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FLEXURAL PROPERTIES OF ACTIVATED CARBON FILLED EPOXY NANOCOMPOSITES

(Sifat Lenturan Nanokomposit Epoksi Berpengisi Karbon Teraktif)

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Abstract

Activated carbon (AC) filled epoxy nanocomposites obtained by mixing the desired amount of nano AC viz., bamboo stem, oil palm empty fruit bunch, and coconut shell from agricultural biomass with the epoxy resin. Flexural properties of activated carbons filled epoxy nanocomposites with 1%, and 5% filler loading were measured. In terms of flexural strength and modulus, a significant increment was observed with addition of 1% vol and 5% vol nano-activated carbon as compared to neat epoxy. The effect of activated carbon treated by two chemical agents (potassium hydroxide and phosphoric acid) on the flexural properties of epoxy nanocomposites were also investigated. Flexural strength of activated carbon-bamboo stem, activated carbon-oil palm, and activated carbon-coconut shell reinforced epoxy nanocomposites showed almost same value in case of 5% potassium hydroxide activated carbón. Flexural strength of potassium hydroxide activated carbon-based epoxy nanocomposites was higher than phosphoric acid activated carbon. The flexural toughness of both the potassium hydroxide and phosphoric acid activated carbon reinforced composites range between 0.79 – 0.92 J. It attributed that developed activated carbon filled epoxy nanocomposites can be used in different applications.

Keywords: epoxy matrix; activated carbon; nanocomposites; flexural properties

Abstrak

Nanokomposit epoksi berpengisi karbon teraktif diperoleh dengan mencampurkan sejumlah nano karbon teraktif iaitu buluh, kelapa sawit tandan buah kosong, dan tempurung kelapa yang dikehendaki daripada biomas pertanian dengan resin epoksi. Sifat lenturan nanokomposit epoksi berpengisi karbon teraktif dengan 1% dan 5% muatan pengisi telah diukur. Dari segi kekuatan lenturan dan modulus, peningkatan ketara diperhatikan dengan jumlah penambahan 1% dan 5% nano-karbon teraktif berbanding epoksi tanpa pengisi. Kesan karbon teraktif menggunakan dua bahan kimia (kalium hidroksida dan asid fosforik) terhadap sifat lenturan terawat juga dikaji. Kekuatan lenturan karbon teraktif-batang buluh, karbon teraktif-kelapa sawit, dan karbon teraktif-tempurung kelapa diperkukuh nanokomposit epoksi menunjukkan nilai yang hampir sama seperti 5% kalium hidroksida karbon teraktif. Kekuatan lenturan kalium hidroksida karbon teraktif berasaskan nanokomposit epoksi adalah lebih tinggi daripada asid fosforik karbon teraktif. Ketahanan lenturan bagi kedua-dua kalium hidroksida dan asid fosforik karbon teraktif diperkukuh komposit epoksi adalah di antara 0.79 – 0.92 J. Ia menyifatkan bahawa pembangunan nanokomposit epoksi berpengisi karbon teraktif boleh digunakan dalam aplikasi yang berbeza.

Kata kunci: matrik epoksi; karbon teraktif; nanokomposit; sifat lenturan

Introduction

Epoxy resin possess long service time, good physical properties, favorable cost–performance ratio in comparison to other thermosets. Epoxy resins have wide range of applications in electronics, transportation in composite structural and furnishing elements. In Malaysia, huge amount of agricultural biomass available which can be converted to useful, value-added products to solve agricultural waste disposal problems. Agricultural biomass such as oil palm empty fruit bunch fibres, bamboo stem and coconut shells are available in Malaysia. However, there are limited studies reported on the utilization of oil palm empty fruit bunch (EFB) fibres, bamboo stem and coconut shells, especially on conversion of EFB into activated carbon (AC)[1-2]. ACs with high specific surface area and pore volumes can be prepared from a variety of carbonaceous materials such as coconut shell [3-4], agricultural wastes [5-7], or industrial wastes [8-9].

AC can be introduced into polymeric materials such as epoxy to modify their properties and to develop composites materials with improved thermal and mechanical properties. AC filled polymer composites are lightweight materials, readily availability in different structural forms and are known to enhance the thermal, mechanical and electrical properties and the ease of fabrication of these composite through compression molding. A combination of epoxy matrix with various types of nano-filler reinforcement materials lead to the development of epoxy nanocomposites which can be used in wide range of potential applications. Several researchers reported on preparation and characterization of nano-filler based epoxy composites [10-12].

This research is extension of previous work deals with preparation and characterization of activated carbon filled epoxy nanocomposites by using activated carbon treated with two chemical reagents, phosphoric acid (H_3PO_4) and potassium hydroxide (KOH) [13-16]. The focus of this study to develop high performance composites materials from biomass (agricultural wastes) materials with aimed to reduce the overall cost of the material preparation by using of inexpensive inorganic fillers. Present study reported flexural properties of chemical reagents (H_3PO_4 or KOH)-treated ACs filled epoxy nanocomposites and compared measured flexural strength, modulus and toughness of chemical reagents (H_3PO_4 or KOH)-treated AC epoxy nanocomposites.

Materials and Methods

Materials

Waste biomass oil palm EFB fibre obtained from Ecofuture Berhad, Malaysia, Bamboo stem supplied by Forest Research Institute Malaysia (FRIM), and coconut shell procured from Anjung Juara Sdn, Bhd, used without further treatments. Potassium hydroxides (KOH) (~85% alkali) and phosphoric acid (H_3PO_4) obtained from R&M Chemical Essex, UK. All chemicals used in this investigation were of analytical grade. Morphological, chemical, and thermal properties of raw materials (oil palm EFB, bamboo stem, and coconut shells) reported in our previous work [15].

Preparation of epoxy nanocomposites

Epoxy nanocomposites filled with activated carbon derived from oil palm empty fibre bunch (AC-EFB), bamboo stem (AC-BS), and coconut shells (AC-CNS) were obtained by mixing the required amount of AC with epoxy resin as reported in our previous work [13].

Characterization

The flexural properties were measured using a three-point bending test method on Instron Universal Testing Machine (Model 5582). The tests were carried out at room temperature at a crosshead speed of 1.15 mm/min as per ASTM D790-02. Rectangular shaped composite samples of 120 mm × 20 mm × 3 mm were tested using a span of 40 mm. An average of at least five tests per sample was performed to report flexural strength and flexural modulus.

Results and Discussion

Abbreviations of Activated Carbons

The chemically activated carbon samples obtained from EFB, BS, and CNS were named AC-EFB, AC-BS, and AC-CNS, respectively. Carbon samples activated by phosphoric acid (H) were named AC-EFB H, AC-BS H, and AC-CNS H and those activated by potassium hydroxide (K) were named AC-EFB K, AC-BS K, and AC-CNS K, respectively.

Flexural Properties

Flexural properties normally will be enhanced with incorporation of fillers, this is in agreement with many researchers [17-20]. Fig. 1 showed flexural strength and modulus of the cured epoxy (0% AC) and nano-activated carbon based epoxy composites with different volume percentage (1, 5, 10% vol) of AC-CNS. In terms of flexural strength and modulus, a significant increment was observed with addition of 1% vol and 5% vol nano-activated carbon as compared to neat epoxy. The value of flexural strength of neat epoxy, 1% vol AC-CNS, and 5% vol AC-CNS nano-epoxy composites were 113 MPa, 123 MPa, and 133 MPa, respectively (Figure 1). The increase in flexural strength of epoxy nanocomposites could be attributed to greater physical interaction in the activated carbon filled epoxy composites. Smaller aggregate size increased the activated carbon surface area interacting with epoxy matrix, which resulted in enhanced flexural properties.

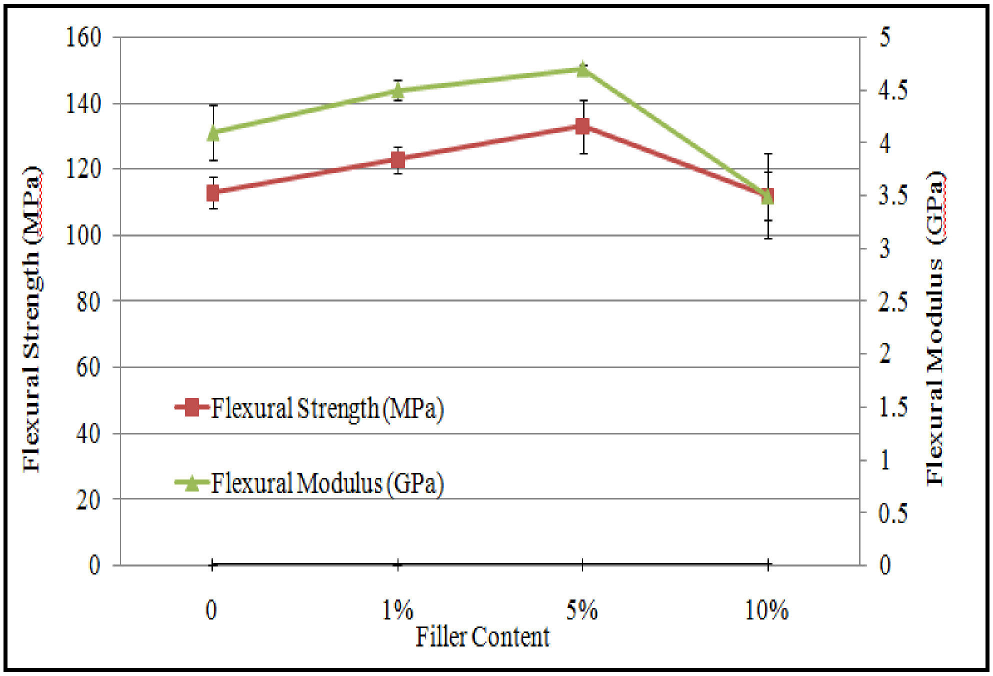

Figure 1. Flexural strength and modulus of AC-CNS K epoxy nanocomposites at 0%, 1%, 5% and 10% loading.

The flexural strength and modulus of nano AC-CNS epoxy composite decreased with increase in AC-CNS content. Earlier reports have shown that at higher filler loading, filler agglomeration and formation of micro void might cause decrease in stress transfer between the matrix and the reinforcing filler [21]. Also, a decrease in the effective cross-section area due to agglomeration of several nano-activated carbon have been reported by other workers [22]. The flexural modulus of the AC-CNS epoxy nanocomposites increased until 5% addition of nano AC-CNS and after that it started to decrease. These results suggest that optimum flexural modulus can be achieved with 5% filler loading above which a sharp deceased was recorded. It was observed that the flexural modulus decreased at 10% AC-CNS which was slightly higher than neat epoxy matrix. Similar study was reported on polyhedral oligomer silsesquioxanes (POSS)/ epoxy composites, it showed decreasing trend of flexural strength and modulus at higher volume of reinforcing materials [23]. The flexural strength of microparticle filled composites reduced with increasing filler content [24], especially in the case where load transfer between matrix and filler was insufficient and interface was weak. The adhesion between resin and nano AC-CNS become weaker as the filler content increases. Consequently, the flexural property of the composite decreased as the nano AC-CNS content increases. Small-size nano AC-CNS possessed more surface area than longer filler particles [25].

It is obvious from above that 1% and 5% AC-CNS reinforced epoxy nanocomposites gave improved and better flexural strength and modulus. These results suggest that optimum flexural strength and modulus were obtained with 5% AC-CNS reinforced epoxy nanocomposites. On this basis, further study mainly concentrates on 1% and 5% AC-BS, AC-EFB, and AC-CNS for both activating agents. Fig. 2 showed flexural strength of AC-BS, AC-EFB, and AC-CNS based epoxy nanocomposites at 1%, and 5% AC by using H_3PO_4 (H), and KOH (K) activating agents. Flexural strength of AC-BS, AC-EFB, and AC-CNS epoxy nanocomposites showed almost same value in case of 5% KOH activated carbon while flexural strength of KOH activated carbon based epoxy nanocomposites was higher than H_3PO_4 activated carbon. It may be due to better quality of interface in KOH activated carbon than in H_3PO_4 activated carbon. It is clear from previous work that that activated carbon obtained by using KOH has the high surface area and uniform surface structure as compared to H_3PO_4 ([16]. Due to that AC obtained by using KOH show improved flexural strength. The adhesion strength and the interfacial stiffness usually play a very important role in the materials capability to transfer stresses and elastic deformation from matrix to fillers [26]. Another important aspect was the significant role of dispersion of nano-particle within matrix. 5% nano-activated carbón-CNS filled epoxy composite gave good dispersion of nano-particle as compared to 10% filler loading [14]. Allaoui et al. [27] advised that it not be helpful to use high filler concentrations to improve the mechanical properties of composites if they are in random distribution and homogeneity of the composite is a critical point for mechanical behavior of the nanocomposites.

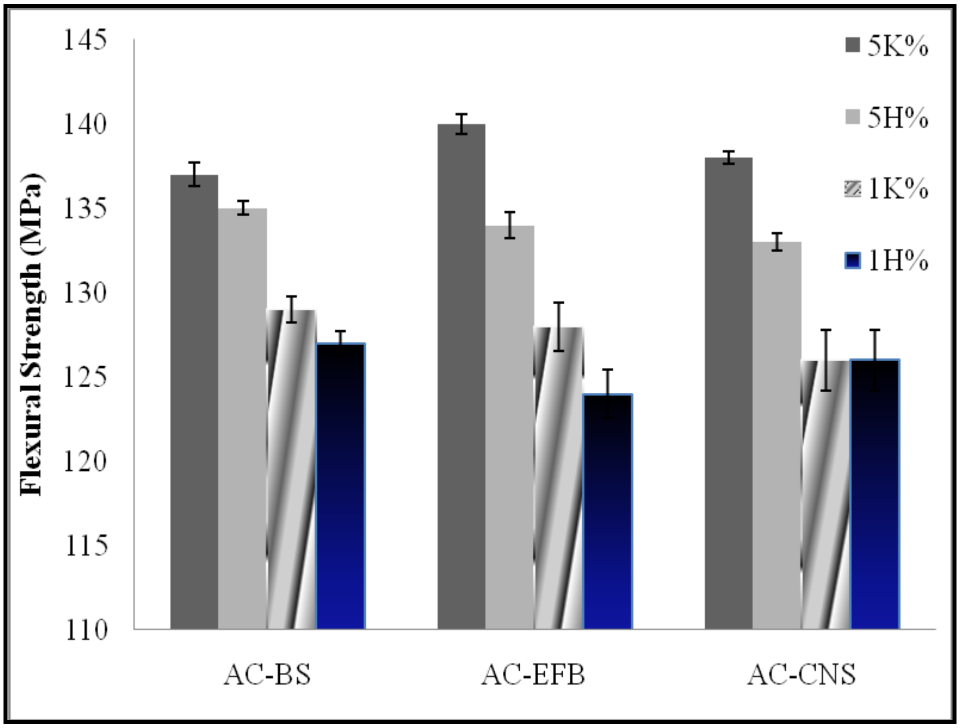

Figure 2. Flexural strength of AC-BS, AC-EFB, and AC-CNS based epoxy nanocomposites at 1% and 5% by using H_3PO_4 (H) and KOH (K) activating agent.

Flexural strength increased in epoxy composites reinforced by AC K possibly due to the presence of the activated carbons at the higher surface area, which may improve the interfacial properties between the matrix and the activated carbon [15-16]. It may also possible to conclude that the compressive strength of the epoxy matrix is improved by the presence of the nano-activated carbon and which ultimately increases the bending strength of the corresponding epoxy nanocomposites. At higher filler loading (10%) in epoxy matrix which may causes agglomeration in the epoxy matrix and propagating crack could encounter a stress concentration locally and then easily causes initiation of failure [28]. Table 1 represents the flexural toughness of various 5% AC filled epoxy composite. The flexural toughness of both the H_3PO_4 and KOH activated carbon reinforced composites range between 0.79–0.92 J. These values are higher than the value obtained for un-reinforced epoxy composite which recorded the value of 0.78 J. Comparison of AC K samples indicate that AC-CNS K gave the highest value of 0.92 J whereas AC-EFB K gives the lowest toughness value of 0.83 J. Similarly for AC H samples, AC-CNS H shows the highest value of 0.84 and AC-EFB H gives 0.79 as the lowest value.

Table 1. Comparison of Fracture Toughness (J) of 5% AC filled epoxy composite.

Composite	Fracture Toughness (J)	STD
Neat Epoxy	0.78	0.04
5%AC-CNS K	0.92	0.06
5%AC-BS K	0.88	0.01
5% AC-EFB K	0.83	0.03
5%AC- CNS H	0.84	0.03
5%AC-BS H	0.82	0.01
5% AC-EFB H	0.79	0.05

The gradient of variation of fracture toughness is not high and the difference between the minimum and maximum values is only 0.5 J as against 0.9 J in case of AC K. Overall, AC-EFB H gives the lowest fracture toughness value while AC-CNS K recorded the highest value of 0.92 J. The value of 0.79 J obtained for AC-EFB H is slightly higher than neat epoxy matrix (0.78 J). The higher values obtained by the addition of 5% of filler suggests increased flexural strength of the composite. The fracture toughness results follow the same trend as quoted by Kim et al. who used carbon black filled epoxy composite[29].

Conclusion

Epoxy nanocomposites developed by reinforcement of AC-EFB, AC-BS, and AC-CNS by hand lay-up technique. Obtained results indicated that 5% AC-CNS display higher flexural strength and modulus compared to 1% and 10% nano filler loading. Flexural strength of AC-BS, AC-EFB, and AC-CNS epoxy nanocomposites showed almost same value in case of 5% KOH activated carbon while flexural strength of KOH activated carbon based epoxy nanocomposites was higher than H_3PO_4 activated carbon. Flexural toughness of both the H_3PO_4 and KOH activated carbon reinforced composites range between 0.79–0.92 J and obtained values are higher than neat epoxy matrix (0.78 J). Overall, AC-EFB by H_3PO_4 gives the lowest fracture toughness value while AC-CNS by KOH recorded the highest value of 0.92 J.

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References

1. Rahman, S. H. A., Choudhury, J. P., Ahmad, A. L. & kamaruddin, A. H. (2007). Optimization studies on acid hydrolysis of oil palm empty fruit bunch fiber for production of xylose. *Bioresource Technology*, 98(3): 554-559.
2. Umikalsom, M. S., Ariff, A. B., Zulkifli, H. S., Tong, C. C., Hassan, M. A. & Karim, M. I. A. (1997). The treatment of oil palm empty fruit bunch fibre for subsequent use as substrate for cellulase production by Chaetomium globosum Kunze. *Bioresource Technology*, 62(1-2): 1-9.
3. Qiao, W., Korai, Y., Mochida, I., Hori, Y. & Maeda, T. (2002). Preparation of an activated carbon artifact: oxidative modification of coconut shell-based carbon to improve the strength. *Carbon*, 40(3): 351-358.
4. Sekar, M., Sakthi, V. & Rengaraj, S. (2004). Kinetics and equilibrium adsorption study of lead(II) onto activated carbon prepared from coconut shell. *Journal Colloid Interface Science*, 279(2): 307-313.
5. Youssef, A. M., Radwan, N. R. E., Abdel-Gawad, I. & Singer, G. A. A. (2005). Textural properties of activated carbons from apricot stones. *Colloid Surface A: Physicochem Engineering Aspects*, 252(2-3)**:** 143-151.

6. Zhang, T., Walawender, W. P., Fan, I. T., Fan, M., Daugaard, D. & Brown, R. C. (2004). Preparation of activated carbon from forest and agricultural residues through CO2 activation. *Chemical Engineering Journal,* 105(1-2): 53-59.
7. Guo, J., Xu, W. S., Chen, Y. L. & Lua, A. C. (2004). Adsorption of NH3 onto activated carbon prepared from palm shells impregnated with H2SO4. *J ournal Colloid Interface Science,* 281(2): 285-290.
8. Ko, D. C. K., Mui, E. L. K., Lau, K. S. T. & Mckay, G. (2004). Production of activated carbons from waste tire-process design and economical analysis. *Waste Management,* 24(9): 875-888.
9. Hayashi, J., Yamamoto, N., Horikawa, T., Muroyama, K. & Gomes, V. G. (2005). Preparation and characterization of high specific surface area activated carbons from K2CO3 treated waste polyurethane. *J. Colloid Interface Sci,* 281(2): 437-443.
10. Mirmohseni, A. & Zavareh, S. (2010). Preparation and characterization of an epoxy nanocomposite toughened by a combination of thermoplastic, layered and particulate nano-fillers. *Materials & Design,* 31(6): 2699-2706.
11. Bakar, M., Kostrzewa, M., Hausnerova, B. & Sar, K. (2010). Preparation and property evaluation of nanocomposites based on polyurethane-modified epoxy/montmorillonite systems. *Advances in Polymer Technology,* 29(4): 237-248.
12. Palencia, C., Mazo, M. A., Nistal, A., Rubio, F., Rubio, J. & Oteo, J. L. (2011). Processing and properties of carbon nanofibers reinforced epoxy powder composites. *Journal of Nanoparticle Research*, 13(11), 6021-6034.
13. Abdul Khalil, H.P..S., Firoozian, P., Jawaid, M., Akil, H.M., Hassan, A. (2013a). Preparation of activated carbon filled epoxy nanocomposites: Morphological and thermal properties. *Journal of Thermal Analysis and Calorimetry*, 113(2): 623-631.
14. Abdul Khalil, H.P.S., Jawaid, M., Firoozian, P., Zainudin, E.S., Paridah, M.T. (2013b). Dynamic Mechanical Properties of Activated Carbon–Filled Epoxy Nanocomposites. *International Journal of Polymer Analysis and Characterization*, 18(4): 247-256.
15. Abdul Khalil, H.P..S., Firoozian, P., Bakare, I.O., Akil, H.M., Noor, A. M. (2010). Exploring biomass based carbon black as filler in epoxy composites: flexural and thermal properties. *Material & Design*, 31(7): 3419-3425.
16. Firoozian, P., Bhat, I.U.H., Abdul Kahlil, H.P.S., Noor, A.M., Akil, H.M., Bhat, A.H. (2011). High surface area activated carbon prepared from agricultural biomass: empty fruit bunch (EFB), bamboo stem and coconut shells by chemical activation with H3PO4. *Material Technology*, 26: 222-228.
17. Teh, P. L., Mariatti, M., Akil, H. M., Seetharamu, K. N., Wagiman, A. N. R. & Beh, K. S. (2008). Effect of Ethanol as Diluent on the Properties of Mineral Silica Filled Epoxy Composites. *Journal of Composite Materials*, 42(2): 129-142.
18. Selvin, T. P., Kuruvilla, J. & Sabu, T. (2004). Mechanical properties of titanium dioxide-filled polystyrene microcomposites. *Materials Letters,* 58(3-4): 281-289.
19. Debnath, S., Ranade, R., Wunder, S. L., Mccool, J., Boberick, K. & Baran, G. (2004). Interface effects on mechanical properties of particle-reinforced composites. *Dental Materials*, 20(7): 677-686.
20. Teh, P. L., Mariatti, M., Akil, H. M., Yeoh, C. K., Seetharamu, K. N., Wagiman, A. N. R. & Beh, K. S. 2007. The properties of epoxy resin coated silica fillers composites. *Materials Letters,* 61(11-12): 2156-2158.
21. Moodley, V. K. (2007). *The Synthesis, Structure And Properties Of Polypropylene Nanocomposites.* MSc., Durban university of technology.
22. Rothon, R. N. (2003). Particulate-Filled Polymer Composites. *In:* ROTHON, R. N. (ed.) *R.N. Rothon.* Second Edition ed. Shawbury, Shrewsbury, Shropshire, SY4 4NR, United Kingdom: Rapra Technology Limited.
23. Rashid, E. S. A., Ariffin, K.., Kooi, C. C. & Akil, H. M. (2009). Preparation and properties of POSS/epoxy composites for electronic packaging applications. *Materials & Design,* 30(1): 1-8.
24. Lee, J. H., Jang, Y. K., Hong, C. E., Kim, N. H., Li, P. & Lee, H. K. (2009). Effect of carbon fillers on properties of polymer composite bipolar plates of fuel cells. *Journal of Power Sources,* 193(2): 523-529.
25. Kuan, H.-C., Ma, C.-C. M., Chen, K. H. & Chen, S.-M. (2004). Preparation, electrical, mechanical and thermal properties of composite bipolar plate for a fuel cell. *Journal of Power Sources,* 134(1): 7-17.
26. Zhang, M., Zeng, H., Zhang, L., Lin, G. & Li, R. K. Y. (1993). Fracture characteristics of discontinuous carbon fibre-reinforced PPS and PES-C composites. *Polymers and Polymer Composites,* 1: 357-365.
27. Allaoui, A., Bai, S., Cheng, H. M. & Bai, J. B. (2002). Mechanical and electrical properties of a MWNT/epoxy composite. *Composites Science and Technology,* 62(15): 1993-1998.

28. Gojny, F. H. & Schulte, K. (2004). Functionalisation effect on the thermo-mechanical behaviour of multi-wall carbon nanotube/epoxy-composites. *Composites Science and Technology*, 64**:** 2303-2308.
29. Kim, B. C., Park, S. W. & Lee, D. G. (2008). Fracture toughness of the nano-particle reinforced epoxy composite. *Composite Structures*, 86(1-3): 69-77.