

# Properties Modification of Fiber Metal Laminates by Nanofillers

R. Eslami-Farsani, S. M. S. Mousavi Bafrouyi

**Abstract**—During past decades, increasing demand of modified Fiber Metal Laminates (FMLs) has stimulated a strong trend towards the development of these structures. FMLs contain several thin layers of metal bonded with composite materials. Characteristics of FMLs such as low specific mass, high bearing strength, impact resistance, corrosion resistance and high fatigue life are attractive. Nowadays, increasing development can be observed to promote the properties of polymer-based composites by nanofillers. By dispersing strong, nanofillers in polymer matrix, modified composites can be developed and tailored to individual applications. On the other hand, the synergic effects of nanoparticles such as graphene and carbon nanotube can significantly improve the mechanical, electrical and thermal properties of nanocomposites. In present paper, the modifying of FMLs by nanofillers and the dispersing of nanoparticles in the polymers matrix are discussed. The evaluations have revealed that this approach is acceptable. Finally, a prospect is presented. This paper will lead to further work on these modified FML species.

**Keywords**—Fiber metal laminate, nanofiller, polymer matrix, property modification.

## I. INTRODUCTION

**D**URING the last decades, composite materials have been subjected to interest fields of researchers. These materials have special characteristics such as low specific mass [1]. The composite materials have some disadvantage such as poor impact and residual strength. The aluminum alloys have poor fatigue life. FMLs combined advantages of metals (such as aluminum alloys) and composites and used them in a hybrid structure simultaneously. It has not only the properties of composites but also aluminum alloys. FMLs have such characteristics as low specific mass, high bearing strength, impact resistance, corrosion resistance and high fatigue life simultaneously. Initial FMLs material, ARALL (Aramid Fiber Reinforced Aluminum laminate) was introduced at Delf University of Technology [2]. Khalili et al. have studied of mechanical properties of steel/aluminum and GRP laminates. It was found that damage tolerance limit and impact damage tolerance of FML is superior when compared to that of GRP [3]. Sadighi et al have reviewed on impact resistance of FML. In term of fiber types, GLARE (Glass Laminate Aluminum Reinforced Epoxy) has proven its significant improvement in the impact resistance over ARALL and CARALL (Carbon Reinforced Aluminum Laminate), as lower permanent deflection, smaller damage width and higher energy absorption are recorded for various type of GLARE [4].

R. Eslami-Farsani, and S. M. S. Mousavi Bafrouyi are with the South Tehran Branch, Islamic Azad University, Tehran, Iran (e-mail: r\_eslami@azad.ac.ir, saleh\_musavi@yahoo.com).

Rajkumar et al. have investigated of tensile and bending of aluminum based hybrid FMLs. They have focused on the effect of strain rate on tensile and flexural behavior of FML. The results have shown that the tensile strength increases with increasing strain rate. However, the flexural strength decreases with increasing strain rate [5]. Then, fatigue life of FML has been investigated by researchers. Yue et al. have worked on fatigue crack growth in FML. Fatigue crack growth rates in notched FML under constant amplitude fatigue loading were investigated experimentally and numerically. The formula to calculate bridge stress profiles of FML was derived based on the fracture theories [6]. Spronk et al. have introduced a methodology to predict the cycle to crack initiation in a notched FML subjected to cyclic loading [7]. Huang et al. have studied delamination extension and fatigue crack growth behavior of GLARE under single overloads [8]. Also Wang et al. have derived an analytical model for fatigue crack prediction containing discretely notched layers [9]. Rans et al. have predicted the influence of temperature on fatigue crack propagation [10].

Hausmann et al. have compared analytical and numerical thermal residual stress models of FMLs [11]. Daghigh et al. have investigated creep behavior of Basalt Fiber Metal Laminates (BFML). It was found that the creep resistance of Basalt Fiber Reinforced Epoxy (BFRE) is higher than Glass Fiber Reinforced Epoxy (GFRE) [12].

Nowadays, composite materials are modified by nanofillers. Herein, a lot of researches have been done [13]-[18]. Graphene has attracted enormous attention in recent years owing to its expectable properties and applications [19]. The researchers have investigated the wonderfully applications of graphene in composite fields [20]-[24]. In recent years, graphene fiber has risen as a new carbonaceous fiber with high expectations in term of mechanical and functional performance. Xu and Gao have reviewed the rapid advances in the promotion of graphene fiber properties [25]. The CNTs have beneficial properties similar to graphene. These can modify the characteristics of polymeric composites [26]-[29]. Kulkarni et al. have numerically and experimentally studied elastic response of carbon nanotube fiber reinforced polymeric composite [30]. Lau et al. have reviewed on nanotube and nanotube/nanoclay in polymer composite materials [31]. Finally, Shokrieh and Rafiee have studied the tensile behavior of an embedded carbon nanotube in polymer matrix with non-bonded interphase region [32].

Ning et al. have studied modifying of interlaminar mechanical properties in CARALL fabricated by carbon fiber reinforced plastic (CFRP) layers combined with aluminum

alloy layers. They have added nanofiller i.e., Vapor Grown Carbon Fiber (VGCF), to the interface of CFRP and aluminum alloy layers. The fracture toughness has been improved [33]. Recently, Kaboodvand et al. have studied the effect of multi-walled carbon nanotubes on the tensile behavior of FMLs. The multi-walled CNTs have been dispersed in epoxy matrix. The tensile strength has been modified 14% by 0.3% wt of CNTs [34].

In this paper, it is intended to investigate the feasibility of properties modification of FMLs by nanofillers. Several important aspects that influence the properties of nanofillers on FML will also discussed in detail.

## II. FMLs

FMLs are hybrid composite structures based on thin sheets of metal alloys and plies of fiber reinforced polymeric materials. FMLs have not only the properties of composites but also metal alloys. Metals are for isotropic, have a high bearing strength and impact resistance and are easy to repair, while composites have excellent fatigue characteristics and high strength and stiffness. The most FMLs with aluminum sheets are ARALL (Fig. 1), based on aramid fiber, GLARE based on high strength glass fibers and CARALL Based on carbon fibers [2].

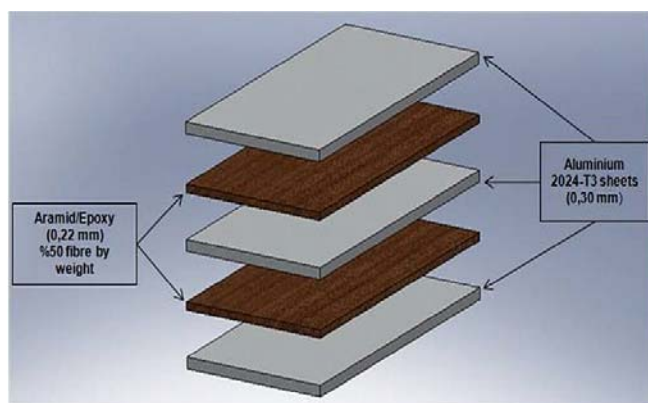


Fig. 1 Schematic presentation of Fiber metal laminate (ARALL) [2]

During past decades, increasing demand of modified FMLs has stimulated a strong trend towards the development of these structures. Hu et al. have investigated FML based on carbon fiber reinforced PMR polyimide. Titanium has been used to avoid galvanic corrosion problem. The titanium sheets were anodized. The process has been caused strong interface between titanium layer and fibre-reinforced polyimide layer (Fig. 2). The results have proved that the FMLs based on carbon fiber reinforced PMR polyimide offered excellent thermal properties. It is revealed that no delamination appears between titanium layer and the fiber-reinforced polyimide layer after 1000 times thermal shock [35].

Iriondo et al. have studied characterization of the elastic and damping properties of traditional FML and FML based on self-reinforced polypropylene (SRPP). The aluminum was employed as reference material in both FMLs. Final Results have shown that the FML based on SRPP offers higher

damping capacity than the traditional FML [36]. Therefore, study on new FML with modified properties is a necessity. Nanofillers have been used for improving and modifying the properties of polymers. Physical properties such as optical, magnetic, electronic, thermal, wear resistance, barrier to diffusion, water resistance or flammability resistance can be strongly affected by nanoparticle dispersion in polymer matrix [37], [38]. So the nanofillers can be used in the polymer matrix of FMLs for modifying the properties.

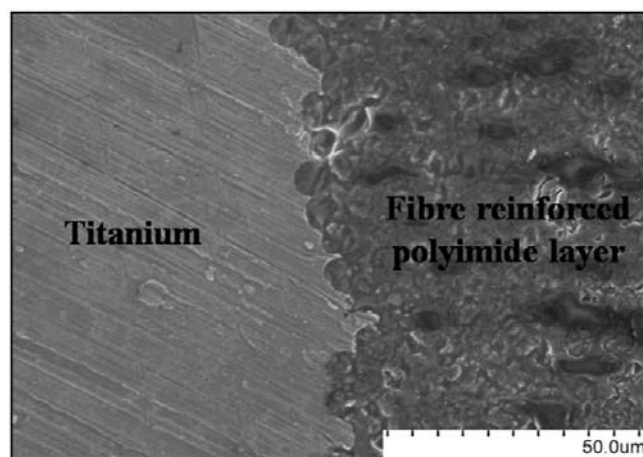


Fig. 2 SEM of the interface between titanium layer and fiber-reinforced polyimide layer [35]

## III. NANOCOMPOSITES

Nowadays, industrial applications of nanomaterials can be found in a wide variety of industries. Nanofillers and nanoreinforcements for polymer composites have in recent years been the subject of intense research and development. Polymer nanocomposites are polymer-matrix composites but contain materials having at least one dimension below about 100nm, wherein the small size offers some level of controllable performance that is different from the expectations developed in the macroworld [39]. Even in the field of nanocomposites, many topics exist including composite reinforcement, barrier properties, flame resistance, bacterial properties, electro-optical properties, and cosmetic applications [40]. Variety of nanomaterials such as nanoparticles, nanoclays, nanotubes, nanofibers, and etc. were used as fillers to enhance the material properties for polymer nanocomposites. The use of epoxy resin (thermosetting polymer) has steadily increased. Due to their low density, good adhesive and mechanical properties, epoxy resins became a promising material for high performance applications. Epoxy-composite materials can be found in various applications on the rise. A recent approach to improve and diversify epoxy resin properties is through the dispersion on nanometer scaled fillers in the epoxy matrix [41].

Some nanofillers such as layered silicates, carbon nanotubes, and graphene can be used to improve the properties of epoxy-composites. In traditional FMLs the epoxy resin is a suitable matrix. Therefore the modifying of epoxy resin by nanofillers is a reasonable method in FMLs.

### A. Graphene

Graphene, an allotrope of carbon in form of a two-dimensional lattice, has attracted attention in recent years due to its exceptional thermal, mechanical, and electrical properties. One of the most promising applications of this material is in polymer nanocomposites. Graphene have been used in polymer matrices as nano-scale filler materials. Graphene Oxide (GO) is produced by the treatment of graphite

using strong mineral acids and oxidizing agents. Production of well-dispersed polymer nanocomposites with GO-derived fillers depends on the exfoliation of GO prior to incorporation into a polymer matrix. Solvent-based exfoliation (Fig. 3), simple peeling process (Fig. 4), ultrasonication, thermal exfoliation technique, chemical vapour deposition (CVD), and etc. have emerged as preferred routes for this step [19], [20].

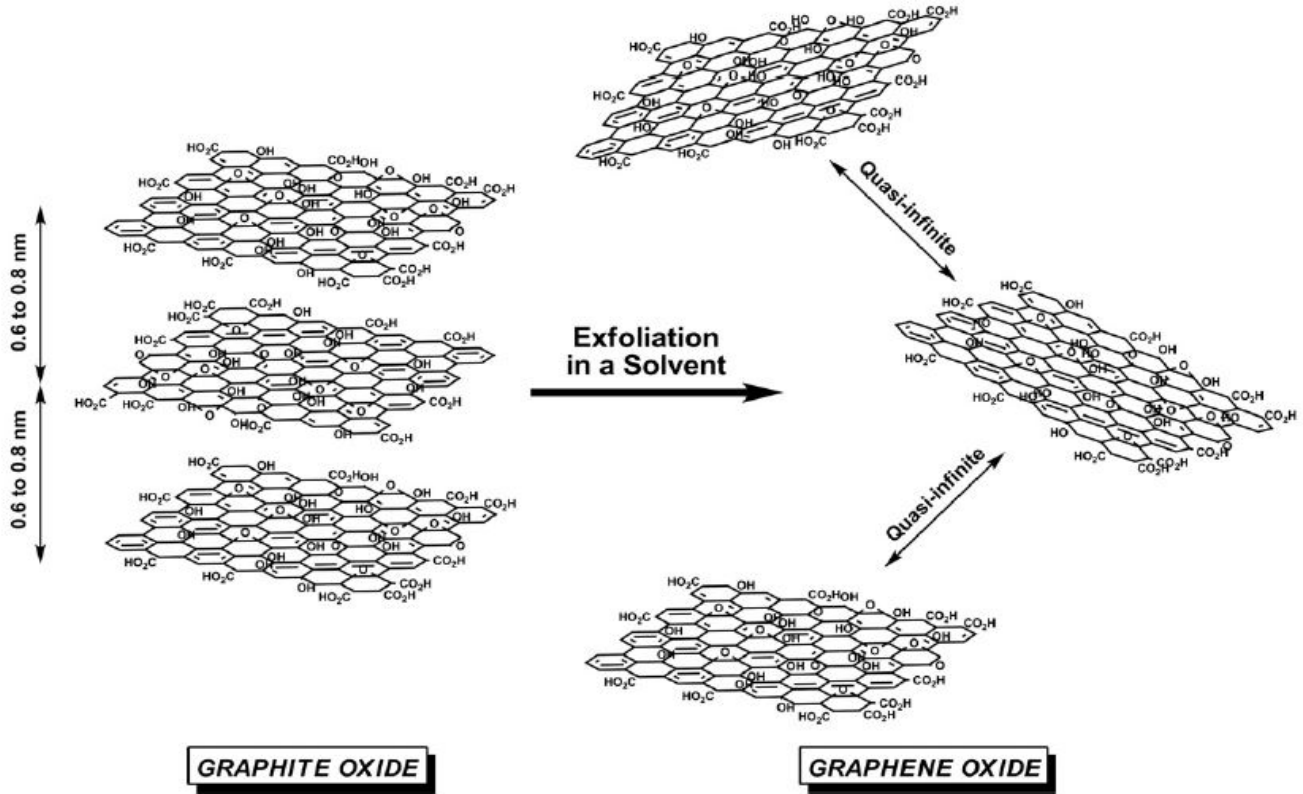


Fig. 3 Schematic illustration the chemical structure of graphite oxide (GO) and the structural difference between layered GO and exfoliated graphene oxide (G-O) platelets [19]



Fig. 4 Scotch tape method of graphene block [20]

Epoxy and its composites are tremendous materials for industrial fields like electrical applications, thermal

applications, high-performance nanocomposites in automobiles and other applications but these composites have some limitations as well. Graphene has shown a new path to overcome these limitations. The studies have shown the dramatically increase in epoxy-graphene nanocomposites research in recent years. A well dispersed state ensures availability of maximum surface area of filler, which will affect the neighborhood polymer chain and, consequently, the properties of the whole nanocomposite. The commonly method to prepare epoxy-graphene nanocomposites is through solvent processing while the mechanical methods is a less effective dispersion strategy (Fig. 5) [20], [24], [42]. Finally, the graphene fibers with increased strength and rich functionalities can be used as proper fillers [25].

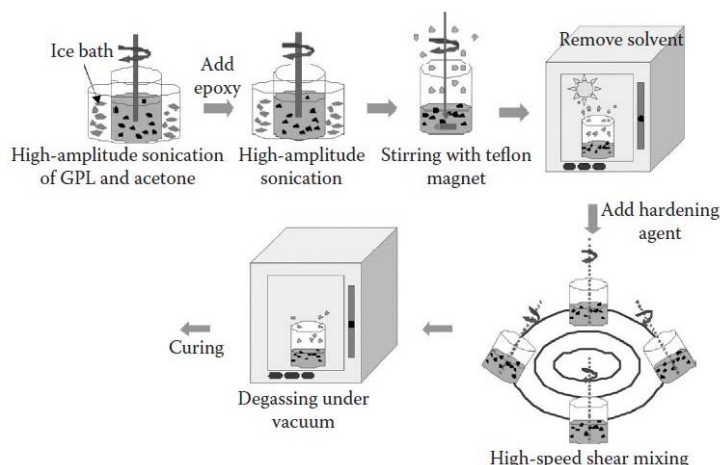


Fig. 5 Illustration of graphene dispersion process [24]

### B. Carbon Nanotubes

Carbon nanotubes, which are allotropes of carbon, are regarded as the ultimate carbon fibers. Their mechanical properties such as stiffness and strength are expected to approach that of an ideal carbon fiber, which has the perfect orientation of defect-free graphene (single layer of graphite) layers along the fiber axis. These graphene layers are also represented to be as thinnest possible layers to form the nanotubes. Carbon nanotube materials are the one of the best example of nanostructures derived by bottom-up chemical synthesis processes. And discovering the multi-walled carbon nanotubes causes an exponential rise in the number of studies on nanocomposites [43]. Fig. 6 illustrated a single-walled carbon nanotube.

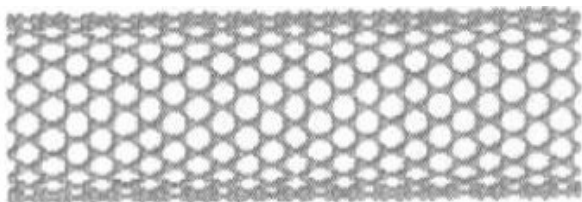


Fig. 6 Schematic presentation of single-walled carbon nanotube [43]

Usually dispersion of carbon nanotubes in the polymer matrix is highly difficult because CNTs agglomerate with each other due to strong Van der Waals force. For functionalization of CNTs there are two methods. These are covalent functionalization and noncovalent functionalization. The covalent functionalization usually realized by oxidizing carbon nanotubes in an acid. And in the noncovalent functionalization polystyrene sulfonate (PSS), poly(4-aminostyrene) (PAS), and etc. were attached on the surface of CNTs [44].

### IV. CONCLUSION

It was found that, the properties modification of polymeric nanocomposites by nanofillers such as graphene and nanotube, is a commonly method. The properties such as fracture toughness, tensile strength, electrical conductivity, wear

resistance, barrier to diffusion, water resistance or flammability resistance can be strongly affected by nanofillers dispersion in polymer matrix. Therefore, the nanofillers can be used in the polymer matrix of FMLs for modifying of the properties.

### REFERENCES

- [1] E.C. Botelho, R.A. Silva, L.C. Pardini, M.C. Rezende, "A review on the development and properties of continuous fiber/epoxy/aluminum hybrid composite for aircraft structures," *Material Research*, vol. 9, pp. 247-256, 2006.
- [2] T. Sinmazcelik, E. Avcu, M.O. Bora, O. Coban, "A review: fiber metal laminates, background, bonding type and applied test method," *Material and Design*, vol. 32, pp. 3671-3685, 2011.
- [3] S.M.R. Khalili, R.K. Mittal, S. Gharibi Kalibar, "A study of the mechanical properties of steel/aluminium/GRP laminates," *Materials Science & Engineering (A)*, vol. 412, pp. 137-140, 2005.
- [4] M. Sadighi, R.C. Alderliesten, R. Benedictus, "Impact resistance of fiber-metal laminates: A review," *International Journal of Impact Engineering*, vol. 49, pp. 77-90, 2012.
- [5] G.R. Rajkumar, M. Kerishna, H.N. Narsimhamurthy, Y.C. Keshavamurthy, J.R. Nataraj, "Investigation of tensile and bending behavior of aluminum based hybrid fiber metal laminates," *Procedia Material Science*, vol. 5, pp. 60-68, 2014.
- [6] M. Yue, X. Zhongchun, X. Xiaofeng, "Fatigue crack growth in fiber metal laminates," *Science China (Physics, Mechanics & Astronomy)*, vol. 57, pp. 83-89, 2014.
- [7] S.W.F. Spronk, I. Sen, R.C. Alderliesten, "Predicting fatigue crack initiation in fiber metal laminates based on metal fatigue test data," *International Journal of Fatigue*, vol. 70, pp. 428-439, 2015.
- [8] Y. Huang, J. Liu, X. Huang, J. Zhang, G. Yue, "Delamination and fatigue crack growth behavior in fiber metal laminates (GLARE) under single overloads," *International Journal of Fatigue*, vol. 78, pp. 53-60, 2015.
- [9] W. Wang, C. Rans, R.C. Alderliesten, R. Benedictus, "Predicting the influence of discretely notched layers on fatigue crack growth in fiber metal laminates," *Engineering Fracture Mechanics*, vol. 145, pp. 1-14, 2015.
- [10] C.D. Rans, R.C. Alderliesten, R. Benedictus, "Predicting the influence of temperature on fatigue crack propagation in fiber metal laminates," *Engineering Fracture Mechanics*, vol. 78, pp. 2193-2201, 2011.
- [11] J. Hausmann, P. Naghipour, K. Schulze, "Analytical and numerical residual stress models for fiber metal laminates-comparison and application," *Procedia Materials Science*, vol. 2, pp. 68-73, 2013.
- [12] V. Daghighi, S.M.R. Khalili, R. Eslami Farsani, "Creep behavior of basalt fiber-metal laminate composites," *Composites Part: B Engineering*, vol. 91, pp. 275-282, 2016.
- [13] K.S. Raghul, D. Nandakumar, R. Jeyakumar, "Mechanical behavior of glass fiber/epoxy modified with nanocomposites: A review," *IJIRSET* vol. 5, pp. 2347-6710, 2016.



- [14] L.F.A. Bernardo, A.P.B.M. Amaro, D.G. Pinto, S.M.R. Lopes, "Modeling and simulation techniques for polymer nanoparticle composites-A review," *Computational Materials Science*, vol. 118, pp. 32-46, 2016.
- [15] M.S.L. Luna, G. Filippone, "Effects of nanoparticles on the morphology of immiscible polymer blends-challenges and opportunities," *European Polymer Journal*, vol. 79, pp. 198-218, 2016.
- [16] P. Li, Y. Zheng, M. Li, T. Shi, D. Li, A. Zhang, "Enhanced toughness and glass transition temperature of epoxy nanocomposites filled with solvent-free liquid-like nanocrystal-functionalized graphene oxide," *Material and Design*, vol. 89, pp. 653-659, 2016.
- [17] W. Osterle, A.I. Dmitriev, B. Wetzel, G. Zhang, I. Hausler, B.C. Jim, "The role of carbon fibers and silica nanoparticles on friction and wear reduction of an advanced polymer matrix composite," *Material and Design*, vol. 93(5), pp. 474-484, 2016.
- [18] M.S. Senthil Kumar, N. Mohana Sundara Raju, P.S. Sampath, L.S. Jayakumari, "Effects of nanomaterials on polymer composites-An expatriate view," *Rev. Adv. Mater. Sci*, vol. 38, pp. 40-54, 2014.
- [19] J.R. Potts, D.R. Dreyer, C.W. Bielawski, R.S. Ruoff, "Graphene-based polymer nanocomposites," *Polymer*, vol. 53, pp. 5-25, 2011.
- [20] J. Wei, T. Vo, F. Inam, "Epoxy/graphene nanocomposites-processing and properties: a review," *RSC Advances*, vol. 55, pp. 73510-73524, 2015.
- [21] T. Ramanathan, A.A. Abdala, S. Stankovich, D.A. Dikin, M.H. Alonso, R.D. Piner, D.H. Adamson, H.C. Schmiepp, X. Chen, R.S. Ruoff, S.T. Nguyen, L.A. Aksay, R.K. Prudhomme, L.C. Brinson, "Functionalized graphene sheets for polymer nanocomposites," *Nature Nanotechnology*, vol. 63, pp. 327-331, 2008.
- [22] T.K. Das, S. Prusty, "Graphene-based polymer composites and their applications," *Polymer-Plastic Technology and Engineering*, vol. 52, pp. 319-331, 2013.
- [23] P. O'Brien, S.H. Krotz, R. Nuzzo, "Polymer-Graphene Nanocomposites," *RSC Nanoscience & Nanotechnology*, vol. 26, 2012.
- [24] P. Mukhopadhyay, R.K. Gupta, "Graphite, Graphene and their polymer nanocomposites," *CRC Press*, vol. 13, 2013.
- [25] Z. Xu, C. Gao, "Graphene fiber: a new trend in carbon fibers," *Materials Today*, 2015.
- [26] M.M. Shokrieh, A. Saeedi, M. Chitsazzadeh, "Evaluating the effects of multi-walled carbon nanotubes on the mechanical properties of chopped strand mat/polyester composites," *Material and Design*, vol. 56, pp. 274-279, 2014.
- [27] K.T. Hsiao, J. Alms, S.G. Advani, "Use of epoxy/multiwalled carbon nanotubes as adhesives to join graphite fibre reinforced polymer composites," *Nanotechnology*, vol. 14, pp. 791-793, 2003.
- [28] K.I. tserpes, N. Silvestre, "Modeling of carbon nanotubes, graphene and their composites," *Springer Series in Material Science*, vol. 188, 2014.
- [29] M.R. Gude, S.G. Prolongo, A. Urena, "Toughening effect of carbon nanotubes and carbon nanofillers in epoxy adhesives for joining carbon fibre laminates," *Intrnational Journal of Adhesion & Adhesives*, vol. 62, pp. 139-145, 2015.
- [30] M. Kulkarni, D. Carnahan, K. Kulkarni, D. Qian, J.L. Abot, "Elastic response of a carbon nanotube fiber reinforced polymeric composite: A numerical and experimental study," *Composite: Part B*, vol. 41, pp. 414-421, 2010.
- [31] K.T. Lau, C. Gu, D. Hui, "A critical review on nanotube and nanotube/nanoclay related polymer composite material," *CompositesPart B: Engineering*, vol. 37, pp. 425-436, 2006.
- [32] M.M. Shokrieh, R. Rafiee, "On the tensile behavior of an embedded carbon nanotube in polymer matrix with non-bonded interphase region," *Composite Structures*, vol. 92, pp. 647-652, 2010.
- [33] H. Ning, Y. Li, N. Hu, "Improvement of interlaminar mechanical properties of CARALL based on nanofiller interface reinforcement and other fabrication techniques," *13<sup>th</sup> International conference on Fracture*, pp. 16-21, 2013.
- [34] E. Kaboodvand, R. Eslami Farsani, H. Khosravi, "Effect of multi-walled carbon nanotube on tensile strength of fiber metal laminates," In *Persian, 1th International conference on Mechanical and Aerospace Engineering*, 2016.
- [35] Y.B. Hu, H.G. Li, L. Cai, J.P. Zhu, L. Pan J. Xu, J. Tao, "Preparation and properties of fiber-metal laminates based on carbon fibre reinforced PMR polyimide," *Composites Part B: Engineering*, vol. 69, pp. 587-591, 2015.
- [36] J. Iriondo, L. Aretxabaleta, A. Aizpura, "Characterisation of the elastic and damping properties of traditional FML and FML based on a self-reinforced polypropylene," *Composite Structures*, vol. 131, pp. 47-54, 2015.
- [37] M. Supova, G.S. Martynkova, K. Barabaszova, "Effect of nanofillers dispersion in polymer matrices: A review," *Science of Advance Materials*, vol. 3, pp. 1-25, 2011.
- [38] U.A. Khashaba, A.A. Aljinaidi, M.A. Hamed, "Nanofillers modification of epocast 50-AI/946 epoxy for bonded joints," *Chinese Journal of Aeronautics*, vol. 27 (5), pp. 1288-1300, 2014.
- [39] R.K. Gupta, E. Kennel, K.J. Kim, "Polymer nanocomposite handbook," *CRC Press*, 2010.
- [40] D.R. Paul, L.M. Robeson, "Polymer nanotechnology: Nanocomposites," *Polymer*, vol. 49, pp. 3187-3204, 2008.
- [41] Y.W. Mai, Z.Z. Yu, "Polymer nanocomposites," *Woodhead Publishing in Materials*, 2006.
- [42] M. Barletta, S. Vesco, M. Puopolo, V. Tagliaferri, "Graphene reinforced UV-curable epoxy resins: Design, manufacture and matrial performance," *Progress in Organic Coatings*, vol. 90, pp. 414-424, 2016.
- [43] V. Mittal, "Polymer nanotube nanocomposites," *John Wile*, 2010.
- [44] J. Cha, S. Jin, J.H. Shim, C.S. Park, H.J. Ryu, S.H. Hong, "Functionalization of carbon nanotubes for fabrication of CNT/epoxy nanocomposites," *Material and Design*, vol. 95, pp. 1-8, 2016.