Studies on Electrically Conductive Composites of Ethylene Propylene Diene Monomer Rubber and Steel Fibers

Anjali A. Athawale, Aparna M. Joshi

Department of Chemistry, University of Pune, Pune 411 007, India

Received 25 February 2010; accepted 27 September 2010
DOI 10.1002/app.33456
Published online 12 January 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: Electrically conductive composites comprised of ethylene propylene diene monomer (EPDM) rubber and steel fibers were prepared by an open mill mixing method. Fibers of two distinctly different lengths (5 mm and several meters) were used and the influence of these fibers on electrical conductivity, mechanical, thermal, and physical properties of the composites was investigated. Composites with different compositions were prepared by varying the loading levels of fibers from 20–100 phr (parts per hundred parts of rubber). Homogeneity of the composites was determined using scanning electron microscopy. Further analysis included the measurement of resistance, hardness, tensile strength, tear strength, rebound resilience, etc. The results of the analysis revealed that the addition of steel fibers rendered conductivity to the otherwise insulating EPDM rubber even at small loading levels, however, the length appears to have negligible effect on conductivity. In case of short fibers, the resistance of composites was observed to decrease from > 40 MΩ (Initial value of EPDM rubber) to 25 KΩ at a loading level of 20 phr with a further significant decrease of the order of 10^3, that is around 18 Ω at 100 phr. Composites with long fibers exhibited resistance in the range of 15 kΩ–70 Ω at loading levels between 30 and 100 phr. The conductivity of the sample is observed to be altered negligibly on ageing. Mechanical properties such as hardness, tensile, and tear strength were observed to be enhanced in case of composites except resilience which decreased by 29 % in comparison to EPDM rubber. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 120: 3036–3041, 2011

Key words: EPDM rubber; steel fibers; open mill mixing; electrical conductivity

INTRODUCTION

Electrically conducting rubber has generated interest in many applications. To dissipate the static charges developed during many dynamic processes such as electrostatic assist printing, fiber weaving, landing of planes etc., conductive rubber is used in different forms either as roller or coating of surface. The static charge generated has to be dissipated, as it may cause spark, fire hazards, damage to machines, electronic devices etc.[1] Elastomers can be made conductive using metals, conducting carbon black or conducting polymers. Metal powders when used are required in higher loadings up to 60–90% so as to attain conducting network within the matrix.[2,3] Lower loading levels yield poorly conducting product while higher loading results in product with poor mechanical properties. Metal oxides can be attacked by atmospheric moisture and conductivity is reduced.[4,5] Carbon black filler gives less thermal stability to the composite; it is also restricted to give intermediate conductivities required for charge dissipation,[6,7] finally, the aesthetics of the components demands pigmented composites. Conductive polymers have restricted their use due to environmental, thermal instability, formation of continuous conducting network in the rubber matrix, and poor mechanical properties.[8–12]

Present work involves the synthesis and characterization of Ethylene propylene diene monomer (EPDM)-Steel fibers conducting composites. From the results, it is observed that composites of EPDM rubber and steel fibers bear advantages such as light weight since the addition of steel fibers barely alters the weight of the composite. Second advantage is that, they exhibit good processing ability and easy control of conductivity. In addition to this, the physical and mechanical properties of rubber are negligibly altered at the optimum loading of steel fibers. Steel fibers form a continuous conducting network in the matrix giving conductivity to the composite as observed from the scanning electron micrographs. The composites can also be pigmented. Pigments are required in very small quantity so that conductivity is not affected. Pigments can be added during mixing of chemicals into rubber on mill. Another advantage of steel fibers is that they can be recycled.

Correspondence to: A. A. Athawale (agbed@chem.unipune.ac.in).

Journal of Applied Polymer Science, Vol. 120, 3036–3041 (2011)

TABLE I
Various Ingredients and Their Quantities in phr Added in EPDM-Steel Fiber Composites

Ingredients							
EPDM (KEP 240)	100	100	100	100	100	100	100
Stearic acid	1	1	1	1	1	1	1
Mercaptobenzimidazole	1	1	1	1	1	1	1
Dicumyl peroxide–40	8	8	8	8	8	8	8
Triallyl cyanurate	2	2	2	2	2	2	2
Steel fibres (5 mm long) (SF)	–	20	30	40	60	80	100
Steel fibres (Continuous length) (LF)	–	–	30	40	60	80	100

EXPERIMENTAL SECTION

EPDM-Steel fiber composites

The EPDM rubber (Kuhmo Products, Taiwan) of density 0.85 g cm^{-3} was used as a base polymer and a compound of EPDM with other chemicals was prepared as mentioned in Table I. All other chemicals and steel fibers were obtained locally. Steel fibers with two different dimensions were used for synthesizing composites. The diameter of steel fibers used was 7–8 micron and lengths were 5 mm and continuous length in several meters named as short fibers (SF) and long fibers (LF), respectively. The composites of EPDM rubber and steel fibers were prepared by mixing EPDM rubber and steel fibers (proportions given in Table I) in a two roll open mill at a temperature of 40–45°C, keeping a fixed particular nip gap as follows - Initially the EPDM rubber was masticated on the mill for 4–5 min followed by the addition of stearic acid and antioxidant—Mercaptobenzimidazole—with an interval of few minutes. This mixture was rolled for 3 to 4 min. Further, Dicumyl peroxide-40 (Dicup 40–a curative) and Triallyl cyanurate (TAC an accelerator) were added and the whole mixture was roll milled for 3–6 min with 5–6 mm nip gap first and then for 4–5 min with 2–3 mm nip gap. After assuring for homogenization of the mixture, steel fibers were added slowly and roll milled for another 5–6 min, without the use of knife cut. Sheets of composite of size 150 mm × 150 mm × 2 mm were compression molded at 170°C for 10 min in a hot compression press. Resistance was measured before and after curing the rubber sheets and also upon heat ageing. Physical, short term and dynamic stress–strain properties of the composites were studied. All the tests were carried out at room temperature. EPDM rubber was used as a binder to steel fibers therefore any other dry bond system was not added in the compound. After vulcanization, the fibers do not separate out if immersed in solvents, which ensure the binding of steel fibers and rubber during polymer cross linking.

Measurement of physical properties

Majority of the testing were done as per the ASTM standards. All the test specimens were cured at 170°C for 10 min in a hot compression press with a pressure of 10.34 MPa, cooled and used after 24 h.[13,14] The curing time of 10 min for the samples was selected as in the present case peroxide was used as a curing agent and the sample thicknesses were relatively small.

Morphological characterization

For SEM analysis samples of the composites (such as 30, 60, 100 phr LF and 20, 60, 100 phr SF) were prepared by pressing the sheets in a hydraulic press and cutting pieces of 0.5 × 0.5 cm size. Samples were coated with Pt followed by analysis using JEOL JSM 6360-A SEM analyzer.

Electrical conductivity

Surface resistance was measured by 2 probe method using a Digital Multimeter (Meco Instruments) by placing the probes at a unit length apart. Resistance was measured before and after curing as well as heat ageing of the samples at 90°C. In rubber elastomers the surface of rubbers may conduct electricity more easily than the bulk hence, surface resistance was measured.[15,16]

Specific gravity

Specific gravity analyses were done by adopting the hydrostatic weighing method as per ASTM standard D297–93. Weights were measured on Contech Instruments weighing balance. Testing was done at room temperature. This method directly gives the specific gravity value, which is the ratio of mass of a unit volume of composite to mass of a unit volume of water.[17]

Short term stress and strain properties

Hardness

Shore A (ASTM standard D 2240) method was used for measuring the hardness of specimens. The specimens were loaded on Type–2 durometer Shore A operating stand. GSE Testing Instrument with indenter of type "A" was used for measuring the hardness. Thickness and diameter of samples were 6 mm each.

Tensile strength

ASTM standard D-412 was followed for testing the tensile strength. A sheet of size 150 × 150 × 2 mm^3 was used to prepare the specimens. Five dumbbell shaped specimens with ASTM standard dimensions were punched in a Die–C. Punching machine was used to get a single impact stroke, the strength of all five specimens were measured, the extreme high and extreme low readings were discarded and average of three specimens were taken. Computerized Tensile Testing machine by Star Testing Systems was used for measuring the tensile strength.

Tear strength

Tear strength testing was done with reference to ASTM standard D-624. Three dumbbells were punched from the sheet of size $150 \times 150 \times 2$ mm^3. Punching machine with a single impact stroke was used to ensure smooth cut surfaces in Die-C with dimensions as per ASTM standard D-624. These specimens were tested and average of them was taken. Tear strength was measured on Star Testing Systems machine.

Dynamic stress and strain properties

Rebound resilience

This test was performed using ASTM standard D 2632-01. The specimen of 12 mm thickness and 6 mm diameter was used. Impact resilience was measured by Vertical Rebound Resiliometer by The Shore Instrument and Manufacturing Co, Mumbai, India.

RESULTS AND DISCUSSION

From the results, it is observed that addition of both short and long steel fibers into the EPDM rubber induces electrical conductivity. The composites are seen to have even distribution of steel fibers. Additionally, the specific gravity, short term stress and strain such as hardness, tensile strength, and tear strength are observed to be higher in composites compared with EPDM rubber. However, the dynamic stress and strain is affected inversely, which shows a lower value.

Morphological characterization

Figure 1 shows the SEMs of the samples containing 30, 60, and 100 phr of LF and 20, 60, and 100 phr of SF respectively. At 30 and 60 phr the fibers appear to be intact with homogeneous dispersion and continuous length in the samples. However, at 100 phr, dense clusters of undispersed fibers are still present suggesting insufficient dispersion in the matrix. In case of 30 phr LF, the fibers are seen to form good connecting network that covers the whole matrix rendering conductivity to the composite, the distribution of steel fibers in 20 phr SF is also found to be uniform making the composite conductive. With 60 and 100 phr of LF and SF, the composites show further denser connecting network of the steel fibers. Polymer rupture due to steel fibers appears to be relatively negligible.

Electrical conductivity

EPDM rubber is basically insulating in nature with the resistance of the order of greater than MΩ.[18] Incorporation of small proportions (20–30 phr) of steel fibers into the rubber reduces the resistance considerably (1000 times) thereby making the composite conducting. The resistance is $\sim$ 35–40 KΩ on addition of 30 and 20 phr of long and short steel fibers, respectively, which decreases further with respect to loading levels. The change is gradual in case of LF [Fig. 2(a)] while it appears to be rapid for SF [Fig. 2(b)]. These results indicate that the composite has a well organized network structure with one component (rubber) being insulating while the other (steel fibers) being conducting, which effectively leads to the induction of conductivity in the matrix. Amongst the two types of fibers, the SF is expected to form a better network in the matrix, hence exhibits relatively higher conductivity at lower loading levels in comparison to LF. Similar reason holds for the differences observed as a function of concentration.

Electrical conductivity after heat ageing

Further, the influence of heat ageing on the conductivity of the samples were investigated [Fig. 3(a,b)]. The results show a marginal increase in resistance with respect to time and temperature that can be attributed to changes in the polymer matrix resulting in micro changes in the conductive paths. However, the data overall point out to a stable conductive composite.

Specific gravity

The enhancement in specific gravity of composites is obvious since the steel fibers constitute a denser matter compared with EPDM rubber. The magnitude of increase is proportional to the concentration of steel fibers being added as observed in Figure 4(a). The graph shows almost a linear nature except at the initial stages. The rise is by 0.5 with 100 phr loading. The length of fibers does not contribute towards the difference.

Hardness

Hardness, which is a resistance to indentation, was measured based on initial indentation and depends upon the visco elastic behavior of the composite. Indentor is the calibrated spring needle,[19] which

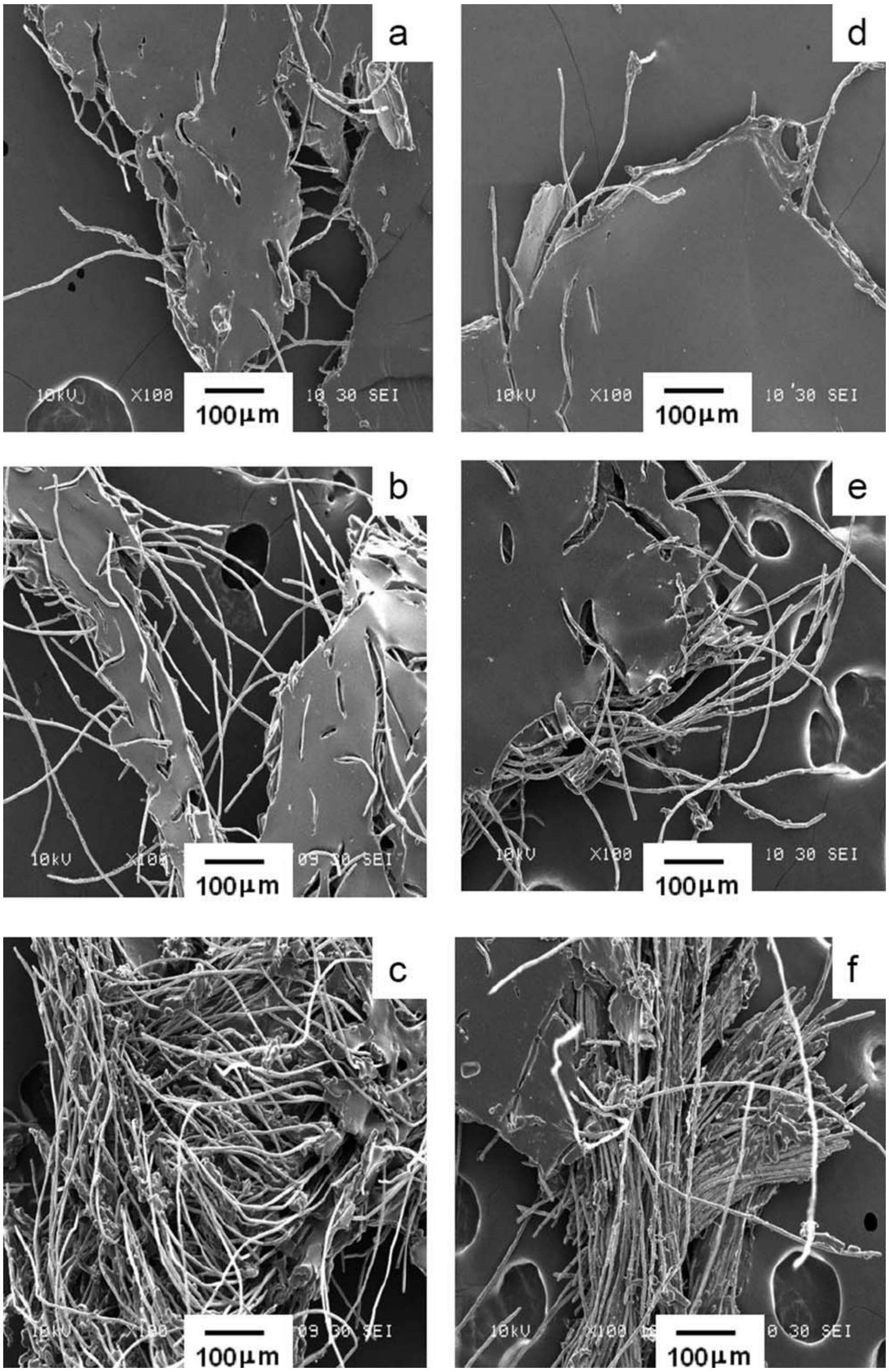

Figure 1 SEM images (a) 30 phr LF; (b) 60 phr LF; (c) 100 phr LF; (d) 20 phr SF; (e) 60 phr SF; and (f) 100 phr SF.

deforms the material for a short period. From Figure 4(b), it is seen that the hardness increases gradually both with long as well as SF. This is because deformation is directly proportional to the elastic content (or range) of a material.[20] Steel fibers being hard, render resistance to deformation thereby increasing the hardness of the composites.

Tensile strength

Tensile strength indicates the maximum tensile stress that a substance can withstand, while stretching without rupture[21] and is a sum of every component added in the composite. The effect of steel fibers on tensile strength has been studied; Figure 5(a), depicts the change in the tensile strength of

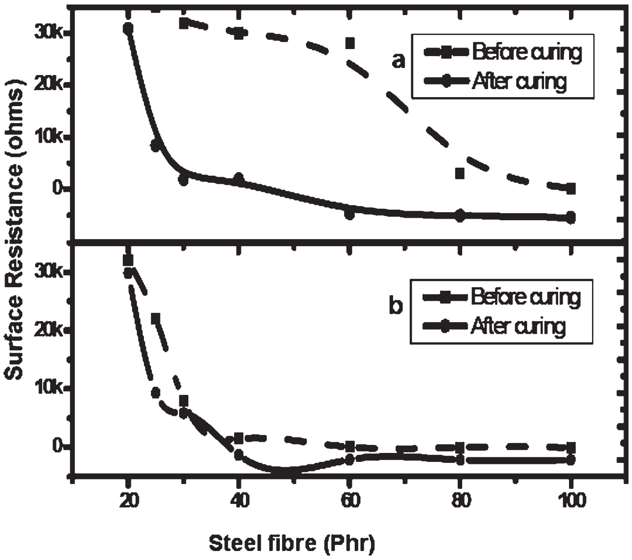

Figure 2 Effect of (a) LF and (b) SF on electrical conductivity of composites.

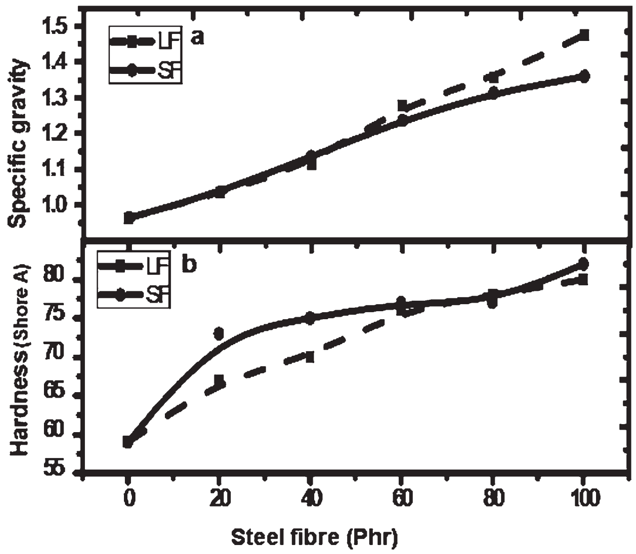

Figure 4 Effect of steel fibers on (a) Specific gravity and (b) Hardness of the composites.

the composites with respect to the parts of steel fibers per hundred parts of rubber added in the composite. The increase is sharper in case of SF in comparison to LF. This is also related to the hysteresis loss. Hysteresis loss increases with fiber loading.[22–24] This is because rubber polymers are visco elastic in nature. Addition of higher fraction of steel fibers leads to an increase in the viscous part of the matrix. On applying force or energy to polymers, they deform. When the applied force is removed, the elastic material regains back its original shape completely. Viscous material or steel fibers retard the deformation. Force required to stretch the steel fibers is more. Therefore, tensile strength is increased with increasing content of steel fibers. Total tensile strength also includes destruction of fiber–rubber interface and friction between fibers and rubber.[25] Tensile strength increases from 1.47 to 2.25 MPa as the steel fiber loading is increased from 20 to 100 phr.

Tear strength

How a tear can be initiated or propagated is very important in case of rubbers. Figure 5(b), shows the effect of LF on tear strength. The trend is quite similar to tensile graph. Tear strength was comparatively same as that of blank compound (without fibers) at lower loading but with higher loading the load required to tear same thickness of specimen was observed to be higher. In case of SF as shown in Figure 5(b), there is a steep rise at 20 phr loading than the blank, it remains unaltered upto 40 phr and

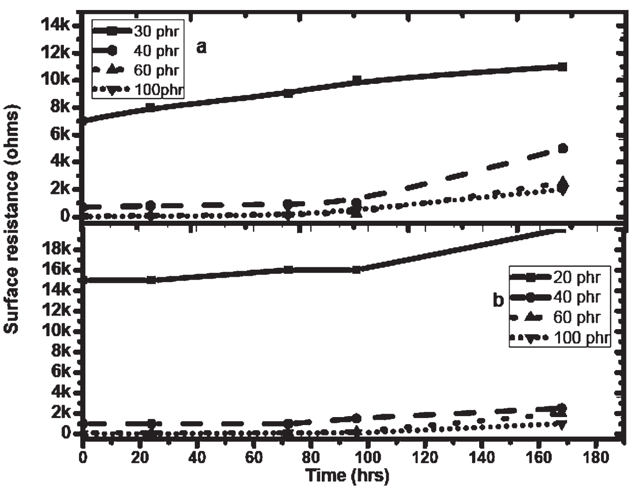

Figure 3 Effect of heat ageing at 90°C on electrical conductivity of (a) LF and (b) SF composites.

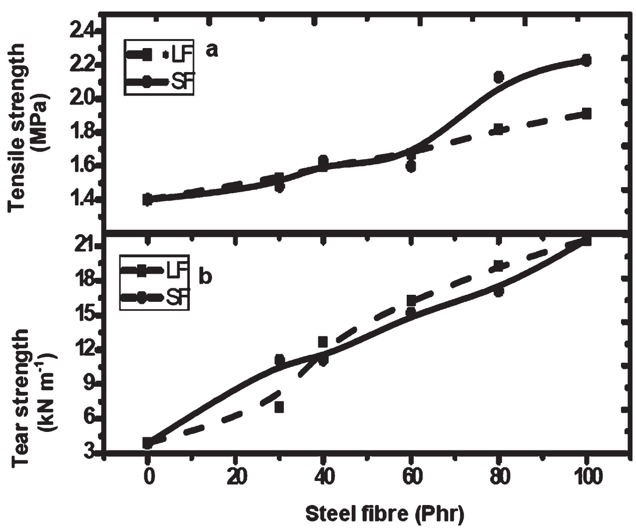

Figure 5 Effect of steel fibers on (a) Tensile strength and (b) Tear strength of the composites.

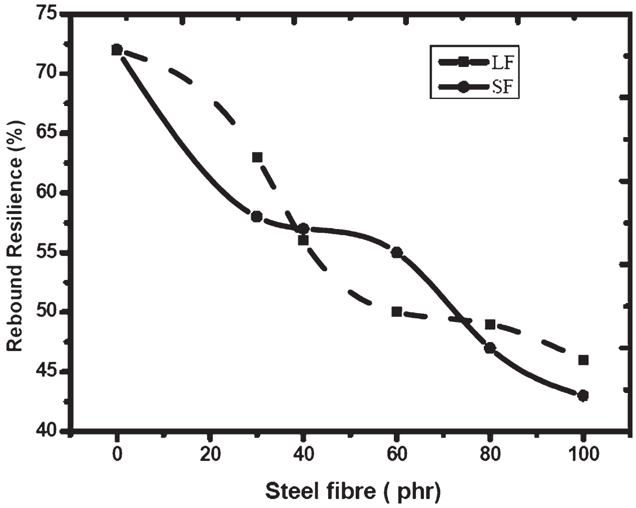

Figure 6 Effect of steel fibers on rebound resilience of the composite.

again rises steeply with degree of loading. LF prevents the material from tearing more than SF.

Rebound resilience

The ratio of energy output to energy input, on rapid recovery of a deformed specimen can be determined through measurement of resilience of the substance. This property is very much related to the elasticity, especially in case of rubbers. The rebound resilience is observed to be an inverse function of fiber loading (both long and SF). As the hardness increases the resilience decreases, so also the elastic behavior.[26] With SF as shown in Figure 6 the difference is in the range of 5% from 20 to 60 phr loading and with LF, there is 25% difference up to 60 phr. As the elastic behavior of composite is reduced due to steel fiber, percentage energy output on sudden impact of energy input is reduced hence rebound resilience is reduced.

CONCLUSIONS

Incorporation of steel fibers into EPDM rubber has significance to combine electrical conductivity with desired mechanical strength and flexibility of elastomer. At all loading levels of steel fibers the composite can be easily cured at specified temperature and specified time retaining the conductivity, heat ageing of the samples lead to a negligible change. The effective phr of long steel fibers and short steel fibers is 30 and 20, respectively. The morphology shows homogeneity of the composites. The extent of change in specific gravity, hardness, tensile strength, tear strength, and rebound resilience does not alter the conductivity of the samples appreciably. Further, at the above loading levels, the composite can be easily molded into any shape, it can be pigmented to improve the aesthetics of the component, and it also retains its visco elastic nature.

Authors thank the support to the project from K.D. Joshi Rubber Industries Pvt. Ltd and Department of Physics, University of Pune.

References

1. Kathirgamanathan, P. Polymer 1993, 34, 1549.
2. Bhattacharya, S. K.; Kathirgamanathan, P.; Dekker, M. Polymer Rep 1986, 1, 265.
3. Gwaily, S. E.; Nasr, G. M.; Badawy, M. M. Egypt J Sol 2001, 24, 2.
4. Baraton, M. I.; Merhari, L. Nanostruct Mater 1998, 10, 5.
5. Sohn, J. H.; Atzeni, N.; Zeller, L. Sens Actuators B Chem 2008, 131, 8.
6. Kathirgamanathan, P.; Fawcett, A. H. R Soc Chem 1991, 34, 2907.
7. Bouguettaya, M.; Vedie, N.; Chevrot, C. Synth Met 1999, 102, 1428.
8. Lindenberger, H.; Schafer, S. D.; Roth, S. Hanack Synth Met 1987, 37, 18.
9. Yigit, S.; Hacaloglu, J.; Akbulut, U.; Oppare, L. Synth Met 1996, 79, 11.
10. Jiang, G.; Gilbert, M.; Hitt, D. J.; Wilcox, G. D.; Balsubramanian K. Compos Part A Appl Sci 2002, 33, 745.
11. Foot, P. J. S.; Kaiser, A. B.; Kirk Othmer Encyclopedia of Chemical Technology; John Wiley and Sons Inc: New York, 2004.
12. Laska, J.; Zak, K.; Pron, A. Synth Met 1997, 84.
13. Brown, R. P. Physical Testing of Rubber, 2nd ed.; Elsevier Applied Science Publishers: London and New York, 1986; Chapter 4.
14. Morton, M. Rubber Technology, 3rd ed.; Van Norstrand Reinhold Company Inc.: New York, 1987.
15. Smith, L. P. The Language of Rubber, 1st ed.; Butterworth-Heinemann Ltd.: Oxford, 1993; Chapter 15.
16. Blythe, A. S. Polym Test 1984, 4, 195.
17. Annual Book of ASTM Standards. ASTM D 2003, 09.01, 297–93.
18. Brown, R. P. Physical Testing of Rubber, 2nd ed.; Elsevier Applied Science Publishers: London and New York, 1986; Chapter 13.
19. Annual Book of ASTM Standards. ASTM D 2003, 09.01, 2240–02b.
20. Loganathan, K. S. Rubber Engineering, 1st ed.; Tata McGraw-Hill Publishing Company Limited: New Delhi, 1998; Chapter 3.
21. Annual Book of ASTM Standards 2003. ASTM D 2002, 09.01, 412–98a.
22. Pramanik, P. K.; Khastagir, D.; Saha, T. N. Composites 23, 3, 183, 1992.
23. Kar, K. K.; Bhowmick, A. K. J Appl Polym Sci 1998, 65, 7, 1429.
24. Bulgin, D. Composites 1971, 2, 3, 165.
25. Smith, L. P. The Language of Rubber, 1st ed.; Butterworth-Heinemann Ltd.: Oxford, 1993; Chapter 5.
26. Annual Book of ASTM Standards. ASTM D 2003, 09.01, 5963–5997.