nanofiller on the mechanical properties of glass and carbon fiber-reinforced hybrid composites.

Properties		Epoxy/CF	CNC/epoxy/CF	Effect [%]	Epoxy/GF	CNC/epoxy/GF	Effect [%]
In-plane shear modulus[a]	[GPa]	3.06	3.46	13.07	2.06	2.56	24.27
In-plane shear strength	[MPa]	88.99	98.61	10.81	73.05	80.62	10.36
Flexural modulus – 0°[b]	[GPa]	30.41	36.71	20.71	13.47	14.59	8.31
Flexural modulus – 90°	[GPa]	28.52	34.29	20.20	11.78	12.94	9.85
Flexural modulus – 45°	[GPa]	8.47	9.45	11.51	6.01	6.91	14.98
Flexural strength – 0°	[MPa]	376.18	475.77	26.47	327.95	342.37	4.40
Flexural strength – 90°	[MPa]	380.61	416.86	9.52	222.03	278.3	25.34
Flexural strength – 45°	[MPa]	189.27	200.32	5.84	133.7	152.92	14.38
Izod impact strength-P – 0°	[kJ/m^2]	110.22	122.97	12.72	208.1	256.13	23.08
Izod impact strength-P – 90°	[kJ/m^2]	152.16	177.69	16.77	228.91	248.44	8.53
Izod impact strength-P – 45°	[kJ/m^2]	113.98	120.09	5.36	167.92	191.76	14.2
Izod impact strength-N – 0°	[kJ/m^2]	65.25	69.57	6.65	106.81	111.94	4.80
Izod impact strength-N – 90°	[kJ/m^2]	63.23	79.92	26.39	94.44	106.39	12.65

[a]The in-plane shear specimens were loaded with a constant speed of 2 mm/min until breaking.
[b]The flexural samples were loaded with a constant speed of 2 mm/min until breaking.

3.4. Thermal behavior

The influence of CNC loading on the T_gs of carbon and glass-fiber reinforced hybrid composites, along with T_g of the epoxy/CNC-4% sample that contains optimum CNC content, were investigated by DSC (Figure 7). Compared to the epoxy/CNC-4% sample, the epoxy/CNC/CF and epoxy/CNC/GF hybrid composites have slightly higher T_gs, which were recorded as 65.1 and 65.2 °C. The inclusion of fibers has restricted the movement of polymer chains that create the interfacial adhesion between fibers and matrix [30, 31]. The good fiber-matrix interaction contributed to not only the enhancement of mechanical properties but also the improvement of the thermal properties of carbon and glass-fiber reinforced hybrid composites.

3.5. Microstructural examination

The SEM analysis was used to identify the fiber-matrix interface properties of the composites by scanning the damaged sample surfaces after the in-plane shear tests (fiber orientation in the tests was ±45°) were examined. The SEM micrographs of neat epoxy/CF composite CNC/epoxy/CF hybrid composite at a magnification of two thousand times, and epoxy/GF composite and CNC/epoxy/GF hybrid composite at a magnification of one thousand times were given as Figure 8, respectively. It was found that the epoxy resins adhered to both carbon and glass fiber surfaces and the load transfers were carried out efficiently between the fibers and epoxy matrix. In both cases, the fiber pullout phenomenon was not apparently observed, whereas fiber breakage was the main damage mode in specimens. On the other hand, the presence of CNC in both epoxy/CF and epoxy/GF hybrid composites caused an increase in friction for the fibers to come out.

To get more detail about the distribution of CNC nanoparticles, the TEM analysis was also utilized for the CNC/epoxy/CF (Figure 9a) and CNC/epoxy/GF (Figure 9b) hybrid composite samples. Based on the TEM images, CNC nanoparticles appeared as dark spherical points in both high and low magnifications. Notably, these points exhibited good dispersion in the epoxy matrix and their diameters varied from 10 to 100 nm, which were clearly visible at higher magnification (Figure 9). Consequently, both SEM and TEM investigations confirmed the existence and random distribution of CNC nanoparticles in the epoxy matrix.

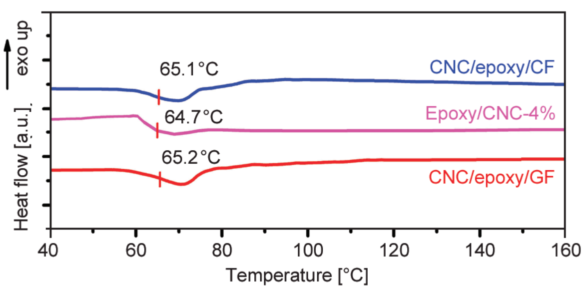

Figure 7. DSC curves of the Epoxy/CNC-4%, and CNC/epoxy/CF and CNC/epoxy/GF hybrid composites.

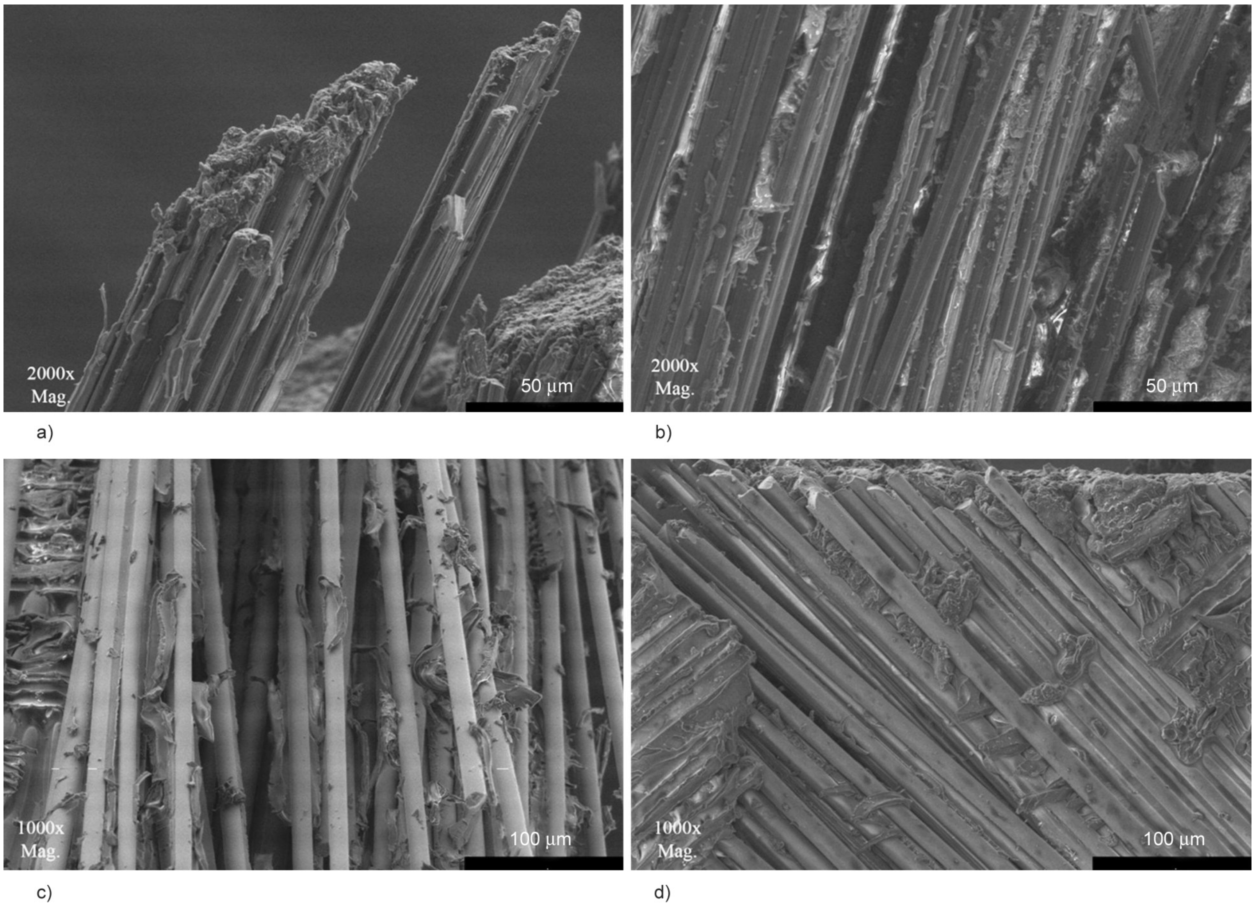

Figure 8. SEM images of neat epoxy/CF composite (a) CNC/epoxy/CF hybrid composite (b), epoxy/GF composite (c) and CNC/epoxy/GF hybrid composite (d).

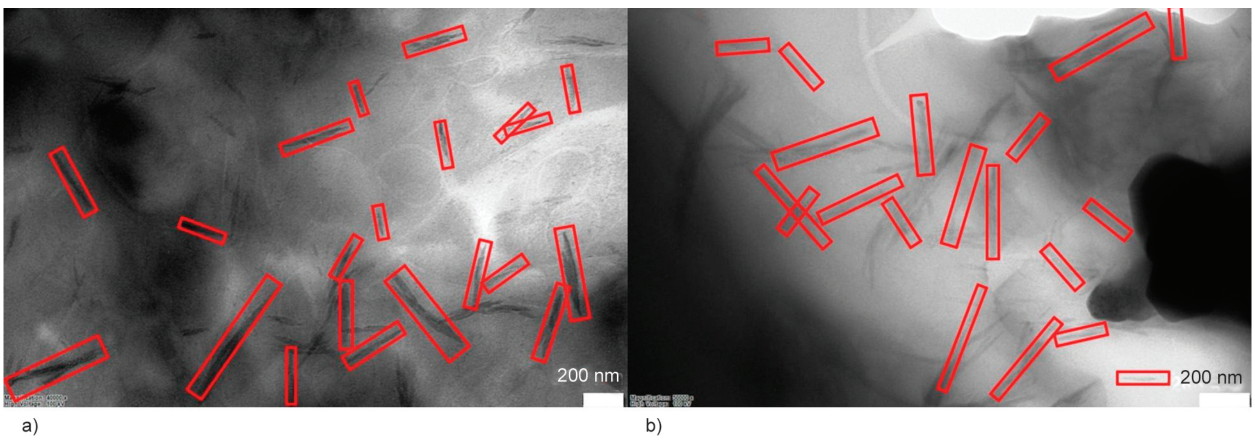

Figure 9. TEM images of CNC/epoxy/CF (a) and CNC/epoxy/GF (b) hybrid composites.

4. Conclusions

In conclusion, the CNC nanoparticles have been successfully mixed with DGEBA-based epoxy resin and utilized as the matrix for glass and carbon fiber fabrics by vacuum bagging technique. Firstly, optimal CNC content was determined with the respective loadings of 0.5, 1, 2, 4, and 6 wt% CNC and it was found to be 4 wt% based on the Izod impact test. The obtained CNC/epoxy/CF and CNC/epoxy/GF hybrid composites demonstrated significant improvements (between 5.36 and 27.18% for CNC/epoxy/CF, and between 4.80 and 25.34% for CNC/epoxy/GF) in the in-plane shear modulus and strength, flexural modulus and strength, and Izod impact strength, respectively, compared to the control composite containing neat epoxy matrix. Furthermore, the glass transition temperatures of CNC/epoxy/CF and CNC/epoxy/GF hybrid composites were recorded higher than the epoxy/CNC-4% sample containing optimum CNC content. The mechanical improvements in CNC/epoxy/CF hybrid composites approximately 0.9% are higher than the CNC/epoxy/GF hybrid composites. Moreover, as the SEM and TEM images confirmed the homogenous distribution of CNC nanoparticles adhering to the entire surface of the fibers in the hybrid composites. The uniform dispersion of the CNC nanoparticles in the epoxy matrix enabled the formation of strong interactions between matrix and fiber providing more efficient load transfer along with enhanced mechanical performance of hybrid composites. Consequently, this work displays a facile modification of epoxy matrix system using CNC nanoparticles as a green additive that allows the preparation of hybrid composites with superior mechanical properties. Considering the abundant availability, low price, biodegradability and environmental harmlessness of CNC, the current study is potentially attractive to develop a library of nanostructured hybrid materials with high-tech applications for a clean future.

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