

Synthesis and Characterization of Hybrid CF/MWCNT_s/Epoxy Resin Composite System

Hany Fouda^{1*}, Lin Guo, Yonghai Yue¹, Ke Chen¹ and Karim Elsharkawy¹

¹School of Chemistry and Environment, Beijing University of Aeronautics and Astronautics, Beijing, China

*E-mail: hanyfouda@buaa.edu.cn

Abstract. In the present investigation, two methods were used for addition multiwall carbon nanotubes (MWCNT_s) into carbon fiber (CF)/epoxy resin composite system. The mechanical properties of the prepared samples were compared to show the best method for addition of MWCNT_s from point of view of mechanical properties. The introduction of carbon nanotubes (CNTs) into fiber reinforced polymer composites has been achieved mainly via two routes: mixing CNTs entirely throughout the matrix (matrix modification) or attaching CNTs onto reinforcing fibers (interface modification). In all previous references the addition of CNTs occur through one route from the two routes but in this research, we introduced MWCNT_s into CF/epoxy resin composite through the two routes at the same time. Three CF composite samples were prepared CF/epoxy resin composite (C1), CF/ 1wt% MWCNT_s /epoxy resin composite (C2) in which MWCNT_s added via one route (epoxy resin system) and the third sample was CF/ 1wt% MWCNT_s / epoxy resin composite (C3) in which MWCNT_s added via two routes (epoxy resin and CF fabric). The result shows that the mechanical properties of C3>C2>C1, for example, the flexural strength of C3 higher than C2 by 19% and C2 higher than C1 by 51% respectively. This is because addition MWCNT_s via two routes increase the ability of good mixing of CNT_s with epoxy resin and good dispersion of CNTs into the CF fabric surface and this leads to improve the interface bonding between the CF and epoxy so improve the mechanical properties.

1. Introduction

Many types of filler can be used with resin matrix composite materials but the most important one is Carbon fiber CF and this is because its excellent engineering properties, such as high specific stiffness and strength, performance to weight ratio, high thermal stability, high conductivity, corrosion resistance and self-lubrication. Most obviously, CF used in the structural application that needs weight reduction of the equipment as vehicles industries due to its high strength to weight ratio. CF reinforced resin matrix composites have a lot of applications as in aerospace applications, turbomachinery, for wind turbines applications in automotive energy systems, fuel cells, compressed gas storage and transportation, antistatic and electromagnetic shielding applications. the carbon fibers reinforced polymer matrix composites is widely used in automobile industries, aerospace, and marine during the last few decades due to their good engineering properties such as high specific strength and stiffness, lower density [1-5]. In The composite material system carbon fibers give strength and stiffness while resin matrix provides the chemical and thermal resistance of the composite [4].

CF structures have crystallized graphitic basal planes with a non-polar surface. CF is chemically inert this is because of the presence of high-temperature carbonization/graphitization step during the



manufacturing process, CF has weak bonding with the matrix surface due to lipophobicity, and excessive smoothness and fewer adsorption characteristics of CF. due to the inertness of CF, improving at the fiber surface is needed in order to improve the interfacial bonding between fiber/matrix for effective stress transfer at the interface [6-7]. Liu and Kumar have studied the existing progress of carbon fiber structure, fabrication, and characterization including the addition of nanotubes in precursor fiber to improve the mechanical properties [8]. However, the fundamental mechanical properties of these composites such as longitudinal and transverse strength and toughness limited by the weak interfacial bonding between reinforcing CF surface and polymer materials. It is a long existing critical and important topic needed to discover for ensuring the continued development of CF reinforced polymer composites for different composites applications [9-14].

According to this, many studies focused on probing an understanding the physicochemical interaction at the fiber/matrix interface. For strong interfacial adhesion, adjusted level of hydrogen bond and Vander Waals forces between the CF and matrix are required during composite fabrication. In addition, the fiber/matrix interfacial adhesion energy should be higher than the cohesion energy of the matrix. The modifications to CF structure made a big difference in improving the mechanical properties of high performance polymeric composites but the focus to control the fiber/matrix interfacial properties is still a major goal [13-14].

From the high-performance thermosetting resins, epoxy resins are the most important with different applications. Epoxy resins have a lot of advantages and show unique properties. One of the most common and important polymers is Epoxy resin due to its wide applications and wide variety of industries. They are composed of polymeric molecules that are converted to a solid by polymerization. The most important advantage of epoxy resins are the ability to be transformed from a low-viscosity liquid (or thermoplastic state) into a hard, tough thermoset. Epoxy systems physically consist mainly from two components: a resin and a hardener. The hardener is the responsible for the chemical reaction, which turns the epoxy resin into a solid, cross-linked network of molecules. Epoxy resin is defined as a thermoset polymer structure with high adhesion properties and cohesive strength because, when cured, it is irreversibly rigid and relatively unchanged by heat. However, the term epoxy can also be used to indicate an epoxy resin thermoplastic or cured state [11].

Many researches about the Composites have been done. When epoxy resins are mixed with high-strength carbon fibers, the product obtained is used in structural applications requiring high strength, high stiffness and low weight. They are of relatively low density and they can be tailored to have stacking sequences to provide high strength and stiffness in the directions of high loading [13-14]. Carbon nanotubes (CNTs) have been widely used in enhancing composites for its large specific surface area, excellent mechanical properties, as well as good compatibility with the polymer [13].

Introducing CNTs to the surface of fibers to enhancement the interfacial properties of composites considered as a hot topic. So far, several methods have been developed to introduce CNTs to the fiber surface, such as chemical grafting and chemical vapor deposition. Both methods could effectively introduce CNTs on the surface of CFs with strong adhesion, but the excessive chemical treatments and the use of high temperature have hindered their practical application. Electrophoretic deposition is another efficient way to introduce and incorporate CNTs in composites. However, it was woven carbon fabric that was mostly used to receive CNTs, in which case CNTs were only deposited on part of the fiber surface rather than all surfaces. Besides, the complicated process also limits its application in industrial scale [15-18].

With comparison to above methods, sizing or coating fibers with CNT-containing sizing agent exhibit excellent advantages for their simplicity and low-cost and show a great potential application in industrial manufacturing of CNT/CF hybrid fibers [9-10]. Generally, we can say that CNT_s are used with CF/polymer composites to produce hybrid CF/CNT_s/ polymer composites in order to improve the mechanical properties of CF/polymer composites [15-19]. Many experimental studies applied before to introduce CNT_s into the CF/Epoxy resin system and from these researches we observed that the addition of CNT_s occur via one route (epoxy resin or CF mat)[20]. Jianwei Zhang, Dazhi Jiang, Su Jua, Hua-Xin Peng, improved mechanical properties of composite material based on carbon fiber (CF)

reinforced epoxy resin by adding a small quantity of multi-walled carbon nanotubes (MWCNTs). 1 wt. % MWCNTs were well dispersed in the epoxy resin and fiber filament wind process were used for manufacturing the CF prepregs. The hybrid MWCNT/CF/epoxy composites were fabricated by laying up technique. Investigation applied on tensile properties, interlaminar shear strength (ILSS) and microstructures for the prepared composites [15].

In this work, a simple and clear method for addition of CNT_s to the CF composite system was produced. This technique depends on adding of CNT_s to the CF/epoxy resin system through the two routes of the composite system (epoxy polymer and CF mat). In this research MWCNTs was introduced into CF and epoxy polymer separately in the first step. Then the MWCNTs/CF and MWCNTs/epoxy polymer were mixed together to get the CF/MWCNT_s/Epoxy Resin Composite. MWCNT_s was introduced into the CF composite system by divided the amount of MWCNT_s into two equal parts the first part added to epoxy polymer using ultrasonic bath sonicator and the second part of MWCNT_s sprayed into the surface of CF surface using spray gun then manufacture hybrid CF/MWCNT_s/Epoxy resin composite using 1% weight ratio from MWCNT_s.

2. Experimental work

2.1. Materials

In this research the used fiber was T300 PAN carbon fiber manufactured from apolyacrylonitrile precursor with moderate mechanical properties (strength; modulus) and with fiber yarn contains 3000 filaments purchased from Kunshan Samson composite material Co., Ltd, China. The polymer used in this study is epoxy resin system with two components A epoxy (EPON 828) cured with B polyamide (versamide 125) hardener. The used resin system purchased from Aladdin Company, China. The resin system was prepared in proportions of 3:1 by weight. MWCNT_s with a particle size 20-45 nm obtained from Aladdin Co., Ltd, China.

2.2. Epoxy system preparation

The used resin system is epoxy resin consists of two parts; part A (EPON 828) polymer and part B (versamide125) as curing agent. The calculated amount of part A were carefully weighed and then added the required amount of part B, then we stirring the mixture for 30 min using magnetic stirrer to prevent air trapping with low velocity rate then put in ultrasonic bath sonicator for 30 min to achieve good mixing.[13]. In case of addition MWCNT_s to CF based composite, MWCNT_s was added first to part A in suitable beaker with stirring for 30 min for to achieve homogenous mixture and for good distribution, then put in ultrasonic bath sonicator for 3 hours to achieve good distribution of MWCNT_s in epoxy, second step by adding part B to the mixture in ultrasonic bath sonicator for 30 min and in this case the epoxy called modified epoxy resin, the last step by use the matrix contain MWCNT_s with CF to produce composite material [21].

2.3. Composite sample preparation

In this paper the hand lay-up technique was used to fabricate the composite material samples. For good impregnation of carbon fiber with epoxy matrix roller and brush were used. First the epoxy resin was prepared as explained in paragraph 2.2. The CF mats treated first with acetone in order to eliminate the sizing. In the case of sample C1 CF mat was cut in to adaptable size of 250×250 mm. the brush was used coating the epoxy resin on the surface of CF. Then put and aligned the epoxy-coated fiber tape together layer above layer then the prepared composite was covered on both the sides with iron sheets and kept between the fixed and movable die of the compression molding machine under 5MPa pressure for 24 hours. In the case of sample C2 the same procedure in step A was used but by using modified epoxy resin instead of normal epoxy resin. In case of sample C3 the half amount of MWCNT_s was added to epoxy resin as illustrated in section 2.2 to prepare modified epoxy resin and the other amount of MWCNT_s was dissolved in 100 ml ethanol and the mixture put in ultrasonic bath soincator for 1 hour then this mixture put in spray gun and sprayed on the surface of CF mat then the

CF mat put in oven at 40 °C for 1 hour to evaporate the ethanol. The both modified CF mat and epoxy were ready to use the same procedure in case of sample C1 prepare the composite sample C3.

2.4. Mechanical testing

The tensile test machine used in this research was universal testing machine (WDW-100), purchased from Fangrui Technology Co., Ltd. Changchun, China) which used to measure all mechanical properties for the prepared samples.

2.4.1. Interlaminar shear strength test results. According to the standard (ASTM D 2344) Short Beam Shear Test Method can be considered as interlaminar shear test method that can be used to determine shear strength of unidirectional lamina. It is used mainly as quality control test and a materials screening. The test is relatively simple and a test can be done fast. We calculated and reported the average value for five specimens of each sample.

2.4.2. Flexural test (the three-point bending test). Flexural strength can be defined as the maximum stress developed when a specimen with bar shape is subjected as single beam to a bending force perpendicular to the bar. The Flexural test was processed according to ASTM D790 with specimen dimensions 12.7mm width, 200mm length and with span to thickness ratio = 32:1. For each sample five specimens were tested and the reported value is the average value for the five specimens.

2.4.3. The tensile test. According to ASTM D3039 the sample specimen with 12.7mm width, 203mm length and with thickness depend on a number of layers was used for the tensile test. We calculated and reported the average value for five specimens of each sample. The Standard Flat shape for tensile specimens is shown in figure 1.

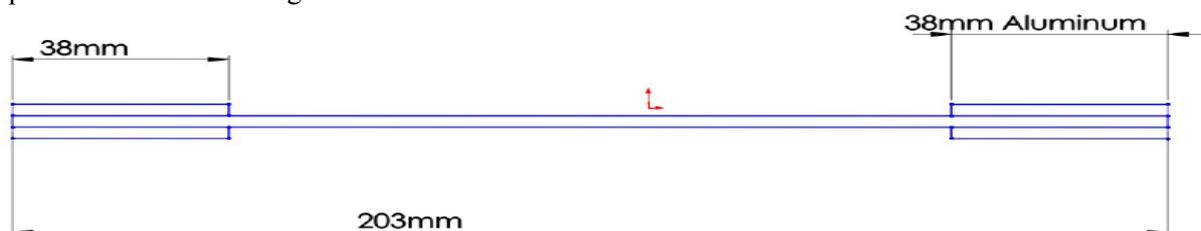


Figure 1. Standard Shape of tensile specimens.

2.5. Samples abbreviations

The abbreviations and formulations of the prepared samples used in this research explained in table 1.

Table 1. Details of sample composition

Sample abbreviations	Composition
C1	6layers[CF+ epoxy resin]
C2	6 layers [CF+ 1wt% MWCNT _S +epoxy matrix] MWCNT _S added via one route.
C3	6 layers [CF+ 1wt% MWCNT _S +epoxy matrix] MWCNT _S added via the two routes.

3. Result and discussion

3.1. Tensile test result

From figure 2 the tensile properties of prepared composite samples can be seen. The figure shows that there are three tensile strength values for three composite samples based on CF and have the same

number of CF layers. The composite sample C1 is based on CF/epoxy resin, the second composite sample C2 is based on CF/1wt% MWCNT_S/Epoxy resin in which CNT_S added via one route and the third composite sample C3 is based on CF/1wt% MWCNT_S/Epoxy resin in which CNT_S added via two routes. The results show that the tensile strength of sample C2 is higher than the tensile strength of sample C1 by 9%. The enhancement achieved due to addition of 1wt% of MWCNT_S in to CF/epoxy resin can be explained due to a stronger resin matrix caused by MWCNT_S reinforcement and enhanced matrix fiber interfacial interactions based on CNT_S incorporation. The results show also that C3 is higher than C2 by 10%. This is because adding of MWCNT_S through two routes in the same time (via epoxy resin and CF mat) increase the possibility for good mixing and good dispersion of CNT_S in to the CF/epoxy resin composite system and this lead to stronger of resin matrix and improve matrix-fiber interfacial interactions so increase the tensile strength.

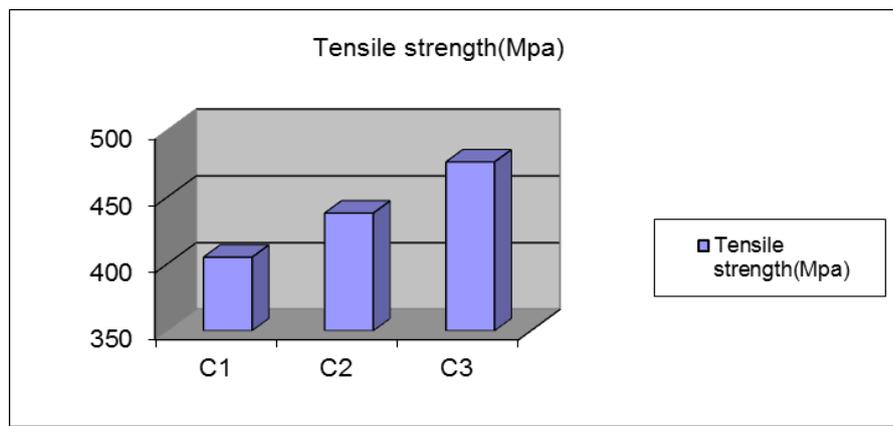


Figure 2. Tensile strength C1, C2 and C3.

3.2. Flexural test result

Figure 3 shows the flexural strength results of the three composite samples. From figure (3), it can be seen that the flexural strength of sample C2 is higher than the flexural strength of sample C1 by 51%. The enhancement achieved due to adding of 1wt% of MWCNT_S into CF/epoxy resin can be due to a stronger resin matrix caused by adding MWCNT_S and enhanced matrix fiber interfacial interactions based on CNT_S incorporation. The results show also that the flexural strength of C3 is higher than C2 by 19%. This is because adding of MWCNT_S through two routes at the same time (via epoxy resin and CF mat) increase the possibility for good mixing and good dispersion of CNT_S into the CF/epoxy resin composite system and this lead to stronger of resin matrix and improve matrix-fiber interfacial interactions so improve the tensile strength.

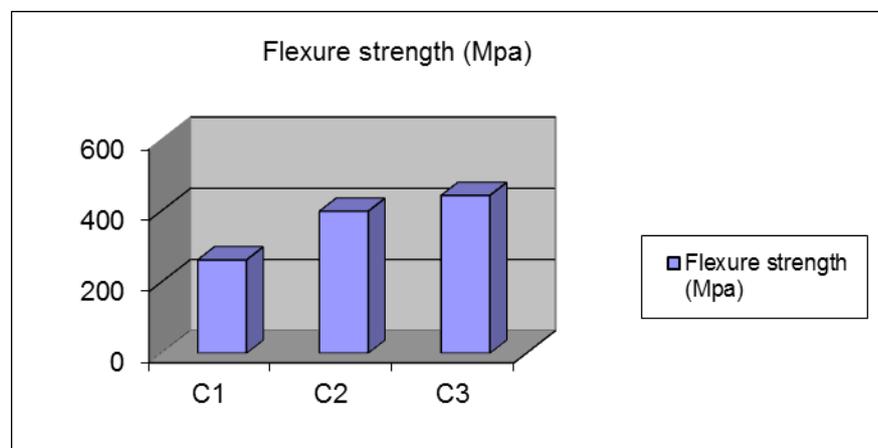


Figure 3. Flexural strength of C1, C2 and C3.

3.3. Interlaminar shear strength test results

Figure 4 shows the interlaminar shear strength of the prepared composite samples. The results show that Interlaminar shear strength of sample C2 is higher than the Interlaminar shear strength of sample C1 by 33 %. The enhancement achieved due to adding of 1wt % of MWCNT_s into CF/epoxy resin can be due to a powerful resin matrix caused by adding MWCNT_s and enhanced matrix fiber interfacial interactions based on CNT_s incorporation.

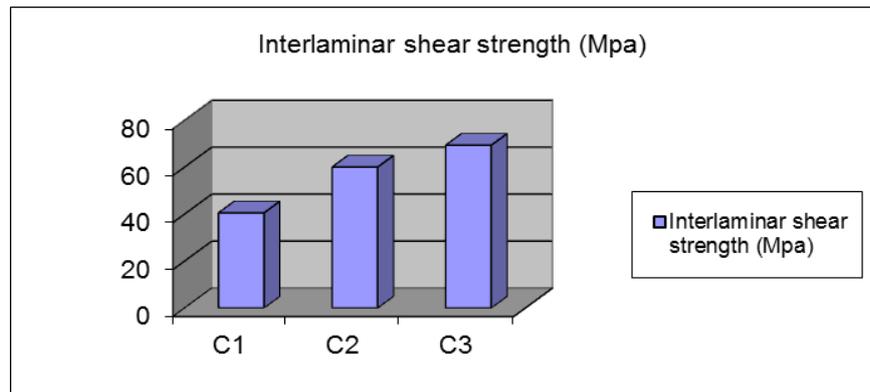


Figure 4. Interlaminar shear strength of C1, C2 and C3.

The results show also that C3 is higher than C2 by 21 %. This is because the addition of MWCNT_s through two routes at the same time (via epoxy resin and CF mat) increase the possibility for good mixing and good dispersion of CNT_s into the CF/epoxy resin composite system and this lead to stronger of resin matrix and improve matrix-fiber interfacial interactions so improve the Interlaminar shear strength.

4. Conclusion

The introduction of CNT_s to the CF/epoxy resin matrix is one of the most effective methods for improving and enhancing the interface between CF and epoxy resin and so improving the structural properties as mechanical characteristics of the CF/epoxy resin composite. It was found that the method of adding CNT_s into the CF composite system play important rule for increasing the interface bonding between CF an epoxy polymer which leads to increase the mechanical properties of the prepared composite. Three composite samples based on CF and epoxy resin were prepared C1[CF/epoxy resin composite], C2[MWCNT_s/CF/Epoxy resin composite] in which MWCNT_s added via epoxy resin and C3[CF/MWCNT_s/Epoxy resin composite] in which MWCNT_s added via CF and epoxy resin. The results show that the mechanical properties of C2 as flexural strength, tensile strength and interlaminar shear strength higher than C1 by 51%, 9 % and 33 % respectively. This is because adding of MWCNT_s to the CF/epoxy resin composite improves the epoxy bonding and the interface bonding between CF and epoxy so improve the mechanical properties. It was found that tensile strength; flexural strength and interlaminar shear strength of C3 higher than that of C2 by 10%, 19% and 21% respectively. This is because adding of MWCNT_s via two routes (epoxy resin and CF mat) improves the probability of good and homogenous mixing with epoxy resin and also good dispersion of MWCNT_s on the surface of CF mat which leads to stronger the epoxy adhesion and improves the interface between CF and epoxy so improve the mechanical properties.

References

- [1] Schwartz M M 1992 *Composite Materials Hand Book* McGraw-Hill.
- [2] Whitcomb J D 1988 *Composite materials testing and design* ASTM.
- [3] Peters ST 1998 *Hand book of composites* CHAPMAN&HALL.
- [4] Soutis C 2005 Fiber reinforced composites in aircraft construction *Prog Aerospace Sci* **41** 143–51.

- [5] Pavia J M F, Santos A N, Rezende M C 2009 Mechanical and morphological characterizations of carbon fiber fabric reinforced epoxy composites used in aeronautical field *Mater Res* **12(3)** 367-374.
- [6] Mallick P K 2008 *Fiber reinforced composites: material, manufacturing and design* **3rd Ed.** New York: CRC Press.
- [7] Edie D D, McHugh J J 1999 *High performance carbon fibers* In: Burchell TD, editor. Carbon materials for advanced technologies, Pergamum p. 119–38.
- [8] Liu Y, Kumar S 2012 Recent progress in fabrication, structure, and properties of carbon fibers *Polym Rev* **52(3–4)** 234–58.
- [9] Sharma M, Gao S, Mäder E, Sharma H, Wei L Y, Bijwe J 2014 Carbon fiber surfaces and composite interphases *Composites Science and Technology* **102** 35–50.
- [10] Pavia M C, Bernardo C A, Nardin M 2000 Mechanical, surface and interfacial characterization of pitch and PAN-based carbon fibers *Carbon* **38(9)** 1323-37.
- [11] Takeichi, Furukawa N 2015 Epoxy Resins and Phenol-Formaldehyde Resins *Reference Module in Materials Science and Materials Engineering Polymer Science: A Comprehensive Reference* **5** 723-751.
- [12] Chung D D L 1994 *Carbon Fiber Composites* Butterworth-Heinemann.
- [13] Gipsen R F *Principles of composite materials mechanics* McGraw-Hill.
- [14] Zhang H, Zhang Z, Breidt C 2004 Comparison of short carbon fiber surface treatments on epoxy composites Institute for Composite Materials, University of Kaiserslautern, and Erwin Schrodinger strasse **58**.
- [15] Zhang J, Ju S, Jiang D, Peng H X 2013 Reducing dispersity of mechanical properties of carbon fiber/epoxy composites by introducing multi-walled carbon nanotubes *Composites Part B: Engineering* **54** 371–376.
- [16] Magdy W 2004 Production technology of composite materials based on polymeric matrices Master, MTC, Egypt.
- [17] Li X, Han X, Tan Y, and Xu P 2007 Preparation and microwave absorption properties of Ni-B alloy-coated Fe₃O₄ particles *Journal of alloys and compounds* 240-245.
- [18] Li X, Yang H, Fu W, Liu S, Zhu H, and Pang X 2006 Preparation of low-density super paramagnetic microspheres by coating glass micro balloons with magnetic nanoparticles *Materials science and Engineering: B* **135** 38-43.
- [19] Rawal S and Brantley J 2013 Development of Carbon Nanotube-based Composite for Spacecraft Components In *Recent Advances in Space Technologies (RAST), 2013 6th International Conference on. IEEE* 13-19.
- [20] Zhao Z, Teng K, Li N, Li X, Xu Z, Chen L, Niu J, Fu H, Zhao L, Liu Y 2017 Mechanical, thermal and interfacial performances of carbon fiber reinforced composites flavored by carbon nanotube in matrix/interface *Composite Structures* **159** 761–772.
- [21] Fouda H, Guo L and Elsharkawy K 2017 Preparation and Characterizations of Composite Material Based on Carbon Fiber and Two Thermoset Resins *MATEC Web of Conferences* **88** 02002.