

# Effect of Graphene Oxide Reduction with L-Ascorbic Acid on Electrical Conductivity and Mechanical Properties of Graphene Oxide-Epoxy Nanocomposites

**Keshavarz, Mohammad Reza; Hassanajili, Shadi\*\***

*Department of Chemical Engineering, School of Chemical and Petroleum Engineering,  
Shiraz University, 71348-51154 Shiraz, I.R. IRAN*

**ABSTRACT:** *In this study, conductive nanocomposites were prepared by dispersing two different types of nanoparticles in the epoxy resin (bisphenol A) matrix. In the first case, Graphene Oxide (GO) was used as the nanoparticle filler, while in the second one, reduced graphene oxide (rGO), which was made using L-ascorbic acid as the reducing agent, was dispersed in the epoxy base. For this purpose, different weight percentages of nanoparticles including 0.25, 0.5, 1, and 2 % were selected to be examined. The prepared samples then were compared with the blank sample in terms of electrical conductivity and mechanical properties involving tensile strength and elastic modulus. According to FT-IR and XRD analyses, it was observed that oxygen functional groups were reduced substantially for the rGO. However, in this case, owing to the transformation of the binary system from polar-polar to polar-nonpolar, rGO could not disperse well in the epoxy matrix. To address this problem, nonylphenol polyethylene was used as a surfactant to provide more suitable dispersion in the epoxy. Results also demonstrated that the electrical conductivity of rGO-epoxy nanocomposite increased dramatically in comparison with both neat epoxy samples and epoxy/GO ones, and the maximum conductivity of  $3 \times 10^{-4}$  S/m (8 orders of magnitude higher than the pristine epoxy resin) was achieved at the rGO percentage of 2%. In addition, mechanical properties (e.g. tensile strength and elastic modulus) were superior in the case of GO and rGO nanocomposites in comparison to that of the blank sample, except for the 2 wt% rGO. Therefore, the best-prepared nanocomposite was the 1wt% rGO sample which improved the electrical conductivity up to 7 orders of magnitude. Likewise, tensile strength and elastic modulus modified 21% and 34 %, correspondingly relative to the neat epoxy sample.*

**KEYWORDS:** *Conductive nanocomposite; L-ascorbic acid; Reduction; Epoxy; Graphene.*

## INTRODUCTION

In the past, polymers were generally used as insulating and structural materials in packaging and electrical insulations where their mechanical and electrically insulating properties were important. In fact, electrical

conduction in polymers was generally regarded as an undesirable phenomenon. Accidentally, in 1977, J. Heeger, Alan G. Mac Diarmid and Hideki Shirakawa discovered that plastics that are commonly considered

---

\* To whom correspondence should be addressed.

+ E-mail: [ajili@shirazu.ac.ir](mailto:ajili@shirazu.ac.ir)

1021-9986/2021/3/731-742

12/\$/6.02

as insulators, could also behave similar to metals. Actually, they manufactured polyacetylene under certain circumstances and thereby succeeded to create electrical conductivity in the polymeric materials [1-3].

Conductive nanocomposites are one of the most important types of conductive polymers, which have various applications at industry including production of chemical or biological sensors, microelectronics devices, fuel cells and catalysts. These nanocomposites can also exhibit remarkable potential in terms of their mechanical property due to their high specific surface area as well as strong nanofiller-matrix adhesion features [4-12].

Nevertheless, recently, many scientists have focused on graphene-based polymer nanocomposites owing to their mechanical, electrical and thermal properties. In fact, graphene can generally enhance thermal, mechanical and electrical properties of graphene-epoxy nanocomposites [13-19]. Actually, GO can be reduced to graphene-like sheets by removing the oxygen-containing functional groups with the recovery of a conjugated structure [20-23]. In this regard, several researchers have dealt with synthesizing graphene oxide (GO)-polymer and reduced graphene oxide (rGO)-polymer nanocomposites through various chemically reducing agents (e.g., sodium borohydride ( $\text{NaBH}_4$ ), octadecylamine (ODA), lithium aluminium hydride, hydrohalic acids (HI), hydrazine, investigating their mechanical, thermal and electrical properties [27-34]. By way of illustration, Toselli et al. prepared GO-epoxy nanocomposite without the use of solvents by direct sonication of graphite oxide (GO) powders using in situ chemically reduction of modified GO powders, where octadecylamine (ODA) was the reducing agent. As a result, they succeeded to improve electrical conductivity of epoxy-rGO nanocomposite around 5 times at 0.5 wt% of rGO in comparison to pristine unmodified resin, but they could not enhance the electrical conductivity [38]. In another work, Tu et al. prepared high-density polyethylene (HDPE)/polypropylene (PP) composites with graphene by melt mixing method. For this purpose, the graphene oxide was thermally reduced (TRG), and exfoliated graphene was used as the surfactant (SEG). According to their results, the electrical conductivity could boost by 10 orders of magnitude at 10 wt % of graphene content [36]. Besides, Mancinelli et al. dispersed graphene oxide nanofillers in the epoxy matrix through two-phase extraction. The measurement of the electrical properties

revealed that adding pristine graphene oxide could not improve the electrical conductivity noticeably, whereas using 0.5 wt % of reduced graphene oxide, which was produced by thermal treatment, the electrical conductivity would enhance 3 times compared to the epoxy sample. Also, polyethylene terephthalate/graphene nanocomposites were synthesized by Zhang et al. [39] through melt mixing method, reporting improved electrical conductivity up to 12 orders of magnitude by adding 3 wt % of graphene.

Therefore, it can undeniably be concluded that in order to promote electrical conductivity of graphene oxide, reduction reactions (deoxygenation) should essentially be taken place regarding the fact that these reactions contribute to increase in charge density of graphene oxide, and consequently higher electrical conductivity [7-15]. Besides, it is of paramount importance to remark that among the existing reducing agents for GO reduction, L-Ascorbic acid shows superiority over others since it can be conducted at low temperatures with low cost. To illustrate, Pei et al. presented a summary of methods through which reduction reactions occur on graphene oxide, and also they stated that L-ascorbic acid is a useful candidate for a simple reduction process at low temperatures [39-43]. Actually, the simplicity of process through using L-ascorbic acid makes it a good choice for large-scale graphene production.

In addition, it should be stated that the epoxy resin can be applied as a coating substance (e.g. as the spray color or liquid) which is resistant to water and chemical materials as well as corrosion and erosion. Moreover, regarding simple curing process of epoxy resin, it is superior to its alternatives such as polyethylene and polypropylene, which require melting process if used as coating materials [44].

Due to the justifications and reasons raised above, L-ascorbic acid is selected as the reducing agent and epoxy resin as the matrix in this investigation to prepare a nanocomposite with high electrical conductivity as well as desirable mechanical properties.

## EXPERIMENTAL MEHODS

### *Materials and instruments*

The main applied materials in this study are: 1-Epoxy resin (bisphenol a-type EPiran-06FL) (from Khuzestan Petrochemical Co. in Iran), 2-Graphene oxide (GO)(purchased from Angstrom Materials Inc Co.), 3-

**Table 1: Applied materials and properties.**

	Specific gravity	Viscosity(cP)@ 25 °C	Purity (%)	appearance
GO(N002-PDE)	2.2	-	98.9	Black powder
L-Ascorbic acid	1.3	-	99	White powder
NPP	1.036	75.0	99.9	Clear liquid
Epoxy(EPiran-06FL)	1.05	9500	99.3	Clear light yellow liquid
Ethanol	0.789	1.095	99.8	Clear liquid

Nonylphenol polyethylene glycol ether (NPP), with chemical formula  $C_9H_{19}C_6H_4(OCH_2CH_2)_n OH$ , (purchased from Kimiagaran Co. at Arak in Iran), 4- L-Ascorbic acid (reagent grade)(from Sigma Aldrich Co.), 5- Ethanol (from Kimia alcohol in Iran). All materials have purity of upper than 98%. Some of their physical properties are summarized in Table 1.

The most important instrument in this study is Hioki (Model: 3455) insulation taster. It has two jaws that subject two sides of intended sample and creates 5kV electrical potential difference, and finally measures electrical resistance and electrical conductivity [49]. Moreover, universal testing machine (Model: 5982, Instron, USA) was used to perform tensile test, FTIR spectrometer (Model: Burker, tensor27) in  $500-4000\text{ cm}^{-1}$  region was used to measure the oxygen functional groups after GO reduction, X-Ray diffractometer (Model: Burker, D8) was employed to evaluate the GO and rGO crystalline structures at the range of  $5-80^\circ$  and finally scanning electron microscope (TESCAN, vega3) images were gained at 20 kV for morphological study.

### Graphene oxide reduction

In this respect, firstly, GO was dissolved in deionized water (0.65g/L). Secondly, mechanical exfoliation method was adopted for ultrasonication (30 min). Thirdly, 5.5 g of L-ascorbic acid was added to the solution as the reducing agent, and then stirred for 72 hrs. Following that, graphene was separated by centrifugation at 8000-10000 r/s. During centrifugation, the solution was washed two times by ethanol and deionized water in order to collect the unreacted L-ascorbic acid. Finally, the solution was put in a vacuum oven for 2 hrs (see Fig. 1).

### Nanocomposite preparation

The rGO-epoxy nanocomposite was prepared by the typical procedure described in Fig.2. In the first place, a

specific amount of rGO was mixed with ethanol, and then sonicated for 30 min. Next, degassed epoxy resin was added to the solution to obtain the concentrations of 0.25, 0.50, 1.0 and 2.0 wt% (weight of GO or rGO /weight of epoxy resin). In the case of rGO-epoxy nanocomposite, 0.01% of NPP (weight of NPP /weight of epoxy resin) was added to achieve better rGO dispersion in epoxy resin, and again the solution was sonicated for 15 min. Fig.3. Shows the role of surfactant between epoxy resin and rGO. Following that, the solution was heated by a heater stirrer for 15 minutes at  $80^\circ\text{C}$  to remove the solvent through evaporation. Subsequently, it was put in a vacuum oven for 2h at  $80^\circ\text{C}$  and -600 mm Hg to evaporate the remained moisture from the solvent. The final mixture was completely mixed with 14 wt % curing agent, poured into molds, and then was immediately put in the oven for 2h at  $80^\circ\text{C}$ .

## RESULTS AND DISCUSSION

### GO and rGO characterization

The GO and rGO structures are characterized by XRD analysis as illustrated in Fig. 3. Accordingly, a sharp diffraction peak can be observed at  $2\theta$  of  $11.5^\circ$ , for the GO. Nevertheless, after reduction, a broad diffraction peak is appeared for the rGO. Considering the fact that the peak width is highly dependent on size of graphite domains existing inside the macroscopic graphene material, rGO with wider peak has smaller crystallite size than GO. This reflects the shrinking of graphite domains and formation of lateral defects upon reduction reaction [8]. Therefore, it can be concluded that reduction has been conducted appropriately [9].

FT-IR measurement was used to explore functional groups on GO and rGO and confirm the reduction reaction (see Fig. 4). As shown in IR spectrum of GO, the presence of C=O (carboxyl) and C-O (carbonyl) groups was confirmed

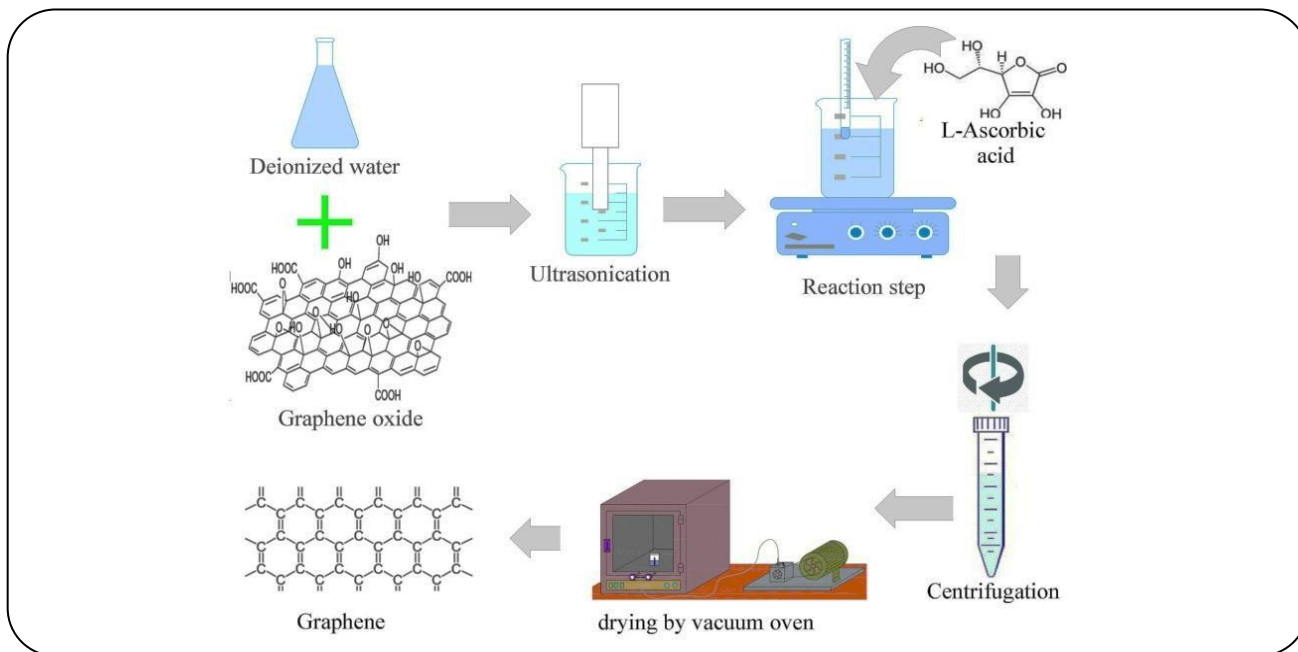


Fig. 1: Graphene oxide reduction process.

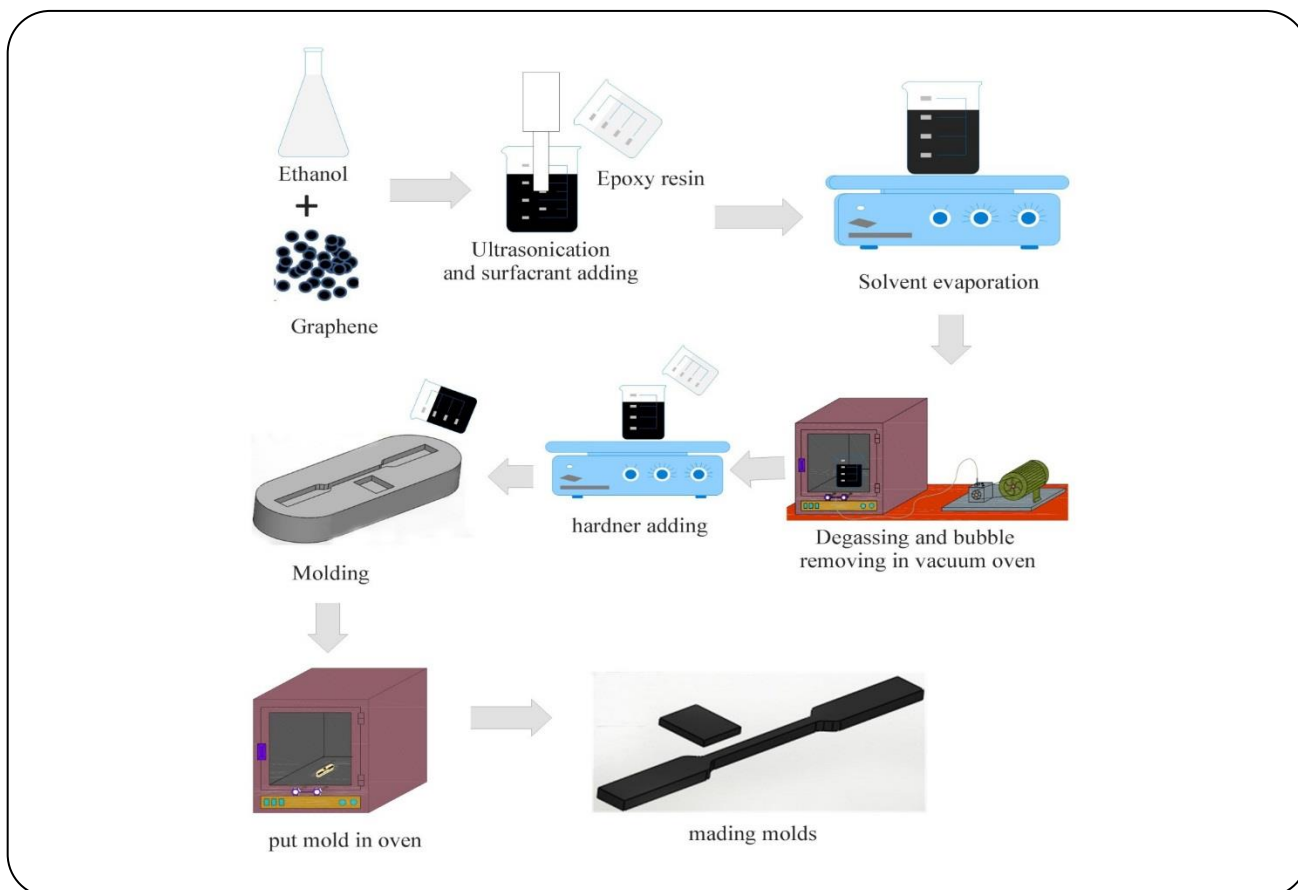
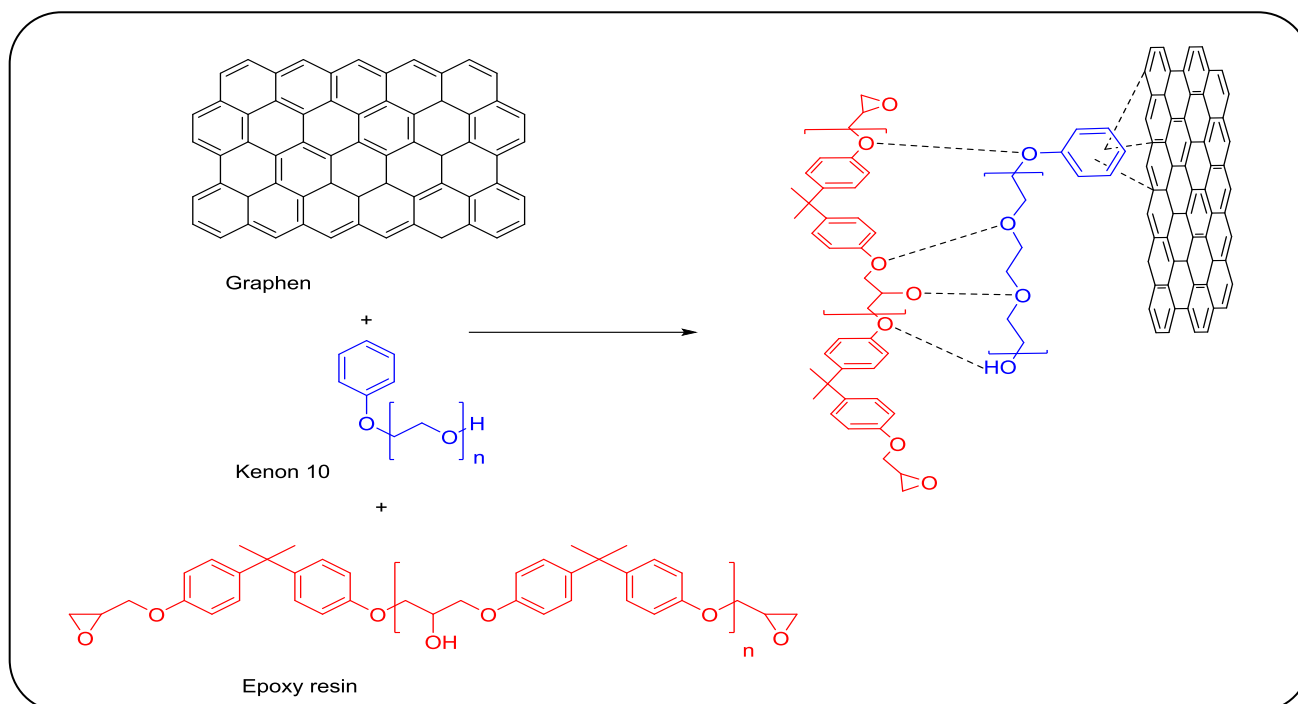


Fig 2: rGO-epoxy nanocomposite preparation.



**Fig. 3:** Surfactant role between the dispersed phase (rGo) and matrix (epoxy resin).

with the peaks at 1726 and 1052  $\text{cm}^{-1}$ , respectively. The broad band at high frequency region (2500-3400  $\text{cm}^{-1}$ ) was assigned to hydroxyl groups of carboxylic acid, carbonyl and residual water molecules in the structure. In addition the peak of stretching vibration of C-O epoxy groups was appeared in the region of 1000-1280  $\text{cm}^{-1}$ . While GO is reduced to rGO by the used chemical reducer, the intensity of abovementioned peaks was reduced and some of them completely eliminated. As shown the reduction of the carboxyl groups (COOH) to hydroxymethyl (CH<sub>2</sub>OH) was proved by the removing of the C=O band at 1726  $\text{cm}^{-1}$  and the appearance of new peaks at band at 2925 and 2853  $\text{cm}^{-1}$ . These changes indicate the successful partial removal of oxygen from the graphene sheets [40].

The intensity of absorbance peaks versus wavenumbers ( $\text{cm}^{-1}$ ) before and after reduction is reported in Table 2. Comparison of data shows the partial removing of oxygen-containing functional groups or GO reduction [40].

Fig. 5a illustrates SEM images of the original GO particles before sonication process in ethanol, in which graphene layers and flakes can clearly be seen. Fig. 5b reveals the differences between rGO and GO; actually, in the case of rGO, the crystals are converted to needle-

shaped particles. Fig. 5c-f shows SEM images of the cross section of GO and rGO nanocomposites. It is observed that the fracture surface of the neat epoxy resin (Fig. 5c) is smooth whereas by adding GO and rGO nanoparticles, the fracture surface becomes rougher with the appearance of cracks on it. The high level of roughness may be related to the good filler/matrix interaction due to the presence of oxygen functional groups on the surface of GO nanoparticles and high surface area in rGO ones [26].

Figs. 5e and 5f show SEM images of the nanocomposites prepared with 0.25 wt. % of rGO in the absence and presence of surfactant, respectively. It can obviously be understood that after the sonication process and separation of GO sheets, these sheets are easily discernible. In addition, it can be seen that a good dispersion of GO nanoparticles is taken place in all regions of epoxy in the presence of NPP surfactant. Figs. 5g and 5h demonstrate the nanocomposite prepared with 1 and 2 wt. % of rGO modified with the surfactant, respectively.

If one compares the morphology of nanocomposites with different weight percentages, it can be concluded that with increase in the weight percentage of the rGO, the crystals show more tendency to agglomerate and flake, which is not desirable. Although the NPP surfactant is used for better dispersion, in the case of 2 wt%

Table 2: FT-IR Absorbance peak vs. functional groups.

	Wavenumbers (cm <sup>-1</sup> )	Absorbance intensity (GO)	Absorbance intensity (rGO)	Absorbance intensity reduction percent (%)
C-O	1052	0.05	0.001	98
C-O-C	1226	0.049	0.006	87
C-OH	1410	0.032	0.003	90
C=C, -OH	1630	0.054	0.009	83
C=O	1726	0.048	0.004	91
O-H, Carboxylic acid, carbonyl, Water	2500-3400	0.125	0.051	59

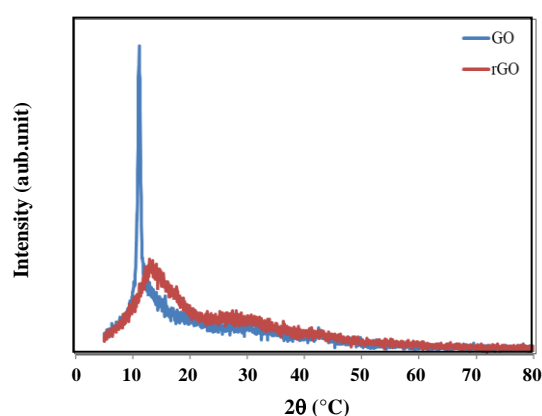


Fig. 4: XRD pattern of GO and rGO.

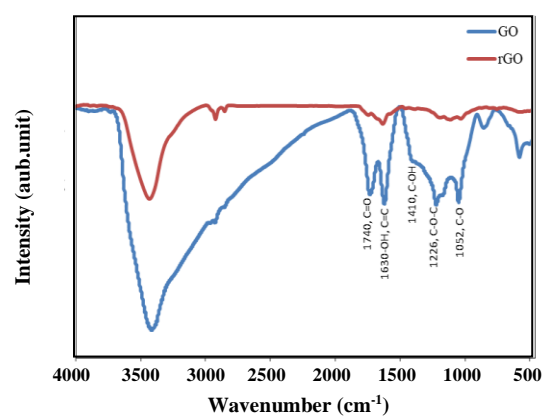


Fig. 5: FT-IR of GO and rGO.

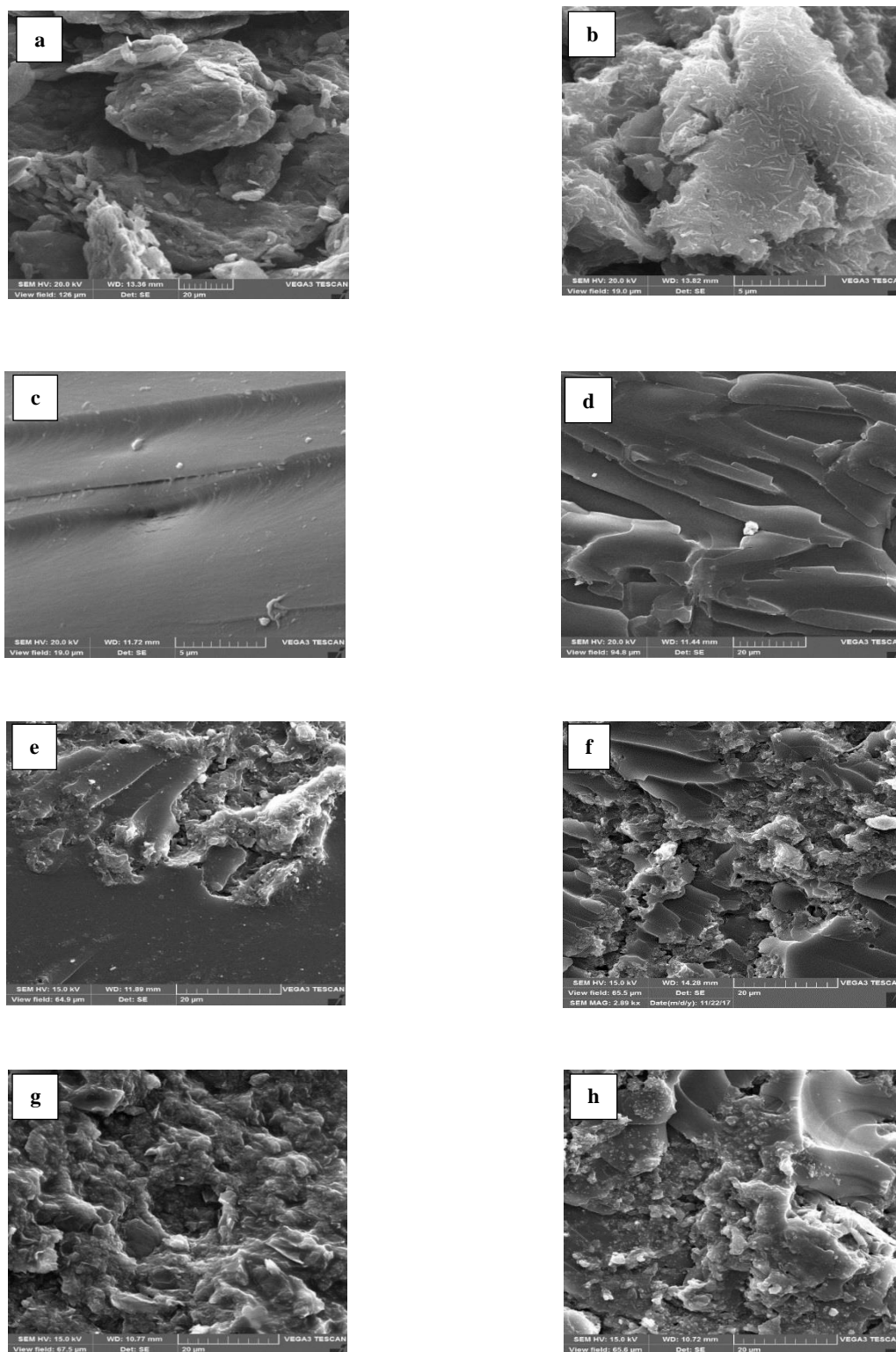
(i.e., highest rGO content), fillers cannot be dispersed well into the polymer mainly due to the poor particle-epoxy interaction. In fact, regarding the transformation of a polar-polar (epoxy-GO) molecular interaction to polar-nonpolar (epoxy-rGO) one, when rGO is loaded instead of GO, uneven and flaked dispersion is plausible. To explain, the more the oxygen groups are removed through reduction process of GO, weaker adhesion to the epoxy matrix is obtained, and consequently weaker dispersion in the matrix is observed. This phenomenon is also seen in the fabrication of nanocomposites with clay as the filler and polyethylene/polypropylene as the matrix phase. Regarding the polar structure of clay particles when polyethylene and polypropylene are used, a suitable dispersion cannot be achieved in the nanocomposite structure. To tackle this problem, adding proper surfactants or modification of nanoparticles can partially enhance the dispersion in the polymeric matrix [42].

### Electrical properties

Electrical conductivity of the prepared nanocomposites is measured using the Megger tester [50]. Fig. 6 shows the electrical conductivity versus weight percentage of filler. Clearly, the electrical conductivity of GO-epoxy composite increases smoothly with the increase in GO content, up to the point that it reaches the maximum conductivity of  $1.11 \times 10^{-11}$  S/m where the GO weight percentage is 2%, implying that it depends slightly on the GO content.

However, the electrical conductivity of rGO-epoxy nanocomposite varies noticeably with the increase in rGO content, and the maximum conductivity of  $3 \times 10^{-4}$  S/m can be obtained at the rGO percentage of 2%.

In fact, with a small increase of filler content from 0.5% to 1% for the rGO case, electrical conductivity changes remarkably from  $2.52 \times 10^{-10}$  to  $3.56 \times 10^{-5}$  S/m, indicating that the percolation threshold is at 0.50 % filler content, while for the GO, no percolation threshold can be seen.



**Fig. 6:** SEM images: (a) Pure GO; (b) rGO; (c) neat epoxy; (d) 1.0 wt% GO; (e) 0.25 wt% rGO; (f) 0.25 wt% rGO+ Surfactant ; (g) 1.0 wt% rGO+ Surfactant; (h) 2.0% wt% rGO+ Surfactant.

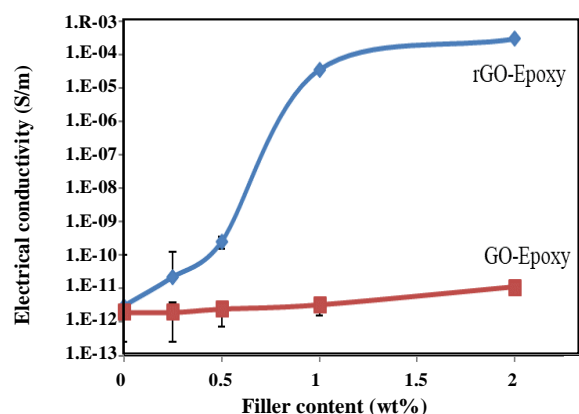


Fig. 7: Comparison between electrical conductivity of GO and r-GO nanocomposite vs. filler content.

Surprisingly, although the rGO sample exhibits poorer dispersion characteristics relative to GO, its conductivity is tremendously higher than that of GO. This can most presumably be attributed to the fact that the positive effect of the reduction reaction (desirable change) can compensate for the negative effect of weak dispersion (undesirable change), and subsequently higher electrical conductivity can be obtained in the case of rGO-epoxy nanocomposites.

### Mechanical properties

Fig. 7 depicts stress-strain curves for GO and r-GO nanocomposites at different filler contents. Accordingly, improvement in tensile strength and elongation at break is achieved as a result of reinforcing the structure with loading of nanofillers. As a result, it is observed that at 1.0 wt% GO and 0.5 wt% rGO the best mechanical properties can be obtained, implying that for each nanofiller, there is an optimum percentage.

Fig. 8a represents variation of tensile strength of nanocomposite versus filler content for both rGO and GO cases. In all samples prepared by GO, the tensile strength is enhanced in comparison with the blank sample (neat epoxy), and it goes up until it reaches a maximum value at 1 wt% filler content, showing 23.2% improvement. Nevertheless, by adding more content (e.g., 2 %), the curve tends to decrease although it is still 4.5 % higher than the blank sample.

To compare, for all filler contents, rGO nanocomposite has lower tensile strength than GO. This can presumably be ascribed to more homogenous dispersion of GO

nanoparticles in the epoxy matrix (see Fig. 5), which contributes to higher mechanical strength.

Another important observation is that even though at the weight percentage of 1%, rGO represents 17% improvement in comparison to the blank sample (neat epoxy), if the content exceeds 1% (i.e., 2 %), the tensile strength deteriorates significantly; i.e., decreases around 23.2 %.

The good tensile property of the composites at lower content of fillers suggests an appropriate interaction of GO and rGO with epoxy and epoxy/NPP surfactant, respectively, which ensures an efficient loading transfer between the matrix and the particles [48].

It is also important to mention that in both cases, probability of agglomeration of graphene layers intensifies with the increase in the content of nanoparticles, resulting in a lower tensile strength at the weight percentage of 2%. In addition, due to the non-polar structure of molecules in the case of rGO, this phenomenon (agglomeration) occurs more intensely, and therefore the tensile strength of rGO-epoxy nanocomposite is even lower than that of the neat epoxy sample. In the other words at 2 wt% loading, filler agglomerates are observed in the prepared composites (Fig. 6d), resulting in a decrease of surface area in contact with the polymer matrix. The graphene agglomerates in the composites can easily lead to cracks initiation and propagation and consequently produce the reduced strength of the composites. But, the highly dispersed graphene fillers which were prepared at lower filler contents are more efficient than the aggregated ones in transferring applied load and reinforcement (see Fig. 8a). The relatively lower tensile strength in rGO-epoxy than that of GO-epoxy composites may be related to their structure difference. Because due to weak van der Waals forces between graphene sheets in rGO, slippage can occur between the mentioned sheets and their orientation in the direction of the tensile load to maximize reinforcement mechanism is limited [49].

Fig. 8b depicts elastic modulus of both nanocomposites versus filler content. Modulus of all GO-epoxy samples is boosted and the maximum value can be seen at 1 wt%, gaining 40.46% improvement in comparison with the blank sample. In the case of rGO-epoxy, similar to what observed for GO case, the elastic modulus increases as the filler content goes up to 0.5 wt%, and remains nearly unchanged for 1 wt%; however, a sharp decrease is



Table 3: Comparison of elongation at break for GO and rGO nanocomposites.

Sample	$\epsilon$ ( $\pm 5\%$ )	Sample	$\epsilon$ ( $\pm 5\%$ )
Blank	11.18	Blank	11.18
0.25% GO	10.76	0.25% rGO+surf	9.48
0.50% GO	9.85	0.50% rGO+surf	10.11
1.0% GO	9.43	1.0% rGO+surf	11.22
2.0% GO	8.95	2.0% rGO+surf	12.58

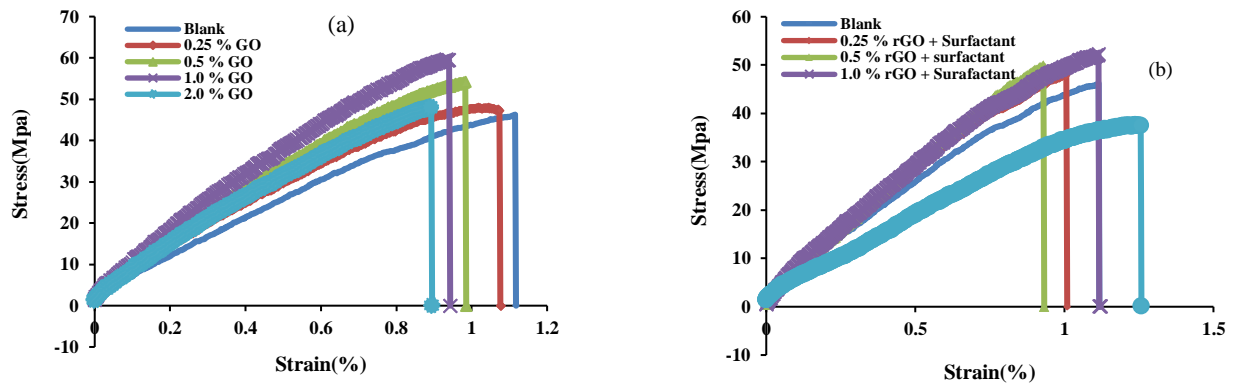


Fig. 8: Tensile Stress-strain curves of epoxy nanocomposites with 0.25, 0.5, 1.0 and 2 wt% of (a) GO and (b) rGO.

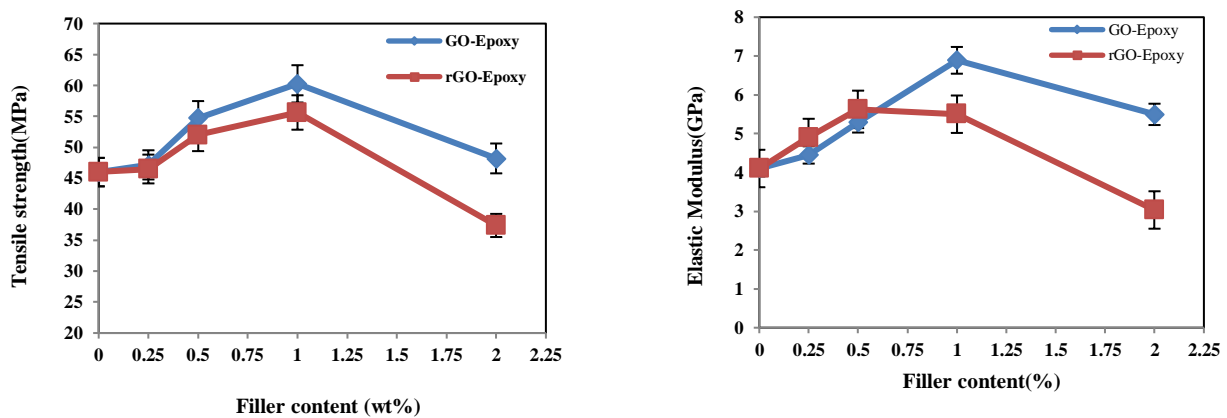


Fig. 9: Comparison between (a) tensile strength and (b) elastic modulus of GO and r-GO nanocomposite vs. filler content.

observed at 2 wt% rGO, meaning 35.24% deterioration in comparison with the blank sample. The main reason for this observation is the decline of adhesion between filler-epoxy resin and the weakness of interphase region after reduction reaction.

As it is shown in Table 3, elongation at break of epoxy nanocomposites decreases with increase in filler content in most cases. At low stresses, because of restricted motion of epoxy chains in the presence of the filler network, epoxy

nanocomposites show high modulus and resistance. Conversely, as the stress increases, weak points become larger, leading to uneven distribution of stress thereby the sample becomes brittle and the strain at break decreases.

Table 3 shows the results of different studies which have been used GO or rGO for enhancing electrical conductivity. As can be seen, the electrical conductivity improvement obtained in this research is higher than others.

**Table 4: Different study on Electrical conductivity enhancement.**

	Year	GO/rGO content in nanocompsit(wt%)	Electrical conductivity enhancement (Order of magnitude)	Reference
This research	2019	2% rGO+epoxy	8	
Wu et al.	2015	3% rGO+SnO <sub>2</sub>	7	[19]
Ghaleb et al	2014	1.0% CNT+GO+epoxy	6	[44]
Monti et al.	2013	3% GO+epoxy	5	[46]

## CONCLUSIONS

In this experimental study, the effects of reduction reaction using L- ascorbic acid as the reducing agent on electrical conductivity and mechanical properties of epoxy-graphene nanocomposite were evaluated. In addition, for more suitable dispersion, nonylphenol polyethylene glycol ether (NPP) is used as a non-ionic surfactant. SEM images represented better dispersion of GO than rGO nanoparticles in the epoxy matrix, indicating better interactions of the polar-polar binary system (rGO-epoxy). Also, the electrical conductivity can be enhanced nearly by 8 orders of magnitude for 2 wt% rGO-epoxy nanocomposites when compared to the neat epoxy.

Besides, tensile strength and elastic modulus can be enhanced in all samples of GO-epoxy nanocomposites. However, mechanical properties become weaker in some cases of rGO-epoxy when compared to the neat epoxy composite due to weak interaction between filler and the polymer matrix and unsuitable dispersion at higher filler loadings. In conclusion, the best-prepared sample contained 1wt% rGO, and its corresponding electrical conductivity was enhanced by 7 orders of magnitude, the tensile strength was modified 21% and elastic modulus was improved 34 %, in comparison to the blank epoxy sample.

## Acknowledgment

The authors would like to express their gratitude to Mr. Omid Khalifeh for his help and advice as to the English Science editor.

Received : Aug. 2, 2019 ; Accepted : Feb. 3, 2020

## REFERENCES

- [1] Shirakawa H., Louis E., Macdiarmid A.G., Chiang C.K., Heeger A.J., [Synthesis of Electrically Conducting Organic Polymers Halogen Derivatives of Polyacetylene](#), *J. Chem. Soc. Chem. Com. (JCSCC)*, **474**:578-580(1977).
- [2] Chiang C.K., Park Y.W., Heeger A.J., Shirakawa H., Louis E.j., Macdiarmid A.J., [Conducting polymers: Halogen doped polyacetylene](#), *J. Phys. Chem.(JPC)*, **69**: 5098-5104 (1978).
- [3] Fatahi A., Liang R., Kaur A., [Photocatalytic degradation using TiO<sub>2</sub>-graphene nanocomposite under UV-LED illumination: Optimization using response surface methodology](#), *Canada. J. Env. Che. Eng*, **7**: 10371-10379 (2019).
- [4] Liang C., Qiu H., Han Y., Gu H., Song P., Wang L., Kong J., Cao D., Gu J., Mater J., [Superior Electromagnetic Interference Shielding 3D Graphene Nanoplatelets/Reduced Graphene Oxide Foam/Epoxy Nanocomposites with High Thermal Conductivity](#), *China. J. Mat. Chem. C.(MKC)*, **7**: 2725-2733 (2019).
- [5] Wei J., Inam F., Vo T., [Epoxy/Graphene Nanocomposite- Processing and Properties: a Review](#), *UK. Roy. Soc. of Chem. (RSC)*, **5**: 73510-73524 (2015).
- [6] Sieradzka M., Binias D., Janicki J., [A Facile Approach to Obtaining PVDF/Graphene Fibers and the Effect of Nano Additive on the Structure and Properties of Nanocomposites](#), *Poland. Pol. Tes.*, **81**: 106229-106237 (2020).
- [7] Gado M. A., [Sorption of Thorium Using Magnetic Graphene Oxide Polypyrrole Composite Synthesized from Water Hyacinth Roots](#), *Iran. J. Chem. Chem. Eng.*, **37(3)**: 145-160 (2018).
- [8] Sharma R., Chadha N., Saini P., [Determination of Defect Density, Crystalite Size and Number of Graphene Layers in Graphene Analogs Using X-Ray Diffraction and Raman Spectroscopy](#), *India. Ind. J. Pur. App. Phy.*, **55**: 625-629 (2017).
- [9] Ghalmi Y., Sayah A., Bahloul A., Nessark B., [Enhancement of the Electrochemical Properties of PbO<sub>2</sub> by Incorporation of Graphene Exfoliated](#), *Iran. J. Chem. Chem. Eng. (IJCCCE)*, **39(2)**: 269-274 (2020).

- [10] Foroughi F., Rahsepar M., Hadianfard M. J., Kim H., Facile Synthesis and Electrochemical Performance of Graphene-Modified Cu<sub>2</sub>O Nanocomposite for Use in Enzyme-Free Glucose Biosensor, *Iran. J. Chem. Chem. Eng.*, **39**(2): 1-10 (2018).
- [11] Balint R., Cassidy N., Cartmell S., Conductive Polymers: Towards A Smart Biomaterial for Tissue Engineering, *UK. Act. Bio.*, **10**: 2341-2351 (2014).
- [12] Ram M., Yavuz O., CO gas Sensing from Ultrathin Nano-Composite Conducting Polymer Film, *USA.Sen. Act. B.(SAB)*, **106**:750-757 (2005).
- [13] Adak B., Joshi M., Butola B., Polyurethane/functionalized-graphene Nanocomposite Films with Enhanced Weather Resistance and Gas Barrier Properties, *India. Com. Par. B. 176: 107303-107315* (2019).
- [14] Ding H., Zhang S., Chen J., Reduction of graphene Oxide at Room Temperature with Vitamin C for RGO-TiO<sub>2</sub> Photoanodes in Dye-Sensitized Solar Cell, *China.Thi. Sol. Fil. (TSF)*, **584**: 1-8 (2015).
- [15] Zhang L., Zhao X., Highly Conductive and Porous Activated Reduced Graphene Oxide Film for High-Power Super Capacitor, *USA.Ame. Che. Soc.(AMS)*, **12**: 1806-1812 (2012).
- [16] Gong J., Yang H., Yang P., Investigation on Field Emission Properties of N-Doped Graphene-Carbon Nanotube Composites, *China, Com. Par. B.(CPB)*, **36**: 250-255 (2015).
- [17] Guo Y., Xu G., Yang X., Ruan K., Ma T., Zhang Q., Gu J., Wu Y., Lu H., Guo Z., Significantly Enhanced and Precisely Modeled Thermal Conductivity in Polyimide Nanocomposites with Chemically Modified Graphene Via in Situ Polymerization and Electrospinning-Hot Press Technology, *J. Mater. Chem. C.(JMCC)*, **6**: 3004 (2018).
- [18] Potts J., Dreyer D., Bielawski C., Ruoff R., Graphene-Based Polymer Nanocomposite, *Poly.*, **52**:5-25 (2011).
- [19] Rafee M., Rafee J., Enhanced Mechanical Properties of Nanocomposites at Low Graphene Content, *ACS nano*, **3**: 3884-3890 (2009).
- [20] Wu Y., Brahma S., Weng S., Chang C., Huang J., Reduced Graphene Oxide (RGO)-SnO<sub>x</sub> (x=0,1,2) Nanocomposite as High-Performance Anode Material for Lithium-Ion Batteries, *Taiwan.*, **38**: 152889-152897 (2019).
- [21] Kim H., Abdala A., Graphene/Polymer Nanocomposites, *Mac.*, **43**: 6515-6530 (2010).
- [22] Ho K.C., Teow Y.H., Mohammad A.W., Ang W.L., Lee P.H., Development of Graphene Oxide (GO)/Multi-Walled Carbon Nanotubes (MWCNTs) Nanocomposite Conductive Membranes for Electrically Enhanced Fouling Mitigation, *J. Mem. Sci. (JMC)*, 552: 189-201 (2018).
- [23] Pei S., Zhao J., Du J., Direct Reduction of Graphene Oxide Films Into Highly Conductive and Flexible Graphene Films By Hydrohalic Acids, *Car.*, **48**: 4466-4474 (2010).
- [24] Li W., Tang X., Zhang H., Simultaneous Surface Functionalization and Reduction of Graphene Oxide with Octadecylamine for Electrically Conductive Polystyrene Composites, *Car.*, **49**:4724-4730 (2011).
- [25] Pei S., Cheng H., The Reduction of Graphene Oxide, *Car.*, **50**: 3210-3228 (2012).
- [26] Li M., Zhou H., Zhang Y., Liao Y., Zhou H., Effect of Defects on Thermal Conductivity of Graphene/Epoxy Nanocomposites, *Car.*, **130**: 295-303 (2018).
- [27] Zhu X., Zhu Y., Nanostructured Reduced Graphene Oxide/Fe<sub>2</sub>O<sub>3</sub> Composite as a High-Performance Anode Material for Lithium-Ion Batteries. *ACS. nano*. 5:3333-3338 (2011).
- [28] Zhixin C., Liu L., Wang X., Poly(Urethane-co-vinyl imidazole)/Graphene Nanocomposites, *Pol. Com. (PC)* 459-466 (2012).
- [29] Chua Ch., Pumera M., Chemical Reduction of Graphene Oxide: A Synthetic Chemistry Viewpoint, *Roy. Soc. Chem. (RSC)*. **20**:20-32 (2013).
- [30] Abulizi A., Okitsu K., Ultrasound-Assisted Reduction of Graphene Oxide to Graphene in L-Ascorbic Acid Aqueous Solution: Kinetics and Effect of Various Factor on the Rate of Graphene Formation, *Ult. son.*21:1174-1181 (2014).
- [31] Pokharel P., Pant S., Effect Of Functional Groups on the Graphene Sheet for Improving the Thermochemical Properties of Polyurethane Nanocomposite, *Com. Par. B. (CPB)*, **78**: 192-210 (2015).
- [32] Zakaria M.R., Kudus M.H., Akil H.M., Thrimizir MZ., Comparative Study of Graphene Nanoparticle and Multiwall Carbon Nanotube Filled Epoxy Nanocomposites Based on Mechanical, Thermal and Dielectric Properties, *Com. Par. B. (CPB)*, **119**: 57-66 (2017).

- [33] Wentzel D., Miller S., Sevostianov H., [Dependence of the Electrical Conductivity of Graphene Reinforced Epoxy Resin on the Stress Level](#), *Int. J. Eng. Sci. (IJEC)*, **120**: 63–70 (2017).
- [34] Moreno J.M., Sanchez V.Y., Hidalgo R.S., Verdejo R., Manchado M.A., Garcia L.F., Blanco C., Mendez R., [Customizing Thermally reduced Graphene Oxides for Electrically Conductive or Mechanical Reinforced Epoxy Nanocomposites](#), *Eur. Pol. J. (EPG)*, **93**: 1–7 (2017).
- [35] Biswas C., et al., [Layer-By-Layer Hybrid Chemical Doping for High Transmittance Uniformity in Graphene-Polymer Flexible Transparent Conductive Nanocomposite](#), *Sci. Rep. (SR)*, **8**: 135-146 (2018).
- [36] Tosseli M., Fabiani D., [In Situ Thermal Reduction of Graphene Oxide Forming Epoxy Nanocomposites and Their Dielectric Properties](#), *Pol. Com. (PM)*, **32**: 294-301 (2014).
- [37] Tu C., Nagata K., Yan S., [Morphology and Electrical Conductivity of Polyethylene/Polypropylene Blend Filled With Thermally Reduced Graphene Oxide and Surfactant Exfoliated Graphene](#), *Pol. Com.*, **42**: 1-8 (2015).
- [38] Zhang H., Zheng W., Yan Y., [Electrically Conductive Polyethylene Terephthalate/Graphene Nanocomposites Prepared by Melt Compounding](#), *Pol.*, **51**: 1191-1196 (2010).
- [39] Fernandez M., Guardia L., Parades J., [Vitamin C Is an Ideal Substitute for Hydrazine in the Reduction of Graphene Oxide Suspensions](#), *J. Phys. Chem. (JPC)*, **114**: 6426-6432 (2010).
- [40] Zhang J, Yang H. [Reduction of Graphene Oxide via L-Ascorbic Acid](#), *Chem. Com.*, **46**: 1112-1114 (2010).
- [41] Mamo M., Sustaita A., [Polymer Composite of Poly \(Vinyl Phenol\)-Reduced Graphene Oxide Reduced by Vitamin C in Low Energy Consuming Write-Once-Read-Many Times Memory Devices](#), *J. Org. Ele. (JOE)*, **14**: 175-181 (2013).
- [42] Pei S., Cheng H.M, [The Reduction of Graphene Oxide](#), *Car.*, **50**: 3210-3228 (2012).
- [43] Nyambo C., Wang D., Wilkie C.A, [Will Layered Double Hydroxides Give Nanocomposites with Polar or Non-Polar Polymers](#), *Pol. Adv. Tec. (PAT)*, **20**: 332-340 (2009).
- [44] Boumaza M., Khan R., Zahrani S., [An Experimental Investigation of the Effects of Nanoparticles on the Mechanical Properties of Epoxy Coating](#), *Thi. Sol. Fil. (TSF)*, **620**:160-164 (2016).
- [45] Ghaleb Z.A., Mariatti M., Ariff Z.M., [Properties of Graphene Nanopowder and Multi-Walled Carbon Nanotube-Filled Epoxy Thin-Film Nanocomposites for Electronic Applications: the Effect of Sonication Time and Filler Loading](#), *Com. Par. B. (CPB)*, **58**: 77–83 (2014).
- [46] Tang G., Jiang Z., Li X., Zhang H., Hong, S., [Electrically Conductive Rubbery Epoxy/Diamine-Functionalized Graphene Nanocomposites with Improved Mechanical Properties](#), *Com. Par. B. (CPB)*, **67**: 564–570 (2014).
- [47] Monti M., Rallini M., Puglia D., Peponi L., Torre L., [Morphology and Electrical Properties of Graphene](#), *Com. Par. A. (CPA)*, **46**: 166–172 (2013).
- [48] Hassanajili Sh., Sajedi M.T., [Fumed Silica/Polyurethane Nanocomposites: Effect of Silica Concentration and Its Surface Modification on Rheology and Mechanical Properties](#), *Iran..Pol. J.*, **25**: 697-710 (2016).
- [49] Tang L., Wan Y., Yan D., [The Effect of Graphene Dispersion on the Mechanical Properties of Graphene/Epoxy Composites](#), *Car.*, **60**: 16-27 (2013).
- [50] Meger test,  
<http://www.megger.com/eu/story/Index.php?ID=839>