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The effect of MWCNTs on the mechanical properties of woven Kevlar/epoxy composites

Iman Taraghi [*1], Abdolhossein Fereidoon [1] and Ali Mohyeddin [2]

[1] *Department of Mechanical Engineering, Semnan University, Semnan, 35131-19111, Iran*
[2] *Department of Mechanical Engineering, Parand Branch, Islamic Azad University, Parand, Iran*

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Abstract. This manuscript presents an experimental investigation on the effect of Multi-walled carbon nanotubes (MWCNTs) addition on the tensile, flexural and impact properties of woven Kevlar fabric reinforced epoxy composites. MWCNTs were dispersed in the epoxy resin by sonication technique and the samples were fabricated by hand layup laminating procedure. Scanning electron microscopy (SEM) was used to characterize the microstructure of produced samples. The effects of adding small amounts (≤ 1%) of MWCNT on the tensile, flexural and impact (Izod) behaviors of laminated composites were analyzed. Results revealed that MWCNTs enhanced the Young's modulus up to 20%, bending modulus up to 40%, and impact strength up to 45% in comparison with woven Kevlar fabric/epoxy composites. It was found that the maximum improvements in mechanical properties were happened for 0.5 wt.% MWCNT.

Keywords: Kevlar fiber; multi-walled carbon nanotube; thermosetting resin; laminated composite; mechanical testing

1. Introduction

Growing industrial demand on strong and light materials gives the polymer composites a very important role in today's modern technologies. The development of new ultra-strong composites is of great interest for various industries such as aerospace, defense, marine, automotive, civil infrastructure, sporting goods, etc. offering high strength and stiffness, dimensional stability, and thermal properties (Chowdhury *et al*. 2006).

Epoxy resins have been widely used as structural adhesives because of their excellent bonding, high-adhesion, low-weight, good chemical resistance, and thermo-mechanical characteristics. However, some of its inferior characteristics e.g., impact strength, resistant to weather, and thermal stability have restricted its use in high performance applications (Alagar *et al*. 2000). Epoxy resin-based composites are used for aerospace, ballistic and engineering hardware components, pollution control equipments, automotive parts, electrical components, and rehabilitation products, etc. Kevlar fiber is a good candidate for reinforcing the polymers owing to its low density and high specific strength (Chen *et al*. 2012). It is competitive with steel in strength, yet has only a fraction

*Corresponding author, M.Sc., E-mail: taraghi.iman@students.semnan.ac.ir

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of steel density (O'Connor *et al*. 2009b). These impressive properties are due to its molecular structure, developed during the production process which is based on liquid crystal technology. The spinning process aligns the molecular chains parallel to the fiber axis leading to a highly ordered structure with a high degree of crystallinity (Lafitte and Bunsell 1982). Thus, Kevlar has a high potential for a variety of high-performance applications including aircraft and automotive industries, ballistic protection armor, helicopter blades, pneumatic reinforcement, and sporting goods. The mechanical properties of Kevlar fibers are related to their particular microstructure characterized by several features such as fibrils, radial pleated sheets and skin-core differentiation (Bencomo-Cisneros *et al*. 2012).

Having been discovered by Iijima (1991), carbon nanotubes (CNTs) received much attention of scientists and industries due to high aspect ratio and excellent mechanical, electrical, and thermal properties (Thostenson *et al*. 2001), (Baur and Silverman 2007). In the last decade, CNTs are considered to be one of the most effective reinforcing fillers in fabricating high strength, light weight polymer composites because of low density (as low as about 1.3 $g.cm^{-1}$), as well as superior strength (up to 63 GPa (Yu *et al*. 2000) and Young's modulus (up to 1 TPa (Wong *et al*. 1977)). CNTs are also incorporated into fiber reinforced composites in order to reinforce the matrix 12 Kim and Hahn (2011) (Abot *et al*. 2008) and/or improve the fiber-matrix interfacial adhesion (Thostenson *et al*. 2002), (Chen *et al*. 2009).

The most common methodologies to disperse the CNTs in either thermoset or thermoplastic resins include direct mixing (Kearns and Shambaugh 2002, Moore *et al*. 2004, Sandler *et al*. 2004) and solvent aided dispersion (Dalton *et al*. 2003, 2004, Lau *et al*. 2005, Miaudet *et al*. 2005, Razal *et al*. 2007, Vaisman *et al*. 2006). Martone *et al*. (2012) employed various mixing techniques including magnetic stirring, roll milling, mechanical agitation or high-energy sonication for both procedures to achieve dispersion and homogenization of CNT within the epoxy resin. They observed that combination of direct mixing and sonication results the lowest density of MWCNTs micron sized aggregates, in comparison with other stirring methods. Performing DMA tests on these samples also showed the highest enhancement in flexural modulus (about 7 percent) for the case of direct mixing-sonication method (Martone *et al*. 2012). Zhou *et al*. (2008) studied the electrical, thermal and mechanical properties of epoxy filled with 0.1-0.4 wt.% CNT. Their mechanical test results showed that the modulus increased with higher CNT loading percentages (up to about 12 percent), whereas the maximum enhancements in strength (about 27 percent) and fracture toughness (about 36 percent) were achieved for 0.3 wt% CNT loading. Hernandez-Perez *et al*. (2008) investigated the effect of 1 wt.% MWCNTs with different aspect ratio on the mechanical, thermo-mechanical and electrical properties of a brittle epoxy resin. They reported that while tensile properties showed very limited improvement, the impact resistance and fracture toughness of the nanocomposites were significantly improved, specially for the nanotubes with higher aspect ratio. Nadler *et al*. (2009) studied the effect of CNT surface functionalisation on the mechanical properties of MWCNT/epoxy composites. They found that only 0.5 wt.% CNT content revealed an reinforcement effect up to + 30% for modulus of elasticity. Montazeri *et al*. (2010) used untreated and acid-treated MWCNTs to reinforce the epoxy. The results of tensile test showed higher Young's modulus values for the composite samples prepared using acid-treated MWCNT. Also the dispersion of acid-treated MWCNT in the epoxy matrix was more uniform compared to their untreated counterparts.

Some previous researches have studied the combinational role of CNT and Kevlar on reinforcing polymers. O'Connor *et al*. (2009a) reported a new method for preparation of Kevlar-CNT composites by swelling the commercially available Kevlar fibers in the suspension of

CNTs in the solvent N-methylpyrrolidone (NMP), and demonstrated the ability of this novel additive for improving the mechanical properties of various polymers. Guo *et al*. (2009, 2010) increased the antiwear and friction-reducing abilities as well as the load-carrying capacity of Kevlar fabric/phenolic composites by air-plasma treatment of Kevlar fabrics. Kang *et al*. (2011) studied the rheological behavior of Magnetic CNTs and Their Application as Kevlar Coating. Wang *et al*. (2011) studied on Kevlar oligomer functionalized graphene for fabricating polymer composites. The organophilic and thermal stability of functionalized graphene were greatly improved as compared with that of graphene oxide. Chen *et al*. (2012) reported that because of high crystallinity, the surface of Kevlar fiber is chemically inert and smooth, making poor its adhesion to the resin matrix. Therefore, to use Kevlar fibers as reinforcement, surface modification is essential to enhance their reinforcing effect (Chen *et al*. 2012). Liu *et al*. (2011) enhanced the wet ability of aramid fibers through increasing the oxygen-containing functional groups on their surface. They showed that the interfacial bonding strength of treated aramid fiber/epoxy resin matrix composites exceeded that of untreated aramid fiber/epoxy resin matrix composites up to 50%. Reis *et al*. (2012) studied the impact response of laminated Kevlar-epoxy composites filled with cork powder and treated with nanoclay particles. Taraghi *et al*. (2014) investigated the effect of MWCNTs on the low-velocity impact response of laminated Kevlar/epoxy composites at ambient and low temperatures. They reported that MWCNTs can improve the performance of these composites under impact loading at ambient and low temperatures.

Regarding to previous studies, there is no report on the effect of MWCNTs on tensile, flexural and impact (Izod) properties of Kevlar/epoxy composites. Therefore, the present research is aimed to produce and investigate the mechanical behavior of MWCNT/Kevlar reinforced epoxy composites dispersion of the nanoparticles in the matrix were characterized using SEM. Standard tensile, flexural and impact tests were performed on the nanocomposite samples with different contents of MWCNT.

2. Materials and methods

There are two main methods to fabricate CNT-reinforced fiber nanocomposites: the dispersion of CNTs in the polymer matrix (Chou *et al*. 2010), or the growing CNTs onto the fiber surfaces (An *et al*. 2012). The former method was used in this study to prepare the required samples.

2.1 Materials

The materials used in this study were the epoxy resin, woven Kevlar, and MWCNT. Epoxy resin was commercially available EPOLAM 2002 from AXSON Technologies. The viscosity of the resin was 700 mPa.s at room temperature and the its glass transition temperature was 65°C. Kevlar fiber woven plain fabrics (AK502) were supplied by COLAN Products. The average fiber areal weight of Kevlar fiber was 175 g/m^2. MWCNTs were produced by US research Nano-materials, Inc by chemical vapor deposition (CVD) method. MWCNTs had Young's modulus of 1200 GPa, tensile strength of 150 GPa, Inside diameters of 5-10 nm, outside diameters of 10-20 nm, and lengths of 10-30 μm.

2.2 Manufacturing nanocomposites

2.2.1 Resin preparation

After drying MWCNT powder in an oven to get rid of moisture, it was mixed with epoxy resin in a high shear laboratory mixer at 2000 rpm for 30 minutes. Afterwards, an ultrasonic processor (Hielscher-UP400S, 20 kHz, 100 W/cm^2) was used to obtain a homogeneous mixture of epoxy resin and MWCNTs. A water bath was used to decrease the temperature during sonication. The MWCNT content was chose between 0 and 1.0 wt%. Since the mechanical mixing introduced air bubbles into the resin, a high vacuum was used for about 30 minutes for removing the trapped air and reaction volatiles from the mixture.

2.2.2 Fabrication of MWCNT/woven Kevlar fabrics/Polymer nanocomposites

The composite sheets were prepared by hand lay-up laminating technique using bi-directional woven Kevlar fabrics. Kevlar fabric sheets of size 33-25 cm weighing 14.58 gr were heated to 150°C in a hot air oven for 2 hours to remove moisture and then cooled to 30°C. Then they were covered by a pre-weighed mixture of epoxy prepolymer resin and a quantity of curing agent (hardener) using rubber roller. The stacking sequence consisted of 8 ply laminates of woven bi-directional Kevlar. Polymerization was achieved at room temperature within approximately 24 hours. The cross ply $[0/90]_{4s}$ laminates were then cured by a compression molding machine at 100 kPa pressure in room temperature. The weight fraction of fiber in the laminated nanocomposites was measured by burn-off test method and the areal density method according to ASTM D3171.

2.3 Tensile, flexural and impact tests

Tensile and flexural tests were carried out using a SANTAM universal testing machine, STM-150 with a 150 kN load cell at ambient temperature to determine the elastic modulus and strength of the laminated nanocomposite specimens. Following ASTM D3039 method, rectangular specimens of 250 mm × 25 mm × 2.6 mm, with epoxy-based adhesive tabs were tested at a loading rate of 2.0 mm/min.

Five samples of each type were tested to determine the average values of Young’s modulus and tensile strength.

Three-point flexural tests were conducted at ambient temperature using a STM-150 universal testing machine with a 5 kN load cell. According to ASTM D790, specimens of 127 mm × 12.7 mm × 2.6 mm with span length of 42 mm were tested at a crosshead speed of 1 mm/min. A flexural stress-strain curve was obtained based on the collected load-deflection data for each sample. Flexural modulus was calculated from the slope of stress-strain plot. Five samples of each type of eight-layered fabric composites were tested and the average values of flexural strength and modulus were determined.

A 2.6 mm thick composite sheet was cut into rectangular bars with width of 12.7 mm and length of 64 mm for notched Izod impact test. A single-edge V-shape notch with 2 mm depth and root radius of 0.25 mm was cut in the middle of the longer edge of each rectangular bar. The impact tests were carried out using an Izod impact tester (CEAST) with a 7.5 J pendulum in accordance with ASTM D256 method. The impact strength was averaged for five samples of each material type.

2.4 Morphological properties

The morphological properties of MWCNT/woven Kevlar fabric/epoxy nanocomposites were evaluated by Taraghi *et al*.(2014) using a VEGA-TESCAN SEM. Figs. 1(a)-(d) show the fracture

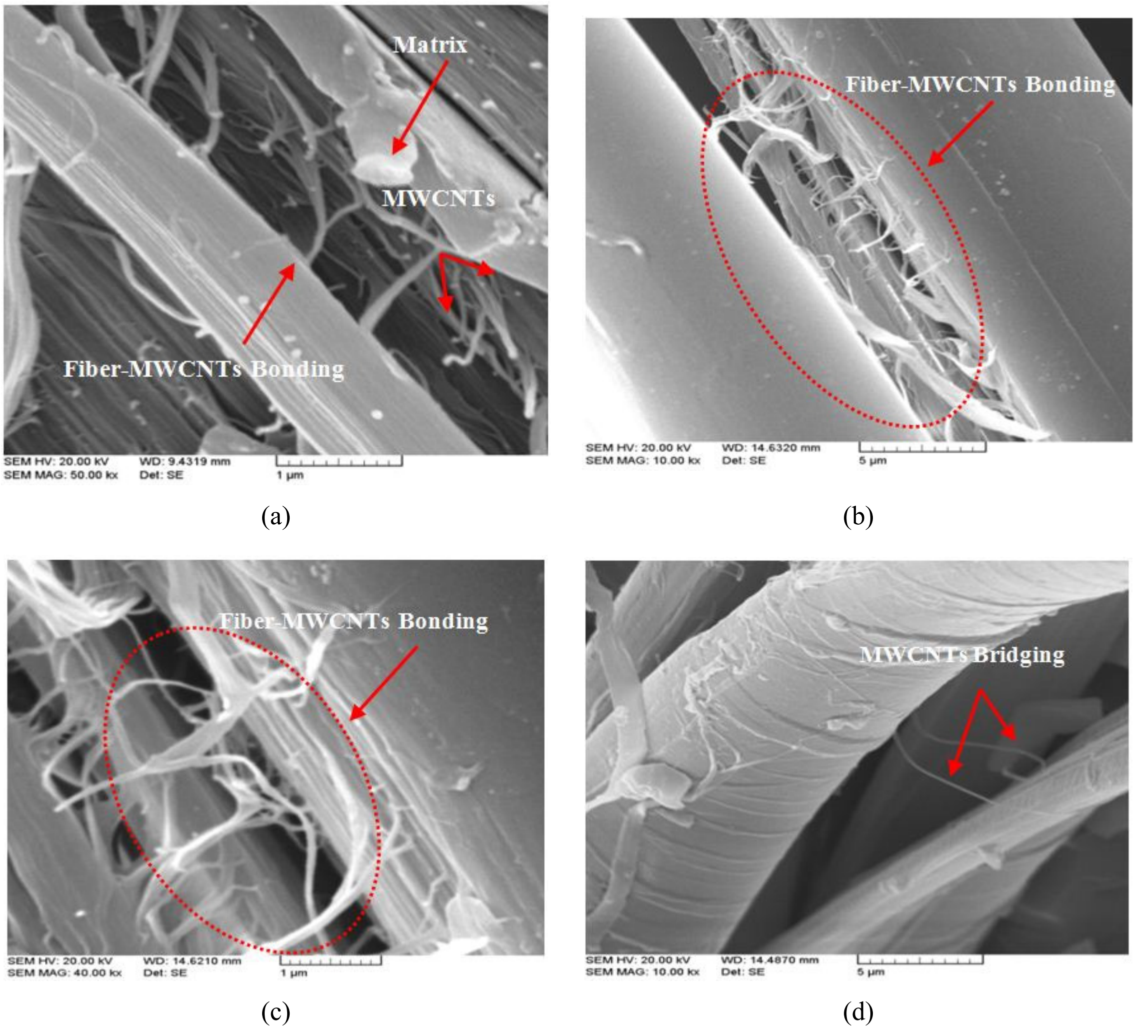


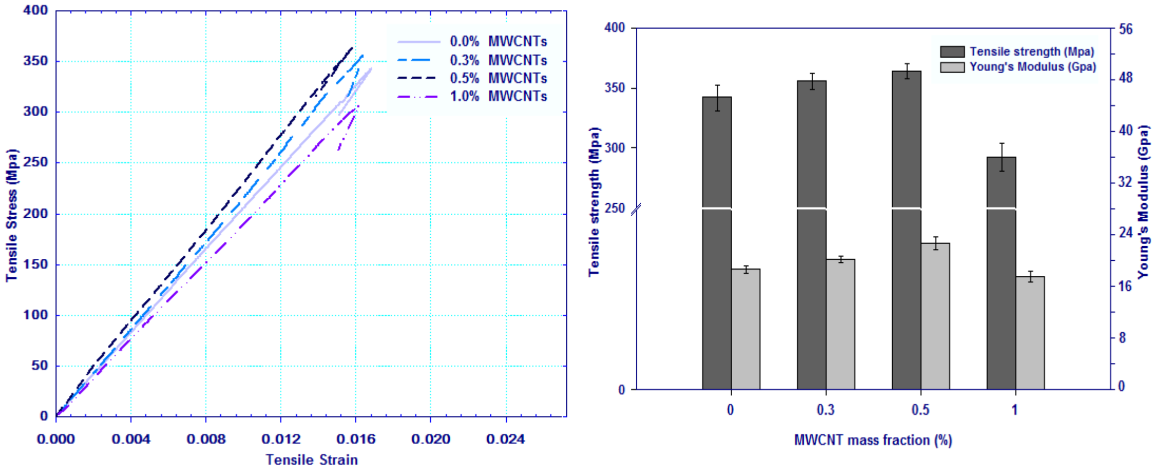
Fig. 1 SEM images of the fracture surface in MWCNT/woven Kevlar fabric/epoxy nanocomposites containing: (a) 0.3 wt.%; (b), (c) 0.5 wt.%; (d) 1.0 wt.% MWCNT

surfaces of Kevlar/epoxy composites reinforced with different contents of MWCNT. The SEM images in Figs. 1(b) and (c) indicate that nanoparticles are well dispersed in the epoxy resin. In Fig. 1(d), because of the agglomeration of MWCNTs, the matrix is absent on the fracture surface of nanocomposites, and only bridging of some nanoparticles between the fibers is clear.

3. Results and discussion

3.1 Tensile properties

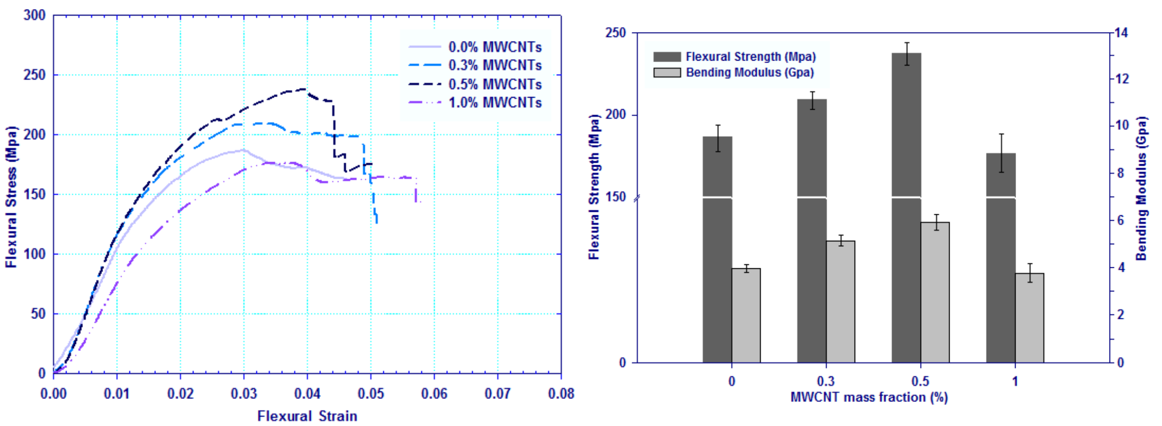
Tensile stress-strain curves are shown in Fig. 2(a). The effects of MWCNT content on the tensile



(a) (b)

Fig. 2 Tensile properties of woven Kevlar fabric/epoxy composites with different MWCNT contents: (a) stress-strain curves; (b) tensile strength and modulus

strength of composite samples are shown in Fig. 2(b) which shows the improvement of strength and modulus by adding MWCNT. As observed, the tensile strength of MWCNT/woven Kevlar fabric/epoxy laminated nanocomposites was raised about 6 percent by adding 0.5 wt.% MWCNTs, but got decreased when the MWCNT content was increased to 1 wt.%. In a similar manner, the tensile modulus of MWCNT/woven Kevlar fabric/epoxy laminated nanocomposites was increased about 20 percent by adding 0.5 wt.% MWCNT, while it was decreased for 1 wt.% MWCNT.



(a) (b)

Fig. 3 Flexural properties of woven Kevlar fabric/epoxy composites with different MWCNT contents: (a) stress-strain curves; (b) tensile strength and modulus

3.2 Flexural properties

The same trend as tensile properties is observed for flexural properties as shown in Fig. 3. Flexural stress-strain curves are shown in Fig. 3(a). The effect of MWCNT content on the flexural strength of composite samples is shown in Fig. 3(b). The flexural strength of MWCNT/woven Kevlar fabric/epoxy got increased about 27 percent by adding 0.5 wt.% MWCNT, but got decreased for higher value of MWCNT content. The flexural modulus was increased about 48 percent by adding 0.5 wt.% MWCNT, whereas it got decreased slightly with increasing the MWCNT content. The aggregation of unmodified MWCNTs at higher contents leading to weak matrix-fiber interfacial interactions is considered as the reason of mechanical properties deterioration.

As was expected, each sample possesses higher modulus and stiffness under tensile loading than under the flexural loading condition. This matter is confirmed by referring to Chamis (1977) in which, typical mechanical properties of some unidirectional continuous fiber composites of epoxy have been reported.

3.4 Impact properties

Izod Impact strength as well as the energy absorbed by various MWCNT/woven Kevlar fabric/epoxy composite samples are compared in Fig. 4. As shown, the impact strength was increased about 45 percent by adding 0.5 wt.% MWCNT. Also the values of absorbed impact energy show the highest impact energy for 0.5 wt.% MWCNT loading (about 27 percent rather than for 0 wt.% MWCNT). This matter contributes to extra energy dissipation mechanisms such as nanotube pull-out and fracture or bridging due to its interaction with polymer matrix. These adhesions could be attributed to effective grafting of MWCNTs on fiber as well as the micromechanical coupling between MWCNTs and epoxy.

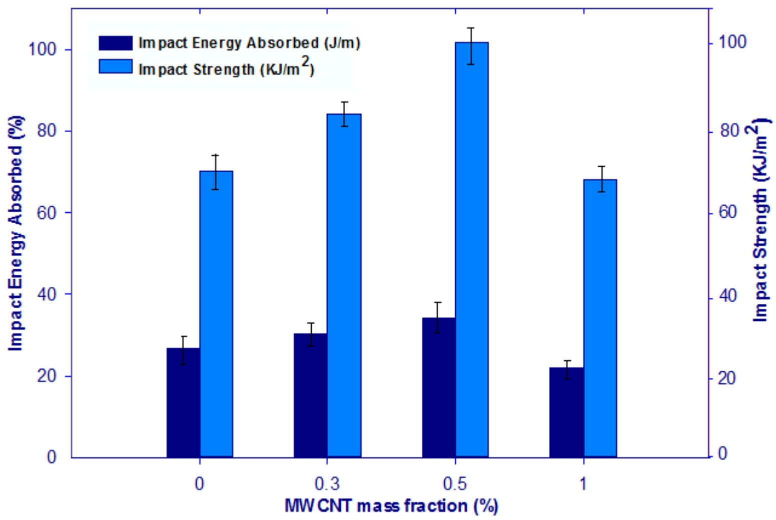


Fig. 4 Impact strength and absorbed impact energy of woven Kevlar fabric/epoxy composites with different MWCNT contents

4. Conclusions

The aim of this study was to evaluate the effect of MWCNTs on the mechanical properties of woven Kevlar fabric-epoxy laminated composites. The current research revealed that MWCNT is an appropriate candidate for tailoring the traditional fiber-reinforced composites. Deterioration in the properties observed for 1 wt.% MWCNT suggests MWCNT loading of 0.5 wt.% as an optimum value. Brief conclusions of the present investigation are as follow:

- Study of morphological properties showed that MWCNTs improves the adhesion of fiber and matrix as well as matrix mechanical properties.
- 0.5 wt.% MWCNT appears to be an optimum loading for MWCNT/woven Kevlar fabric/epoxy nanocomposites from the viewpoint of mechanical properties.
- Enhancements as much as 6% in strength and 20% in modulus were achieved from tensile mechanical tests.
- Flexural tests resulted improvements in strength and modulus up to about 27% and 48%, respectively.
- Impact tests resulted improvements in strength and absorbed energy up to about 45% and 27%, respectively.

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