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# Ball milled short carbon fiber for reinforced epoxy composites

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## ABSTRACT

Epoxy-Carbon fiber composites are fabricated. The carbon Fibers are first cut using a mechanical chopper. The short carbon Fibers are also subjected to ball milling. Composites are made of ball milled and non-ball milled Fibers. Composites with varying carbon fiber content (1 wt % - 2 wt %) were made. An increase in the mechanical properties is registered with increasing loading of carbon Fibers in the epoxy matrix. Mechanical properties are measured by 3 point bending test for and shore scleroscope. The electrical properties of the composites are also studied. An increase is seen in the electrical properties as well with increase in fiber loading due to the higher conductivity of the carbon Fibers. Ball milled carbon fiber composites show better properties than the normal fiber composites. This is attributed to the better dispersion of ball milled carbon Fibers in the epoxy matrix and preventing agglomeration of the Fibers. The fracture surface is also analyzed by scanning electron micrography to ascertain the fracture mechanism. 

## INTRODUCTION

Composite materials are being used increasingly as replacement for traditional materials owing to superior properties and the ability for the properties of these materials to be tailored to meet specific requirements. Composite materials look to bring together the best properties of both the filler and matrix in a synergistic manner to produce high-performance materials for advanced applications[1-7].

Polymer composites are finding applications in a wide range of fields like electronics[8,9], energy applications[10,11], bio-based applications[12,13] and sensory devices[14-17].

Epoxy is regarded as a highly useful material in engineering, finding uses in adhesives[18], electronic packaging material[19,20]. It has inherent properties such as good strength and modulus, corrosion resistance, and high thermal stability which make it an attractive material to be used as matrix material, mostly with carbon based materials (Carbon nanotubes and Carbon Fibers)[21,22] and some other materials such as silicates or clays[23,24].

Epoxy is prone to crack initiation which imparts it a brittle nature[25,26]. In the current study we have used short (non-continuous) carbon Fibers (SCF) for reinforcement of Epoxy matrix. Carbon Fibers (CF) show attractive properties such as high specific strength and stiffness, lower density compared to other filler materials, higher thermal stability and high electrical conductivity, finding uses in automotive, aerospace, marine and sports materials[27-29]. In the Epoxy/carbon fiber composite carbon Fibers provide strength to the composite improving the mechanical properties of the composite. Though in theory outstanding mechanical properties can be achieved with the addition of carbon Fibers to the epoxy composites, practically there are some challenges to overcome. The mechanism for improving the mechanical properties of epoxy is the transfer of load to the carbon Fiber. To exploit this property two challenges have to be overcome, (i) good interfacial bonding between matrix and reinforcement material and (ii) even distribution of reinforcement material through the matrix. In this work we have focussed on the even distribution of carbon fiber in the epoxy matrix, this is done to achieve to distribute the load over a larger area, resulting in the composite being able to withstand higher loads before mechanical failure.

In the current study we have first cut the carbon fiber in to smaller fragments using a mechanical chopper and then used high energy ball milling to disentangle the Fiber, break down aggregates and separate the fiber into single strands as far as possible. Ball milling has been heavily used in homogenizing reinforcement materials in metal matrix composites[30-32]. These processes were done to achieve high dispersion levels of carbon fiber in the epoxy matrix with a view to maximize the mechanical and electrical properties of epoxy/carbon fiber composites.

## EXPERIMENTAL

### Materials and methods

Unidirectional carbon fibers T300 ( Tensile strength 3400 MPa, modulus 230 GPa, diameter 5-7µm, and density 1.76 g/cm$^3$) were used as reinforcing material. Epoxy resin LY556 was chosen as matrix material and Hy5200 was used as curing agent. Carbon fiber was obtained from Toray Industries inc. (Tokyo, Japan). Epoxy and curing agent (hardner) were obtained from Huntsman Corporation,

### Fabrication of epoxy/carbon fiber composite

Uni-directional carbon fibers were chopped into 1mm short fibers by mechanical chopper. Chopped fibers were milled in centrifugal ball mill (Retsch PM-100) for 30 minutes in the presence of ethanol, ball to fiber ratio was kept at 5:1, zirconia balls were used. Desired amount of Liquid epoxy(LY-556) resin was drawn into the beaker. Chopped carbon fibers were added with epoxy resin and initially dispersed through sonication in water bath at 50$^o$C for 1 hr. Thereafter resin was stirred on magnetic stirrer for 24 hours at 60$^o$C. Premixed fibers were again dispersed using sonicator. Curing agent (HY5200) was added to blended resin in 23:100 ratio. Curing agent was mixed for 30 mins at room temperature. Resin was then poured in open die having 10 cavities having dimensions 70 X 12.7 X 3.2 mm$^3$. The resin was cured first at 100$^o$C for two hours and then at 150$^o$C for three hours in an air circulating oven. Die was coated with silicone wax polish to prevent adhesion of cured resin with metal before pouring the resin. Five specimens were prepared for each composition. Composites containing ball milled SCF (1wt% and 2wt %) and non ball milled (1wt% and 2wt %) with Epoxy matrix were prepared. Mechanical and electrical properties of the composites were evaluated and compared.

### Shore hardness

Shore hardness of composites was computed on shore scleroscope rebound hardness device. It measures hardness associated to material elasticity. In this device hardness is calculated from the rebound height from impact of diamond tipped small conical hammer inside a glass tube. Higher the hardness higher is the rebound height. Measurement is done by ASTM D2240 durometers.

### Flexural properties

Flexural strength and modulus were measured in three point bending method and were loaded to failure on Instron model No. 440. Final extension and stress values were noted. Flexural strength, modulus was calculated by:

$\sigma_f = 3pl/2bt^2$

$E_f = l^3p/4bdt^3$

where b and t are width and thickness of the specimen, while l is span length. d and p are maximum deflection and maximum load encountered until failure. $\sigma_f$, $e_f$ are flexural strength and modulus. Span length to thickness was 13:1 according to ASTM (D-790) standards and cross head speed was 0.5mm/min.

### Electrical conductivity

The electrical conductivity of the composite specimens was measured by 4-point contact method. Kiethley 224 programmable current source was used for providing current. The voltage drop was measured

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by Keithley 197 A auto ranging micro volt DMM. Current was passed along the length of the specimens and the voltage drop was measured across different points separated by unit length. Each sample reading was averaged over 10 to 15 readings.

$$\sigma_c = \frac{L}{RA}$$

Where "*L*" is the length of the composites "*R*" is resistance and "*A*" is cross-sectional area of the composite normal to direction of current flow

### Microstructure analysis

Specimens' fractured surface was examined by Scanning electron microscope (SEM, model LEO 440) for microstructure studies.

## RESULTS AND DISCUSSION

### Morphological and microstructure analysis

Figure 1 shows the neat Epoxy and Epoxy/CF composites specimens. It is apparent from Figure 1 that there is change in colour of epoxy after addition of CF; the level of dispersion is also evident as the black colour is equally distributed in the specimen.


**Figure 1 : shows the samples of Neat Epoxy and Epoxy-SCF composite**

Figure 2(a) shows the scanning electron micrograph of neat epoxy composite. Figure 2(b) represents Epoxy/SCF (1wt %) composite material, and Figure 2 (c) shows the microstructure of Epoxy/SCF. The SCF can be seen differentially in the epoxy matrix, Figure 2(b) shows a much lower concentration of SCF as compared to Figure 2(c) as there is a higher concentration of SCF in the later. The SCF are distributed equally throughout the Epoxy matrix indicating the high level of dispersion of the ball milled SCF in the epoxy matrix. This confirms that ball milling of the SCF and sonication of SCF in the epoxy matrix is effective in aiding dispersion of carbon fibers in the epoxy matrix. Figure 2(d) takes a closer look at the surface of the epoxy matrix displaying the SCF impregnated in the epoxy matrix. The diameter of the fiber as seen in the micrograph is 5-7μm. Ball milling was done to ensure that the SCF would not form any aggregates and that the fibers would be well separated so that they do not agglomerate inside the matrix, the figures show that SCF Fibers have separated into single fibers and dispersed well.

### Mechanical properties

5 samples were prepared for mechanical testing neat epoxy (NE), Epoxy-SCF (1wt %) (E-SCF1), Epoxy-SCF (1wt %) ball milled (E-SCF1 BM), Epoxy-SCF (2wt %), Epoxy-SCF (2wt %) ball milled (E-SCF2 BM).

### Hardness

The hardness of the samples was calculated using Shore A hardness scale shown in Figure 3(a). Shore scleroscope measures the degree of elasticity of a material, the higher the value the lower the degree of elasticity and more rigid the material. Neat Epoxy has the highest level of elasticity and the lowest value of 80. A gradual increase is seen in the composites with the addition of carbon fiber. The highest value was that of E-SCF2 BM, which is 89 an 11% increase in the hardness on addition of 2% fiber. This was expected as carbon fibers have higher strength and increase the rigidity of the material and make the material harder. The hardness of the material was tested on 3 spots on the sample; near the centre, and 2cm from the centre on each side. There was minimal fluctuation in the hardness of the sample at all 3 points, indicating that the carbon fibre is equally distributed throughout the epoxy matrix.

### Flexural strength and modulus

Figure 3(b) and (c) show the Flexural strength and

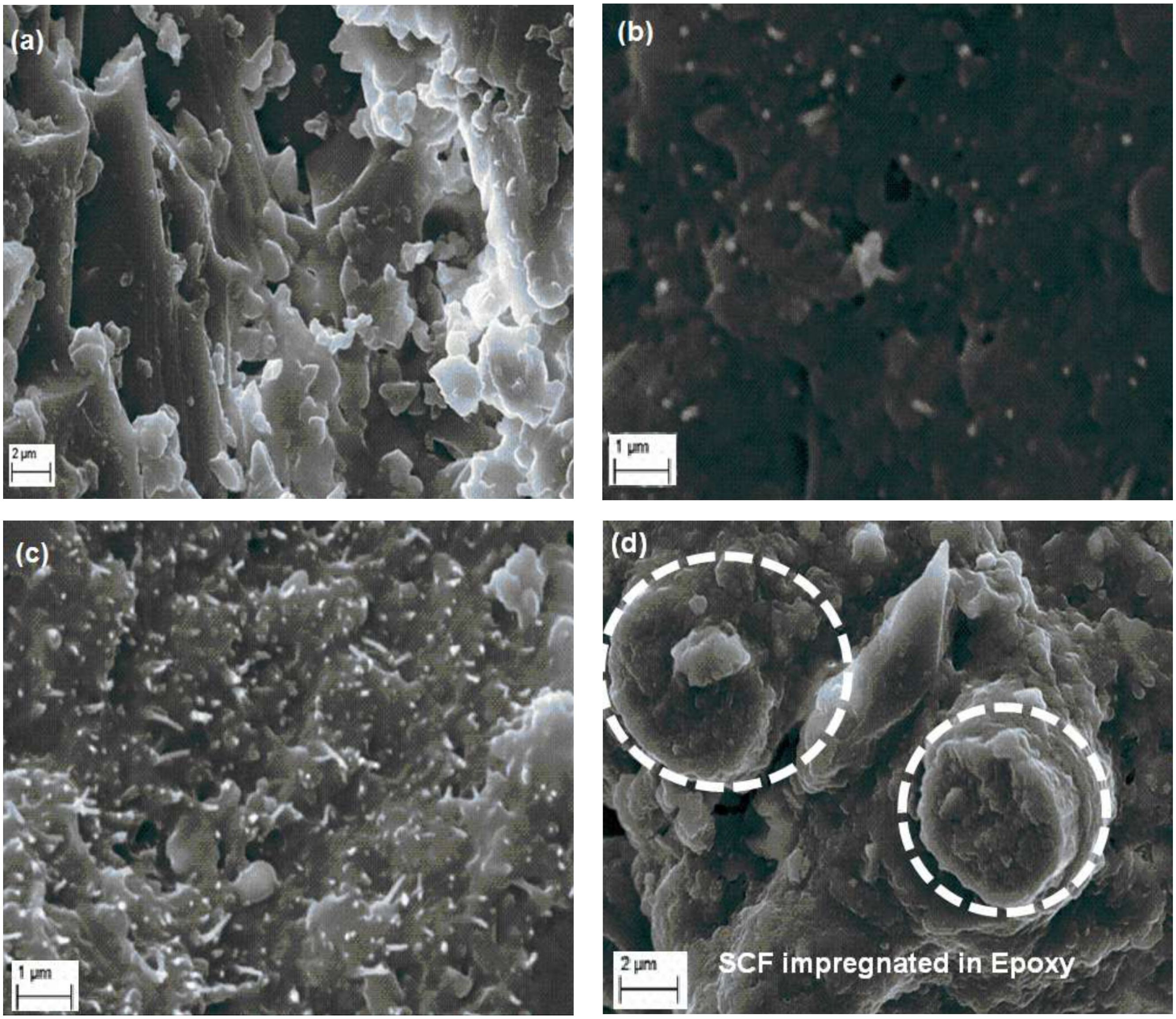


**Figure 2 : Scanning electron micrograph of : (a) Neat epoxy (b) Epoxy-SCF (1wt %) (c) Epoxy-SCF (2wt %) and (d) SCF impregnated in Epoxy**

Flexural modulus respectively. The trend of increasing strength and modulus with increasing SCF content continues with the flexural properties of the material. Neat epoxy shows a flexural strength of 96 MPa and that of E-SCF2 BM is 126 MPa that is a 31% increase in strength over pure matrix material. Whereas with unmilled SCF the increase in flexural strength is 111 MPa, which is a 15% increase in the strength of the material with addition of 2wt% SCF. This is a substantial increase in the flexural strength of the fiber. In composite materials reinforced by carbon fibers the load bearing is done by the reinforcement fibers. Load is transferred from the matrix to the fiber, when the fiber breaks it leads to the failure of the composite. Another important feature is the interfacial bonding between the fiber and the matrix, if the interfacial bonding is week the transfer of load is not optimal which results in the cracking of the matrix and decreased mechanical properties. Concentration of the reinforcement fibers within the matrix results in stress concentration at a few points in the composite and leads to composite failure at lower loads. In the current work we have tried to overcome these problems. We have used a mechanical chopper to decrease the length of the carbon fibers and then used ball milling to separate the fibers. This process ensured the equal dispersion of the carbon fibers in the epoxy matrix, ensuring when the load is applied to the composite material the load is equally distributed throughout the matrix

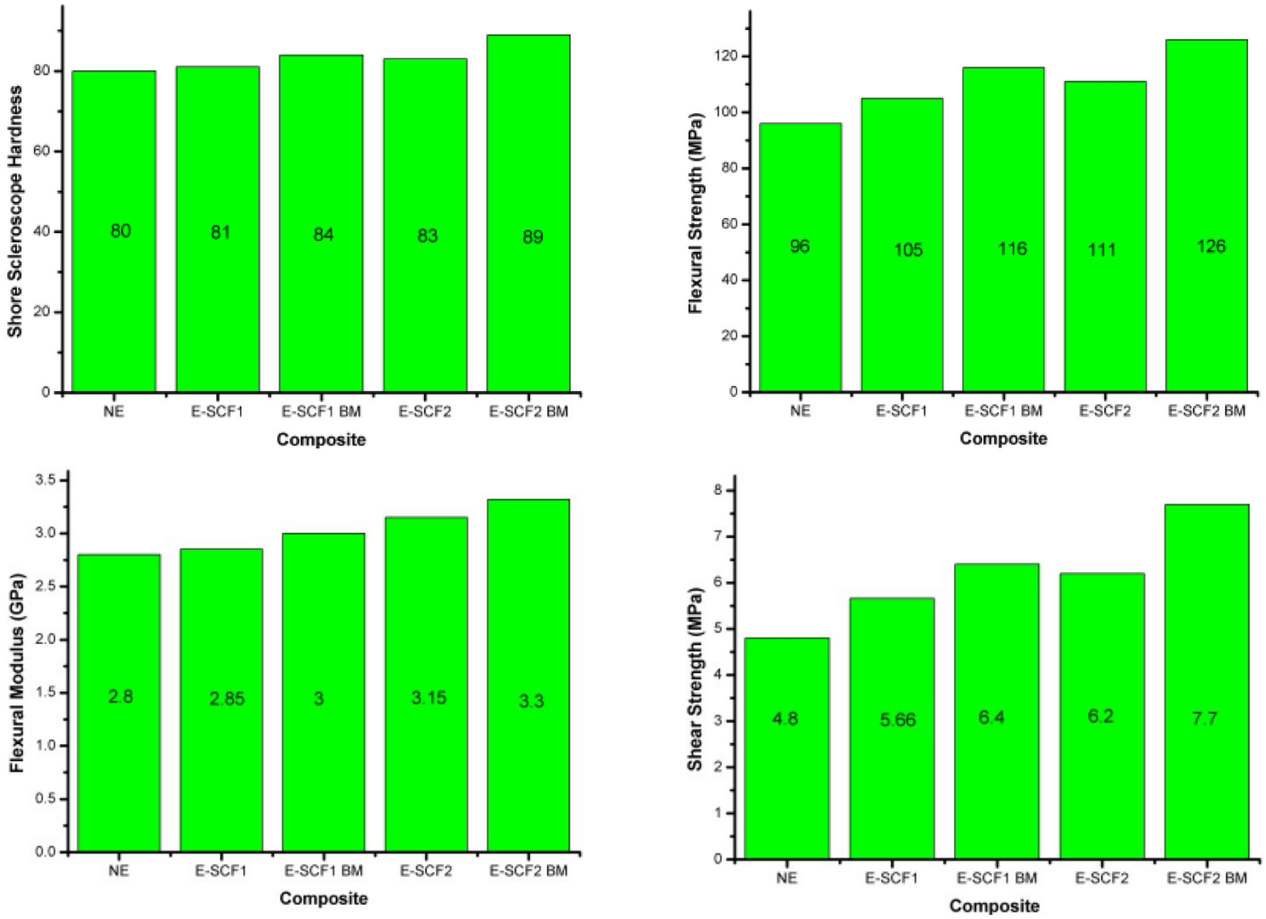


**Figure 3 : shows mechanical properties of composites : (a) Hardness (b) Flexural strength (c) Flexural modulus and (d) Shear strength**

and higher loads can be sustained by the composite before failure. The transfer of load from one fiber to another is also ensured, when one fiber fails the load is transferred to the next fiber and this continues till the composite fails. The chopping and ball milling of the fibre also results in increase of the surface area of the reinforcement material in the matrix, leading to a larger area of interface between the fiber and the epoxy matrix. This leads to better transfer of load to the reinforcement material in the composite and increase in interfacial bonding strength between the matrix and reinforcement material. We attribute the higher flexural strength of the material to the above discussed factors.

The ultimate increase in the modulus of the composite is 17%. The extension at breaking is the same, meaning that the flexibility of the sample remains same; increase in the modulus is only attributed to the increase in flexural strength by the addition of carbon fiber to the matrix.

## Shear strength

The same increase is observed in shear strength of the material. Shear strength was calculated using the following formula:

$$\tau = 0.75P_m/bh$$

where $\tau$ = shear strength (MPa), $P_m$ = Maximum load recorded (N), b= width of sample (mm) and h= height of sample (mm). The results are displayed in Figure 3(d). Highest shear strength is seen for E-SCF2 BM (7.7 MPa) which shows a 60% increase over NE. The shear strength for non-ball milled materials is lower when compared to ball milled SCF (6.2MPa) a 30% increase in strength.

## Strengthening mechanism

It is seen consistently throughout the mechanical properties that there is an increase in the mechanical properties on addition of SCF, but when the SCF has been ball milled it shows a higher increase in the mechanical properties than with the normal SCF. Ball milling helps in separating the Carbon Fibers and stops them from forming agglomerates. The infiltration of the fibers in composites was improved by carefully ball milling the SCF so as to reduce the diameter of the fiber aggregates. Further, due to ball milling it is easier for the fibers to disperse inside the epoxy matrix when they are added to the matrix under magnetic stirring. The even dispersion helps in distributing the load equally throughout the composite material. When the carbon fiber is not dispersed equally the load is concentrated to one point and crack initiation occurs breaking the composite material and decreasing the mechanical properties of the material. Micro cracks forming in the matrix to due to the load are better dissipated as the SCF is dispersed equally in the matrix, the carbon fibers tend to form bridges between the cracks and hold together the matrix resulting in increase in mechanical properties. Another reason for the improvement in mechanical properties could be that ball milling leads to roughening of the surface of the fibre, the rough surface of the fibre helps in better interfacial bonding with the matrix and also prevents pull-out of the fibre due to increased friction, hence leading to an improvement in mechanical properties.

## Microstructure of cracked surface and failure mechanism

The main mechanisms of failure of composites are stress concentration, resin matrix cracking, fiber pull out and debonding, the main mechanism of composite failure is assumed to be delamination[33,34]. Figure 4(a) & (b) are representative of the cracked surface of the composite. It can be seen in Figure 5(a) that the fiber has been pulled out of the matrix, which also can be seen in Figure 4(b). The pulled out fiber has left grooves in the epoxy matrix and one pulled out fiber can be seen in the figure as well. In Figure 4(a) it can be seen that there is minimal matrix residue on the pulled out fiber, which shows that the interfacial bonding between the fiber and the matrix was weak, the epoxy matrix is seen to be detaching in both figures suggesting that the major mode of composite failure is delamination. In the non-ball milled material we assume that stress concentration due to uneven dispersion of fiber has lead to matrix cracking along with delamination and hence displays lower mechanical properties than the composites with ball milled SCF.

## Electrical properties

Figure 5 shows the electrical conductivity of the composites. Neat epoxy didn't show any electrical conductivity as it is an insulator. With the increase in SCF loading there is increase in the electrical conductivity, which is expected as Carbon fiber has very high electrical conductivity. The electrical conductivity of the ball milled SCF composites is higher than those of normal SCF composites. Ball milling disentangles the fibers and straightens the fibers as the fiber length decreases further, these properties lead to easier transport of elec-

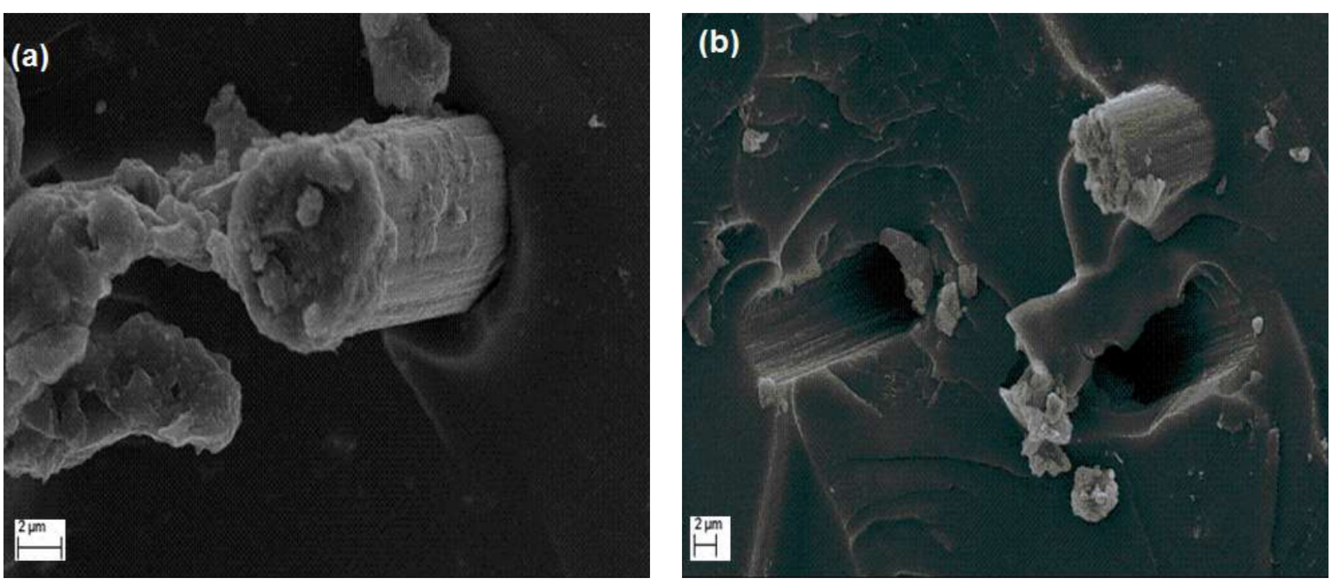


**Figure 4 : SEM of fractured surface (a) pulled out fiber and (b) Grooves left by pulled out Fiber**

trons between the fibers. At higher loadings of SCF there are increased points if contact between the fibers and hence the electrons can transfer at higher rate throughout the composite material leading to lower resistivity and better electrical conduction.

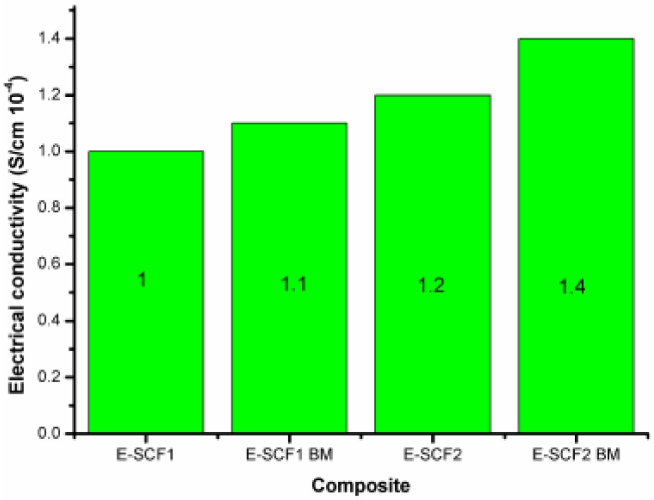


**Figure 5 : Electrical conductivity of the composite materials**

## CONCLUSION

In this work we have observed that increase in SCF loading in the Epoxy matrix leads to better mechanical and electrical properties of the composite materials. We have also established that at these percentages ball milling of the SCF results in better properties. This is possible due to the better dispersion of the SCF in the matrix and prevention of SCF Fibers due to their intrinsic attraction. We have established that the major causes of composite failure in our composites are due to delamination and fiber pull-out. More work is needed to improve interfacial bonding of the fiber with the matrix. We conclude that SCF is an ideal material to improve properties of Epoxy composites and that ball milling has a significant effect on improvement of the properties of the composite material.

## REFERENCES

**[1]** K.T.Lau, M.Lu, L.Chun-ki, H.Y.Cheung, F.L.Sheng, H.L.Li; Compos.Sci.Technol., **65**, 719–725 **(2005)**.

**[2]** J.Cho, J.Y.Chen, I.M.Daniel; Scr.Mater, **56**, 685–688 **(2007)**.

**[3]** X.L.Xie, Q.Liu -X, R.K.Y.Li, X.P.Zhou, Q.X.Zhang, Z.Z.Yu, Y.W.Mai; Polymer, **45**, 6665–6673 **( 2004)**.

**[4]** D.Zhang, A.B.Karki, D.Rutman, D.P.Young, A.Wang, D.Cocke, T.H.Ho, Z.Guo; Polymer, **50**, 4189–4198 **(2009)**.

**[5]** Z.Guo, T.Y.Kim, K.Lei, T.Pereira, J.G.Sugar, H.T.Hahn; Compos Sci Technol, **68**, 164–170 **(2008)**.

**[6]** Z.Guo, S.Park, S.Wei, T.Pereira, M.Moldovan, A.B.Karki, D.P.Young, H.T.Hahn; Nanotechnology **18**, 335704 **(2007)**.

**[7]** D.Zhang, R.Chung, A.B.Karki, F.Li, Z.Young Guo; J.Phys.Chem., **C114**, 212–219 **(2010)**.

**[8]** P.Vacca, G.Nenna, R.Miscioscia, D.Palumbo, C.Minarini, D.D.Sala; J.Phys.Chem., **C113**, 5777–5783 **(2009)**.

**[9]** Z.Guo, S.Park, H.T.Hahn, S.Wei, M.Moldovan, A.B.Karki, D.P.Young; J.Appl.Phys., **101**, 09M511 **(2007)**.

**[10]** V.L.Pushparaj, M.M.Shaijumon, A.Kumar, S.Murugesan, L.Ci, R.Vajtai, R.J.Linhardt, O.Nalamasu, P.M.Ajayan; Proc.Natl.Acad.Sci., USA, **104**,13574–13577 **(2007)**.

**[11]** P.Kim, N.M.Doss, J.P.Tillotson, P.J.Hotchkiss, M.J.Pan, S.R.Marder, J.Li, J.P.Calame, J.W.Perry; ACS Nano, **3**, 2581–2592 **(2009)**.

**[12]** D.S.Koktysh, X.Liang, B.G.Yun, I.Pastoriza-Santos, R.L.Matts, M.Giersig, C.Serra-Rodrýguez, Liz-L.M.Marzan, N.A.Kotov; Adv.Funct.Mater, **12**, 255 **(2002)**.

**[13]** S.Liu, L.Wei, L.Hao, N.Fang, M.W.Chang, R.Xu, Y.Yang, Y.Chen; ACS Nano, **3**, 3891–3902 **(2009)**.

**[14]** T.Shimada., K.Ookubo, N.Komuro, T.Shimizu, N.Uehara; Langmuir, **23**,11225–11232 **(2007)**.

**[15]** Z.Wen, S.Ci, J.Li; J.Phys.Chem. **C113**, 13482–13487 **(2009)**.

**[16]** Z.Guo, H.T.Hahn, H.Lin., A.B.Karki, D.P.Young; J.Appl.Phys., **104**, 014314 **(2008)**.

**[17]** Z.Guo, S.Park, H.T.Hahn, S.Wei, M.Moldovan, A.B.Karki, D.P.Young; Appl.Phys.Lett.; **90**, 053111 **(2007)**.

**[18]** Carlberger, Thomas, Anders Biel, Ulf Stigh; International journal of fracture, **155(2)**, 155-166 **(2009)**.

**[19]** Liu, Wanshuang, Zhonggang Wang, Li Xiong, Linni Zhao; Polymer, **51(21)**, 4776-4783 **(2010)**.

**[20]** Raza, Mohsin Ali, Aidan Westwood, Chris Stirling; Carbon, **50(1)**, 84-97 **(2012)**.

**[21]** R.J.Sager, P.J.Klein, D.C.Lagoudas, Q.Zhang, J.Liu, L.Dai, J.W.Baur; Composites science and technology, **69(7)**, 898-904 **(2009)**.

**[22]** S.Kang, S.I.Hong, C.R.Choe, M.Park, S.Rim, J.Kim; Polymer, **42**, 879 **(2001)**.

**[23]** M.Zanetti, G.Camino, D.Canavese, A.B.Morgan, F.J.Lamelas, C.A.Wilkie; Chem.Mater, **14**, 189–193 **(2002)**.

**[24]** J.Zhu, A.B.Morgan, F.J.Lamelas, C.A.Wilkie; Chem.Mater, **13**, 3774 **(2001)**.

**[25]** Tadaharu Adachi Mayuka Osaki, Wakako Araki, Soon-Chul Kwon; Acta Materialia, **56**, 2101–2109 **(2008)**.

**[26]** Young-Kuk Choi, Koh-ichi Sugimoto, Sung-Moo Song, Yasuo Gotoh, Yutaka Ohkoshi, Morinobu Endo; Carbon, **43**, 2199–2208 **(2005)**.

**[27]** F.Rezaei, R.Yunus, N.A.Ibrahim; Mater Des., **30**, 260–263 **(2009)**.

**[28]** M.K.Seo, S.J.Park; J.Colloid.Interface.Sci., **330**, 237–242 **(2009)**.

**[29]** Z.W.Xu, L.Chen, Y.D.Huang, J.L.Li, X.Q.Wu, X.M.Li; Eur.Polym.J., **44**, 494–503 **(2008)**.

**[30]** Singhal, Sunil Kumar, Maneet Lal, Indu Sharma, Rakesh Behari Mathur; Journal of Composite Materials,. doi:10.1177/0021998312443397, **(2012)**

**[31]** A.Esawi, K.Morsi; Composites Part A: Applied Science and Manufacturing, **38(2)**, 646-650 **(2007)**.

**[32]** Kollo, Lauri, Marc Leparoux, Christopher R.Bradbury, Christian Jäggi, Efraín Carreño-Morelli, Mikel Rodríguez-Arbaizar; Journal of Alloys and Compounds, **489(2)**, 394-400 **(2010)**.

**[33]** J.Li, M.L.Sham, J.K.Kim; Compos Sci Technol, **67**, 296–305 **(2007)**.

**[34]** M.F.Hibbs, M.K.Tse, W.L.Bradley; Toughened composites, ASTM STP, 937. American Society for Testing and Materials, 115–30 **(1987)**.