

Effect of Carbon Nanotube on Adhesion Strength of E-Glass/Epoxy Composite and Alloy Aluminum Surface

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Abstract: In a metal and fiber reinforced epoxy hybrid with composite structure, weakest point is interface of metal surface and fiber patch (Adhesive layer). In this study single lap joint in some specimens with different weight fraction of multi wall carbon nanotubes in composite part, tested in functionalized and non-functionalized situation and shear strength between polymer matrix and alloy aluminum investigated. Based on obtained results, Samples contain non- functionalized carbon nanotube showed a positive effect on shear strength in comparison with specimens contain the same amount of functionalized nanotube. It was completely obvious from test results, shear strength between composite patch and aluminum 2024-T3 surface has been increased with CNT while aluminum has not been chemically affected by CNTs.

Key words: Epoxy hybrid • Adhesive layer • Carbon nanotube and matrix • Single lap joint • E-glass/Epoxy Composite

INTRODUCTION

Recently, nanoparticles have been attracting increasing attention in the composite community because they are capable of improving the mechanical and physical properties of polymer matrix [1- 3].

Recent advances in composites manufacturing technologies provide capable solutions to the production of complex, hybrid and large composite parts. Repairing metal structure like damaged aircrafts with polymer matrix patch is a new application for these materials because they can provide many advantages such as low cost, high strength to weight ratio, fewer processing requirement and formability. But low shear strength after adhering to aluminum surface, is serious problem in this technique. Shokrieh and Omid [4] used combination of carbon and glass fiber with epoxy adhesive to reinforce central cracked Al 2024-T3 plate. Ultimate strength was increased only 25% because of sudden dispatching in adhesive layer. in similar research Okafor *et al.* [5] tried to design single side repairing patch to validate simulation results, pre-cracked Al 2024-T3 plates were reinforced with boron/Epoxy patch, were provided. An increasing about 42% in specimens with composite patch was observed in comparison to specimens without patch but disbanding was occurred in adhesive layer as it was predicted.

After carbon nanotube (CNT) discovery by Iijima [6] and their remarkable mechanical properties [7] they were used for polymer matrix reinforcement and mechanical properties optimization but most of them concentrated on achieving use of CNT challenge and opportunities.

The addition of amount of carbon nanotubes and carbon nanofibers can enhance the matrix-dominated properties of composites, such as tension, compression, stiffness, fracture toughness and interlaminar shear strength [8-12].

Qain and Dickey [9] dispersed multi wall carbon nanotube (MWCNT) throughout polystyrene matrices by a simple solution-evaporation method, about 36% to 42% and 25% increases in elastic modulus and break stress, respectively were indicated by tensile test. Also it was determined there is a significant load transfer across the nanotube-matrix interface. Ajayan *et al.* [13] investigated load transfer mechanism in CNT/epoxy composite. They showed MWCNTs are more effective in comparison because during load transfer to MWCNTs, only the outer layer is stressed in tension whereas all layers respond in compression. Advani *et al.* [14] experimentally checked CNT effect on adhesion between carbon fiber reinforced composite parts. They found, only 1 wt% of MWCNT in adhesive epoxy layer, shear strength has been increased to 32% by increasing CNT to 5wt% shear strength was

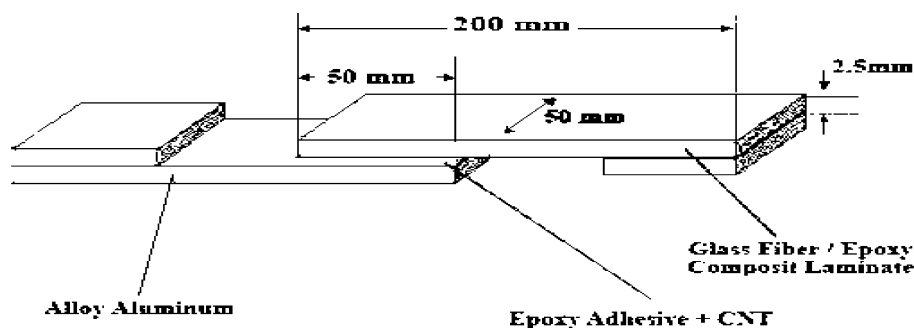


Fig. 1: The Configuration of the shear test (ASTM-D5868-01)

arised to 45.6 %. Tai *et al.* [15] entered epoxy resin to in CNT network by a melt mixing method. Ultimate strength and Young's modulus were dramatically increased 97% and 48% respectively. Schults and Gojny [16] reported CNT effect on epoxy resin thermo mechanical properties. Results showed by increasing amount of CNT, thermal properties have been increased; also functionalized effects are more noticeable. Schults [17] tested the epoxy composite fracture toughness with different kind of carbon nanotube (SWCNT, MWCNT, functionalized-MWCNT), by dispersing 0.5 wt% of SWCNT in L130i (an epoxy resin), composite fracture toughness was increased to 43% because of their high aspect ratio (L/D). Research has not been limited to epoxy/CNT composite. Gojny *et al.* [18] used mini calendar to disperse single wall carbon nanotube throughout epoxy resin (L135i) then with resin transfer molding (RTM), CNT/epoxy injected into glass layer. Simple tensile test in fiber showed there are no noticeable changes in tensile strength, but young's modulus has been increased. Zhu *et al.* [19] with short composite bending test found CNT influence on interlayer shear stress (ILSS) in E-glass fiber/epoxy composite. 20% to 45% increases in ILSS were observed in test results.

In this experimental study, tried to determine CNT influence on adhesion strength between glass fiber/epoxy composite and alloy aluminum 2024-T3 plate in order to find a solution for hybrid composite problems with carbon nanotubes. Adhesion in a single lap joint in combined Al 2024-T3 and glass composite was evaluated according to ASTM D5868 [20].

Experiment: Test specimen preparation, contains three steps: (i) metal surface preparation (ii) composite patch preparation and (iii) adhering composite and Aluminum.

Metal Section Preparation: Alloy Aluminum 2024-T3 has been used as metal part in single lap joint test specimen.

A sheet of Al 2024-T3 with thickness of 2.54 mm was cut to strips of 200 mm by 50 mm according to ASTM D5868. As shown in Figure (1).

Aluminum surface should be prepared for adhering composite part, so modified FPL- Etch was used [22]. In this method involved degreasing with Methyl Ethyl Ketone (MEK), water break test for inspection of cleaning procedure, sand blast abrasion with aluminum-oxide sand, dry tissue wipe to remove contaminant caused by sand blast, an alkaline rinse at 60-70°C, a hot water rinse, etching in a sulfo-chromic solution and air drying and then followed with a treatment of γ -glycidoxypyltrimetoxy silane, applied as a 1% aqueous solution by brushing for 10 min. the silane solution was allowed to hydrolyze for 1 hour and then used within the next 2 hour.

Composite Patch Preparation: In order to evaluate CNT effect on fiber reinforced polymer composite, multi wall carbon nanotube (MWCNT) should dispersed in Matrix. CVD technique has been used from Research Institute of Petroleum Industry (RIPI) to produced MWCNT. MWCNT outer diameter is 23 nm and inner diameter is 11 nm also these CNT functionalized with Oxidation and Ultraviolet ray technique. Figure (2) showed carbon nanotube TEM image before and after functionalized.

Multi wall carbon nanotube (MWCNT) and functionalized MWCNT (-COOH) with three different weight fraction of total resin and hardener weight (0.5, 1, 1.5 Wt %) have been dispersed directly into HY560ch (a polyamide hardener from Huntsman) with ultra sonic device for 30 min at 60% power. Final production has been mixed with LY 564 (an epoxy resin from Huntsman) for 10 min at 700 RPM. 7 layer of unidirectional E-glass fiber have been cut in dimension of 250 mm \times 60 mm, modified resin has been injected into resin by hand lay up method to satisfy 50% fiber volume fraction.

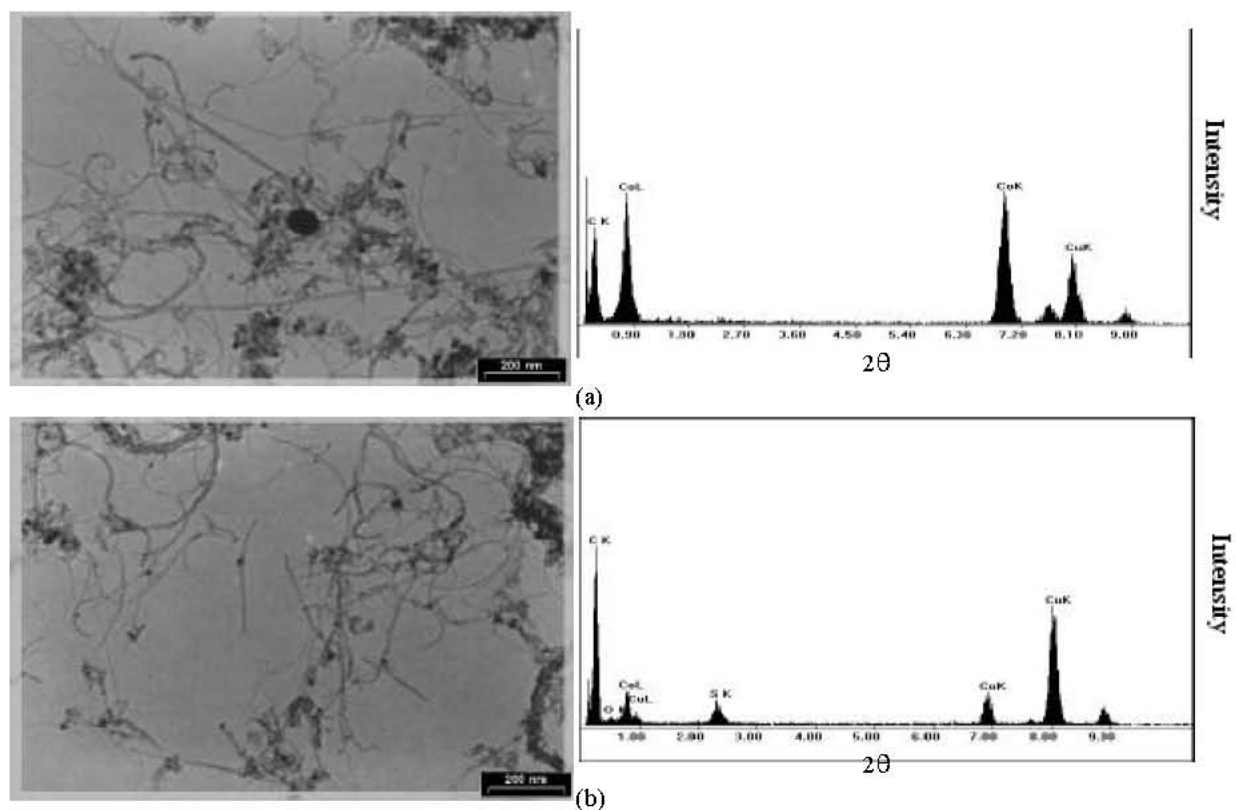


Fig. 2a-b: a) EDX analysis of the acid treated samples and Carbon nanotube micrograph for 0.5 wt% before functionalization, b). after functionalization

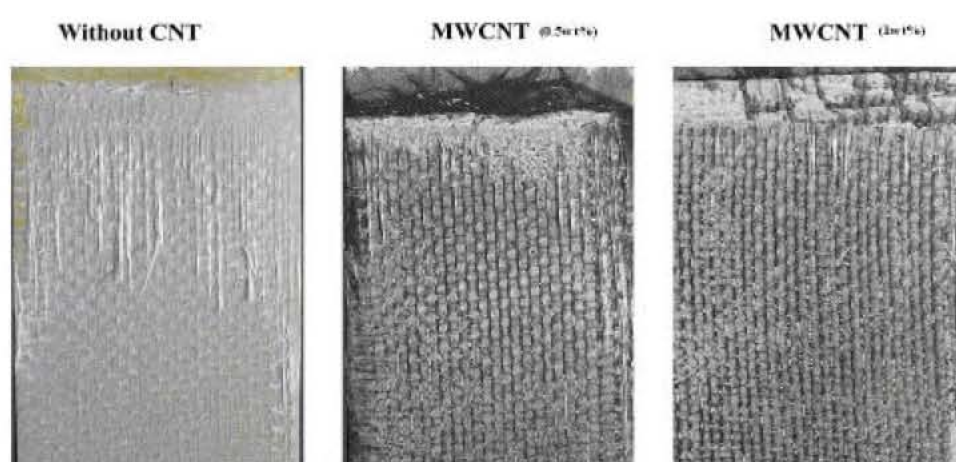


Fig. 3: Prepared single lap joint test specimen

Adhering Composite and Aluminum: From bubble exit vacuum bagging technique [20] brushing modified resin on prepared surface of Aluminum 2024-T3, then put the E-glass wet layers on Aluminum to overlap an area with dimension of $50_{mm} \times 50_{mm}$ in order to fix composite part on aluminum surface. Specimens have been left for 15 hour at $50^{\circ}C$ under vacuum bag for

curing composite patch and adhesive. Additional part has been cut by rotational saw disk. Figure (3) shows a prepared specimen.

Test: Due to ASTM D5868-01, the two ends of sample were pulled away from each other at relative speed of 12.7 mm/min. specimens were tested by using a

Zwick/Amsler tensile and Fatigue test machine with computer data acquisition. (Fracture & fatigue Laboratory of Mechanical Department of Iran University of Science & Tech).

RESULTS AND DISCUSSION

As shown in Figures 2, the dark points are metal particles and amorphous carbon materials. With acid treatment, the metal catalyst was eliminated, the closed tube was opened and then the length of nanotubes was shortened.

In agreement with TEM investigation, EDX analysis of the acid treated samples showed a decrease in the metal catalyst (cobalt). Agglomerates of nanotubes are because of strong van der Waals forces between nanotubes.

Test results saved as load-displacement curve as shown in Figure (4).

As shown in Figure (4) it can be seen that tensile modulus and strain at break with increasing CNT increased and strain at break for nano composites in comparison with specimens without CNT decreased.

For normalizing results the pick tensile test has been divided by lap area and the average shear strength of the bonding area was obtained. Note that to check repeatability, shear test of each MWCNT and MWCNT-COOH was repeated three times. Figure (5) shows the average shear strength of specimens with different MWCNT and MWCNT-COOH weight fractions.

Maximum load was measured from load-displacement curve Figure (4) and ILSS calculated from equation (1) [20]:

$$F^{abs} = 0.75 \frac{P_m}{b \times h} \quad (1)$$

Where F^{abs} is relatively inter layer shear stress, P_m is maximum load, b is width and h is specimen thickness.

Effect of Aluminum Surface Preparation: As shown in Fig. (5) Shear strength in adhesive layer in prepared aluminum surface in compare with unprepared one increased from 3.7 Mpa to 13.8 Mpa. Microscopic images were used to determine separation mechanism. Adhesive layer on prepared Aluminum surface, even after composite dispatching showed resin penetration into mechanical and chemical etching holes of surface; so connection area between adhesive layer and aluminum surface was increased, thus failure was occurred inside adhesive layer while completely separation at interface of adhesive layer and Aluminum surface caused. Lower shear strength in addition γ -glycidoxypolytrimetoxo silane usage FPL method [20] for aluminum surface preparation is another reason for shear strength increasing because it acts as a link which reacts from epoxy head with LY564/HY560 and on the other side (silane head) compose with hydroxide on aluminum surface.

Figure (6a) shows the Microscopic image of bound area; no surface treatment. Figure (6b) shows the Microscopic image of bound area; modified FPL surface preparation caused epoxy adhesive remained on lap area after separation.

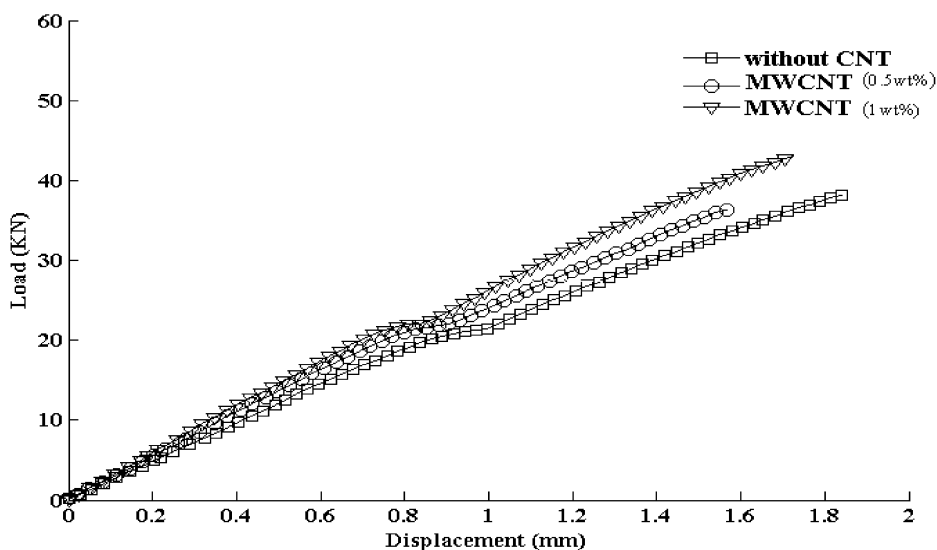


Fig. 4: Average shear strength of the adhesion specimens

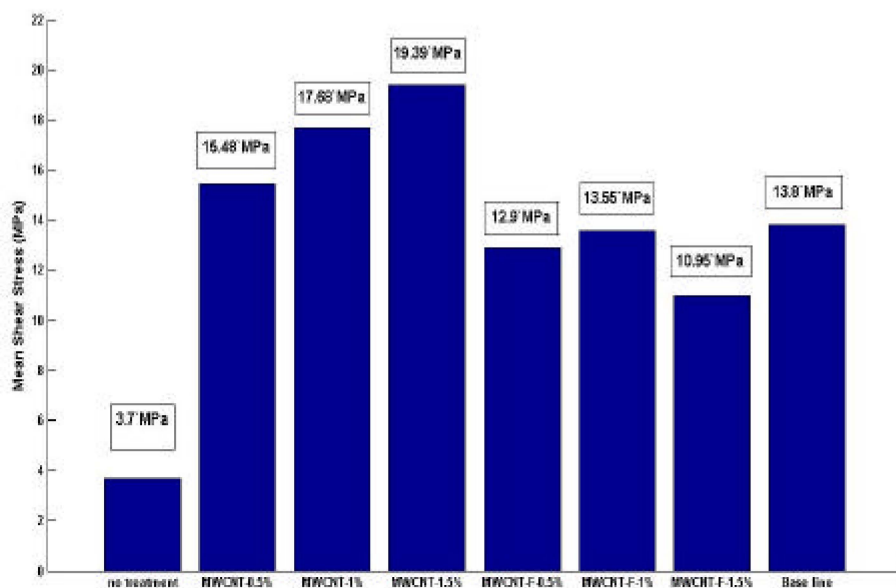


Fig. 5: The average shear strength of specimens with different MWCNT and MWCNT-COOH weight fractions

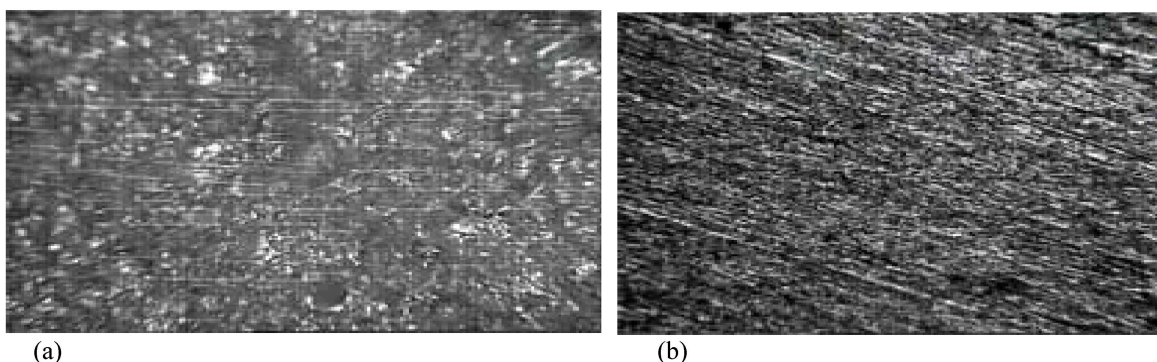


Fig. 6: Microscopic image of bound area; (a). no surface treatment, (b). modified FPL surface preparation caused epoxy adhesive, remained on lap area after separation

Effect of CNT: Fracture toughness and adhesion between fiber and resin are considered as main parameters in composite inter layer shear stress. By using MWCNT with 1.5 weight fraction, adhesion was increased more than 40% from reference test (adhesive layer without CNT) and shear strength decrease to 10.98 MPa when functionalized CNTs (MWCNT-COOH) were used. Surface observation showed that lap area was covered with epoxy adhesive layer or E-glass fiber.

In bending, composite layer tries to slice, so interface of resin and fiber acts as an important role in shear strength. By injecting matrix into fibers and CNT was dispersed, penetrated between layers and increase interface of fibers and resin. So ILSS that will be directly depended on CNT length will be increase. CNT acts as a bridge and block the movement of layer. After CNT

distortion, it poll out from layer and failure will be start. Non functionalized CNT because of their production technique have longer length in comparative to COOH-MWCNT.

When MWCNT were used, not only epoxy layer remained on lap area on aluminum surface but also it contains noticeable amount of fiber which belongs to first composite layer but in specimen with MWCNT-COOH failure was occurred in boundary of adhesive layer and aluminum surface so average shear strength decreased (As shown in Figure (7)).

As shown in Figure (8) fibers failure observed with MWCNT, but in specimens with MWCNT-COOH occurred along to layer in E-glass fiber system, thus interlayer adhesion was decreased finally maximum shear strength obtained in lower amount.

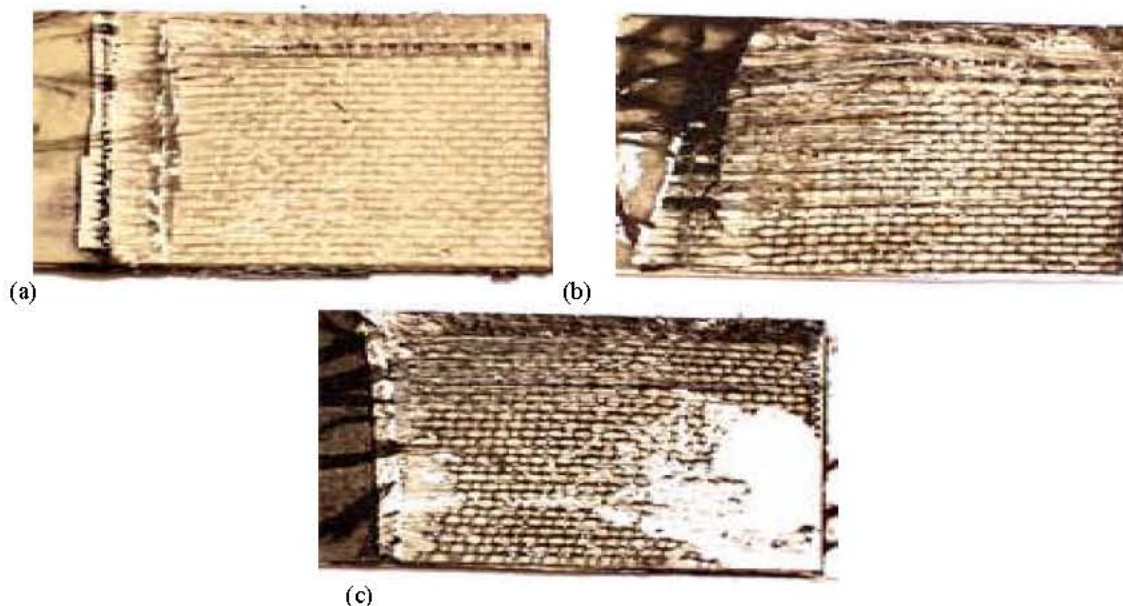


Fig. 7: Bound area after separation; (a). without CNT, (b). MWCNT 1.5 Wt%, (c). MWCNT-COOH 1.5 Wt%

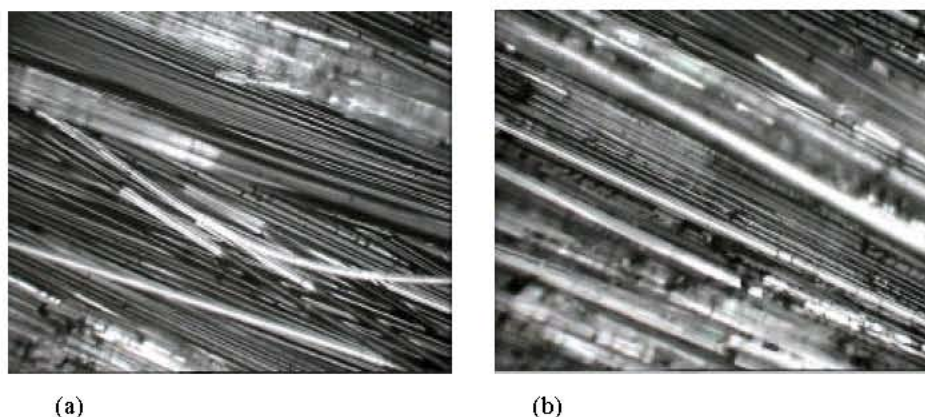


Fig. 8: Failure mode; (a). fibers were failed beside MWCNT, (b). failure occurred between two layer

CONCLUSION

Shear strength in adhesive layer in prepared aluminum surface in compare with unprepared one increased from 3.7 MPa to 13.8 MPa. So Aluminum surface preparation acts an important role on increasing adhesion between composite patch and aluminum surface.

This experimental study demonstrates the benefits of using MWCNT in bonding E-glass/epoxy composite and Aluminum plates. experiments showed that by adding 1.5 wt% MWCNT in the epoxy adhesive, shear load has been effectively transferred from the adhesive to the E-glass fiber system in the composite laminates and improved the average shear strength of the adhesion by

40.5% but functionalized MWCNT synthesized with oxidation method has not been recommended in this manner.

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