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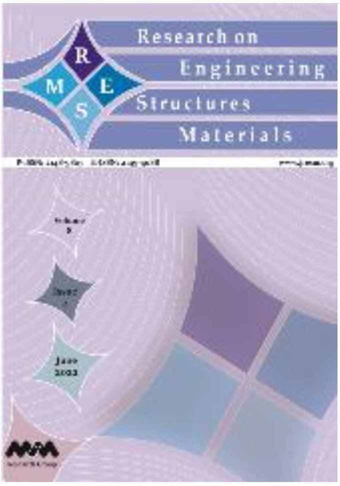

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Research Article

Tensile and charpy impact properties of CNTs integrated PET/ Glass Fiber thermoplastic composites with commingled yarn

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Abstract

Within this study, the non-crimp fabrics (NCF) with commingled yarns that they contained hybrid structures in which two different materials in the form of fibers were mixed, which consisted of polyethylene terephthalate (PET)/glass fiber (GF), were coated with multi-walled carbon nanotubes (MWCNTs) (weight percentages were 0 and 0.9%) and modified multi-walled carbon nanotubes (MWCNTs-Carboxylic acid (COOH)) (weight percentages were 0 and 0.9%) to fabricate hybrid composites. Three types of composite materials were prepared (pure polyethylene terephthalate/glass fiber (PET/GF), PET/GF with MWCNTs and PET/GF with MWCNTs-COOH) and they were tested against tensile and Charpy impact loadings. The effects of MWCNTs contents on the micro-structure and morphology of the composites were reported by using a scanning electron microscope (SEM), fourier transform infrared spectroscopy analysis (FTIR) and optical microscopy (OM). The specimens with MWCNTs-COOH exhibited an enhancement of 33% tensile strength, 23% tensile modulus and 8% Charpy impact energy compared to the samples without MWCNTs-COOH. It can be concluded that even a small mass fraction of MWCNTs was capable of improving the mechanical performance of the glass fiber reinforced PET matrix composites. In other words, due to the presence of the carbon nanotubes on the fiber surface helped to improve interfacial adhesion in the fabricated composites.

1. Introduction

Scientists and researchers used the nanomaterials in the producing of composite materials [1]. Carbon nanotubes (CNTs) are today considered to be the most important nanomaterials. CNTs are used in numerous applications like nanotechnology, optics, water treatment, electronics and other levels of the materials science. In addition, the tensile strength of CNTs is close to 100 times higher than those in steel. CNTs are made from thin, long cylinders of the atomic layer of graphite, with diameters usually measured in nanometers [2-5]. The incorporation of nanotubes can improve the properties of composites very well [6, 7]. MWCNTs consist of several concentric graphene tubes installed inside the other with a diameter in the nanometer scale [8, 9]. In addition, MWCNTs have a higher grade of stability and stiffness compared to single-WCNTs [10].

Thermoplastic consumption is approximately 80% from the total plastic consumption. These materials permit manufacturing of composite materials in short times that makes them possible to produce large quantities [11]. Polyethylene terephthalate (PET) is used as a container for food and liquids. PET material is hard, strong material that absorbs a little water and it has resistance to impact, moisture, alcohols and solvents [12, 13, 14].

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Glass fibers (GF) are made from extremely fine fiber of glass. The principal advantages of the glass fibers are high tensile, low cost, high chemical resistance properties [15, 16].

Some studies were done on the commingled fiber reinforced textile composites. Effect of comingling techniques on mechanical properties of natural fibre reinforced cross-ply thermoplastic composites were studied by Abidin et al. [17].

Many researchers studied the improvement of the mechanical properties of thermoplastic and thermoset composites through adding CNTs. Gojny et al. reported the investigation of the effect of various types of CNTs on the mechanical properties of epoxy matrix materials [18]. Shazed et al. reported that the Young's modulus increased by 104% and the tensile strength improved by approx 64%, due to the presence of CNTs in the polypropylene composites [19]. The mechanical properties of CNTs integrated carbon fiber reinforced thermoplastic composites was investigated by Liu et al. [20]. They obtained an improvement of 27.0% of the impact toughness after using CNT/CF hybrid fiber in the fabrication process. Hao et al. studied the enhancing of the mechanical properties of the poly ether ketone/zinc oxide nanocomposites [21]. Glass fiber–epoxy composites with boron nitride nanotubes for enhancing interlaminar properties in structures were studied by Rahmat et al. [22].

There are many studies on improving interface properties of the CNTs integrated thermoplastic composites. Demircan et al. reported the effects of carbon nanotubes (0.0, 0.7, 0.9, and 1.1-wt %) on the mechanical properties of glass fiber thermoplastic composites [23]. The difference from the previous article [23] was that they used a low melting point temperature PET (LPET) polymer with one type of MWCNTs, they didn't conduct a Charpy impact test on specimens and they didn't modelled the tensile properties of the composites. The obtained results of our study can be inspiring for many upcoming studies in this field.

2. Experimental

2.1. Composite Constituents

The specifications of the NCFs (Metyx Composites Corporation, Istanbul/Turkey), MWCNTs and the modified MWCNTs-COOH (Ege Nanotek Kimya Sanayi, Izmir/Turkey) are given in Table 1, Table 2 and Table 3 respectively.

There are glass fiber types such as A, C, E, S etc. E-glass commonly used in most of the composite applications. We used E glass type in our research.

2.2. Fabrication Method

The NCFs with the dimensions of 200 mm x 200 mm were prepared in ten layers. Two kinds of solutions with the MWCNTs and modified MWCNTs (MWCNTs-COOH) were prepared using ethanol. At first, the MWCNTs distributed throughout the ethanol by using a magnet bar, stirred by a magnetic field. After that, an ultrasonic bath was used to disperse the MWCNTs in the ethanol.

Both faces of NCFs were coated by using the solution of MWCNTs and ethanol as shown in Figure 1 (a). The MWCNTs coated fabrics were symmetrically left in the mold cavity of the hot press machine $[90^o/0^o]_{10s}$ (Figure 1 (b)). The top views of schematic representation of the NCF layers were shown in Figure 1 (c).

The hot press machine was used to produce the NCF composites. The molding temperature and pressure were 205 ^oC and 22 bar, which were the same used values for all fabricated specimens. ASTM D3171-99 standard was used to calculate weight and volume fractions of the samples (Table 4).

Table 1. The specifications of non-crimp fabric (NCF)

	Biaxial yarn 0° (warp) fibers	Biaxial yarn 90° (weft) fibers	Binding fibers
Weight composition	60% E glass fibers 40% PET fibers	60% E glass fibers 40% PET fibers	100% polyester
Color	Natural white	Natural white	Natural white
Weight (g/m2)	380	380	5.0
Yarn count (Tex)	525	525	7.6

Table 2. The specifications of multi-walled carbon nanotubes (MWCNTs)

Parameter	Value
Outer diameter (nm)	10–20
Interior diameter (nm)	5–10
Length (μm)	10–30
Surface area (m^2/g)	>200
Color	Black
Ash	Mass<%1.5
Electrical conductivity (S/cm)	>100
Density (tap) (g/cm^3)	0.22
Density (true) (g/cm^3)	2.1

Table 3. The specifications of modified multi-walled carbon nanotubes (MWCNTs-COOH)

Parameter	Value
Content of-COOH (wt %)	2
Outer diameter (nm)	10–20
Interior diameter (nm)	5–10
Length (μm)	10–30
Surface area (m^2/g)	>200
Color	Black
Ash	Mass<%1.5
Electrical conductivity (S/cm)	>100
Density (tap) (g/cm^3)	0.22
Density (true) (g/cm^3)	2.1

Table 4. Weight and volume fractions of composites for the tested specimens

Weight percentages of MWCNTs (MWCNTs wt-%)	Weight percentages of glass fibers (GF wt-%)	Volume percentages of glass fibers (GF vol-%)	Density (g/cm^3)	Thickness (mm)
0.0 (Pure GF/PET)	63.38	44.47	1.824	3.42
0.9 (GF/PET with MWCNTs)	59.86	42.00	1.824	3.64
0.9 (GF/PET with MWCNTs-COOH)	63.85	45.64	1.858	3.42

2.3. Characterization

Tensile and Charpy impact tests were performed on the fabricated specimens in the 0 degree directions. Figure 2 (a) and (b) show the specimen geometries for tensile and

Charpy impact characterization tests. The tensile tests (INSTRON 5982 100KN (USA)) were conducted on the samples with the dimensions of the 160 mm x 20 mm x thickness and the thickness of the aluminum tab was 0.2 mm according to ASTM-D3039 standard (Figure 2 (a)). The linear displacement speed in the tensile test was 1 mm/min.

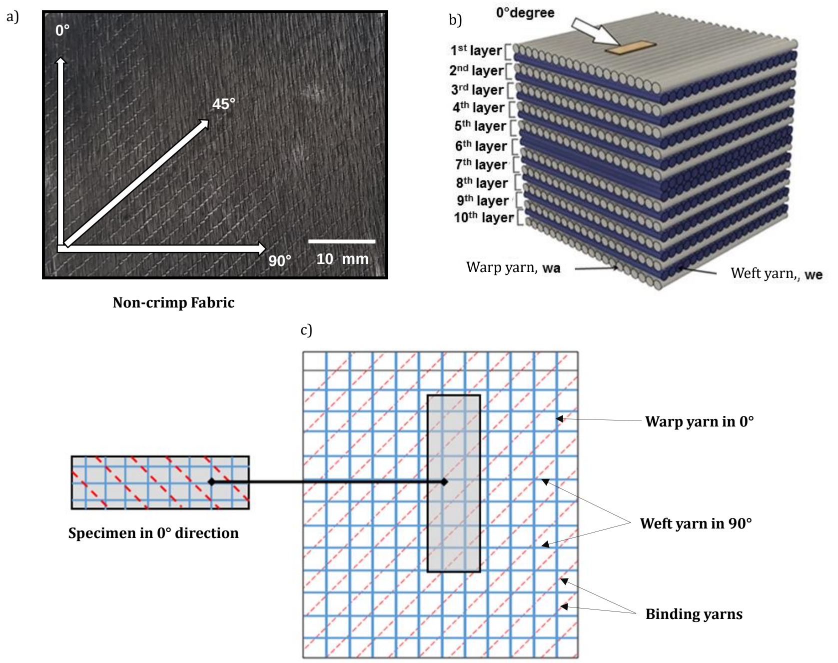
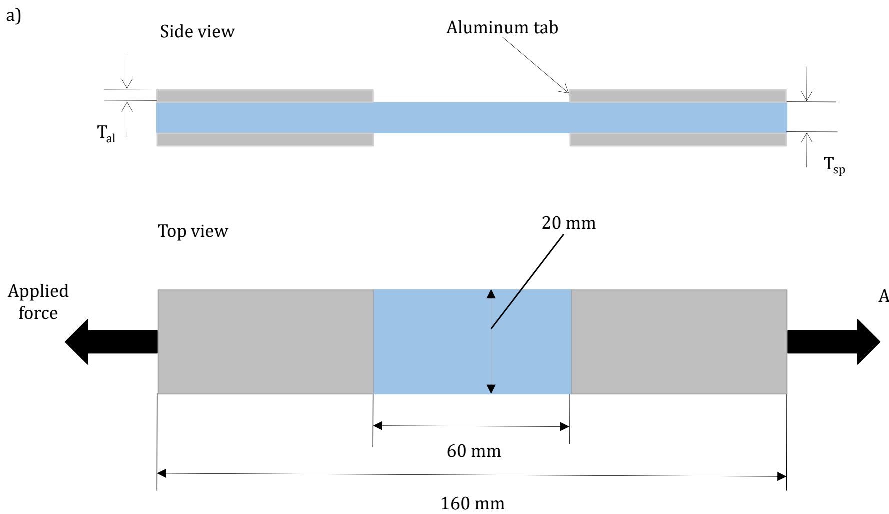
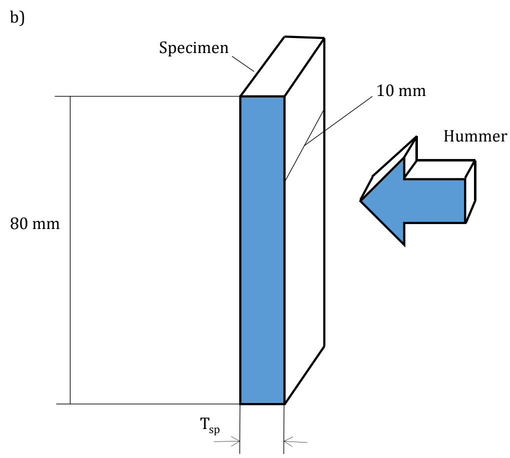


Fig. 1 (a) Real image of MWCNTs coated biaxial (0°/90°) NCF, (b) side view of schematic representation of symmetrical stacking of biaxial (0°/90°) NCF layers and (c) top view of schematic representation of symmetrical stacking of biaxial (0°/90°) NCF layers



(a)



(b)

Fig. 2 (a) The sample dimensions of the tensile test, Tsp: the thickness of the specimen, Tal: the thickness of the aluminum tab, (b) The sample dimensions of the Charpy impact test

ALSA ZBC 2000, Turkey type of machine was used in the Charpy impact tests. The all types of fabricated unnotched samples were tested in size of 80 mm x 10 mm x thickness (Figure 2 (b)). The tensile and Charpy impact test set ups were shown in Figure 3.

(a) (b)

Fig. 3 (a) Tensile, (b) Charpy impact test set ups

3. Results and Discussion

3.1. Surface Characteristics of CNT-coated Fibers

Figure 4 (a) to (c) show the scanning electron microscope photographs of the commingled yarn (PET fiber/glass fiber). The homogeneous carbon nanotubes distributions on the surfaces of the glass fibers were seen in Figure 4 (a) and (b). The CNT bridging and pullout were influenced by the homogeneous carbon nanotube distribution on fiber surface. Figure 4 (c) shows the presence of a few CNTs agglomerations on the coated the fiber.

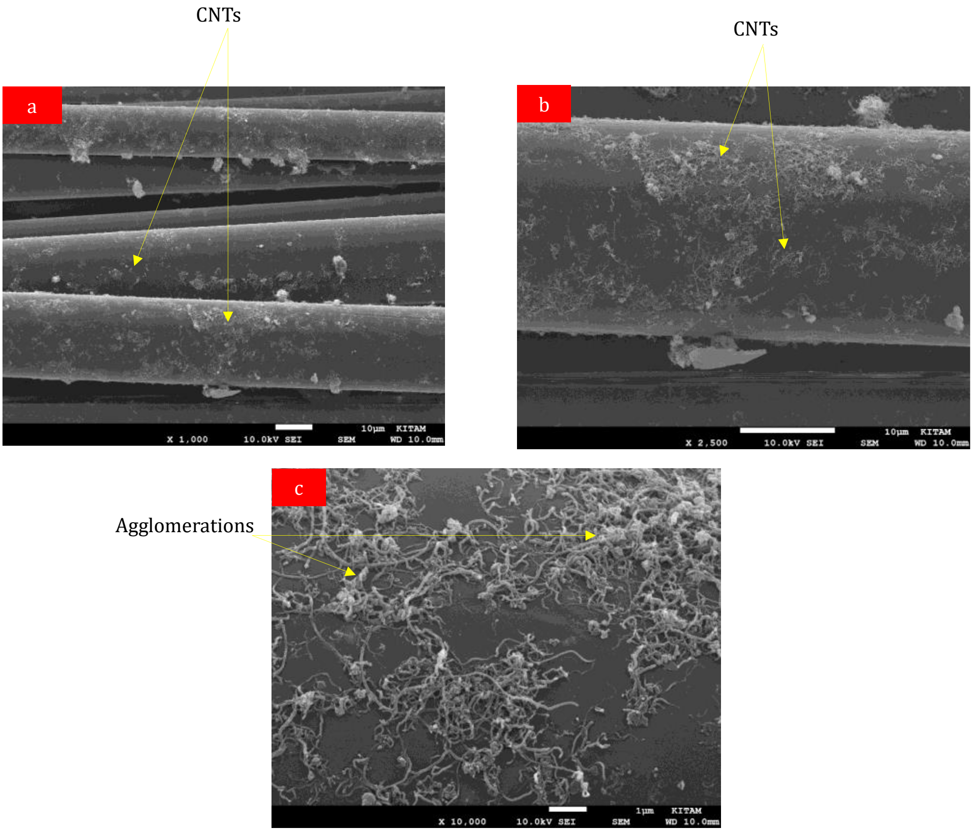


Fig. 4 (a), (b) and (c) SEM images of surface morphologies of PET-glass commingled fibers after grafting CNTs. SEM: scanning electron microscopic; PET: polyethylene terephthalate; CNTs: carbon nanotubes

The obtained data from FTIR analysis of pure MWCNTs, PET/GF and PET/GF+MWCNTs are shown in Figure 5. Figure 5 (a) (FTIR spectrum of the MWCNTs) shows the presence of the hydroxyl groups at 3731 cm^{-1}. The bands in the spectrum at 2989 and 2904 cm^{-1} are induced by C–H stretching vibrations on the nanotube surface, which reveal the –CH3 absorption [24, 25]. The appearing of the peaks and its positions depending on the bonds types of the structure. The production process of nanotubes is responsible for the generation of the functional groups on the nanotube surface [26]. The PET monomer consists of: 2 esters, 1 aromatic ring and 1 ethyl functional groups.

Group of terephthalate are bound with the ethyl group to form a PET monomer. The functional groups at the PET monomer consist of several bonds such as: C – O, C – H, C – C and aromatic ring. The main peak from the PET structure at 1715 cm^{-1} was the C=O group of terephthalic acid ester [27, 28] (Figure 5 (b)). The bands at 872 cm^{-1} and 1408 cm^{-1} refer to the groups of silanol on the surface of the fiber and the C-O groups. After grafting of the MWCNTs on the non-crimp PET/GF (Figure 5 (c)), any changes on the intensities and the characteristics of the spectrum were not seen, just a physical interaction between the MWCNTs and NCFs. Our results agreed well with the results of FTIR characterization of

Demircan et al. [23]. It can be noted that the most of the peaks that appeared in the article of Demircan et al. [23] were seemed in our experiment of FTIR test.

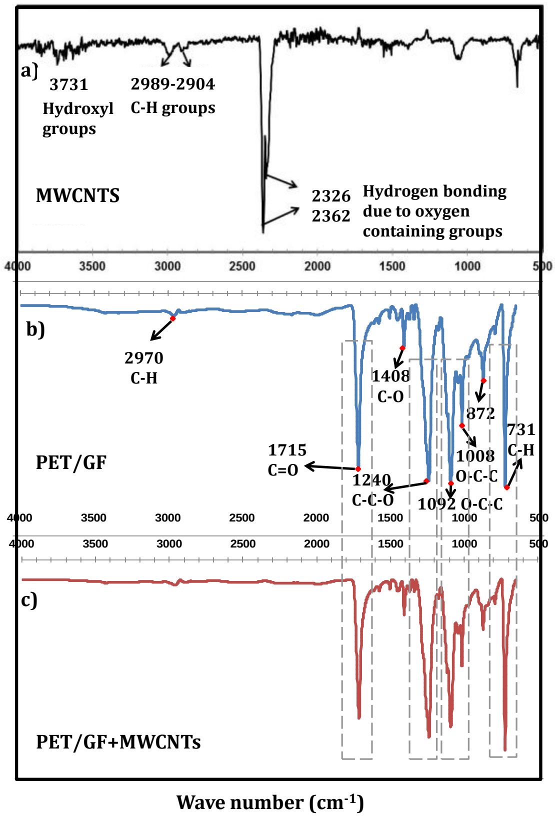


Fig. 5 (a), (b) and (c) Results of FTIR characterization

3.2. Tensile Properties

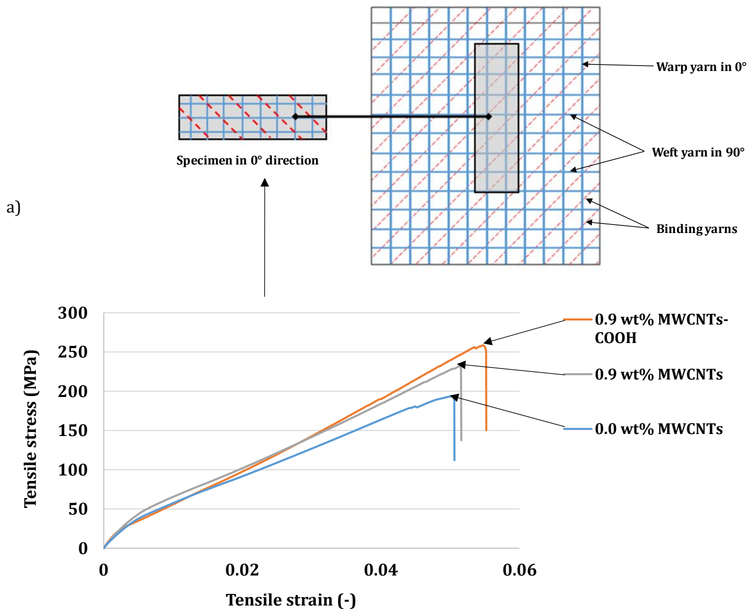
The tensile properties were calculated from the excel table of the test results. The tensile strength was the value of the maximum stress on the stress-strain graphs. Tensile modulus were calculated from the slopes of the curves of the stress-strain diagrams. The presented stress-strain diagram (Figure 6 a) was corresponding to the longitudinal direction of the loading.

The tensile properties of the fabricated composites with 0.9-wt % MWCNTs and 0.9-wt% MWCNTs-COOH showed higher value compared to the 0.0-wt% MWCNTs and 0.0-wt% MWCNTs-COOH composites. An improvement of approximately 17% and 23% tensile modulus and 20% and 33% tensile strengths were obtained from 0.9-wt% MWCNTs and MWCNTs-COOH compared to the samples without MWCNTs (Figure 6 (b)). It was expected that the possible cause for having the enhanced tensile properties for the samples with MWCNTs was the presence of the carbon nanotubes on the fiber surface.

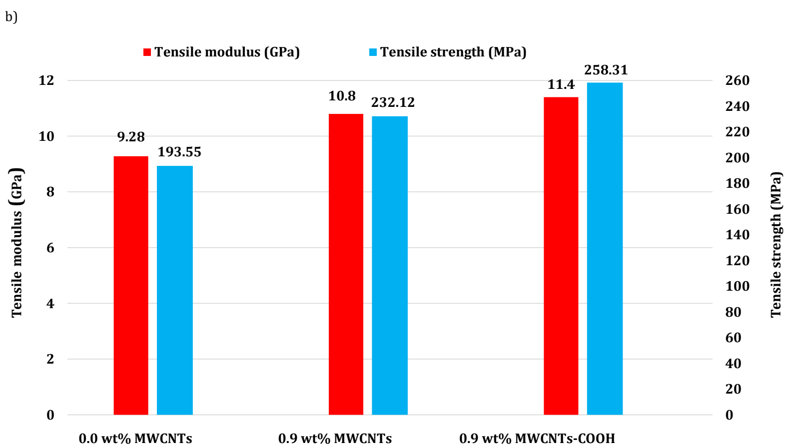
3.3. Laminate Theory

The laminate theory was used for the calculation of the tensile modulus of the composite. The modulus of the composites were increased by adding CNTs (Table 5). The tensile modulus of the composites with MWCNTs-COOH (25.4 GPa) was highest compared to the composites with MWCNTs (22.7 GPa) and pure GF/PET (21.4 GPa). Since the modulus of the composites were increased by adding CNTs from the experiments, the results from the laminate theory agreed well with the results from the experiments.

Effect of the binding yarns on the calculated tensile modulus could not be reflected in the laminate theory, due to this reason the modulus results of the laminate theory of the composite structures were much higher than the experimental results [29].



(a)



(b)

Fig. 6 (a) Stress–strain curves from tensile test, (b) Results of the tensile modulus and strength of the specimens

Table 5. The modulus results from tensile test and laminate theory

Sample	Modules from experiment E_{exp} [GPa]	Modules from laminate theory E_{Lam} [GPa]	Difference between E_{Lam} and E_{exp} [%]
0.0 (Pure GF/PET)	9.28	21.4	+130.6
0.9 (GF/PET with MWCNTs)	10.8	22.7	+110.2
0.9 (GF/PET with MWCNTs-COOH)	11.4	25.4	+122.8

3.4. Charpy Impact Properties

The fabricated specimens with 0.0-wt% MWCNTs had 4.49 J Charpy impact absorbed energy, whereas that was 5.50 J with 0.9-wt% MWCNTs and 4.85 J with 0.9-wt% MWCNTs-COOH (Figure 7).

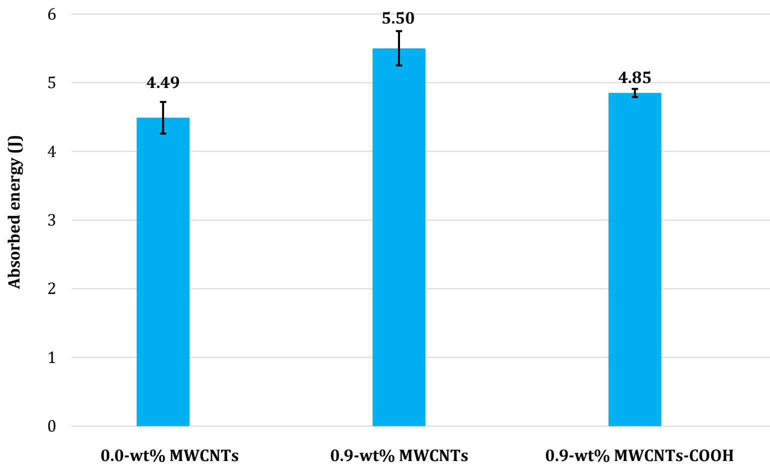


Fig. 7 Charpy impact absorbed energy of the specimens

The obtained results in our study clearly showed an enhancement of the Charpy impact absorbed energy in the fabricated composite materials combined with nanomaterial (MWCNTs and MWCNTs-COOH) compared to the fabricated composite without nanomaterial in 0°.

The Charpy impact properties of the fabricated composites with 0.9 wt-% MWCNTs and 0.9-wt% MWCNTs-COOH had higher value compared them to the 0.0-wt% MWCNTs in 0°. An enhancement of about 22% and 8% Charpy impact absorbed energies were gained from 0.9-wt% MWCNTs and MWCNTs-COOH compared to the fabricated samples without MWCNTs.

3.5. Results of Fracture Aspects of Composites

The SEM images of the through thickness parts of the samples containing 0.9-wt% of MWCNTs from the tensile test is shown in Figure 8 (a) and (b). It is believed that each of the following factors like, fiber bridging, CNT pullout and CNT bridging contributed to the fracture toughness of the specimens.

We can see in Figure 9 (a) and (b) the fracture aspects from the OM of tensile tested through thickness parts of the samples in the 0°. In Figure 9, it was shown some force application. This represents the direction of the force from the tensile test. It is a tensile force. We applied the force on the specimens on the longitudinal direction of the fabricated plates. The prominent failure mechanism was mode I in Figure 9a. The failure modes of the tensile tested composite samples with 0.0-wt% MWCNTs were matrix cracks, transverse cracks and delaminations (Figure 9 (a)). The absence of delaminations and few numbers of fiber and matrix cracks in the composite with 0.9-wt% MWCNTs (Figure 9 (b)) refers to enhanced mechanical properties of those composite structures.

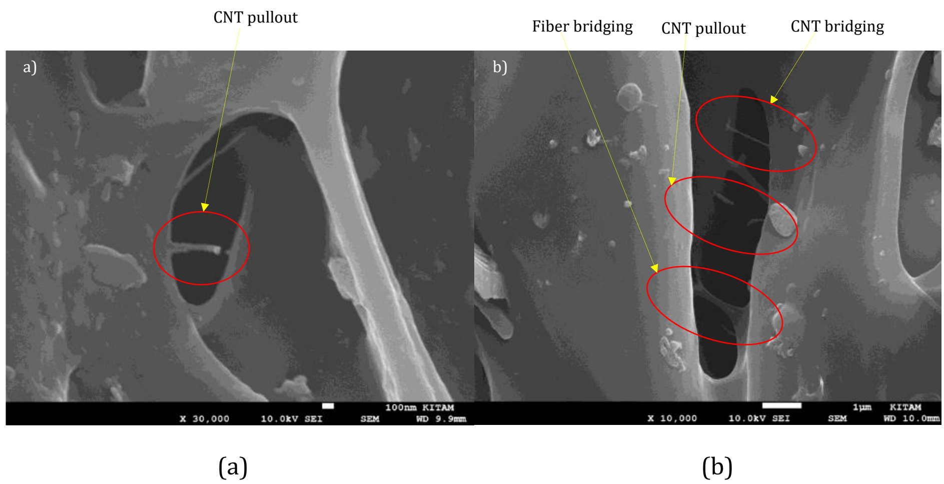
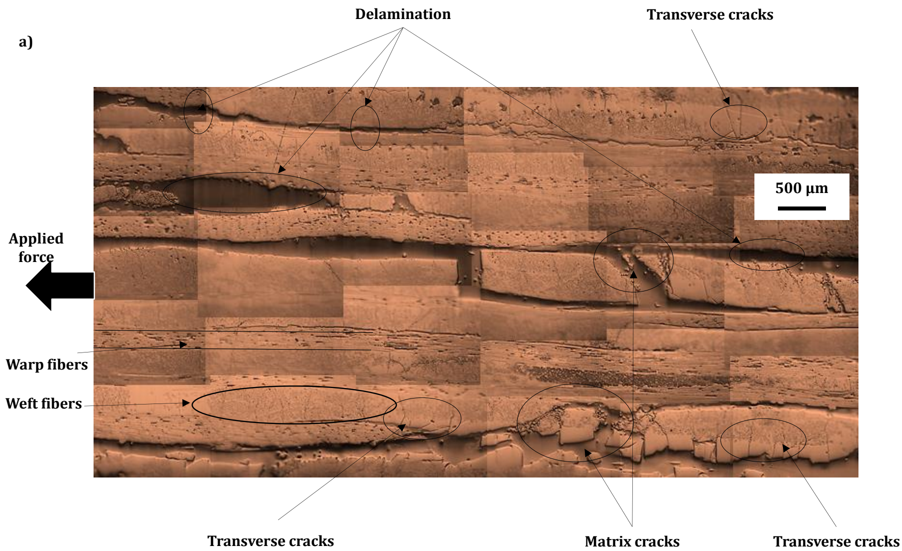
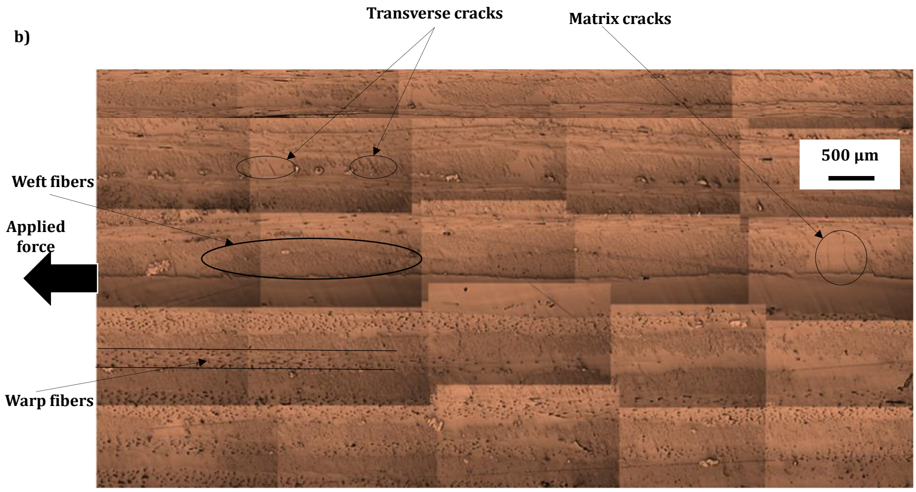


Fig. 8 (a) and (b) SEM images of fractured specimens with MWCNTs from tensile test



(a)



(b)

Fig. 9 (a) and (b) Fracture aspects of optical micrograph from tensile tested specimens (a) 0.0-wt% MWCNTs and (b) 0.9-wt% MWCNTs. MWCNTs: multi-walled carbon nanotubes

4. Conclusions

Within this study, the NCF with the commingled yarn of the polyethylene terephthalate (PET)/glass fiber (GF) were coated with MWCNTs and MWCNTs-COOH (the weight percentages were 0 and 0.9%) to fabricate hybrid thermoplastic composites. Three different types of thermoplastic composite materials were prepared (pure polyethylene terephthalate/glass fiber (PET/GF), PET/GF with MWCNTs and PET/GF with MWCNTs-COOH). The effects of MWCNTs contents on the micro-structure and morphology of the composites were investigated by using a SEM, FTIR analysis and OM.

The homogeneous carbon nanotubes distributions on the surfaces of the glass fibers as well as the presence of a few CNTs agglomerations on the coated the glass fibers were seen from the SEM images of the CNTs coated fibers.

Our study showed the Charpy impact and tensile properties of the fabricated PET/glass fiber reinforced thermoplastic composites with 0.9 wt% MWCNTs and 0.9 wt% MWCNTs-COOH demonstrated higher values compared to the 0.0 wt% MWCNTs fabricated composites. Furthermore, each of the following factors like, fiber bridging, CNT pullout and CNT bridging contributed to the fracture toughness of the specimens. Morover, from the tensile tested OM images of MWCNTs integrated glass fiber reinforced specimens, the absence of delaminations and few numbers of fiber and matrix cracks were appeared.

FTIR results showed that after grafting of the MWCNTs on the non-crimp PET/GF, any changes on the intensities and the characteristics of the spectrum were not seen, just a physical interaction between the MWCNTs and NCFs.

Considering the improvement in tensile modulus, tensile strength, Charpy impact strength, it can be concluded that even a small mass fraction of MWCNTs was capable of enhancing the mechanical performance of the glass fiber reinforced PET matrix composites. Our future research will be investigating the effect of nano materials on the Short Shear Beam characteristics of the thermoplastic and thermoset composite materials.

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