

MECHANICAL AND PHYSICAL PROPERTIES OF EPOXY COMPOSITES CONTAINING CFRP POWDER WASTES

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Abstract

In this research, carbon powder wastes obtained from the cutting process of laminate composites have been incorporated into epoxy matrix phase in order to improve the mechanical characteristics. Physical and mechanical properties and thermal behavior have been analyzed. Results show that carbon powder wastes incorporated to new epoxy matrix phases act basically as reinforcement. This allows for the recycling of the residues as well as improves some properties of the composites.

1. Introduction

The industry of composites generates undesirable wastes and contaminants into the environment, which need to be treated so that they would cause minimum damage prior to disposal. The disposal of solid wastes, such as resin remains, remnants of sheets and other components, has been a problem all around the world and landfilling is not a desirable option due to the future environmental protection costs and associated problems [1,2].

Polymer–matrix composites dominates the market, among which thermoset composites account for more than two thirds. Two major application sectors (based on value) are automotive industry (over 30%) and aerospace industry (over 20%). Extensive recycling activities have been conducted, and various technologies yet to be commercialized [3,4], have been developed basically in three categories: mechanical recycling, thermal recycling, and chemical recycling. But Land-fill disposal is nowadays the most common way for waste composite material causing an environmental problem because of the great volume of material long life of these wastes. Furthermore, governments and local councils are limiting or completely banning the disposal of composites by land fill-in [5]. For example, the European Union recently set limits on the amount of composites placed in land-fill and Germany has imposed a complete ban.”

The company Carbontek S.L. [6] has began to add the powder waste from the cutting process of carbon fibre composites into the matrix phase, in order to (i) reduce the volume of generated wastes and (ii) improve the mechanical behaviour of the new composites, which were used to fabricate diving carbon fins. The results are presented in this paper, which evaluates the effect of the incorporation of different percentages of this powder waste on the

thermal and mechanical properties of recycled compounds. Results also indicate that the powder from the cutting process of carbon fibre components can be used as a new reinforcing phase in hybrid composites improving the mechanical characteristics of such materials.

Most of the discussed treatments, mechanical recycling [7], pyrolysis [1], chemical recycling [8], require quite expensive processing of the waste. In this paper a proven method with cheap process for CFRP recycling wastes is proposed [9].

2. Materials and Methods

The carbon powder wastes (CPW) were obtained from the cutting process of laminate composites. CPW were incorporated into the epoxy matrix phase. Fig. 1 shows a pair of diving carbon fins after the cutting and assembly process.

The cutting process take place inside a hood and the generated powder is collected by the use of filters. Figures 2 and 3 show, respectively, the leftovers and the collected powder after grinding. Usually, the leftovers are taken directly to landfill. The powder from the filter was classified by sieving, in order to remove the large particles which could hinder the mixing procedure. The passing material under 1.25 mm was used. No further treatment is required, since the main objective is to avoid increasing manufacturing costs.



Figure 1. CarbonTek carbon fins for diving, freediving and spearfishing.



Figure 2. Rest of cutting of the carbon fiber components.



Figure 3. Collected powder after cutting carbon fiber components.

The ratio by weight of epoxy resin/carbon fibre has been determined by combustion in an inertial atmosphere. It has been found that in a week production of a specific model of carbon fins, the percentage of pure carbon fibre in the CPW ranges between 15.1% and 15.8%.

The geometrical characteristics of the carbon tubes have been determined by SEM. Figure 4 and 5 show the carbon tube dimensions: 30 μm in length (a) and 6 μm in diameter (b). Based on the microstructural analysis the mean of length and diameter was found at 45 μm and 6 μm , respectively.

Once the CPW was characterized, two different additions into the epoxy matrix phase were carried out and characterized. The epoxy resin (SR 8500) was supplied by [10] Sicomin company [11] with two different hardeners, SD 8605 and SD 8601 in order to achieve a fluid viscosity of 1500 mPa·s, using the weight proportions of 100:25:10. The CPW additions were 10 %wt. and 20 %wt. The CPW were mixed during 8 minutes, with subsequent curing period

of 24 h at 22±5 °C and followed by 6 h at 60±2 °C. Table 1 shows the mixtures of epoxy resin with different percentages of CPW.

Code	Description	Epoxy SR 8500	Hardener SD 8605	Hardener SD 8601	Waste CPW
ER-0W	Epoxy resin	100.0	25.0	10.0	0
ER-10W	Epoxy resin with 10% waste	92.6	23.1	9.3	10
ER-20W	Epoxy resin with 20% waste	85.2	21.3	8.5	20

Table 1. Description and mix design proportions by weight.

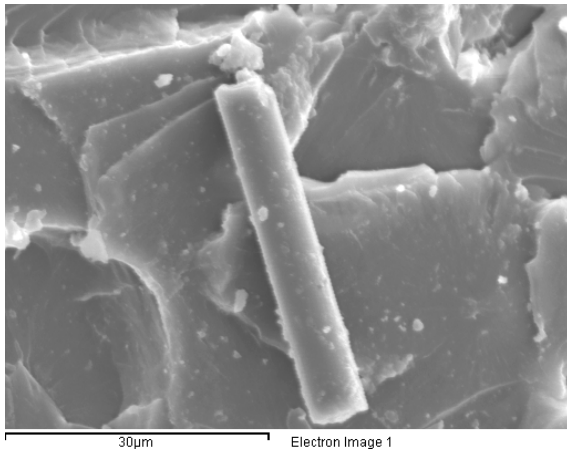


Figure 4. Microanalysis of the powder: 30 mm length carbon tube.

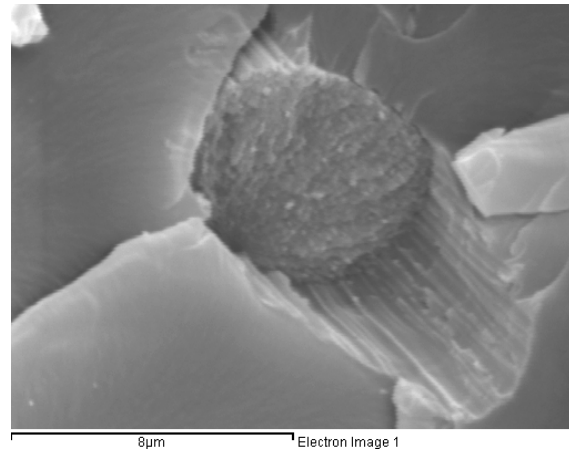


Figure 6. Microanalysis of the powder: 6 mm diameter carbon tube

2.1. Physical properties and thermal behavior

Three prismatic specimens of 40×40×160 mm for each experimental condition were used to determine the bulk density of the composites.

TGA and DSC were carried out using a TGA-DSC SETARAM SETSYS Evolution equipment, using a heating rate of 20 °C/min in an inert atmosphere.

Mechanical properties

2.2. Mechanical properties

The three-point bending test [12] was carried out based on the recommendations of EN 1015-11 [13]. Six prismatic samples (40×40×160 mm) for each experimental condition were tested. The flexural tests were done without strain record so flexural modulus results are not presented. Twelve cubic specimens of 40 mm were manufactured and tested under compression according to EN 1015-11 [13]. In addition, three other samples were used to measure the modulus of elasticity in compression with strain gauges, following the recommendations of UNE-83316 [14]. A fractographic evaluation by SEM was conducted in order to assess the flexural failure surfaces.

3. Results and discussions

3.1. Manufacturing

The mixing of epoxy resin and CPW up to 20 %wt. provided a remarkable increase in the fluid viscosity. Due to the significant loss of fluidity, additions over 20 %wt. have not been considered. However, the possibility of adding a percentage greater than 20 %wt. of CPW into the epoxy matrix is not discarded. In that case, a longer time of mixing or sonication process [15] is demanded to ensure proper distribution/dispersion of the residue in the matrix phase. The use of SR 8500 resin demands the use of a higher percentage of SD 8601 hardener (with less viscosity), consequently, large amount of wastes can be added with no effect on the material's viscosity.

3.2. Physical properties and thermal behavior

The bulk densities of the composites are $1.135 \pm 0.003 \text{ g/cm}^3$ for the epoxy resin, $1.148 \pm 0.004 \text{ g/cm}^3$ for the ER-10W and $1.191 \pm 0.002 \text{ g/cm}^3$ for the ER-20W; it is possible to note that, as the amount of CPW increases, so does the bulk density. Figure 8 shows the thermogravimetric analysis up to 1000 °C of the considered formulations compared with the TG curve of the CPW.

Figure 9 shows the differential scanning calorimetry (DSC) analysis up to 1000 °C of the formulations studied. The first exothermic peak corresponds to the onset of decomposition which is situated nearly at 275 °C. However, the major peak, corresponding to the total decomposition, is dependent on the level of CPW addition.

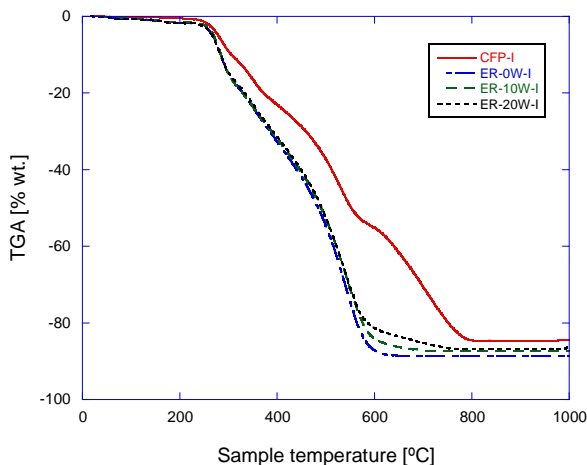


Figure 8. TGs up to 1000 °C of the considered formulations compared with the CPW.

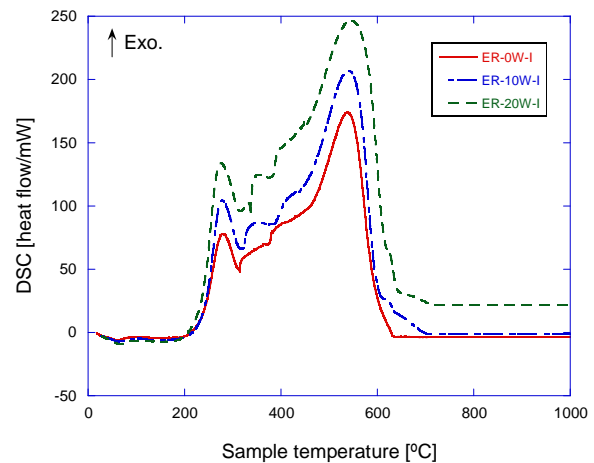


Figure 9. DSCs up to 1000 °C of the considered formulations.

All samples started to decompose around 275 °C (first stage); this mass loss corresponds to the epoxy resin matrix, which is presented in all formulations. The thermal conductivity and the increase of the heat diffusion of the incorporated CPW [16] has no significant effect on the onset temperature of decomposition. However, the final decomposition took place between 600 °C and 800 °C, depending on the level of CPW addition. The increase of temperature and the loss of mass at total decomposition are proportional to the percentage of CPW in the composite. The TGA curves of pure epoxy and reinforced composites reveal that the decomposition of materials shifted at high temperatures with increasing CPW given that

carbon offers a stabilizing effect against the decomposition of the epoxy matrix. Similar effects have been observed in carbon nanotube composites, which might be attributed to radical capture by the nanotube surface and by the interactions between the carbon fibres [17-19].

3.3. Mechanical properties

Table 2 shows the results of the compressive strength, flexural strength and modulus of elasticity. It is found that the CPW addition increases all mechanical properties. The addition of 10 %wt. of CPW (1.54 %wt. of carbon with respect to total weight) provided a 6% percent increase in the compressive strength. When 20 %wt. of CPW is added (3.08 %wt. of carbon) the compressive strength is nearly 20% higher than the strength of the pure resin. So it possible to conclude that, as a result of the waste incorporation, more effective load transfer to the epoxy matrix is achieved. The strain at the compressive failure is 3.1 % for the ER-0W, 3% for the ER-10W and 2.86% for the ER-20W.

Code	Compressive strength [MPa]	Flexural strength [MPa]	Compressive modulus [GPa]
ER-0W	98.42	3.424	3.179
ER-10W	104.52	3.961	3.486
ER-20W	118.29	4.477	4.137

Table 2. Results of the mechanical tests.

It is generally accepted that the increase of the filler volume fraction into resin-based materials enhance the flexural behaviour [20], assuming that the integrity is maintained between the resin matrix and filler particles [21]. In this work, the flexural strength increased considerably more than the compressive strength. The flexural strength increased nearly to 14% and 30%, respectively, when 10 %wt. and 20 %wt. of CPW were added, compared to the pure resin data. By contrast, the large shift in compression compared to tension indicates that the nanotubes carry more strain in compression than in tension because those oriented at an angle to the loading direction are easily loaded in compression [22]. The difference in the response in tension and compression must be due to the load transfer behaviour of fiber in tension compared to that in compression because of the relatively weak bonding between the fibre layers, load is not transferred to the inner layers [23].

The modulus of elasticity exhibited a direct correlation with the CPW additions. The addition of 10 %wt. and 20% wt. of CPW provided an increase of modulus of elasticity of nearly 10% and 30%, respectively. This result is in agreement with other studies [20,24] in which it is found that the particle incorporation increases the compound stiffness.

Figure 10 and 11 show the specimens after the compressive and flexural tests. Samples of compressive strength show a correct distribution of load during the test. The specimens tested in tension were observed with a scanning electron microscope. The fractured surfaces of composites with 0 %wt., 10 %wt. and 20 %wt. waste incorporation show a rise in the surface roughness with CPW additions, as well as a few dimples which reveal ductile characteristics.

River lines and the fracture surface between the river lines are relatively smooth, which are the characteristics of brittle fracture behaviour [25], indicating that the carbon tubes of the CPW can improve the toughness of the composite. The pulling out of fibres confirms that the bond between the old epoxy on the CPW and the new epoxy matrix is adequate.

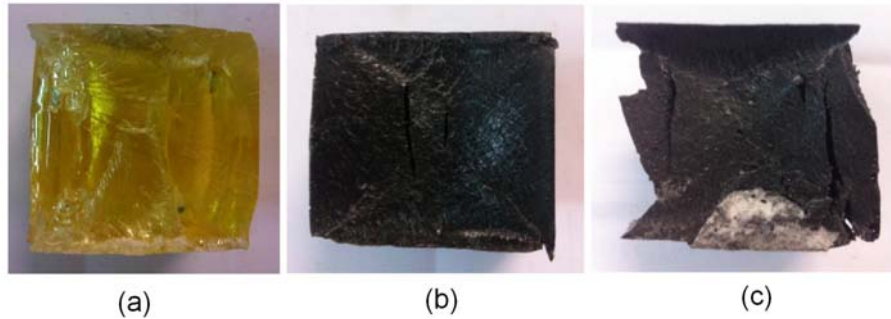


Figure 10. Specimens of (a) ER-0W (b) ER-10W and (c) ER-20W specimens after the compressive strength test.

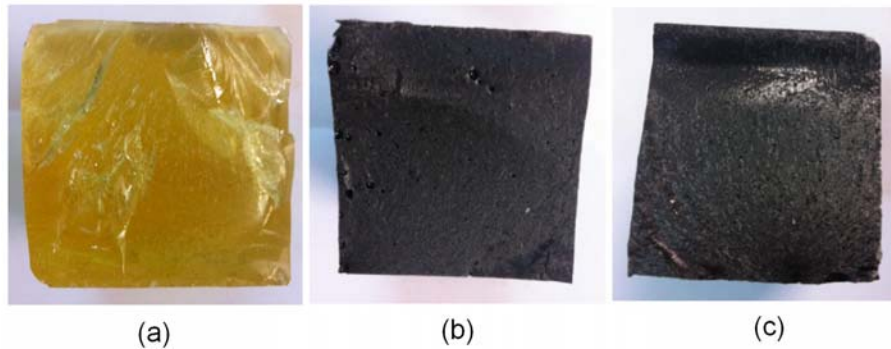


Figure 11. Fracture surface of a flexural strength test of (a) ER-0W (b) ER-10W and (c) ER-20W specimens.

The addition of increasing amount of CPW provide a more rigid and stronger composite because the increased reinforced surface implies that the path of the crack tip is distorted by the CPW fiber, making the stress and crack propagation more difficult. Also, the fracture surfaces show good bond between the epoxy resin of the CPW and the new matrix. So the CPW incorporation has a similar effect that the incorporation of carbon filler in an epoxy matrix. However, to ensure a proper bond between the new matrix and CPW, it must not be contaminated in surface.

4. Conclusions

Based on this research, the following conclusions can be obtained:

- The incorporation of CPW to the epoxy resin provides a loss of fluid viscosity. It is not recommended that the CPW addition exceed 20% wt. However, this flow loss can be compensated using resins or hardeners with more fluidity. The CPW addition has changed the epoxy resin to black colour even when small amount is added.
- The CPW addition into the epoxy resin provides a slight increase in the density of the composites and there is no effect on the initial temperature of decomposition. However, the carbon tubes provided a stabilizer effect increasing the temperature of total decomposition.

- The incorporation of CPW improves the characteristics of the composite, especially the compressive strength and flexural strength, which proportionally increased with the CPW addition.

As final conclusion, it was possible to incorporate a powder residue from the cutting process of carbon fibre composites in order to reinforce a new epoxy matrix phase. This allows for the recycling of the residues as well as improves some properties of the composites.

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References

- [1] Pickering SJ. Recycling technologies for thermoset composite materials—current status. *Composites Part A: Applied Science and Manufacturing* 2006 8;37(8):1206-15.
- [2] Khale D, Chaudhary R. Mechanism of geopolymerization and factors influencing its development: A review. *J. Mater. Sci* 2007;42:729-46.
- [3] Hadeg-recycling. Available at: <http://www.hadeg-recycling.de/>. 2014
- [4] ELG Carbon Fibre Ltd. <http://www.recycledcarbonfibre.com/>. 2014.
- [5] Feih S, Boiocchi E, Mathys G, Mathys Z, Gibson AG, Mouritz AP. Mechanical properties of thermally-treated and recycled glass fibres. *Composites Part B: Engineering* 2011 4;42(3):350-8.
- [6] CarbonTek S.L. <http://www.carbontek.es>. 2014.
- [7] Yang Y, Boom R, Irion B, van Heerden D, Kuiper P, de Wit H. Recycling of composite materials. *Chemical Engineering and Processing: Process Intensification* 2012 1;51(0):53-68.
- [8] Dang W, Kubouchi M, Sembokuya H, Tsuda K. Chemical recycling of glass fiber reinforced epoxy resin cured with amine using nitric acid. *Polymer* 2005 2/24;46(6):1905-12.
- [9] Thomas C, Borges PHR, Panzera TH, Cimentada A, Lombillo I. Epoxy composites containing CFRP powder wastes. *Composites Part B: Engineering* 2014;59:260-8.
- [10] SR 8500 epoxy resin Technical file. <http://www.mcmc-uk.com/prod-data-sheet/sr-8500-860x-uk.pdf>. 2012.
- [11] SICOMIN Epoxy Systems. <http://www.sicom.in.com>. 2012.
- [12] RILEM CPT PC-8. Method of test for flexural strength and deflection of polymermodifiedmortar. TC 113. London ; 1995.
- [13] EN 1015-11:2000. Methods of test for mortar for masonry - part 11: determination of flexural and compressive strength of hardened mortar.
- [14] UNE-83316. Concrete tests. Determination of the modulus of elasticity in compression. 1996.

- [15] Gkikas G, Barkoula N-, Paipetis AS. Effect of dispersion conditions on the thermo-mechanical and toughness properties of multi walled carbon nanotubes-reinforced epoxy. *Composites Part B: Engineering* 2012 9;43(6):2697-705.
- [16] Ciecierska E, Boczkowska A, Kurzydłowski K, Rosca I, Van Hoa S. The effect of carbon nanotubes on epoxy matrix nanocomposites. *Journal of Thermal Analysis and Calorimetry* :1-6.
- [17] Choi Y, Sugimoto K, Song S, Gotoh Y, Ohkoshi Y, Endo M. Mechanical and physical properties of epoxy composites reinforced by vapor grown carbon nanofibers. *Carbon* 2005 8;43(10):2199-208.
- [18] Troitskii BB, Troitskaya LS, Yakhnov AS, Lopatin MA, Novikova MA. Retardation of thermal degradation of PMMA and PVC by C60. *European Polymer Journal* 1997;33(10-12):1587-90.
- [19] Goh HW, Goh SH, Xu GQ, Pramoda KP, Zhang WD. Dynamic mechanical behavior of in situ functionalized multi-walled carbon nanotube/phenoxy resin composite. *Chemical Physics Letters* 2003;373(3-4):277-83.
- [20] Wetton RE. Dynamic mechanical method in the characterisation of solid polymers. *Polym.Test.* 1984;4(2-4):117-29.
- [21] Ikejima I, Nomoto R, McCabe JF. Shear punch strength and flexural strength of model composites with varying filler volume fraction, particle size and silanation. *Dental Materials* 2003 5;19(3):206-11.
- [22] Lourie O, Wagner HD. Evaluation of Young's modulus of carbon nanotubes by micro-Raman spectroscopy. *J.Mater.Res.* 1998;13(9):2418-22.
- [23] Schadler LS, Giannaris SC, Ajayan PM. Load transfer in carbon nanotube epoxy composites. *Appl.Phys.Lett.* 1998;73(26):3842-4.
- [24] Masouras K, Silikas N, Watts DC. Correlation of filler content and elastic properties of resin-composites. *Dental Materials* 2008 7;24(7):932-9.
- [25] Yu Z, You S, Baier H. Effect of organosilane coupling agents on microstructure and properties of nanosilica/epoxy composites. *Polymer Composites* 2012;33(9):1516-24.