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PRODUCTION AND CHARACTERIZATION OF NANOCOMPOSITES WITH GRAPHENE AND GRAPHENE OXIDE

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Abstract: The aim of this study was to i.) obtain Graphene (G)/epoxy, Graphene Oxide (GO)/epoxy, G/carbon fiber reinforced plastic (CFRP), GO/CFRP nanocomposites by G and GO addition to epoxy and CF matrices and ii.) investigate the effect of G and GO addition on tensile strength of the nanocomposites produced. The graphene was not homogeneously distributed in polymer matrices; therefore, the G was converted into the GO using hummers method. The characterization GO was carried out by SEM, EDX, FTIR and X-RD analysis, which confirmed the successful production of the GO. The G/epoxy, GO/epoxy, G/CFRP and GO/CFRP nanocomposites were obtained by adding G and GO to epoxy and CF at a rate of 0.1, 0.2, and 0.3%. The effect of G and GO on tensile strength of the nanocomposites was determined using a tensile test. The addition of G and GO at 0.2% rate yielded the best results. The effect of GO on tensile strength was better compared to that of the G.

Keywords: Hummers Method, Graphene, Graphene Oxide, CFRP, Nanocomposite

Nanokompozitlerin Grafen ve Grafen Oksit ile Üretimi ve Karakterizasyonunun İncelenmesi

Öz: Bu çalışmanın amacı, epoksi ve CF matrislerine G ve GO ekleyerek G/epoksi, GO/epoksi, G/CFRP, GO/CFRP nanokompozitlerini elde etmek ve bu nanokompozitlerin çekme mukavemeti üzerine G ve GO'nun etkisini araştırmaktır. Bu amaçla, polimer matrislerde G homojen olarak dağılmadığından, G hummers yöntemiyle GO'ya dönüştürülmüş ve elde edilen GO'nun karakterizasyon analizi yapılmıştır. SEM, EDX, FTIR, X-RD analiz sonuçları, GO'nun başarıyla elde edildiğini göstermiştir. Daha sonra epoksi ve CFRP'ye %0,1, %0,2 ve %0,3 oranında G ve GO eklenmiş ve G/epoksi, GO/epoksi, G/CFRP, GO/CFRP nanokompozitleri elde edilmiştir. G ve GO'nun bu nanokompozitlerin çekme mukavemeti üzerindeki etkisini karşılaştırmak için çekme testi uygulanmıştır. En iyi sonuç %0,2 oranında G ve GO ilavesi ile elde edilmiş ve GO'nun çekme mukavemeti üzerindeki etkisinin G'ye göre daha iyi olduğu gözlemlenmiştir.

Anahtar kelimeler: Hummers Metodu, Grafen, Grafen Oksit, CFRP, Nanokompozit

1. INTRODUCTION

Graphene (G), carbon allotrope discovered in 2004 by Novoselov in the University of Menchester, England (Pan et al., 2012). Graphene was known only as a single layer carbon allotrope at the beginning of its discovery, but later, the multi-layered structure of graphene was identified.

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Multi-layered graphene's attract the attentions (Goncalves et al., 2012) due to the superior mechanical, thermal and conductivity properties such as specific surface area (2630 m^2 g^{-1}), high internal mobility (200000 cm^2 v^{-1} s^{-1}), high young module (~ 1.0 TPa), thermal conductivity (~ 5000 $Wm^{-1}K^{-1}$) and optical transparency (~ 97.7%) (Li et al., 2013; Zhu et al., 2010). The graphene is the most durable material of the world with 130 GPa tensile strength and 1 TPa young module, and is the thinnest material of chemistry, biotechnology and material science (Kuila et al., 2012; Kausar et al., 2016). The graphene's are widely used in various sectors such as space, aerospace, automotive and paper industries, fuel cells, solar and lithium-ion batteries, super capacitors, electronic devices and biosensors (Ibrahim et al., 2021; Sainbury et al., 2017; Deshmukh et al., 2014).

The graphene is easily produced with low cost (Mahmoud, 2011). Several methods have been used in the production of graphene while from top to bottom and the bottom-up production methods were recommended. The bottom-up method includes epitaxial growth, chemical vapor deposition (CVD) and micromechanical exfoliation. The from top to bottom method is more effective and more nanographene plates can be produced with the exfoliation (Shah et al., 2015). The most promising application of the nanomaterials is the production of polymer nanocomposite materials (Kernin et al., 2019) such as epoxy, polystyrene, polypropylene, polyethylene terephthalate, polyaniline, nylon and polymethyl methacrylate (Tang et al., 2013). In situ polymerization, melt blending and solution mixing methods are employed by adding graphene or other nanoparticles to produce the nanocomposites.

The melt blending method is preferred in the production of nanocomposites, because the method does not require solvents and is environmentally friendly and low-cost (Kim et al., 2011). The graphene rate used in polymer matrix is very low (wt. 0.05%), while polymer nanocomposites are lighter, more robust and have superior mechanical, thermal and electrical properties than the polymer composites (Kernin et al., 2019). The characteristics of polymer nanocomposites depend on homogeneous distribution of the nanoparticles in a polymer matrix, and the strength of the bond formed in the interface of the polymer matrix (Park et al., 2016). However, the graphene has a high propensity to agglomerate due to the hydrophobic nature, high specific surface area, Van der Waals bond and π-π^* transitions (Kausar et al., 2016). When the graphene is agglomerated, a homogeneous distribution cannot be obtained in the polymer matrix, and the properties of the material obtained may not be improved as desired. Therefore, the graphene should be activated to provide homogeneous distribution in the polymer matrix. The graphene is converted to graphene oxide (GO), which has a larger a specific surface area, to obtain an active material. The GO is a layered material produced by oxidation of graphite and has two-dimensions (Li et al., 2018; Shah et al., 2015). The GO is produced mostly by oxidation from pure graphite using Brodie, Staudenmair and Hummer methods (Shah et al., 2015). More and low-cost GO can be obtained by the hummers method (Çelik et al., 2018). The layers in the GO contrary to pure graphite are oxidized intensively by binding the hydroxyl and epoxide functional groups to side of each layer, and the hydroxyl groups to top and bottom of the layer. The functional groups make the GO layers more hydrophilic, and allow to swell more and disperse in water (Li et al., 2018).

The GO reinforced polymer nanocomposites are used in energy storage, gas sensors, electronic devices and super capacitors (Wang et al., 2016). The polymer reinforced with the GO can homogeneously be dispersed in the matrix, and a stronger bond is formed in the intermediate layers. Large surface area and functional groups of the GO help dispersion of GO in the polymer matrix and formation of stronger covalent bonds with the matrix. The GO is readily soluble in organic solvents such as, acetone and tetrahydrofuran (THF) (Rastompour et al., 2016). However, the exact structure of the GO is still not clear and some models have been introduced to define the structure of GO. The structures were Hofmann's, Ruess's, Sholtz's & Boehm's and Lerf's & Klinowski's models (Shah et al., 2015; Çelik et al., 2018), which all developed based on the oxidation of graphite (Çelik et al., 2018).

Many studies have been conducted recently to investigate production of polymer nanocomposite by the synthesis of the GO. In this study, the graphene was converted to the GO. Characteristics of the GO produced were determined. In addition, the tensile strengths of the nanocomposites obtained by graphene and GO addition to the polymer at different ratios were measured. The G/CFRP and GO/CFRP nanocomposites were fabricated considering the reinforcement ratio of the highest tensile strength of the nanocomposite The effect of G and GO addition on CFRP composite was analysed.

The effect of polystyrene GO on thermal stability of reinforced nanocomposite was studied by Rostampour et al. (2016). The microstructure and mechanical properties of the nanocomposites produced by GO addition into the polymer were investigated by Zhang et al. (2020). The GO was used as a coating material by Wang et al. (2017) who also studied the effect of GO as a coating material on crack formation. Sridharan et al. (2016) investigated the effect of exfoliating graphite on the electrical conductivity of the nanocomposite. Mechanical and electrical properties of the nanocomposites produced by the addition of G have been determined by Sun et al. (2021) and Sofla et al. (2019). The availability of G/GO in different matrix nanocomposites was determined by Lalabadi et al. (2019). Deshmukh et al. (2014) studied the effect of GO addition on properties of polyvinylchloride (PVC) nanocomposites. The effect of different rates of GO reinforcement on nanocomposites was determined by Kang et al. (2017) and Colak et al. (2020). Önder et al (2012) investigated wet synthesis of chemically derived graphene using hydrazine hydrate reduction of the GO The researchers stated that the method favours the ex-situ synthesis of Pd nanoparticles with certain particle size and dispersion. The aim of this study was to investigate the effect of G and GO addition to epoxy and CF matrices on the tensile strength of G/epoxy, GO/epoxy, G/CFRP, GO/CFRP nanocomposites.

3. MATERIAL AND METHOD

3.1. Materials

The materials needed for GO synthesis by the modified hummers method are powdered graphene (<20µm), hydrogen peroxide (H_2O_2, 37.5%) and sulfuric acid (H_2SO_4, 95-97%) were obtained from Sigma Aldrich company. Sodium nitrate ($NaNO_3$) and potassium permanganate ($KMnO_4$) were obtained from Merck company. In addition, the CF fabric with a density of 300 gr m^{-2} was used to produce CFRP composites. The CF fabric used in the study is shown in Figure 1.

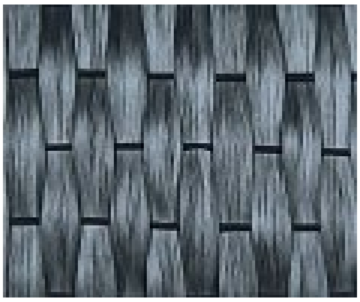


Figure 1.
Carbon fiber fabric

3.2. Synthesis of Graphene Oxide (GO)

In the first step of the synthesis, 5 g of powder graphene and 2.5 g of sodium nitrate were weighted at precision scale, placed into a reaction flask and 115 mL of H_2SO_4 was added. The mixtures were stirred in an ice bath for 1 hour and 15 g of $KMnO_4$ was slowly added into the mixture. The temperature of the mixture was less than 5 °C. The mixture was removed from the ice bath and stirred by heating for 2 hours at less than 50 °C. The mixture turned into grey color.

In the second stage of the synthesis, 500 mL of deionized water was slowly added into the mixture. The color of mixture turned into dark brown. Then, 8.4 mL of hydrogen peroxide was added in drops till the color of mixture turned into light brown. The mixture was stirred for another 2 hours. The mixture having light brown color was filtered by whatman filter paper and pH was adjusted to 7.0 by washing with pure water. This operation lasted ten days and pH was checked. Then, the GO material was placed as a thin layer on glassware and dried at 50 °C temperature for 24 hours. The color of GO turned into black as dried. The processes of converting G into GO using the Hummers method are given in Figure 2.

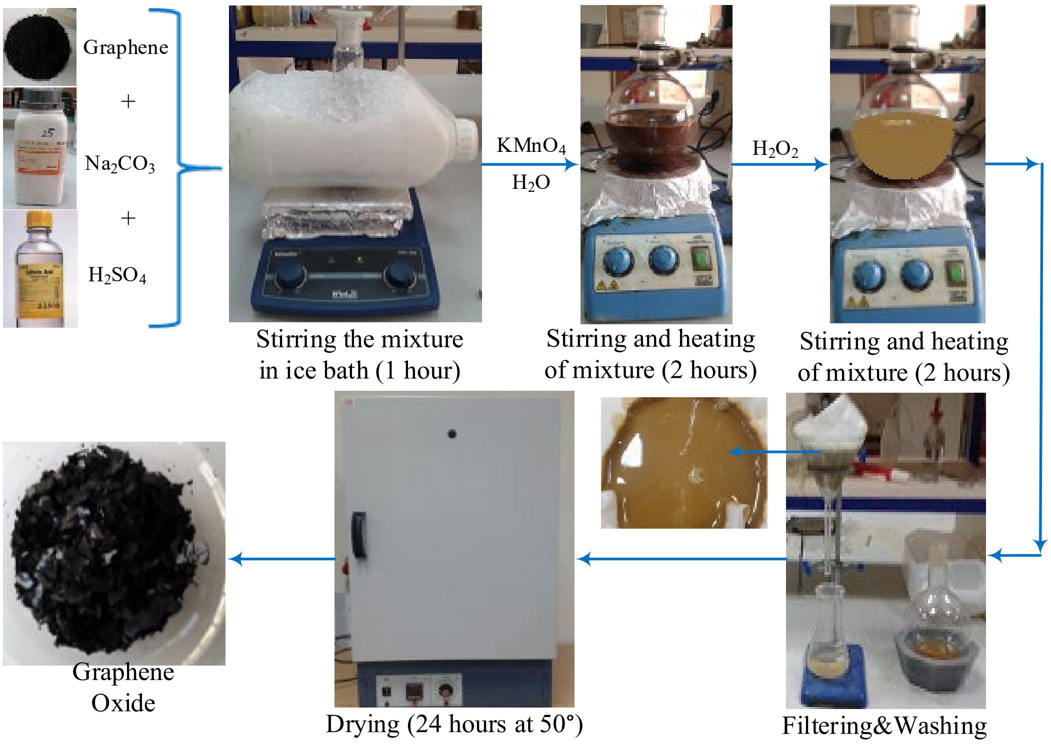


Figure 2.
The synthesis of GO from G by the Hummers method (Çelik et a.l, 2018)

3.3. Addition of Graphene and Graphene Oxide to Polymer Matrix

The synthesized GO was placed in a beaker and tetrahydrofuran (THF) was added on the GO. The beaker was placed in a sonicator and was waited for 1 hour at 60 °C. The GO with THF was removed from the sonicator, placed in a reaction flask and different rates of polymer were added. The THF was removed by evaporating and the GO was homogeneous distributed in the polymer. Similar processes were repeated for the G, which was homogeneous distributed in the polymer. In order to achieve a homogeneous distribution, the operations given the flow diagram in Fig. 3 was carried out.

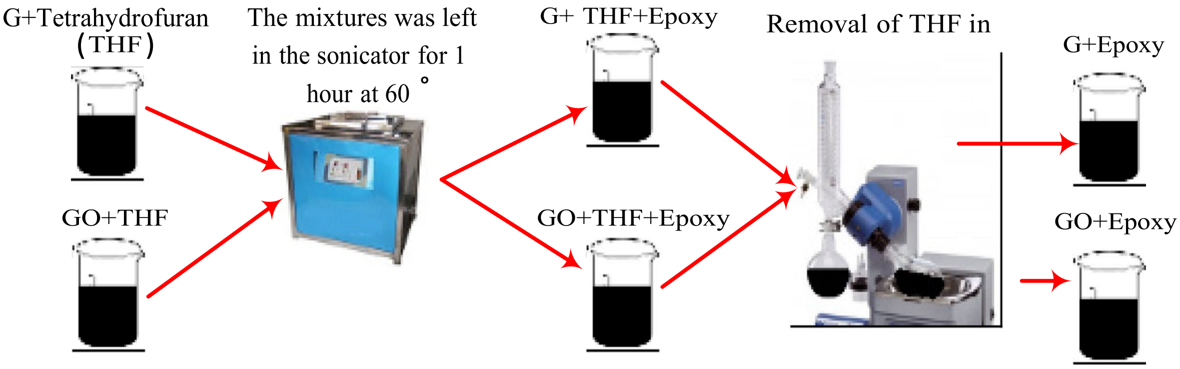


Figure 3.
Synthesis of G and GO reinforced epoxy nanocomposites (Çelik et al., 2018)

3.4. Production of Polymer and CFRP Nanocomposites

The mixtures of G and GO reinforced polymer matrices obtained following removal of THF were weighed using a precision scale to bring the desired proportions and the polymer was added into it**.** The nanocomposite polymers were cured by pouring into a 180 x 250 mm mold. Tensile specimens were prepared from the composites according to ASTM standards, and placed to a tensile tester to determine the tensile strength. Nanocomposite polymer with the highest tensile strength was added to carbon fiber fabric by hand lay-up method. The CFRP nanocomposites were produced by using a total of 4 carbon fiber layers. The polymer and CFRP nanocomposites were also produced without using G and GO and the reinforcement ratios of composites are given in Table 1.

Table 1. The reinforcement ratios of composites produced in the study

Material	Reinforcement Ratio (%)		Material	Reinforcement Ratio (%)
Polymer	-		CFRP	-
G/Polymer	0.1, 0.2 ve 0.3		G/CFRP	0.2
GO/Polymer	0.1, 0.2 ve 0.3		GO/CFRP	0.2

The tensile strength of the nanocomposites was measured in the tensile tester following the production of CFRP nanocomposites (Figure 4).

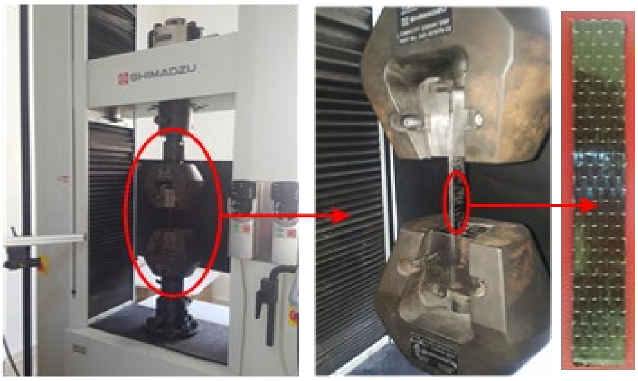
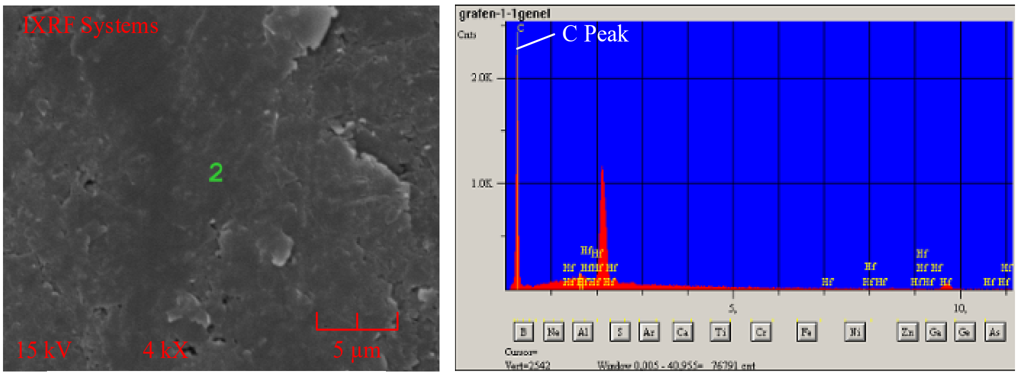


Figure 4.
Measurement of tensile strength in a test specimen

4. RESULTS AND DISCUSSION

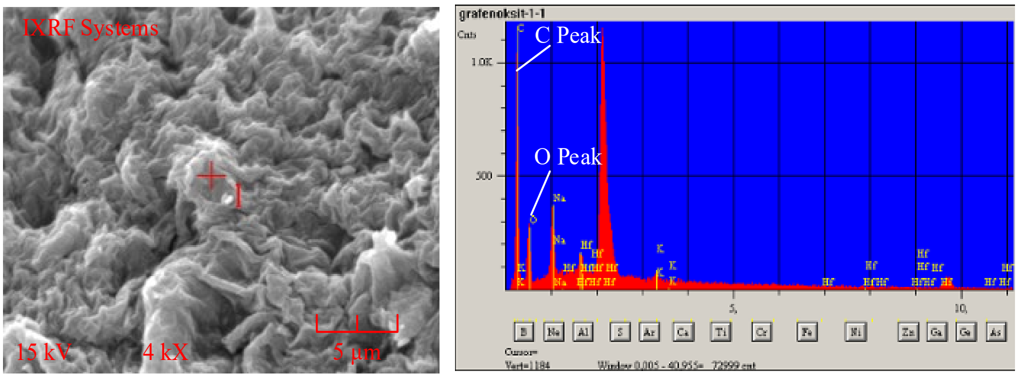
4.1. Characterization of the G and GO nanoparticles

The SEM images and the EDX results of the G and GO are shown in Figure 5. The layered structure of GO and the non-layered structure of G were easily identified in the SEM image (Figure 5a and 5c). The structure of G does not have a functional group in was as indicated by a single peak of carbon in the EDX chart (Figure 5b). Carbon and oxygen peaks in EDX chart of the GO confirmed the conversion of G into the GO (Figure 5d). The FTIR spectrum showing the functional groups in the GO structure are shown in Figure 6.



a) The SEM image of graphene

b) The EDX chart of graphene



c) The SEM image of graphene oxide

d) The EDX chart of graphene oxide

Figure 5.

The SEM images and EDX charts of G and GO nanoparticles

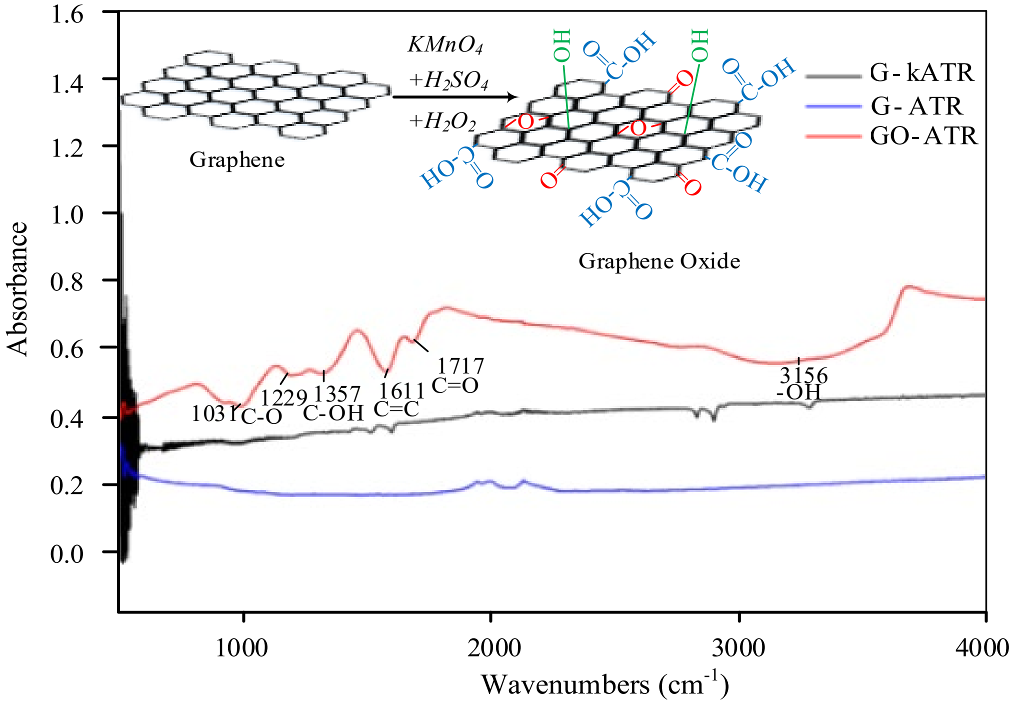


Figure 6.

The FTIR, ATR and kATR spectrums of G and GO nanoparticles

The C-O stretching of carboxyl groups was identified at 1717 cm^{-1}, and the O-H stretching of carboxyl and hydroxyl groups were observed at 3156 cm^{-1}. The C-O stretching of epoxy groups, which are the principal functional groups of the oxidized graphene, and alkoxy groups was observed at 1229 cm^{-1} and 1031 cm^{-1}, respectively. The result confirmed the formation of the GO. In addition, the band between 3000 cm^{-1} and 3700 cm^{-1} is associated with the O-H stretching in the spectrum. The presence of oxygen-containing groups indicates oxidation of the G. Polar groups with hydrogen bonds, especially between surface hydroxyl groups, G and water molecules, indicated the hydrophilic structure of the GO. The XRD spectrum of the G and GO nanoparticles is given in Figure 7.

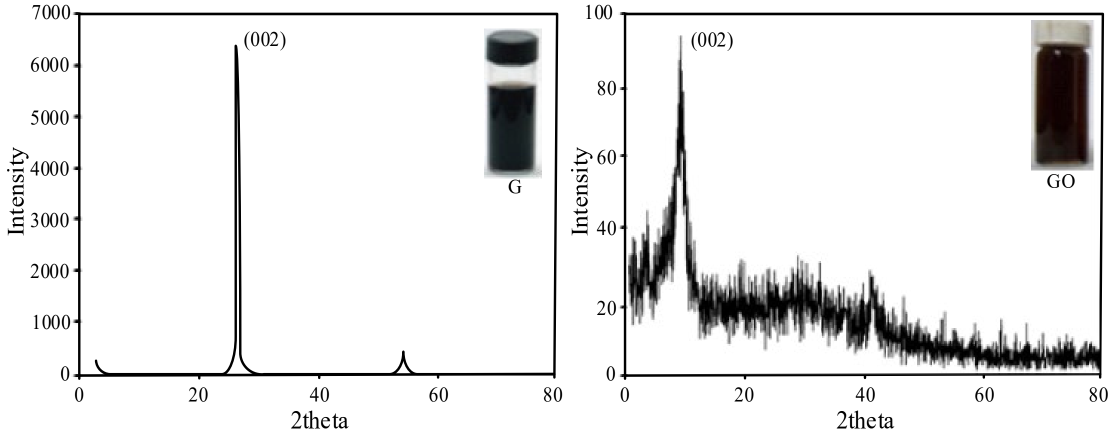


Figure 7.

The XRD spectrum of G and GO nanoparticles (λ=1.54 A°)

Characteristic XRD peaks for G and GO in 2ϴ are 26.4623° and 10.5465°. The characteristic peak of G disappeared in GO following the oxidation of G. The characteristic peak of GO, consistent with the literature, indicated as the formation of GO. The d-spacing between planes was 0.33 0.83 nm for G and GO, respectively. The d-spacings revealed that the functional groups were connected to the structure and water entered between the layers. The functional groups in the GO structure were carboxyl and epoxy groups, which larger than the size of carbon. Therefore, the layers moved away from each other, and caused to increase the interlayer space.

4.2. Tensile strength of nanocomposites with the G and GO

Mechanical properties of reinforced nanocomposite and unreinforced polymers obtained by addition of G and GO were determined. The tensile force-displacement graph obtained in tensile test of the nanocomposites is given in Figure 8.

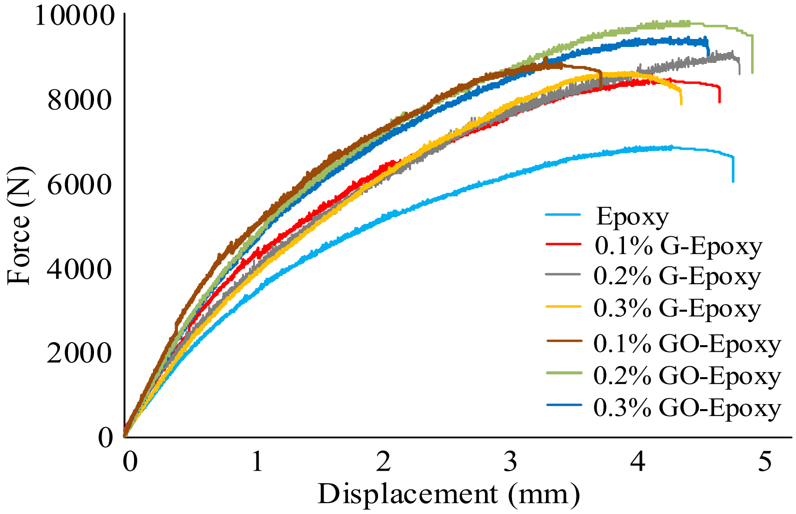
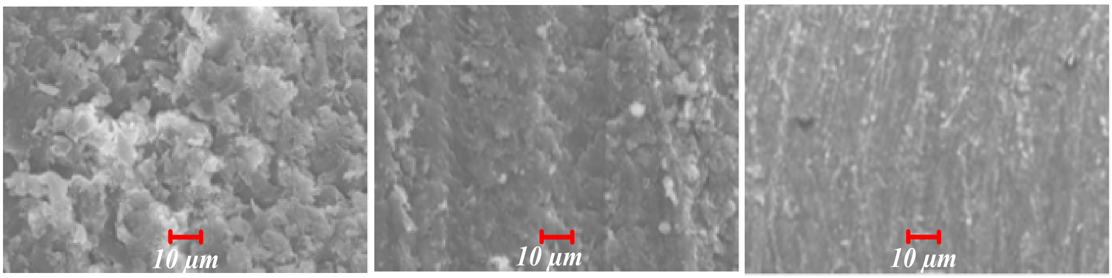


Figure 8.
The Reinforcement type and reinforcement ratio depended on pulling curves

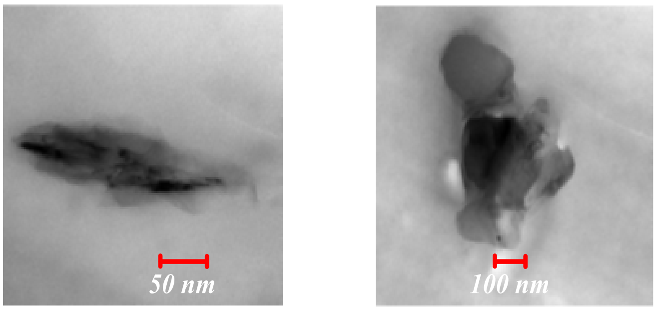
The most important parameter in tensile strength of the nanocomposites was the reinforcement ratio of G and GO nanoparticles (Figure 9). The tensile strength of pure epoxy was 6875 N, while the tensile strengths after 0.1, 0.2 and 0.3 wt% addition of G to epoxy were 8500, 9125 and 8625 N, respectively. The tensile strength of composites increased up to 0.2 ratios. In contrast, the tensile strength of nanocomposites in 0.3% addition decreased compared to the 0.2% addition. Similar results were reported by Le and Huang [30], who obtained the highest strength for nanocomposites having 0.2% reinforced material. The researchers also stated that reinforcement addition more than 0.2% caused a decrease in the tensile strength of nanocomposites. The tensile strength of nanocomposites with 0.2% G increased up to 33% compared to the tensile strength of epoxy materials. The tensile strengths of the nanocomposites with the addition of 0.1, 0.2 and 0.3% of GO to the epoxies were obtained 9125, 9750 and 9625 N, respectively. The tensile strength of the nanocomposites proportionally increased with the addition of GO content into nanocomposites. However, the tensile strength of nanocomposites decreased in GO additions higher than 0.2%. The tensile strengths of nanocomposites with 0.2 G and GO addition were 33 and 42% higher compared to the tensile strength of epoxy materials.



a) The SEM image of unreinforced epoxy

b) The SEM image of nanocomposites reinforced with G

c) The SEM image of nanocomposites reinforced with GO



d) The TEM image of nanocomposites reinforced with 0.2% G

e) The TEM image of nanocomposites reinforced with 0.3% G

Figure 9.
The SEM ve TEM images of produced composites

The SEM image of pure epoxy revealed that the pure epoxy had a gappy and rough surface (Figure 9a). The surface characteristics indicated that the pure epoxy has a weak crack resistance and impact strength. The SEM image of nanocomposite after G addition to the epoxy matrix is shown in Figure 9b. The gaps in epoxy structure were closed with G addition and more rigid structure was obtained. However, the G was not homogeneously dispersed in the epoxy and deep channels were formed on the surface. The SEM image of nanocomposite after GO addition is shown in Figure 9c. The number of superficial randomly spread cracks on nanocomposite surface was lower compared to the nanocomposites with G added. This situation caused high energy need during fracture formations, as well as formation of rough fracture surface, which leads to impact strength and fracture toughness.

The TEM images nanocomposite matrix with 0.2% G are shown in Figure 9d. The G was not homogeneously dispersed in the epoxy due to the agglomeration tendency of nanoparticles. Nonhomogeneous distribution of G negatively affected the properties of the material. Therefore, the G was activated to obtain a homogeneous distribution in the epoxy matrix, and converted into the GO. The TEM images of nanocomposite matrix with 0.3% G are given in Figure 8e. The graphene agglomerated in the epoxy matrix and was homogeneously dispersed. The G was fractured in places where the epoxies were mixed (Figure 9e). Mechanical tests revealed that the fractures adversely affected the mechanical properties of the nanocomposites.

4.3. Tensile strength of CFRP nanocomposites with G and GO

The highest tensile strength in nanocomposites with G and GO was obtained in 0.2% reinforment rate. Therefore, G/CFRP and GO/CFRP nanocomposites were produced by 0.2% addition of G and GO to the epoxy. The tensile tests were carried out to determine the mechanical strength of CFRP composite, 0.2% G/CFRP and 0.2% GO/CFRP nanocomposites. The results of tensile test for the composites are given in Figure 10.

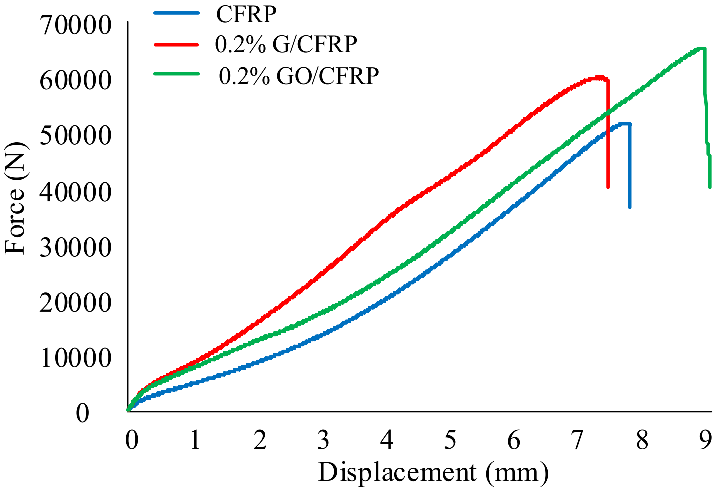
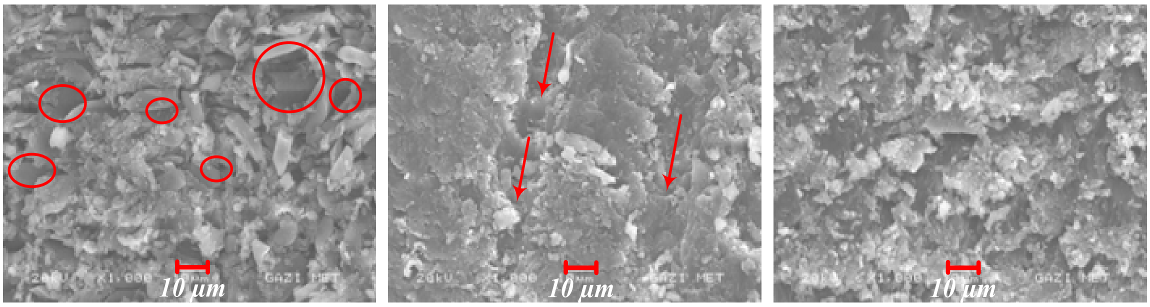


Figure 10.
The Tensile test of CFRP, 0.2% G /CFRP and 0.2% GO/CFRP composites

Addition of G and GO to the CFRP composite increased the tensile strength of G/CFRP and GO/CFRP nanocomposites. The tensile strength of the CFRP composite was 51810 N. The tensile strength of CFRP composite significantly increased with 0.2% G and GO addition. The tensile strength of CFRP composite with 0.2% G and GO were 60060 and 65450 N, respectively. The tensile strength of nanocomposites with 0.2% G/CFRP and GO/CFRP increased up to 16 and 26%, respectively compared to that of the CFRP composite. The SEM images of CFRP composite, G/CFRP and GO/CFRP nanocomposites are given in Figure 11.



a) The SEM images of CFRP composites

b) The SEM images of G/CFRP nanocomposites

c) The SEM images of GO/CFRP nanocomposites

Figure 11.
The SEM images of produced CFRP composites and nanocomposites

Fibers of CF in the epoxy resin are shown in Figure 10a. The gaps, observed between the resin and CF at some points, indicated a weak adhesion between the matrix and CF. However, the addition of G to the CFRP composites increased the stability of structure by increasing the cohesion between the CF and the resin. But, some channels on the nanocomposite increased the surface roughness, which contributed the breaking energy. The gaps between the CF fibers were filled with the addition of GO filled; thus, the adhesion between CF and resin increased. The mechanical test results confirmed the increase in mechanical properties of the composite.

5. CONCLUSION

Synthesis of the GO from the G by the Hummers method was carried out to ensure homogeneous distribution of G in polymer matrices. The chemical and mechanical properties of G and GO nanoparticles were determined. The G and GO nanoparticles were added to the epoxy and CFRP composites, and nanocomposites with G and GO were produced. The prominent results of the study are as follows;

1. Layered structures were observed in the GO structure. Both oxygen and carbon peaks were observed in EDX elemental analysis. The vibration frequencies of carboxyl, epoxy, hydroxyl and alkoxy functional groups which are essential for GO structure were determined in the FTIR spectrum. The characteristic peak of G disappeared in XRD analysis following the oxidation of G. The aforementioned evidences confirm that GO was successfully obtained from the oxidation of G.
2. The TEM results showed that G in the polymer was agglomerated, therefore, the G was converted into GO to obtain a homogeneous distribution in the polymer.
3. Addition of G and GO to epoxy improved the tensile strength of nanocomposites. The effect of GO on tensile strength of nanocomposites was higher than that of the G. The tensile strength of nanocomposites with 0.2% G and GO was higher than the other reinforcement ratios. The tensile strength of nanocomposites with 0.2% G and GO increased up to 33 and 42%, respectively, compared to the tensile strength of the epoxy materials.
4. The tensile strength of G/CFRP and GO/CFRP nanocomposites was higher than that of the CFRP composites. The tensile strengths of nanocomposites with 0.2% G/CFRP and 0.2% GO/CFRP increased up to 16 and 26% ratios, respectively, compared to the tensile strength of CFRP composite.

CONFLICT OF INTEREST

The author acknowledges no conflict of interest or common interest with any institution /organization or person.

AUTHOR CONTRIBUTIONS

Nihayet Koçyiğit contributed full and sole responsibility in determining and managing the conceptual and design processes, data collection, data analysis and interpretation, drafting, critical analysis and final approval of the intellectual content.

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