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RESEARCH ARTICLE

Impact Performance Improvement of Multiscale Hybrid Fiber Reinforced Polymer Composites with CNT

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ABSTRACT

Improving the interface properties of carbon nanotubes increases the mechanical performance of fiber-reinforced polymer matrix composites. Studies on different fiber types and different polymer matrix materials present promising results in literature. The effect of carbon nanotube (CNT) additives on impact performance of fiber reinforced polymer matrix composites produced by vacuum infusion method and drop weight impact test applied. Glass and carbon 1 m^2 fiber fabrics were divided into 9 equal square pieces and placed on top of each other to make them multi-layered structure. Fiber reinforcements were produced using vacuum infusion method with epoxy resin. 0.5% of the total composite weight was added to CNT with same production parameters and intraply hybrid composite containing glass, carbon and aramid fibers was also produced. Samples were produced from the composite plates and the drop weight impact test was performed with 50 J impact energy in accordance with ASTM D7136 standard. While this increase could be observed in glass fiber and carbon fiber reinforced composites, the impact energy absorption performance in carbon fiber reinforced composite increased more than 100%. CNT increased the impact performance of multi-layer fiber reinforced polymer matrix composites.

Keywords: Carbon nanotubes, Polymer Composite, Drop Weight Impact

1 Introduction

The evolution of various industries has led to the formation of new material performance expectations. Conventional materials have been replaced by the composite material concept with a combination of these materials. The concept of composite was first formed by polymer matrix composites during mid-20th century. While the first technical representatives of this concept started with glass fiber and polyester applications, with the widespread use of carbon fiber reinforcements, polymer composites have become increasingly widespread [1]. Polymer matrix composites, which had limited applications in the past, they are now applied in many areas such as automotive, aviation, medical, construction and defense industry. The most obvious example of composite applications is in aviation [1]. In civil aviation, Boeing is using fiber-reinforced composite polymer matrix composites about 50% in the 787 Dreamliner and, while Airbus is using fiber-reinforced composite polymer matrix composites about 53% in the body parts of A350 XWB nowadays [1], [2].

Composite materials are a type of material created by combining two or more types of materials without a chemical interaction. In composite materials, matrix's materials performance is desired to be increased with reinforcements. While matrix is traditionally classified as metal, ceramic and polymer according to its type, reinforcement materials additionally can be classified with their size, shape and distribution to matrix. The main reason why composite materials are preferred is to reinforce a material (matrix) desired to be used in various places with other material types to improve the material properties and increase their performance [3]- [5].

When it comes to composite materials, polymer matrix composites are preferred to be used intensively because of their lightness, high corrosion resistance, high hardness, high strength, easy manufacturability, and ability to fill the mold well. With these features, they are preferred to be used in renewable energy and aerospace. With the new polymers and new reinforcement materials produced day by day, polymer matrix composites have an increasing need, production and usage rate compared to other conventional materials. The need for polymer composites reveals a new and ever-increasing need for material performance, the concept of hybrid composite, in which more than one reinforcement material is used in the composite structure instead of a single reinforcement material [6]- [8].

Hybrid composites are created by hybridizing reinforcement or matrix materials in certain compositions have an effect that increases material performance, such as being lighter than conventional composites, obtaining high mechanical properties. Four different hybrid composite composites can be defined in the literature and these are; "interply hybrids" containing different types of additives in the same layer, "intraply hybrids" with different reinforcement materials in multi-layer structure, "interply-intraply hybrids" with different reinforcements in the same layer in multilayer structure and different types of reinforcement in different layers, and they can also be classified as resin hybrids, where more than one resin is used in the composite structure [9], [10]. In addition to hybridization, matrix performance can be increased by matrix modification. Additives to the matrix can be filler additives, conductive additives, and flame-retardant additives. In the modification of reinforcement materials, applications such as reinforcing with impregnation, functionalization, grafting on the surface can be made. These modifications, both in the matrix and in the additive, not only add new properties to the composite such as non-flammability, thermal and electrical conductivity, but also increase its mechanical performance [11].

With their high mechanical and physical properties, nano-sized additives as a reinforcing material are widely used in current applications with their good area/volume ratio and therefore good interaction with the matrix and homogeneous dispersibility. One of these additives is carbon nanotubes (CNTs), which are allotropes of carbon. With their superior mechanical and physical properties, CNTs, for example, have a high modulus of up to 1 TPa and a tensile strength of more than 10 GPa [12], [13]. Thanks to such high mechanical and physical properties, CNTs are also used as additives in composite materials [14], [15].

In the literature, there are studies involving the incorporation of carbon nanotubes into fiber-reinforced polymer matrix composites. Kostopoulos *et al.* [16] introduced MWCNT to carbon reinforced polymer composites (CFRP) and analyzed MWCNT's effect on fracture toughness under different impact energies. With the addition of MWCNT, fracture toughness improved in Mode I and Mode II. Similarly, Christoph *et al.* [17] introduced vertically aligned CNT (VACNT) to CFRP and they showed improvement in applied force to the composite under impact up to 13%. On the other hand, Kara *et al.* [18] investigated the impact properties of MWCNT reinforced CFRP and 0.3% MWCNT by mass increased impact energy up to ~28% in the structure. Additionally, Tehrani *et al.* [19] added 2% MWCNT by mass to CFRP with epoxy matrix in their study. Result of the impact test have shown that the contribution of MWCNT to the epoxy matrix increased the impact energy absorption performance of the composite by ~21.3%. Also, Mahdi *et al.* [20] produced a CFRP with epoxy and incorporated with MWCNT to define the effect on impact performance of the composite in their study. The impact test results showed that under different impact energies with 30 J, 40 J and 50 J, MWCNT improved absorbed impact energy performance of CFRP up to ~7%. Furthermore, in the study of Inam *et al.* [21], an amino-modified double-walled CNT (DWCNT) was added to CFRP with epoxy and DWCNT to the epoxy resin was up to 0.1% by weight. In the impact tests, CNT contribution to the composite structure increased the absorbed impact energy performance. Moumen *et al.* [22] investigated the effect of CNT additives in different weight ratios on the mechanical performance of CFRP with epoxy. In the study, 0.5%, 1%, 2% and 3% CNT additives by weight were used. According to the applied impact test results, they observed that the composite structure of CNT improved the impact energy absorption performance regardless of CNT amount.

In glass fiber reinforced polymer composites (GFRP), Peng *et al.* [23] coated CNT on glass fiber by 1% by mass. Impact tests have shown that such multi-scale MWCNT-GFRP interaction effectively develops better impact strength performance. Alhazov and Zussman [24] added polyvinyl butyral (PVB) between multilayer glass fibers in their study, and the impact performance of the composite increased with 0.5%, 1% and 1.5% CNT by weight contributed to the PVB. It was observed that the energy absorption performance was increased by 341% with addition of 1.5% CNT by weight. On the other hand, Tasyurek and Kara [25] produced pre-stretched glass fiber reinforced epoxy polymer matrix composites and incorporated them with 0.5% and 1.0% CNT by weight to the epoxy matrix. In the study, impact test was applied with 5 J, 10 J and 15 J and with amount of CNT increase, the rebound energy also increases. It has been observed that the resistance to low velocity impact was increased with CNT addition to composite. Liao *et al.* [26] added 0.021% by weight of flame-synthesized CNT (F-CNT), CVD synthesized CNT (S-CNT) and single-walled CNT (SWCNT) to glass fiber reinforced vinyl ester polymer matrix composites. All CNT added composite structures were found to have better impact performances compared to non-CNT additive composites. Nor *et al.* [27] added 0.5% by weight MWCNT to a hybrid multilayer epoxy matrix composite produced with glass fiber and bamboo and they showed that CNT addition increased maximum force about 36%.

In addition to glass and carbon fiber individually, aramid fibers are widely used in many ballistic applications due to their proven fracture toughness performance. Aramid's use with CNT incorporation and other fiber types more likely with glass and carbon fibers to create hybrid composites are also studied. Sukanya *et al.* [28] added 0.5% and 1% by weight MWCNT to aramid fiber reinforced epoxy matrix polymer composites in their study and it was found that the pure aramid composite is inferior if compared to CNT incorporated aramid reinforced polymer composites in terms of impact performance. For composite without CNT, the structure absorbed ~5 J, but the structure with 0.5% MWCNT absorbed ~12 J, the structure with 1% MWCNT absorbed ~10 J of impact energy. It was seen that the 0.5% CNT added aramid fiber reinforced structure exhibited the best impact energy absorption performance. Badawy and Khashaba [29] added 0.5% by weight CNT to epoxy polymer matrix composites consisting different glass and aramid fiber fabrics and their mechanical performances were investigated. After the applied impact tests of 20 J, the absorbed energy with help of CNT increased 11.4% in pure epoxy matrix, and glass and aramid fiber which reinforced with CNT, 1.6% and 1.5% performance increase were observed respectively. On the other hand, Uzay *et al.* [30] produced interply and intraply aramid-carbon fiber reinforced epoxy polymer matrix hybrid composites and examined the Charpy impact performance. Within the scope of their study, carbon, glass and aramid containing intraply composites tested with Charpy impact test and test results showed that hybridization in intraply aramid-carbon composite gave better results than others. Interply hybrid laminates showed 63.4% and 17.6% higher results compared to pure carbon and pure aramid composites respectively. Intraply hybrid composites showed 71.6% and 23.5% higher results compared to pure carbon and pure aramid composites.

In this study, the effect of CNT on the impact performance of multilayer fiber reinforced polymer matrix composite was investigated. For this purpose, glass and carbon fiber fabrics with the same weave system and similar areal density properties were preferred as fiber reinforcement. Multilayer fiber reinforced polymer composites were produced using vacuum infusion method with and without CNT additives. In addition, the intraply hybrid multilayer polymer composite containing glass, carbon and aramid fibers in the same structure was produced with or without CNT additives using the same parameters. Samples in accordance with the ASTM D7136 standard were produced from the composite plates and the drop weight impact test was performed with 50 J impact energy.

2 Materials and Methods

2.1 Manufacturing of Composites

In this study, multilayer fiber reinforced polymer matrix composite plate materials were produced using vacuum infusion method. Glass, carbon, aramid fiber fabrics as fiber reinforcement were supplied from Dost Kimya A.S. (Istanbul/Turkey) as twill 2/2. Glass fiber 300 g/m², carbon fiber 245 g/m² and aramid fiber 300 g/m² were preferred as areal density (g/m²). Kevlar 49 was preferred as aramid. Epoxy resin was used as the polymer matrix. Hexion's LR285 epoxy resin and LH285 curing agent were also supplied from Dost Kimya A.S. (Istanbul/Turkey). In the study, carbon nanotubes (MWCNTs) were applied on glass, carbon and aramid fibers. For MWCNTs, carbon nanotubes from Ege Nanotek (Izmir/Turkey) with an inner diameter of 2-6 nm, an outer diameter of <8 nm, a length of 10-35 µm and a purity of >96% were preferred.

Glass, carbon and aramid fiber fabrics were produced as fiber reinforced epoxy matrix polymer composite with epoxy resin in 50:50 weight ratio. In addition, glass, carbon and aramid fibers were produced as plates consisting of an intraply hybrid structure with multiple layers. First, 100 × 100 cm² (1 m²) fiber fabrics were divided into 9 pieces of 33 × 33 cm². MWCNTs were applied on the separated pieces with a spray paint gun isopropyl alcohol mixture at 0.5% of the total mass of the composite plate. After the application, the alcohol was removed from the fabrics and the coating of MWCNTs on the fabrics was completed. Layer placement for 9 coated 33 × 33 cm² fiber fabrics, were applied as $(0_{glass})_9$, $(0_{carbon})_9$, $(0_{glass}/0_{carbon}/0_{aramid})_3$ and produced by vacuum infusion method.

2.2 Specimens and Drop Weight Impact Test

A total of 6 composite plates made of glass, carbon, and hybrid structure containing aramid fibers alongside glass and carbon fibers, and their CNT reinforced composites were sized on a diamond-headed marble cutting machine in accordance with ASTM D7136/D7136M-15 standard's sample sizes under room temperature (Figure 1-2).

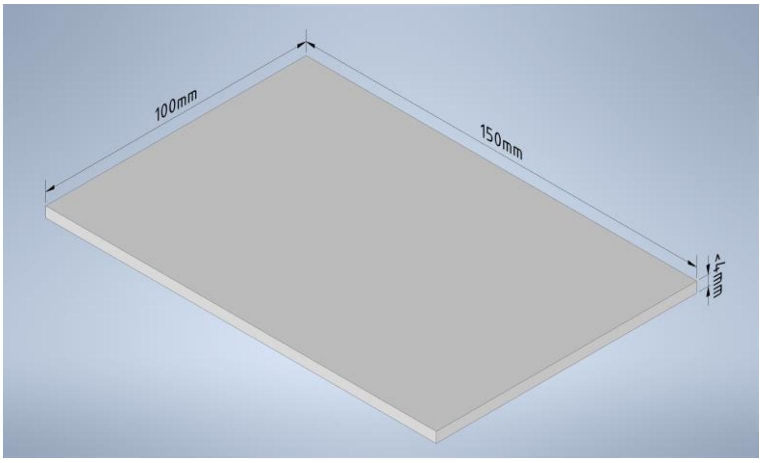

Figure 1: *Sample dimension specifications of the ASTM D7136/D7136M-15 standard.*

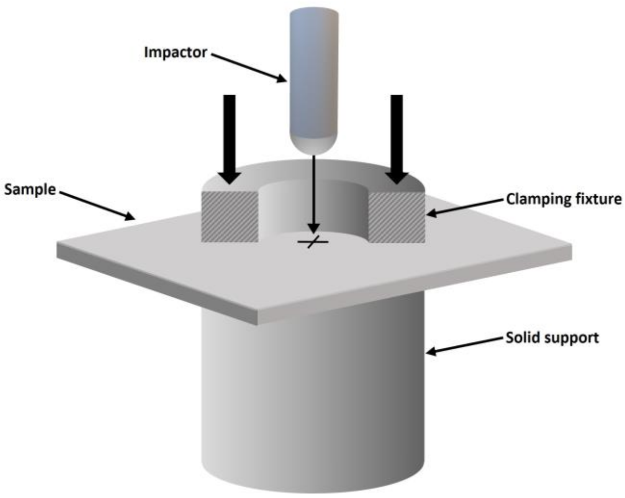

Figure 2: *Low-velocity impact tester and test mechanism.*

3 Results and Discussions

In this study, carbon and glass reinforced epoxy matrix composites were manufactured with CNTs addition with 0.5 wt.%. In addition, carbon, glass and aramid reinforced hybrid composite were manufactured with the same amount of CNTs. ASTM D7136 standard and the drop weight impact test was applied to the samples with 50 Joule impact energy (Figure 3-5, Table 1). The highest performance for impact energy was 24.08 J in hybrid fiber reinforced structure, 22.1 J in composites with glass fiber, and the lowest impact energy performance was 14.4 J in composites with carbon fiber. More than 100% increase in impact energy performance (31.77 J) was detected in carbon fiber reinforced composites when CNT was introduced to structure. Although the contribution of CNT additive to the performance in glass fiber reinforced composite structure was minimal, the increase was up to 24.64 J. However, energy absorption performance decreased to 23.32 J in composites with hybrid fiber reinforcement with addition of CNT (Table 1, Figure 4). On the other hand, deflection/displacement (mm) at maximum energy absorbed, deflection was 12.75 mm in composites with glass fiber, 11.27 mm in composites with carbon fiber and 12.79 mm hybrid structural fiber reinforced composites. It was observed that the highest displacement was 12.4 mm in the composites with hybrid structure in other fiber reinforced composites, followed by 13.57 mm in glass fiber and 17.55 mm in carbon fiber reinforced composites. With the CNT additive, an increase in the displacement of carbon and glass fiber reinforcements has been achieved. Decrease in displacement was observed in hybrid fiber reinforced composite (Table 1, Figure 4). When the impactor was perforating the sample, a linear energy absorption and displacement relationships were obtained. CNT addition into composites with carbon and glass fibers showed linear increase behavior but due to more than two different fiber types in same structure, such incompatibility occurred in hybrid composite, and this caused low impact performance.

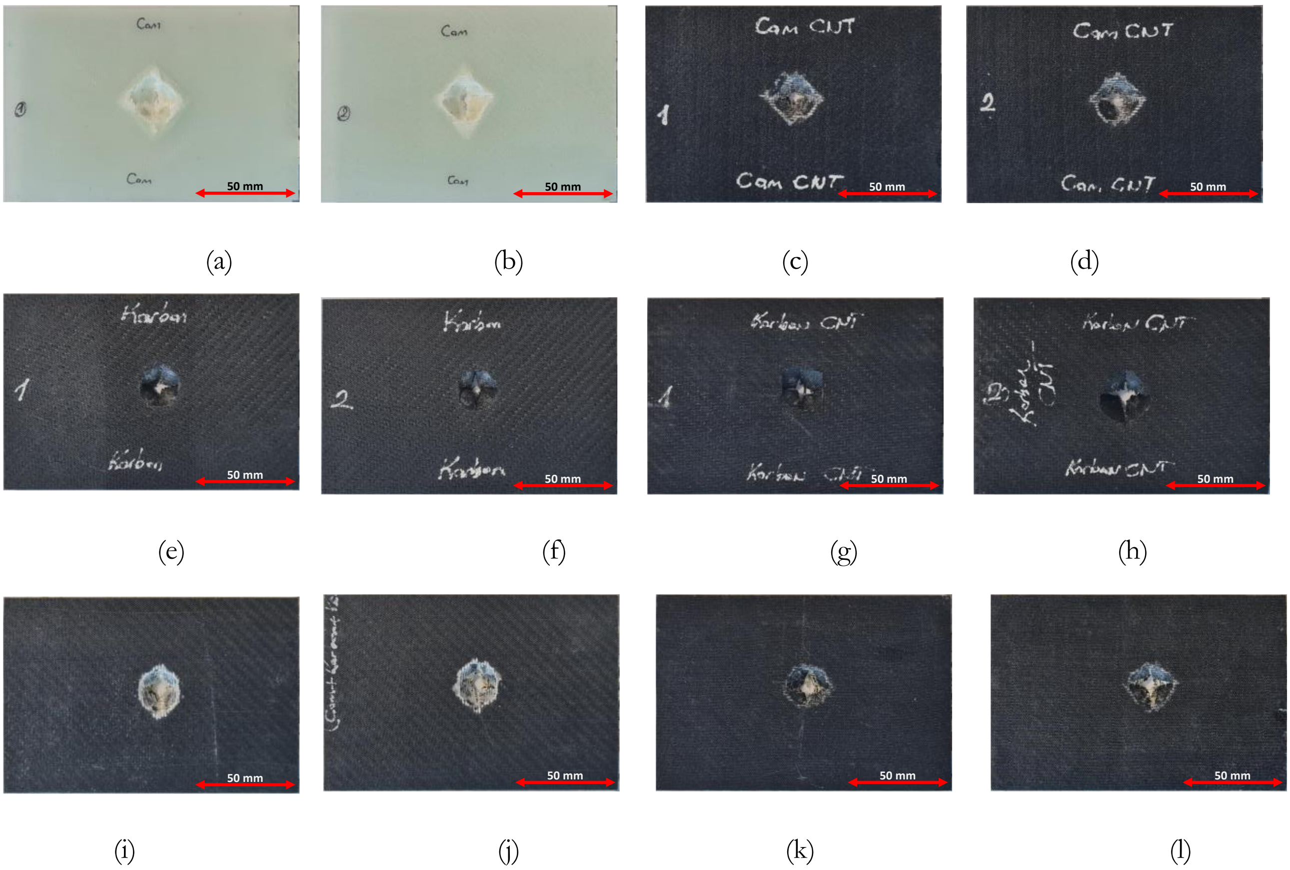

Figure 3: *Samples of CNT added glass (a,b), carbon (e,f), and hybrid (i,j) and non-CNT added glass (c,d), carbon (g,h), and hybrid (k,l) structured fiber fabric reinforced polymer matrix composite material samples after drop weight impact test applied.*

Under 50 Joule impact energy, 4613.82 N in composites with glass fiber, 3098.02 N in composites with carbon fiber and 5084.45 N in composites with hybrid structure were obtained. When CNT was brought, the impact force performance of the carbon fiber reinforced composites increased more than 100%, while there was no visible change in the contact force performance, on the contrary, a decrease in the contact force (2947.88 N) was observed. In glass fiber reinforced and hybrid fiber reinforced composite structures, the contact force performance of the CNT additive increased (4980.51 N and 5569.50 N) (Table 1, Fig. 6). It was also seen that the highest time (ms) was spent in hybrid structure composite with 4.49 ms. In composites with glass fiber, it was 4.42 ms and the lowest time in composites with carbon fiber with 3.98 ms. The time spent on carbon and glass fiber reinforcements with CNT additives increased to 6.47 ms and 4.81 ms, respectively. In the hybrid fiber reinforced composite, the time spent decreased by 4.32 ms (Table 1, Fig. 5).

The data of the damaged and perforated area by the impact device was also examined (Table 1). It was observed that the CNT reinforcement increased the impact energy of the glass fiber reinforced composite from 22.10 J to 24.64 J, while the area of the damage area increased from about 384 mm² to 404 mm². This change was similarly seen in carbon fiber reinforced composite. The impact energy increased from 14.4 J to 33.77 J, while the area of the damage area increased from 335.5 mm² to 465 mm². It is seen that the reinforcement of the CNT to the composite helps structure to absorb the impact energy better by creating a larger damage area in the structure and thus the damage spreads over a larger area. In the composites with hybrid structure, the impact energy absorption performance remained nearly same with the CNT reinforcement in the damped energy, and when the damaged structures were examined, the change was from 384.5 mm² to 386 mm². CNT's effect on impact performance wasn't significant to notice on composites with hybrid structure because incorporation of more than two different type of fibers to same composite brought more incompatibility to composite. Incompatibility can be seen with different fracture occurrences observed during SEM micrographs (Figure 6). On the other hand, uniformly occurred fractures with impact can be seen with glass and carbon fiber reinforced composites (Figure 7-8).

Table 1: *Energy (J), force (N), time (ms), displacement (mm), area of perforation (mm²) and chaga in area (%) data for the drop weight impact test applied to glass, carbon and hybrid fiber reinforced composite material structures.*

Composite	CNT amount (wt%)	Average Max. Energy (J)	Average Max. Force (N)	Average Displacement at E_{max} (mm)	Average Time at F_{max} (ms)	Average Area of Perforation (mm²)	Change in Area with CNT (%)
Glass	0	22.10	4613.82	12.75	4.42	384	+5.2
	0.5	24.64	4980.51	13.58	4.81	404	
Carbon	0	14.40	3098.02	11.27	3.98	335.5	+38.6
	0.5	31.77	2947.88	17.55	6.47	465	
Hybrid	0	24.08	5084.45	12.79	4.49	384.5	+0.4
	0.5	23.32	5569.50	12.40	4.32	386	

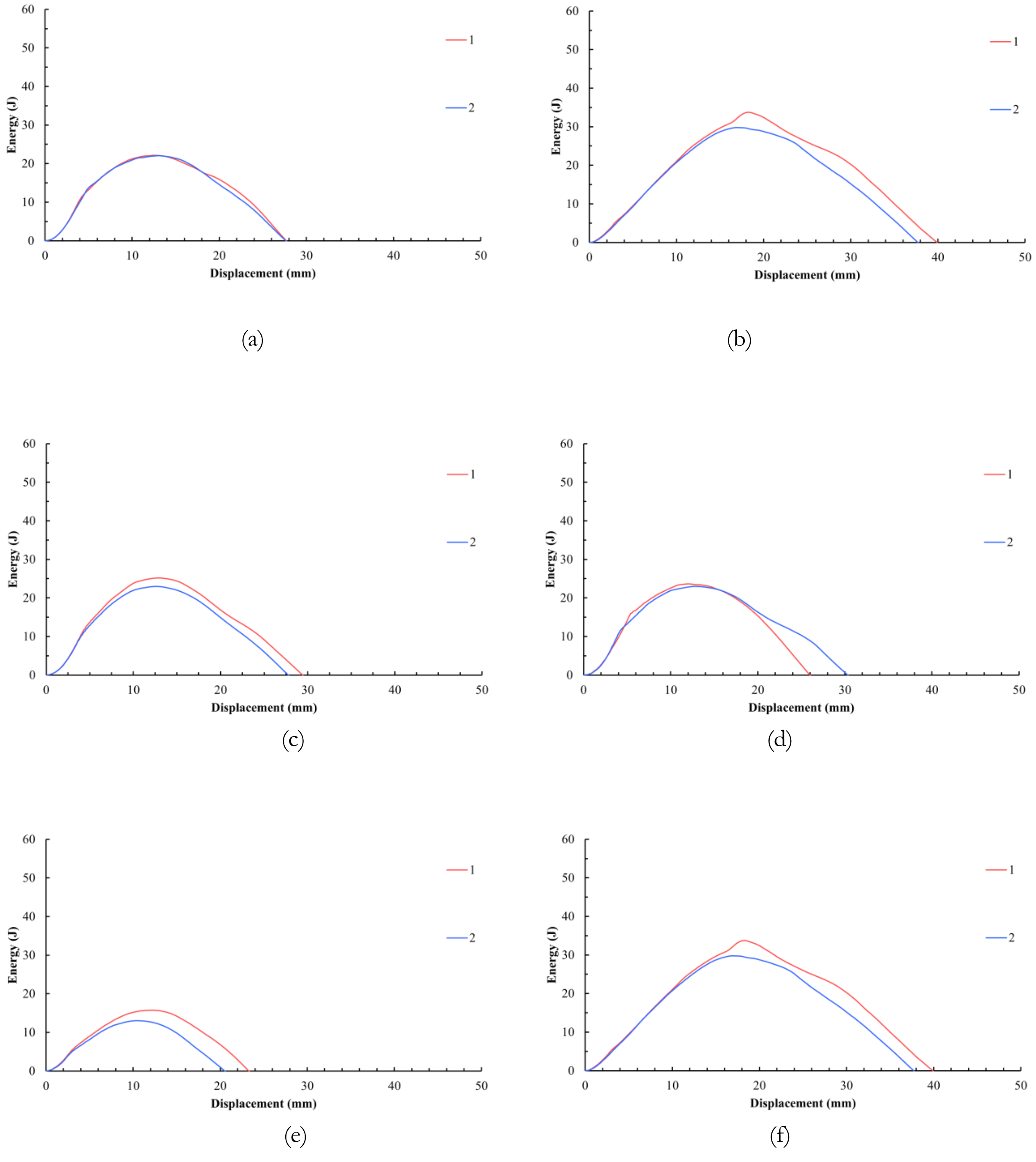

Figure 4: *Energy (J) – Displacement (mm) properties of glass (a), carbon (c), hybrid (e) reinforced epoxy matrix polymer composites and CNT added glass (b), carbon (d), hybrid (f) reinforced epoxy matrix polymer composites after drop weight impact testing.*

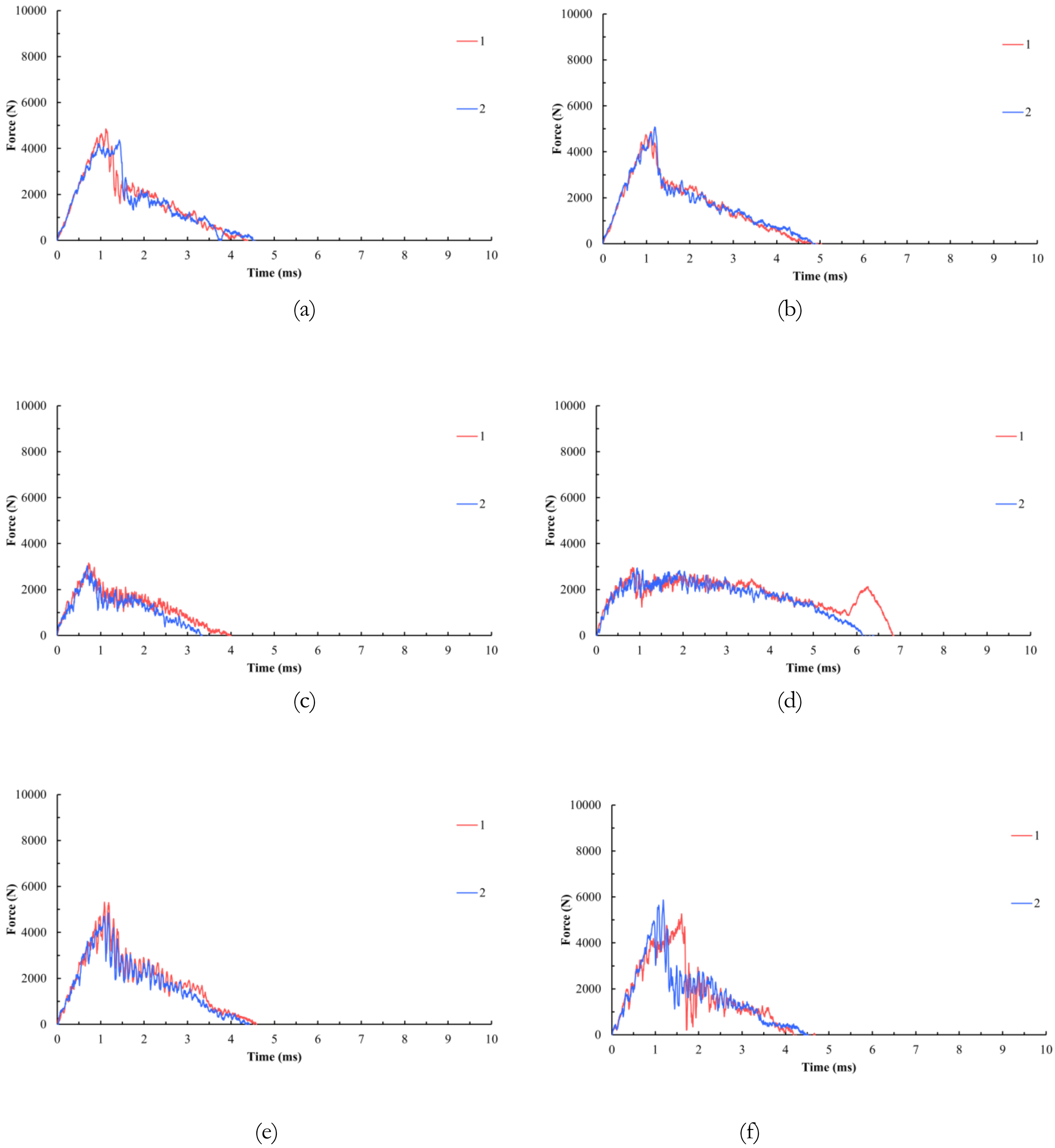

Figure 5: *Force (N) – Time (ms) properties of glass (a), carbon (c), hybrid (e) reinforced epoxy matrix polymer composites and CNT added glass (b), carbon (d), hybrid (f) reinforced epoxy matrix polymer composites after drop weight impact testing.*

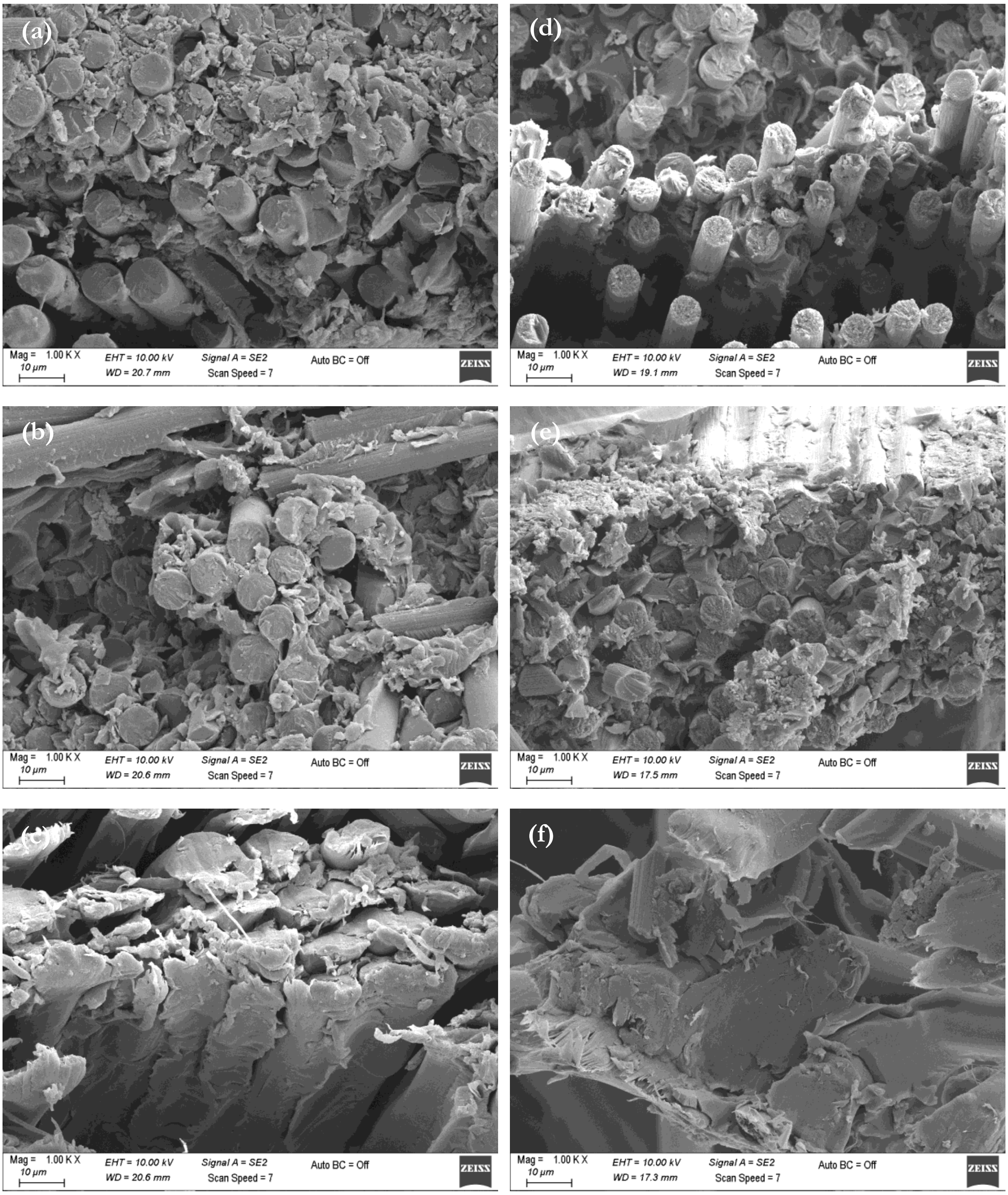

Figure 6: *SEM images of hybrid multi layered fiber reinforced CNT added (a,b,c) and CNT not added (d,e,f) polymer composite samples from fracture area for x1.0K magnifying. Glass fibers (a,d), carbon fibers (b,e) and aramid fibers (c,f) were positioned as glass-carbon-aramid from top to bottom with 3 layers in order to complete total 9 layers for both CNT added and CNT not added hybrid fiber reinforced polymer matrix composites.*

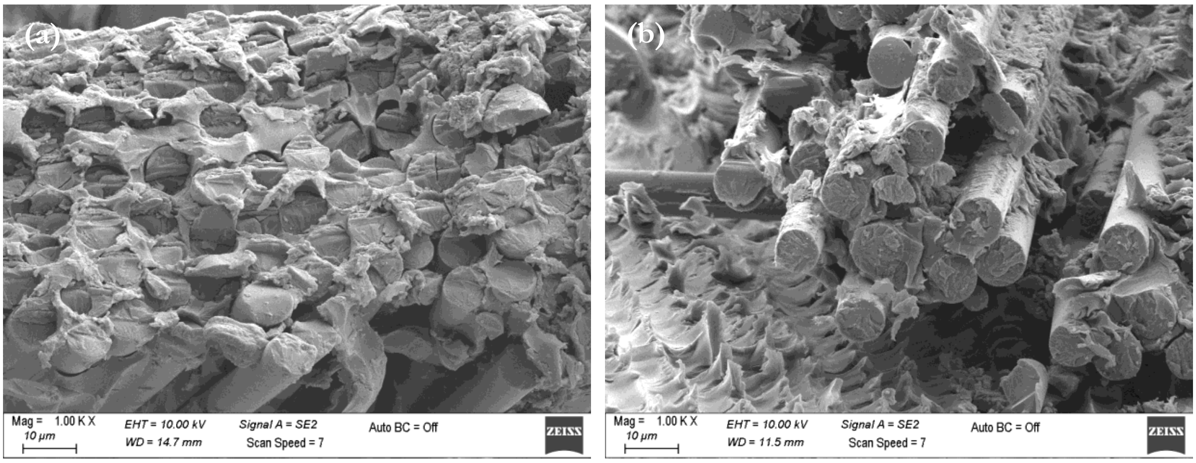

Figure 7: *x1.0K magnifying SEM images of multi layered glass fiber reinforced CNT added (a) and CNT not added (b) polymer composite samples from fractured areas.*

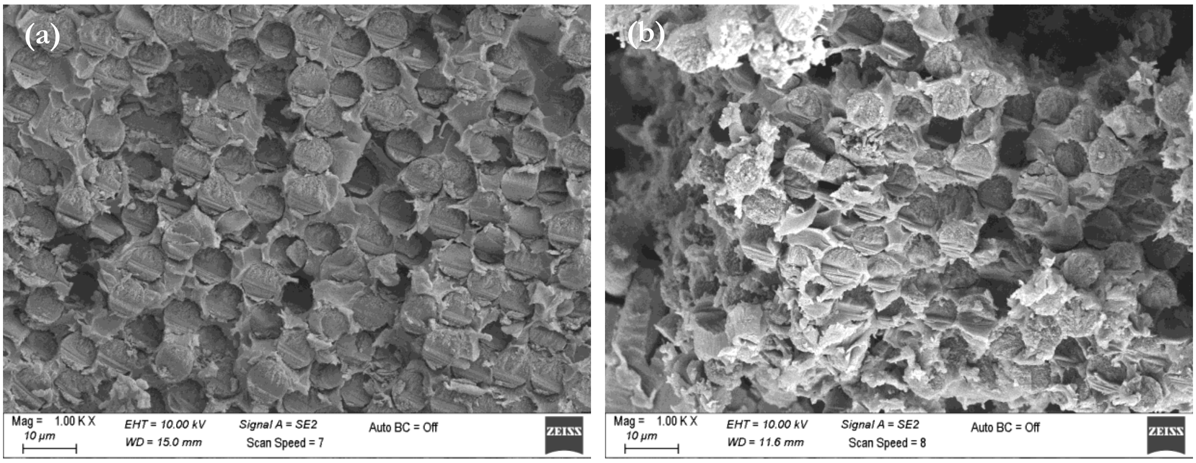

Figure 8: *x1.0K magnifying SEM images of multi layered carbon fiber reinforced CNT added (a) and CNT not added (b) polymer composite samples from fractured areas.*

4 Conclusions

In this study, the impact performance of a multi-layer fiber-reinforced polymer matrix composite incorporating carbon nanotubes (CNT) was investigated. Glass, carbon, and aramid fiber fabrics with identical weaves and similar areal density properties were employed as reinforcements. Alongside multi-layer composites consisting solely of glass and carbon fibers, intraply hybrid composites were manufactured, integrating glass, carbon, and aramid fibers. The production of multi-layer fiber-reinforced polymer composites involved the vacuum infusion method, both with and without the addition of CNTs. Samples were manufactured according to the ASTM D7136 standard, and a drop-weight impact test was conducted, delivering an energy input of 50 J. The composites with hybrid structure displayed the highest impact energy (J), followed by the composites with glass fiber, while the composites with carbon fiber demonstrated the lowest impact energy. Specifically, the impact energy performance increased by 120.7% for composites with carbon fiber and 11.5% for composites with glass fiber, whereas the composites with hybrid structure experienced a 3.2% decrease. Concerning displacement (mm), composites with carbon fiber exhibited the most significant increase at 55.7%, followed by composites with glass fiber at 6.5%. In contrast, composites with hybrid structure showed a 3.1% decrease. The highest contact force (N) occurred in the composites with hybrid structure, followed by the composites with glass fiber and the composites with carbon fiber. The introduction of CNT reinforcement resulted in a 5.2% increase in the damage area for composites with glass fiber and a significant 38.6% increase for composites with carbon fiber. In summary, CNT reinforcement enhanced the absorption of impact energy in the composite, leading to a larger damage area and more widespread distribution of damage.

5 Declarations

5.1 Competing Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

5.2 Publisher's Note

AIJR remains neutral with regard to jurisdictional claims in published institutional affiliations.

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