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The Impact of Filler Content on Mechanical and Micro-Structural Characterization of Graphite-Epoxy Composites

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Abstract

In this study, the characteristics of Graphite/Epoxy Composites (GECs) are evaluated from mechanical perspectives. Different weight percentages of graphite were used (0 - 7 wt%) for tensile and hardness experiments. Then the findings were discussed to ascertain the optimum mixing ratio of the graphite with the epoxy. The primary finding of this study is that the graphite weight fraction has a substantial impact on the composites' mechanical performance. At a low percentage (1 wt%), the graphite has little influence on the tensile behaviour. An intermediate weight percentage of the graphite is considered optimum for mechanical performance in the epoxy composites as it slightly reduces the tensile properties and significantly improves the hardness. Micrographs of the fractured surface of specimens showed many signs that clearly explained why fractures had occurred. For instance, when graphite/epoxy composite contained a low proportion of graphite, the cleavage failure was very easy to observe because there was no sign of aggregation or the detachment of fillers.

Keywords

Graphite, Epoxy, Composites, Microstructural, Mechanical, Tensile, Hardness

1. Introduction

Polymeric composites have turned out to be a superb substitute for metal materials in several industrial applications [1]. Among the commonly used polymers in various industrial applications is epoxy. Epoxy resins are a class of thermosetting polymers having epoxide end groups. They can provide a better combination of distinctive properties than other thermosetting resins do; hence, they fulfil a wide range of demands in situations where fibres and epoxy resins intermix to form composites [2]. Epoxy resins provide high resistance, proper electrical insulation, low shrinkage, excellent adhesion, and high bending strength to deterioration from solvents and chemicals; furthermore, they exist in several physical forms [3]. Comprehending the features and properties of such materials has become an attractive field of investigation for various research areas in science and engineering. In order to find the required properties, the reinforcements in polymeric composites need to be selected early in the design stage. Most of the studies carried out on polymeric composites in the literature centered on their mechanical properties and the characterization of polymeric composites, but considering the tribological behavior of the material under particular conditions is vital, and so is having components that are appropriately designed [4] [5] [6]. The optimization of tribological performance and mechanical properties requires a precise selection of composite reinforcement. When the friction coefficient is low, graphite fillers are considered among the best particles of the solid lubricant, which can produce a self-lubricating film [7]. However, the amount of graphite needs to be optimized because it may impair properties, such as tensile strength, hardness, compressive power, conductivity, water absorption, degradation, etc. [7] [8] [9] [10]. In other words, the correlation between tribological as well as mechanical properties and the amount of filler requires attention.

In view of the above, the present study focuses on the effect of the graphite content on the mechanical behavior of epoxy composites in an attempt to understand the correlation between them and learn the optimum amount of graphite to include.

2. Materials Selection and Preparation

Test specimens of graphite/epoxy composite were prepared to carry out experimental mechanical tests. The prepared specimens were made up of Kinetix (H160 medium) and epoxy resin (R246TX) hardener, which formed the resin matrix. It was provided by "Australian Calibrating Services Pty. Ltd" (Melbourne, Australia). The graphite filler used in this study was 92% pure, and was 45 μm in size, as obtained from Chem-supply Pty Ltd, Australia. Hou, Hu [11] reported that bigger sizes of graphite (>45 μm) may help to break down the microstructure of the composite. Therefore, the present study focuses on graphite of 45 μm in size to preserve consistent mechanical properties. The resin mix was produced by blending the epoxy and the hardener in a ratio of 3:1, according to the industrial Standard. Different volume fractions of graphite (1, 3, 5, and 7 wt%) were added to the prepared resin mixture to compose the samples of the study, as shown in **Table 1**, and stored for a while until the mixture reached the right consistency.

The NE and GE specimens were prepared for mechanical testing, according to "ASTM D638-99" [12]. **Figure 1** illustrates the standard dimensions (in specimen

Table 1. Designation of the GECs.

Material	Graphite (G) wt%	Matrix (E) wt%
NE	0	100
GE1	1	99
GE3	3	97
GE5	5	95
GE7	7	93

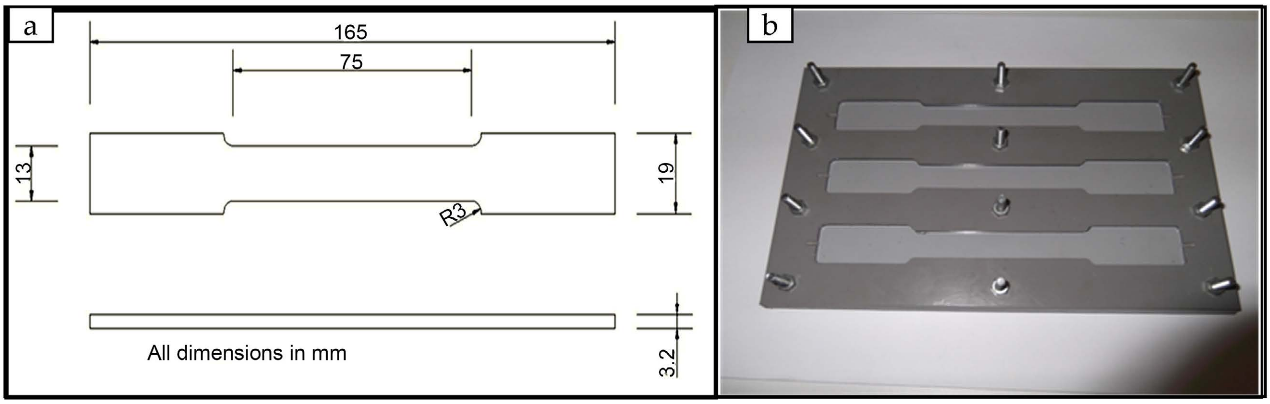

Figure 1. (a) Specimen dimensions of tensile (b) Used mould for tensile test.

geometry) and the mould used. The resin mixture was poured into the mould and when it had solidified, the specimen was peeled off the mould and cured for 24 hours under the same conditions in the atmosphere. The samples were cured in the oven again at a temperature of 50˚C for 24 hours.

3. Experimental Procedure

Under ambient conditions, "Modulus of elasticity" (E) and "Tensile strength" (S) were measured according to ASTM D638-99 [12], utilizing the Test Star "Material Testing System" (MTS 810) equipped with 100 KN. All trials with a 50 mm gauge length and a 1 mm/minute crosshead speed were performed. Durometer type D was used to determine the hardness according to ASTM D2240 [13]. The same two tests were repeated and the average values were obtained for each set of five samples. The morphology of the failed (fractured) composite surfaces was examined using the SEM to identify and analyse the principal features of failure. SEM (Philip XL–30) was used to categorize the damage features and fracture mechanisms of the failed samples after each test.

4. Results & discussion

4.1. Tensile and Hardness Properties of Graphite/Epoxy Composites

4.1.1. Stress-Strain Diagram, Ultimate Tensile Strength, and Modulus of Elasticity (E)

Figure 2 indicates the average tensile stress-strain curves of NE and GE composite samples. Moreover, Figure 3 presents the summary of UTS ("Ultimate tensile strength") and the *E* of the NE and GE composite samples and their standard deviation.

Figure 2 shows that the trend of stress is in general almost the same for all the composites since there is a definite region of elastic deformation and a slight area of plastic deformation. In other words, the composites exhibited brittle failure regardless of the graphite percentage. The reason behind this is that both materials were brittle, since the epoxy is a thermoset material, and the graphite is also considered to be brittle, according to Berto, Lazzarin [14]. However, as regards strain, there is no remarkable difference between the composites. In contrast, the UTS shows deterioration with the addition of the graphite fillers. This deterioration may have resulted from the low interaction in the graphite fillers and the epoxy matrix (Sengupta *et al.*) Sengupta, Bhattacharya [15] comprehensively reviewed the mechanical properties of several polymer composites based on graphite fillers and confirmed that the addition of more than 4% graphite

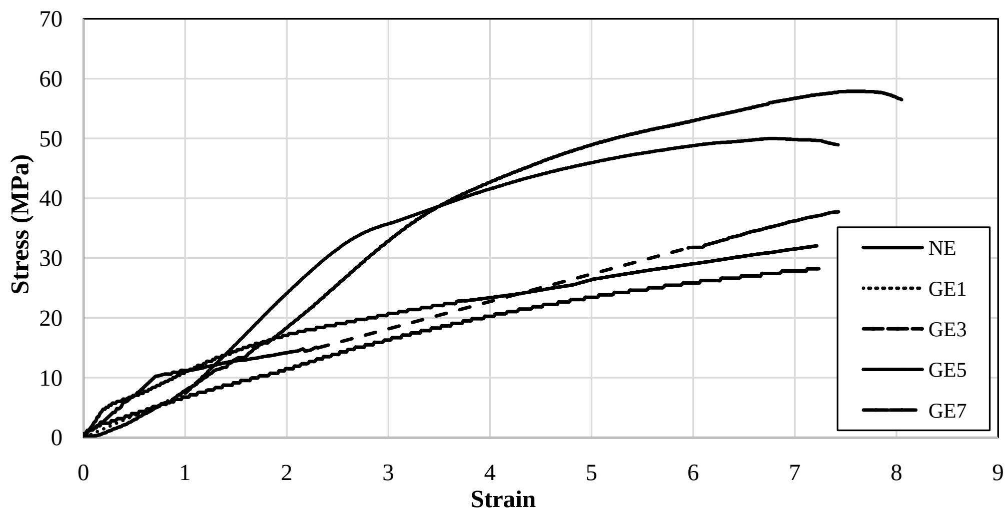

Figure 2. Stress-strain diagrams of graphite/epoxy composites.

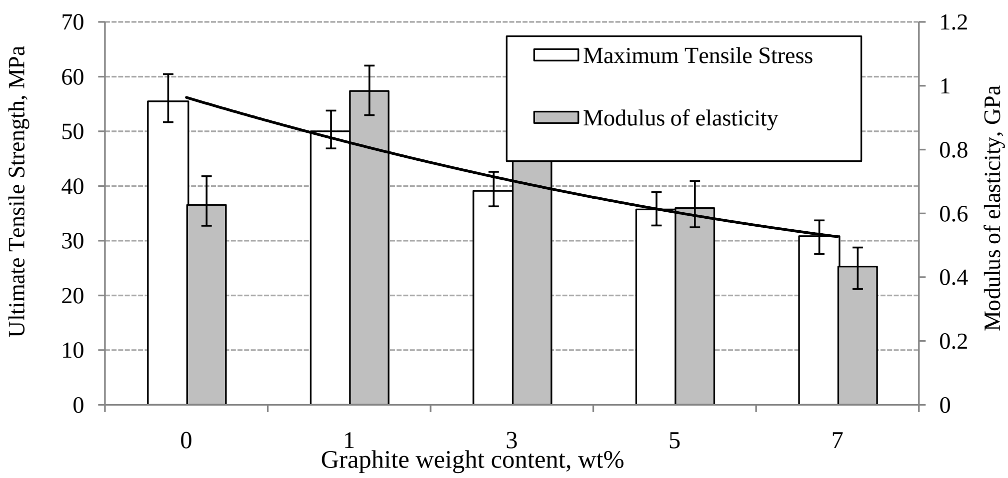

Figure 3. Ultimate TS and modulus of elasticity of GECs.

significantly reduces the UTS of various polymer composites like PLA [16], HDPE [17], PMMA [18], EVA [19] and PPE [20]. In these studies, the interaction, distribution, orientation, and size of the graphite have a major impact on the mechanical properties. Furthermore, **Figure 3** illustrates the UTS and the E of GEC samples. This figure demonstrates a reduction in the UTS of the GECs since there is a 45% reduction of UTS at the graphite amount of 7 wt%. Such a reduction is not desirable from the mechanical point of view. Therefore, a slight reduction in the TS could be considered in the design. At 1 wt% and 3 wt% of graphite, the UTS is reduced by about 10% and 20%, respectively, since the UTS reduces from 55 MPa to 50 MPa and 47 MPa, respectively. However, **Figure 3** also reveals that the existence of the graphite in the composites improves the E up to a specific graphite concentration. The Modulus of Elasticity increases with increased amounts of graphite content to around 3 wt%, whereas it declines when the graphite content exceeds 3 wt%. That is to say, the optimum graphite percentage is found to be around 3 wt%., which contributes to achieving the highest Modulus of Elasticity. The explanation for this finding is that as the graphite content increases the viscosity of the fluid also increases' this weakens the dispersion quality that determines the prospect of forming agglomerations and increasing their size in the process of solidification.

4.1.2. Shore Hardness (SHD)

The Shore hardness of the composite is illustrated in **Figure 4**. This indicates the impact of the graphite content on hardness. **Figure 4** indicates that raises in the percentage of graphite increase the composite hardness. The lowest Shore hardness (82.2 SHD) was found with a graphite content of 0 wt%, while the highest Shore hardness (84.7 SHD) was found with the content of graphite at its highest, 7 wt%. The hardness increases monotonically with the graphite concentration. This behaviour may be due to the higher hardness in graphite than in epoxy, as noted by Suherman, Mahyoedin [21].

To a certain extent, the addition of graphite has a positive effect on some mechanical properties while it may have drawbacks for others. In the next section, the impact of graphite content on the microstructure of the composite and the relationship between the microstructural changes and mechanical properties of the composite are discussed.

4.1.3. Fracture Behaviour of the Epoxy Composites

The micrographs of the failed NG samples after the test are shown in **Figure 5**. There are irregular fracture features and no obstacles or initiators for the cracks. The figure clearly reveals a cleavage failure and a river-like pattern, which represents the nature of the thermoset epoxy. The existence of the shear lips and the detachments (bright and reflective facets) means that the material resisted the shear loading and detachments in the molecules occurred. Such failure has been reported by some published works [22] in which a plain strain fracture mechanism was evident.

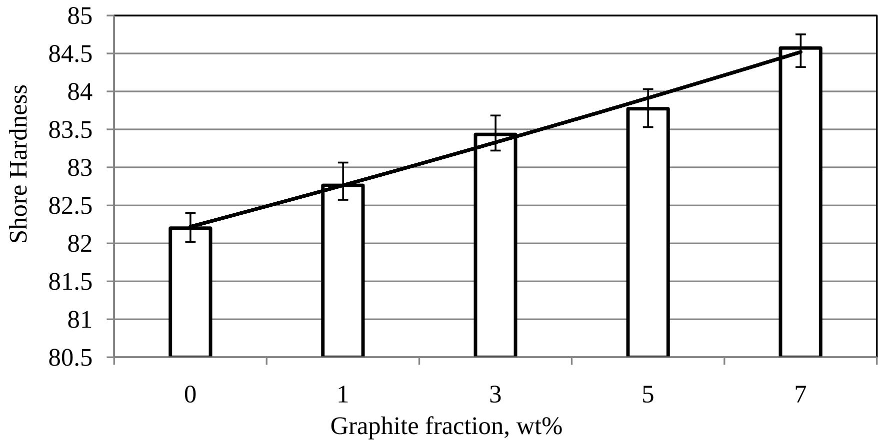

Figure 4. Shore D hardness of graphite/epoxy composites.

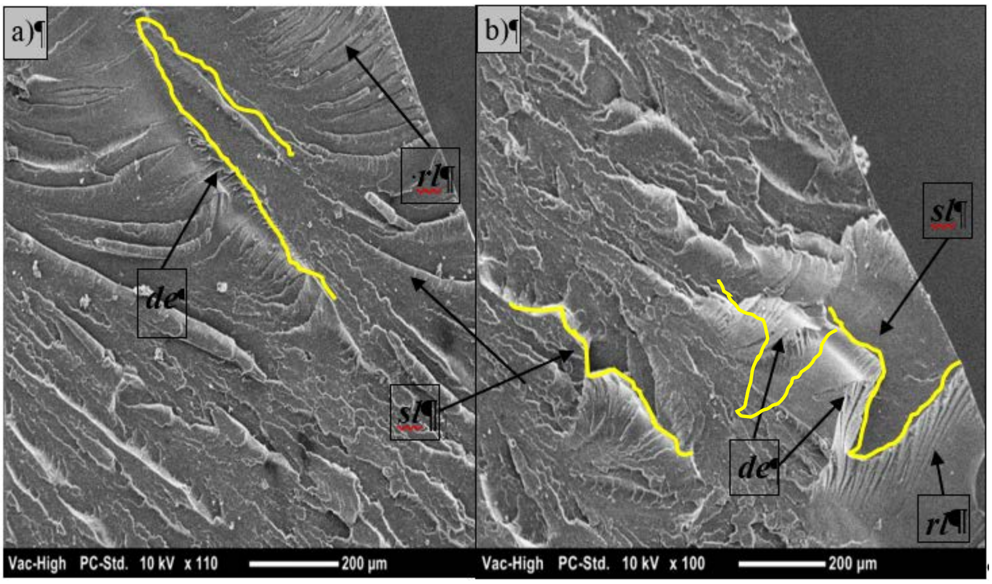

Figure 5. Micrographs of the NEC after tensile testing. (sl = shear lips, de = detachment, rl = river-like pattern).

The micrographs of the failed GE1 samples are shown in **Figure 6** (note the different magnifications). There are apparent differences between the micrographs of the NE and the GE since there are fewer brittle failure features in the GR than in the NE; that is, no acute fracture appears on the surface. In **Figure 6**, there is a river-like pattern and stretching, indicating resistance to the load. Shear lips are obvious and micro-cracks are apparent due to the presence of tenuous agglomerations of graphite. In other words, the graphite interface with the epoxy seems to be acceptable compared to the descriptions in the literature on the graphite pallet [15] and nano-clay [23] [24] [25], since there are no voids [26] or debonding [27] of the fillers at this weight fraction of the graphite. However, some researchers have reported that such fillers can initiate cracks [28] [29] [30]. In the present work, there is no sign of crack initiation because the fraction of graphite is low.

The micrographs showing the fractured samples of the epoxy composites with 3 wt% are shown in Figure 7. The figure indicates signs of shear lips surrounded by debris, which seems to come from the graphite fillers. At a higher magnification, Figure 7(a) and Figure 7(b) display the graphite at a size of 10 - 20 µm. The present work has a filler size of 45 µm. Figure 7 shows signs of stretching, indicating an excellent resistance to the load. Further, there is no evidence of voids, and no detachment of fillers is observed. This represents a filler's good interfacial adhesion with the matrix at this low weight content of the graphite. It should be noted here that, in the graphite/epoxy composite fabrication process, an ultrasonic machine was used for 1 hour before the solidification process began, to assist the dispersion of the graphite and get rid of the bubbles. This technique contributes to a better homogenization of composites than other techniques in the literature.

At the highest proportion of graphite in the epoxy composites (5 wt%), the micrographs of the fractured samples display clusters and aggregation of graphite

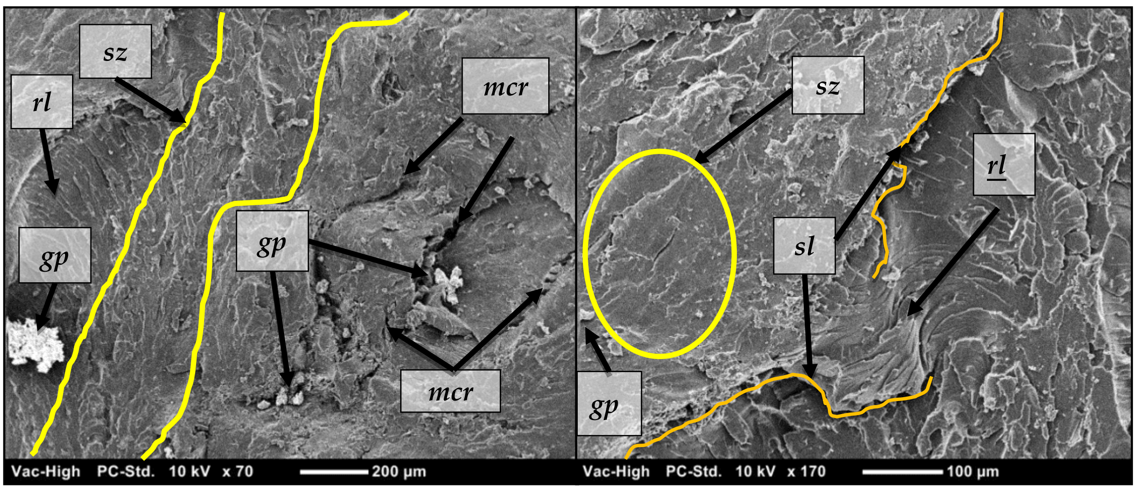

Figure 6. Micrographs of the 1% graphite/epoxy composites after tensile testing. (cr = micro-cracks, sl = shear lips, rl = river-like pattern, sz = stretching zone, gp = graphite particle).

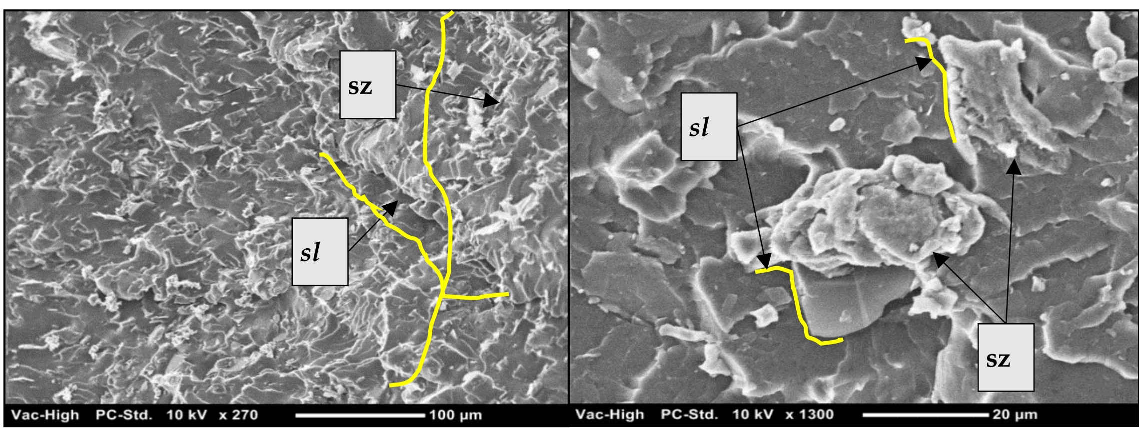

Figure 7. Micrographs of 3% GECs after tensile testing. (sl = shear lipsrl, sz = stretching zone, gp = graphite particle).

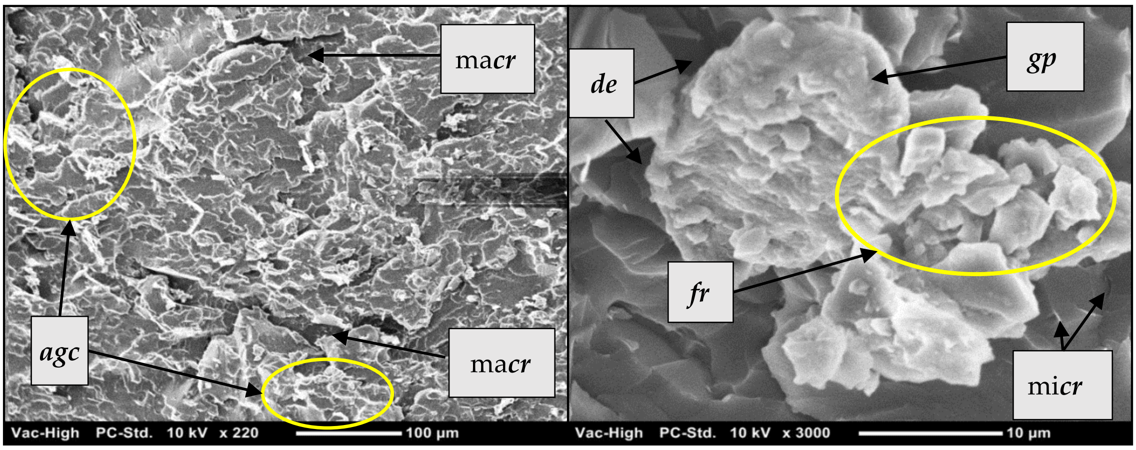

Figure 8. Micrographs of 5% GECs after tensile testing. (gp = graphite particle, de = debonding, ag = aggregation, macr = macro-crack, micr = micro-crack, fr = fragmentation).

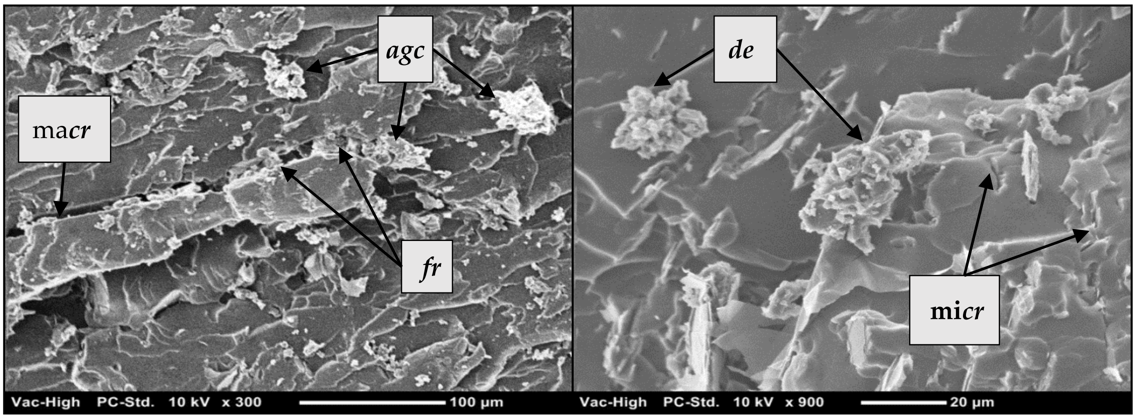

Figure 9. Micrographs of 7% GECs after tensile testing. (de = debonding, ag = aggregation, macr = macro-crack, micr = micro- crack, fr = fragmentation).

(Figure 8). It appears that large amounts of aggregated graphite significantly damage the microstructure of the composites, leading to debonding and the inception of a fragmentation process. Despite the use of ultrasonics in the fabrication process, the large percentage of graphite in the composites affected the quality of the composite mixing and the integration of the fillers with the resin in the curing process. This correlates with published works on nano-clay/epoxy [24], graphite pallet/epoxy [15] and graphite/polyester composites [15]. It is highly pronounced at 5 and 7 weight percent of graphite in the epoxy composites, as shown in Figure 8 and Figure 9. In this figure, micro-and macro-cracks can be seen. These may have been initiated by the poor interface between the large aggregated amount of graphite and the resinous regions.

5. Conclusions

The primary results of this study may be summarised in a few points:

There is a significant impact of the weight fraction of the graphite on the mechanical and performance properties of the composites. At a low percentage of graphite (one weight percent), there is not much influence on the tensile behaviour and a slight improvement to the hardness performance of the epoxy composites. An intermediate wt% of the graphite is considered optimum for the mechanical and performance properties in the EC since it makes a slight reduction in the tensile properties and modulus of elasticity and a significant improvement to the hardness.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Yousif, B. and El-Tayeb, N. (2007) On the Effect of Woven Glass Fabric Orientations on Wear and Friction Properties of Polyester Composite. *Surface Review and Letters*, **14**, 489-497. https://doi.org/10.1142/S0218625X07009487

[2] Begum, K. and Islam, M.A. (2013) Natural Fiber as Subtitute to Synthetic Fiber in Polyemrs Composites. Shahjalal University and Technology, Bangladesh.

[3] Mallick, P.K. (1993) Fiber-Reinforced Composites: Materials, Manufacturing, and Design. 2nd Edition, Rev. and Expanded Edition, Mechanical Engineering, M Dekker, New York, 566 p.

[4] Yousif, B. and Chin, C. (2012) Epoxy Composite Based on Kenaf Fibers for Tribological Applications under Wet Contact Conditions. *Surface Review and Letters*, **19**, Article ID: 1250050. https://doi.org/10.1142/S0218625X12500503

[5] Shalwan, A. and Yousif, B. (2013) In State of Art: Mechanical and Tribological Behaviour of Polymeric Composites Based on Natural Fibres. *Materials & Design*, **48**, 14-24. https://doi.org/10.1016/j.matdes.2012.07.014

[6] Williams, J. (2005) The Influence of Repeated Loading, Residual Stresses and Shakedown on the Behaviour of Tribological Contacts. *Tribology International*, **38**, 786-797. https://doi.org/10.1016/j.triboint.2005.02.006

[7] Sun, W.-C., Zhang, P., Zhao, K., Tian, M.-M. and Wang, Y. (2015) Effect of Graphite Concentration on the Friction and Wear of Ni-Al_2O_3/Graphite Composite Coatings by a Combination of Electrophoresis and Electrodeposition. *Wear*, **342-343**, 172-180. https://doi.org/10.1016/j.wear.2015.08.020

[8] Alizadeh, T. and Hamedsoltani, L. (2014) Graphene/Graphite/Molecularly Imprinted Polymer Nanocomposite as the Highly Selective Gas Sensor for Nitrobenzene Vapor Recognition. *Journal of Environmental Chemical Engineering*, **2**, 1514-1526. https://doi.org/10.1016/j.jece.2014.07.007

[9] Etmimi, H.M., Mallon, P.E. and Sanderson, R.D. (2013) Polymer/Graphite Nanocomposites: Effect of Reducing the Functional Groups of Graphite Oxide on Water Barrier Properties. *European Polymer Journal*, **49**, 3460-3470. https://doi.org/10.1016/j.eurpolymj.2013.08.004

[10] Yu, G.-C., Wu, L.-Z. and Feng, L.-J. (2015) Enhancing the Thermal Conductivity of Carbon Fiber Reinforced Polymer Composite Laminates by Coating Highly Oriented Graphite Films. *Materials & Design*, **88**, 1063-1070. https://doi.org/10.1016/j.matdes.2015.09.096

[11] Hou, Y., Hu, P., Zhang, X. and Gui, K. (2013) Effects of Graphite Flake Diameter on Mechanical Properties and thermal shock behavior of ZrB_2-nanoSiC-Graphite Ce-

ramics. *International Journal of Refractory Metals and Hard Materials*, **41**, 133-137. https://doi.org/10.1016/j.ijrmhm.2013.02.015

[12] American Society for Testing and Materials (2000) D638-99. Standard Test Method for Tensile Properties of Plastics. American Society for Testing Materials, Philadelphia.

[13] American Society for Testing Materials (2005) Standard Test Method for Rubber Property—Durometer Hardness. ASTM International, West Conshohocken.

[14] Berto, F., Lazzarin, P. and Ayatollahi, M.R. (2013) Brittle Fracture of Sharp and Blunt V-Notches in Isostatic Graphite under Pure Compression Loading. *Carbon*, **63**, 101-116. https://doi.org/10.1016/j.carbon.2013.06.045

[15] Sengupta, R., Bhattacharya, M., Bandyopadhyay, S. and Bhowmick, A.K. (2011) A Review on the Mechanical and Electrical Properties of Graphite and Modified Graphite Reinforced Polymer Composites. *Progress in Polymer Science*, **36**, 638-670. https://doi.org/10.1016/j.progpolymsci.2010.11.003

[16] Narimissa, E., Gupta, R., Bhaskaran, M. and Sriram, S. (2012) Influence of Nano-Graphite Platelet Concentration on onset of Crystalline Degradation in Polylactide Composites. *Polymer Degradation and Stability*, **97**, 829-832. https://doi.org/10.1016/j.polymdegradstab.2012.02.010

[17] Zheng, W., Lu, X. and Wong, S.C. (2004) Electrical and Mechanical Properties of Expanded Graphite-Reinforced High-Density Polyethylene. *Journal of Applied Polymer Science*, **91**, 2781-2788. https://doi.org/10.1002/app.13460

[18] Ramanathan, T., Abdala, A., Stankovich, S., Dikin, D.A., Herrera-Alonso, M., Piner, R.D., *et al.* (2008) Functionalized Graphene Sheets for Polymer Nanocomposites. *Nature Nanotechnology*, **3**, 327-331. https://doi.org/10.1038/nnano.2008.96

[19] George, J.J. and Bhowmick, A.K. (2008) Ethylene Vinyl Acetate/Expanded Graphite Nanocomposites by Solution Intercalation: Preparation, Characterization and Properties. *Journal of Materials Science*, **43**, 702-708. https://doi.org/10.1007/s10853-007-2193-6

[20] Celik, C. and Warner, S.B. (2007) Analysis of the Structure and Properties of Expanded Graphite-Filled Poly(Phenylene Ether)/Atactic Polystyrene Nanocomposite Fibers. *Journal of Applied Polymer Science*, **103**, 645-652. https://doi.org/10.1002/app.23772

[21] Suherman, H., Mahyoedin, Y., Septe, E. and Rizade, R. (2019) Properties of Graphite/Epoxy Composites: The In-Plane Conductivity, Tensile Strength and Shore Hardness. *AIMS Materials Science*, **6**, 165-173. https://doi.org/10.3934/matersci.2019.2.165

[22] Kanchanomai, C. and Thammaruechuc, A. (2009) Effects of Stress Ratio on Fatigue Crack Growth of Thermoset Epoxy Resin. *Polymer Degradation and Stability*, **94**, 1772-1778. https://doi.org/10.1016/j.polymdegradstab.2009.06.012

[23] Jawahar, P., Gnanamoorthy, R. and Balasubramanian, M. (2006) Tribological Behaviour of Clay—Thermoset Polyester Nanocomposites. *Wear*, **261**, 835-840. https://doi.org/10.1016/j.wear.2006.01.010

[24] Alamri, H. and Low, I.M. (2012) Effect of Water Absorption on the Mechanical Properties of Nano-Filler Reinforced Epoxy Nanocomposites. *Materials & Design*, **42**, 214-222. https://doi.org/10.1016/j.matdes.2012.05.060

[25] Shivakumar, H., Renukappa, N., Shivakumar, K. and Suresha, B. (2020) The Reinforcing Effect of Graphene on the Mechanical Properties of Carbon-Epoxy Composites. *Open Journal of Composite Materials*, **10**, 27-44. https://doi.org/10.4236/ojcm.2020.102003

[26] Tang, L.-C., Wan, Y.-J., Yan, D., Pei, Y.-B., Zhao, L., Li, Y.-B., *et al.* (2013) The Effect of Graphene Dispersion on the Mechanical Properties of Graphene/Epoxy Composites. *Carbon*, **60**, 16-27. https://doi.org/10.1016/j.carbon.2013.03.050

[27] Kim, H.S. and Khamis, M.A. (2001) Fracture and Impact Behaviours of Hollow Micro-Sphere/Epoxy Resin Composites. *Composites Part A*: *Applied Science and Manufacturing*, **32**, 1311-1317. https://doi.org/10.1016/S1359-835X(01)00098-7

[28] Delucchi, M., Ricotti, R. and Cerisola, G. (2011) Influence of Micro- and Nano-Fillers on Chemico-Physical Properties of Epoxy-Based Materials. *Progress in Organic Coatings*, **72**, 58-64. https://doi.org/10.1016/j.porgcoat.2011.02.018

[29] Gultekin, D., Uysal, M., Aslan, S., Alaf, M., Guler, M.O. and Akbulut, H. (2010) The Effects of Applied Load on the Coefficient of Friction in Cu-MMC Brake Pad/Al-SiCp MMC Brake Disc System. *Wear*, **270**, 73-82. https://doi.org/10.1016/j.wear.2010.09.001

[30] Tang, L., Xiong, J., Wan, W., Guo, Z., Zhou, W., Huang, S., *et al.* (2015) The Effect of Fluid Viscosity on the Erosion Wear Behavior of Ti(C,N)-Based Cermets. *Ceramics International*, **41**, 3420-3426. https://doi.org/10.1016/j.ceramint.2014.10.141