24th ABCM International Congress of Mechanical Engineering
December 3-8, 2017, Curitiba, PR, Brazil

COBEM-2017-2920
CNT AND GO EPOXY NANOCOMPOSITES- AN EXPERIMENTAL COMPARISON OF MECHANICAL PROPERTIES

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Abstract. *In this study, the mechanical properties of epoxy nanocomposites reinforced with graphene oxide (GO), non-functionalized carbon nanotubes (CNTs) and multi-walled amino functionalized carbon nanotubes (CNT-Am) were evaluated at two different weight fractions of 0.25 and 0.5%. The mechanical properties measured were ultimate tensile strength and Young's modulus in tensile tests and flexural strength and Young's modulus in flexural tests. Results show that nanocomposites reinforced with GO have more rigidity, presenting a higher tensile Young's modulus (up to 48%) and a lower flexural Young's modulus (-40%) than its CNT reinforced counterpart (up to 44% in tensile tests and -15% in bending) at 0.5% weight fraction. Also, non-functionalized CNTs have decreased both tensile and flexural properties of the epoxy matrix.*

Keywords: *carbon nanotubes, graphene oxide, nanocomposites, epoxy resin, mechanical properties.*

1. INTRODUCTION

Lately, researches related to nanoscience and nanotechnology have attracted significant interest from scientific and industrial communities, since the ability to manipulate materials at the nanoscale has opened a wide range of potential solutions in a variety of fields. Particularly, the development of nanocomposites has become of great interest in materials science. By dispersing strong and stiff nanomaterials in a polymer matrix we can obtain high-performance and lightweight composites that can be tailored to specific applications. Nanomaterials are generally regarded as high potential fillers to improve mechanical properties of polymers even at low content (Gojny *et al*., 2005; Wang and Liew, 2015).

Carbon nanotubes (CNTs) and graphene are carbon allotropes that have been frequently added to epoxy resin to increase its mechanical, electrical and thermal properties. The use of reinforcing fillers in epoxy resins can extend the range of its potential applications: adhesives, coatings, potting compounds, encapsulates, structural materials, liquid crystal displays (Liao *et al*., 2004; Eitan *et al*., 2003). But the aggregation of these nanomaterials can hinder the mechanical properties of nanocomposites: CNTs entangle and agglomerate easily due to its large aspect ratio and graphene platelets tend to restack (van der Waals and strong π-π interactions), making its dispersion a key note in the fabrication process (Disfani and Jafani, 2013; Li *et al*., 2008; Yang *et al*., 2011). Also, attaining a good interfacial interaction between nanofiller and polymer is crucial for an effective load transfer to occur (Li *et al*., 2013; Farrash *et al*., 2017).

To improve dispersion and secure the interfacial interaction, a usual approach is the chemical modification of the filler surface prior to its dispersion into the prepolymer (Xie *et al*., 2005; Zhu *et al*., 2003; Abdalla *et al*., 2007). For CNTs, amine functionalization can weaken the tube interactions (Farrash *et al*., 2017; Meng *et al*., 2008; Cividanes *et al*., 2013). For graphene, it is common to use an oxidized form (known as graphene oxide, GO), in which the oxygen-containing groups avoid the restacking of layers and can also interact chemically with the epoxy matrix; GO retains some of the layer structure of graphite but with a much larger and irregular spacing, presenting a C:O ratio between 2.1 and 2.9 (Hummers and Offeman, 1958). All these nanomaterials possess excellent mechanical properties due to their sp^2 carbon bonding network. But the key point is which of these fillers is best suited to efficiently assure load transfer from the polymer matrix in nanocomposites.

To address this matter, we obtained GO by a modified Hummers' method, CNTs by chemical vapor deposition and functionalized CNTs by acid and amino treatments. Nanocomposites based on epoxy resin were fabricated with these nanomaterials at weight fractions of 0.25 and 0.50 %. Mechanical properties of the neat resin and nanocomposites were compared through tensile and flexural tests.

2. EXPERIMENTAL

CNTs were obtained by chemical vapor deposition (CVD) and annealed at 1800 °C to remove impurities. They were functionalized by two chemical treatments: firstly, with a mixture of sulphuric and nitric acids followed by ethylenediamine. The amino functionalized CNTs were designated as CNT-Am. GO was synthesized by a modified Hummers' method (Wu *et al*., 2009).

Test specimens were prepared in two different shapes according to tensile and flexural test requirements. They were made without nanoparticles (i.e. neat epoxy resin), with GO and with CNT-Am. Nanocomposites were prepared with two different weight fractions of filler (0.25 % and 0.5 %). In addition, a set of non-functionalized (only thermally treated by the annealing process) CNTs nanocomposites was prepared with 0.25 % wt. The bisphenol-A-based thermosetting epoxy (DGEBA) used in this work was Araldite GY-260 and the diaminodiphenylmethane (DDM) hardener was Aradur 972, both supplied by Huntsman. Hardener was used at 27 % weight fraction of the neat resin for all samples.

To achieve a more homogeneous dispersion, nanomaterials were each mixed with acetone by bath sonication for 30 minutes. Then the solutions were poured on the epoxy resin at 65 °C (to reduce viscosity) and the mixture underwent tip sonication (Hielscher UP200Ht 500 W, frequency of 42 kHz) with 40% of the total power (200W) and duty cycle of 100% (continuous mode) for 30 minutes. As the sonication process heats up the mixture, its temperature was kept stable by a jacketed beaker connected to a thermal bath.

The resulting mixtures were degassed under vacuum at 80 °C for 30 hours to eliminate acetone and avoid bubble defects in the final nanocomposites. The hardener was added to each mixture and heated to 90 °C to melt down. The final mixture was poured over silicon molds designed to the shapes required for mechanical tests. Curing process was held in two stages: at 80 °C for 1 hour and at 120 °C for 2 hours, according to the manufacturer's instructions. Figure 1 illustrates this process schematically. The specimens were carefully measured and any excess material was removed by sanding. Specimens that could not meet the measuring tolerance for testing were discarded.

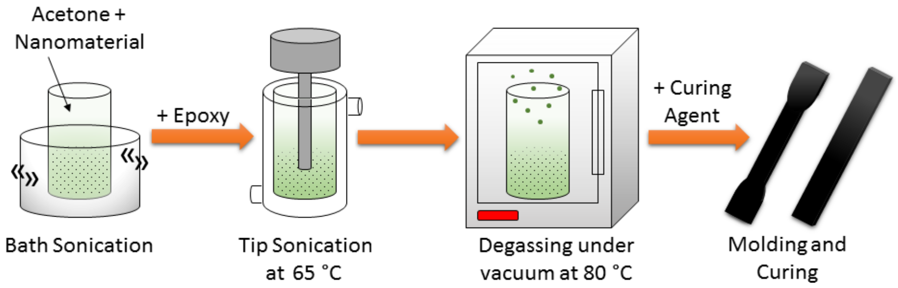

Figure 1. Schematic showing the dispersion of nanomaterials in the epoxy matrix by solution mixing with high amplitude ultrasonic agitation.

The neat resin and the nanocomposite samples were fabricated as dog-bone-shaped specimens for uniaxial tensile testing. Their measures are according to ASTM D638 type I for reinforced composites (Fig. 2). Tensile tests were performed in an Instrom 5500R with a load cell of 30 kN. Specimens that didn't break within the gage sensor range or that broke outside of the narrow cross-sectional test section (dimension L in Fig. 2) were discarded.

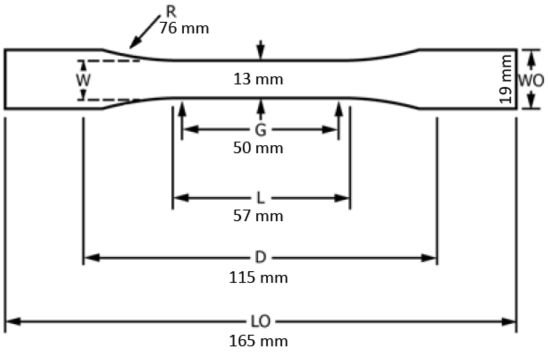

Figure 2. Type I specimen dimensions for rigid and semirigid plastics and reinforced composites for thickness of 3.2 ± 0.4 mm.

Samples were also fabricated as bar-shaped specimens for flexural tests on a three-point loading system applied to a simply supported beam. Their measures are according to ASTM D790, a flat bar of 127 x 12,7 x 3,2 mm. Flexural tests were performed in a MTS E45.105 with 6 mm radii cylindrical loading nose and supports, and a support span of 3.8 mm. All mechanical tests were performed at 0.25 mm/min strain rate and considering the nanocomposites as isotropic materials.

SEM images were obtained in a Tescan VEGA3 and in a FEI Magellan 400 L. It was used to examine CNT-Am and GO morphology as well as the fracture surfaces of the nanocomposites. Samples were mounted on a standard specimen holder using double-sided carbon conductive tape with the fracture surfaces toward the electron beam. For fracture surface images, samples were exposed to liquid nitrogen and broken with the help of pliers. Actual samples from mechanical tests could not be used due to contamination from the moment of breakage, since they break quite violently and fall off from the testing machines.

3. RESULTS AND DISCUSSION

Figure 3 shows SEM micrographs of non-functionalized CNT, functionalized (CNT-Am) and GO. Non-functionalized CNTs can be seen in blocks of vertically aligned tubes with approximately 300 µm length. CNT-Am shows free tubes with some tangling, but it is clear that the basic structure of the tubes from non-functionalized CNTs was preserved. The appearance of GO resembles naturally wavy nanosheets morphology type, weekly held as a foam-like structure that can be separated into smaller or even individual groups by sonication in acetone. GO is naturally observed to agglomerate, and also due to the process of SEM imaging, which required the sample to receive a few drops of ethanol and be quickly dried in order for the nanomaterial to adhere to its sample holder. The surface of GO is not perfectly flat, presenting a rich-wrinkled morphology that can be seen in samples prepared by CVD and arc discharge as well (Su *et al*., 2011; Kumar *et al*., 2013).

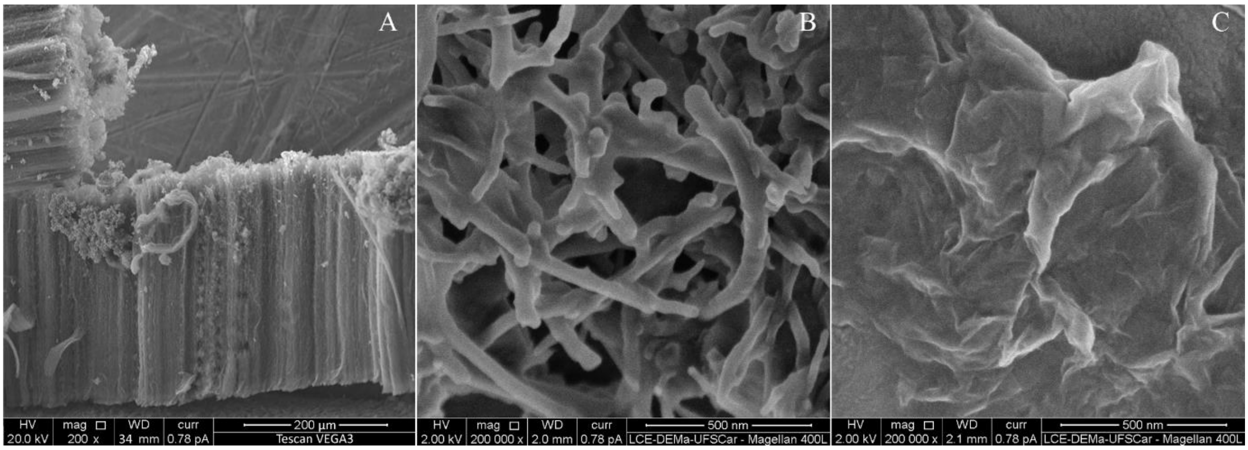

Figure 3. SEM images of: (a) non-functionalized CNT produced by CVD; (b) amino functionalized CNT-Am; and (c) GO obtained by a modified Hummer's method.

The results of uniaxial tensile testing of the neat epoxy and nanocomposite samples are shown in Table 1. Results show that mechanical strength is very dependent on the presence of nanomaterials in the composites. The incorporation of CNT-Am and GO increased the tensile properties of the epoxy resin making it more rigid, which can be attributed to a good dispersion in the matrix and to the strong adhesion of these nanofillers to the epoxy resin (Cividanes *et al*., 2017), while non-functionalized CNTs decreased its UTS and Young's modulus. It's clear that adding 0.25 % wt. of GO into the epoxy resin yields the most effect on the tensile Young's modulus and ultimate tensile strength (UTS) of the nanocomposite although the addition of 0.50 % wt. of GO resulted in the highest tensile properties observed in this study. Lower filler contents can ensure a relatively uniform dispersion (Rafiee *et al*., 2009) and is one possible explanation to the smaller increase in tensile properties observed at 0.50% weight fraction for CNT-Am and GO when compared to the same nanomaterials at 0.25% weight fraction.

Table 1. Comparison of tensile mechanical properties of nanocomposites (ASTM D638). Average results of 5 specimens per group.

Sample group	**Filler content wt. (%)**	**Mean ultimate tensile strength (MPa)**	**Comparison with neat resin (%)**	**Mean Young's modulus (GPa)**	**Comparison with neat resin (%)**
Neat resin	0	46.39 ± 0.67	-	3.39 ± 0.13	-
Non-functionalized CNT	0.25	42.60 ± 3.45	- 8.17	2.97 ± 0.41	- 12.41
CNT-Am	0.25	63.02 ± 1.36	+ 35.85	4.66 ± 0.51	+ 37.57
	0.50	65.58 ± 2.86	+ 41.36	4.88 ± 0.32	+ 44.13
GO	0.25	65.39 ± 1.92	+ 40.97	4.86 ± 0.39	+ 43.46
	0.50	67.94 ± 1.87	+ 46.46	5.04 ± 0.37	+ 48.96

Overall, the toughening effect of GO in the epoxy matrix is superior to CNTs. This is in agreement with the observed flexural properties of studied nanocomposites, which decreased according to the increase in rigidity of samples as seen on tensile tests. Table 2 reports the flexural strength and in-bending Young's modulus obtained for the neat resin and its nanocomposites.

Table 2. Comparison of flexural mechanical properties of nanocomposites (ASTM D790). Average results of 5 specimens per group.

Sample group	**Filler content wt. (%)**	**Mean flexural strength (MPa)**	**Comparison with neat resin (%)**	**Mean Young's modulus (GPa)**	**Comparison with neat resin (%)**
Neat resin	0	174.92 ± 17.89	-	3.15 ± 0.56	-
Non-functionalized CNT	0.25	164.40 ± 12.89	- 6.01	3.01 ± 0.23	- 4.17
CNT-Am	0.25	159.39 ± 19.16	- 8.88	2.85 ± 0.65	- 9.32
	0.50	152.02 ± 27.75	- 13.09	2.65 ± 0.22	- 15.64
GO	0.25	122.50 ± 35.71	- 29.96	2.08 ± 0.48	- 33.95
	0.50	105.23 ± 17.61	- 39.84	1.89 ± 0.30	- 40.06

The non-functionalized CNTs have presented a negative effect on tensile properties, due to a bad dispersion and its association with void-like defects in the epoxy matrix, which happen when the filler is unable to chemically interact with the matrix, therefore resulting in inefficient or non-existent load transfer. It also decreased the epoxy matrix flexural properties, probably because its presence may cause some hindrance to the linking of polymer chains, making them slip more easily and amounting to lower flexural properties.

GO nanocomposites presented higher tensile properties and lower flexural properties than CNT-Am reinforced samples, leading to the assumption that GO had a stronger interfacial adhesion with DGBA matrix and/or more homogeneous dispersion. According to Yu *et al*. (2016), the presence of many polar moieties such as carboxylic acid, amide, and amine groups can facilitate the dispersion of nanoparticles into the epoxy matrix because they actively interact with the epoxy via hydrogen or covalent bonding. Therefore, the dispersion in CNT-Am and GO nanocomposites could be considered equivalent to some extent, and the higher increase in tensile properties seen for GO specimens can be attributed to a stronger interfacial adhesion. According to Tiwari *et al*. (2017), the high surface area of GO increases the interfacial interaction area thus improving the load transfer. Also, the improved adhesion of GO can be justified by the atomic concentration of oxygen (35.1%) and nitrogen (1.1%) in this nanomaterial, against 5.1% oxygen and 0.9% nitrogen found in CNT-Am as seen by XPS investigations previously reported (Moraes *et al*., 2017).

Although there is a harsh difference in atomic concentrations of oxygen between CNT-Am and GO, tensile properties were not that different. Comparing their respective nanocomposites at same filler content (0.25%) to the neat resin, there was an increase of 37.57% of tensile Young's modulus for CNT-Am, while GO increased it by 43.46%. To elucidate this point, it is suggested that when terminal amine groups on CNT-Am react with the epoxide groups of the resin, CNT-Am can participate in the formation of the polymer crosslink network like the curing agent (Yu *et al*., 2016) in a more efficient way than other oxygen-containing functional groups attached by simple oxidation (acid treatments). In addition, Zehua and Guaijan (2012) evaluated epoxy-based nanocomposites reinforced with pristine multi-walled CNTs and CNT functionalized with EDA, TETA, and DDA; they concluded that the interfacial strength between filler and matrix enhances more the mechanical properties than a good dispersion.

Zhang *et al*. (2013) have studied the tensile properties of functionalized graphene and CNTs at 5% wt. in poly(ether sulfone) based nanocomposites, reporting an increase of approximately 30% in tensile Young's modulus for CNT reinforced composites and of approximately 70% for graphene reinforced composites. Yu and coworkers (2016) used amino functionalized GO to improve mechanical properties of epoxy and obtained an increase of 26% in tensile strength. Rafiee *et al*. (2009) compared the tensile properties of CNTs and reduced GO epoxy nanocomposites, finding that reduced GO out-performs the CNTs as fillers increasing the tensile strength of the epoxy matrix in approximately 40%, while the latter increased it only 14%.

Other works in literature reveal different results. Tang *et al*. (2013) have studied the effect of reduced-GO in epoxy composites, observing an increase in both tensile and flexural properties at different filler loadings of up to 0.2% wt. Despite being able to increase both sets of properties, the tensile Young's modulus increased only approximately 6%, and the flexural modulus by 14% when compared to the neat resin, which can be due to two main factors: the weak interfacial adhesion between reduced-GO and the epoxy matrix (Tang *et al*., 2013) and also the fact that weak van der Waals forces exist between the individual sheets of the graphene agglomerates, where slipping between the sheets may occur (Chen and Lu, 2012) and the graphene agglomerates may lead cracks to initiate and propagate easily, therefore producing a reduced strength in nanocomposites (Zaman *et al*., 2012).

In another work, Kavita *et al*. (2016) have modified a novolac epoxy resin with thermoplastic poly(vinyl butyral) (PVB) to achieve more flexibility and used several different weight fractions (0.1 – 0.5%) of acid functionalized CNT to counter the reduction in the thermal stability caused by the addition of PVB. In this case, a reduction of up to 60% in UTS and tensile Young's modulus was obtained due to the presence of PVB, but the expected gain in flexural properties was not met; flexural modulus decreased up to 65% and flexural strength decreased up to 70%. Investigations showed heterogeneous distribution of PVB in the continuous novolac epoxy matrix; also, the presence of CNTs was observed to be greater in the PVB phase compared to the epoxy phase, which led to agglomerates and inferior mechanical properties at higher CNT loadings.

Figure 4 shows SEM images of cryogenic fracture surfaces of neat epoxy resin and non-functionalized CNTs, CNT-Am and GO nanocomposites at 0.25% wt. The fracture mechanism observed for the neat resin and nanocomposites was governed by brittle failure. The neat resin shows a smooth, quite featureless fracture surface with low deviation in crack growth, which is an indicative of the brittleness of the sample (Konnola *et al*., 2015). For non-functionalized CNTs sample, brittle striations and several pullout nanotubes were observed. This is a characteristic behavior of tubes that were not chemically attached to the epoxy matrix (Luo *et al*., 2012). It is clear to see the entanglement of CNTs reflected in a cluster of non-homogeneous dispersion. At the moment of breakage, these CNTs acted like a void defect on the polymer, lowering its toughness and resulting in an early breakage.

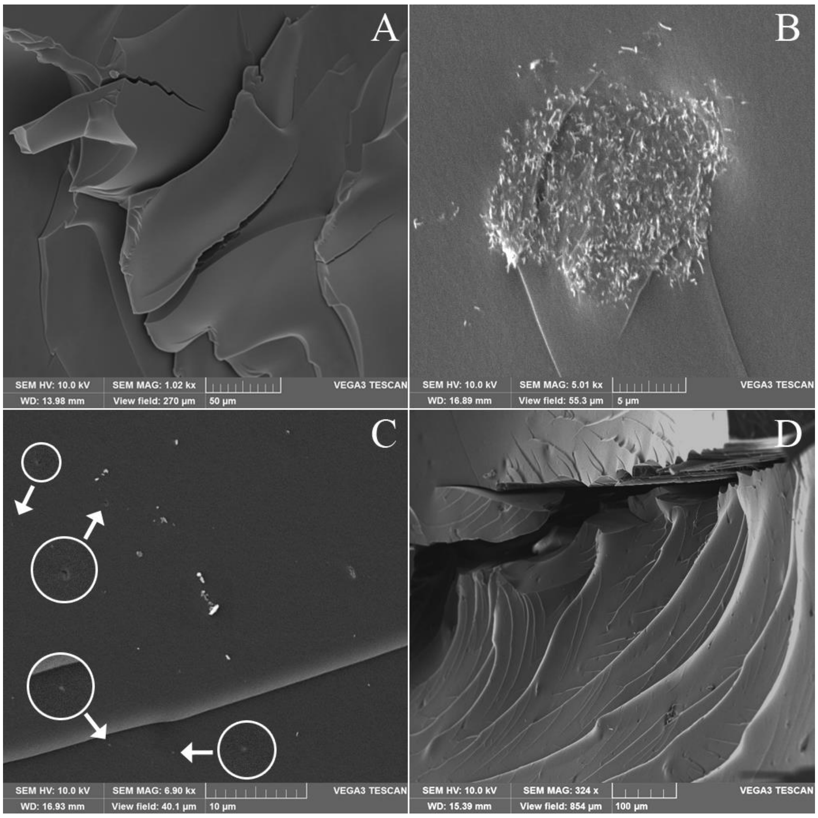

Figure 4. SEM images of cryogenic fracture surface of (a) neat epoxy resin; (b) non-functionalized CNT; (c) CNT-Am and (d) GO nanocomposites at 0.25 % wt.

For CNT-Am nanocomposite it is possible to see a few holes suggesting that the tubes broke along with the epoxy resin due to their chemical bonding. Also, no clusters of entangled CNTs were observed. It was not possible to precisely identify GO particles in their nanocomposite. According to Rafiee et al., at low weight fractions of graphene (below 0.5%) it becomes challenging to observe its dispersion by SEM analysis due to the planar geometry of this molecule and the epoxy coating on the graphene, which allows only the exposed platelet edges to be discernible (Rafiee *et al.*, 2009). However, discernible changes could be seen when compared to the neat epoxy fracture surface. The rougher surface is likely result of an enhanced mechanical interlocking with the polymer chains (due to the wrinkled sheet morphology and surface functionality), leading to a better adhesion between the GO filler and the epoxy matrix than CNTs (Ramanathan *et al.*, 2008).

4. CONCLUSIONS

In this work, we have studied the synthesis and functionalization of carbon nanotubes and graphene oxide. Morphology of non-functionalized CNT, CNT-Am and GO was observed by SEM images. These materials were compared as fillers for DGEBA epoxy resin by tensile and flexural mechanical tests, and GO nanocomposites presented higher UTS and tensile Young's modulus, as well as the highest decrease in flexural properties, which is expected once the material becomes more rigid. Non-functionalized CNTs have presented a negative effect on both tensile and flexural properties, due to its inability to chemically bond with the epoxy resin and the hindrance of crosslinking chains. CNT-Am performance can be attributed to its amino functionalization which improves dispersion and enables the tubes to bond with the polymer crosslink network. The outstanding tensile properties seen on GO nanocomposites can be explained by the high surface area of this filler as well as the massive atomic concentration of oxygen in it. It is suggested that amino functionalized GO could reach even higher toughness. The results presented can be considered beneficial for the manufacture of structures and components with higher strength-to-weight ratios.

5. ACKNOWLEDGEMENTS

The authors gratefully acknowledge FAPESP and CNPq for financial support and LAS/INPE, LCE/DEMa/UFSCar for collaboration.

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