Advanced Composite Materials
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/tacm20

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Published online: 12 May 2014.

To cite this article: Jeong U. Roh, Han Sang Kim & Woo Il Lee (2014) Isotropic conductivities in chopped carbon fiber composites using expanded polypropylene, Advanced Composite Materials, 23:5-6, 409-420, DOI: 10.1080/09243046.2014.915096

To link to this article: http://dx.doi.org/10.1080/09243046.2014.915096

Advanced Composite Materials, 2014
Vol. 23, Nos. 5–6, 409–420, http://dx.doi.org/10.1080/09243046.2014.915096

Isotropic conductivities in chopped carbon fiber composites using expanded polypropylene

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(*Received 7 February 2014; accepted 10 April 2014*)

The aims of this study were to fabricate lightweight composites having isotropic electrical and thermal conductivities. The composites consist of expanded polypropylene (EPP) and chopped carbon fibers. They were randomly mixed together and fusion-bonded using steam at a high temperature with a high saturation vapor pressure. The quasi-isotropic electrical conductivities and isotropic thermal conductivities of the composites were achieved by the secondary expansion of the EPP. The densities, electrical and thermal conductivities of the composites with volume fractions of carbon fibers ranging from 4.2 to 8.5% were in the ranges of 0.2–0.3 g/cm^3, 1.1–105.8 × 10^{-4} S/cm, and 0.51–0.86 W/mK, respectively. The thermal conductivities of the composites were shown to increase monotonically with increasing volume fractions of carbon fibers, which is in agreement with the theoretical values using the series model.

Keywords: carbon fiber; expanded polypropylene; isotropic conductivity

Introduction

Lightweight and multifunctional materials are of interest in many industrial fields.[1–7] Electrical and thermal conductivities can be useful for applications such as the heat sink or electromagnetic wave shielding. Multifunctional materials can be made by mixing two or more materials, or by structural inducement.[1,6–14] To obtain isotropic properties, powder type fillers, for example, carbon black, carbon nanotube, and graphene have been used. The fillers of powder form are easy to realize isotropic properties. However, using fillers has a disadvantage which is the difficulty in filler dispersion in the matrix. Rod-shape fillers may result in better electrical and thermal conductivities than the powder-type fillers. However, it may result in anisotropic properties after the process because the rod-shaped filler with a high aspect ratio will inevitably have orientation depending on the direction of the mold flow or orientation biased towards the in-plane direction due to the shape of the filler even in the absence of mold flow. Consequently, electrical or thermal conductivities of the composite between the in-plane and the out-of-plane directions may become quite different.[10–15] In order to enhance the conductivities in the out-of-plane direction and thus to realize isotropic conductivities, expanded polypropylene (EPP) and carbon fibers were used in this study.

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The EPP is a foam type of semicrystalline polypropylene resin comprising many cells and has good impact characteristics.[16–19] It is widely used in packaging, thermal and sound insulation products.[16,17] One of the interesting characteristic of the EPP is the additional expansion during the fusion-bonding process by introducing steam (heating medium) into the mold. This is called the secondary expansion.[19,20] Unlike the compression molding or the injection molding,[13] the second expansion of EPP beads in the mold may force the carbon fibers to be randomly oriented along the tight gaps between the beads which may result in the isotropic properties as illustrated in Figure 1. In these composites, continuous paths through the carbon fibers in contact with each other can be formed in random directions. For this reason, carbon fiber/EPP composites were fabricated and their electrical and thermal conductivities were evaluated to assess the carbon fiber distributions in the composite and the resulting isotropic characteristics.

Experimental details

Materials

The following materials were used in the preparation of the samples: EPP (B 4.5P grade, Howtech Co., Ltd) was used as the matrix material. For the reinforcement, chopped carbon fibers with 6-mm length (T700SC, Toray Corp.) were used. The density of the EPP was decreased to 0.138 g/cm^3 from 0.2 g/cm^3 by the secondary expansion during the fusion-bonding process. The densities of the EPP and the carbon fiber/EPP composites are shown in Table 1. The EPP is the insulator. The electrical conductivity of the carbon fiber is 625 S/cm.[21] The thermal conductivities of the EPP and the carbon fiber are 0.061 and 9.408 W/mK, respectively.[21,22]

Preparation of the samples

Prior to the molding process, dry mixing of the chopped carbon fiber and EPP was conducted using a rotating mixer at 200 rpm for 30 min as shown in Figure 2. The mixture was then molded in a mold cavity. In order to achieve a high fusion-bonding efficiency of the EPP beads and secondary expansion in the mold cavity, the molding of the EPP beads in the mold cavity was done using the steam-chest molding process.[16,17] Prior to setting the mixture in the mold, the mold was preheated to

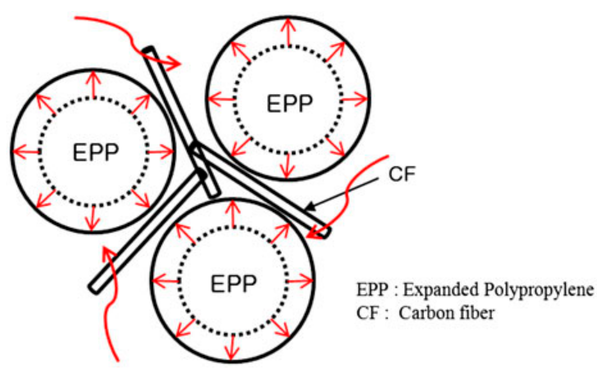

Figure 1. Illustration of carbon fibers positioned between the EPP beads.

Table 1. Densities of the EPP and carbon fiber/EPP composites after secondary expansion.

Items (vol.%)	Density (g/cm^3)
EPP (100%)	0.138
Carbon fiber (4.2%)/EPP (95.8%)	0.207
Carbon fiber (5.2%)/EPP (94.8%)	0.224
Carbon fiber (7.3%)/EPP (92.7%)	0.260
Carbon fiber (8.5%)/EPP (91.5%)	0.280

125 °C. The mixture was then set in the preheated mold cavity. After closing the mold, steam of 130 °C with a pressure of 640 kPa was injected from the bottom (fixed) mold and the upper (moving) mold. The steam was injected into the mold until the pressure gage on the mold indicated 440 kPa. This pressure was maintained for 4 s without opening the vent on the mold. Then, the vent on the mold was opened to release the steam pressure. Cool water (20 °C) was subsequently injected into the mold to cool the product. After the water in the mold drained, the sample was taken out of the mold. This cycle helps to create the fusion between the EPP beads.[18] After the above steaming cycle, the steam was injected from both sides of the mold. It helps to reduce pores on the surface of the molded EPP part.[17] The dimensions of the mold cavity were 30 cm (length) × 30 cm (width) × 5 cm (height), which were the same as those of the fabricated samples. The samples were then dried in a forced convection oven (Model FC-1D-2, Universal Scientific Co., Ltd) at 55 °C for 24 h.

For the comparison group, the sample was prepared without secondary expansion of EPP. The dimensions of the mold cavity were 5 cm (length) × 5 cm (width) × 5 cm (height). The mixture was poured in the preheated mold cavity at 125 °C. After closing the mold, the temperature of hot plate was set to 150 °C for 30 min. The sample was then taken out of the mold after the mold was cooled down to room temperature.

Measurement of electrical conductivity

The electrical conductivities of the carbon fiber/EPP composite specimens were measured in accordance with ASTM D4496 using a multi-meter (Fluke 187, Fluke Corp.) in the directions of in-plane and out-of-plane as shown in Figure 3. Specimens for measuring the electrical conductivity were machined to a cube of 1 cm^3 with a water jet cutter. Prior to the measurement, all surfaces of the samples were trimmed using a razor to create a flat surface. Silver paste was then painted onto their surfaces to ensure good contact between the surfaces of specimen and the copper electrodes. Due to the large deviation in electrical conductivity at different points on the surface of a specimen from the four-terminal measurements, the DC volume electrical resistivity was measured using the two-terminal technique in this study.[11] The electrical conductivity was recorded after a 5-min wait to obtain a stationary value. The volume electrical conductivity was calculated using the following equation.

$$\sigma = \left(\frac{RLW}{t}\right)^{-1} \quad (1)$$

where σ is the volume electrical conductivity, R is the measured resistance, L is the length of the specimen, W is the width of the specimen, and t is the thickness.

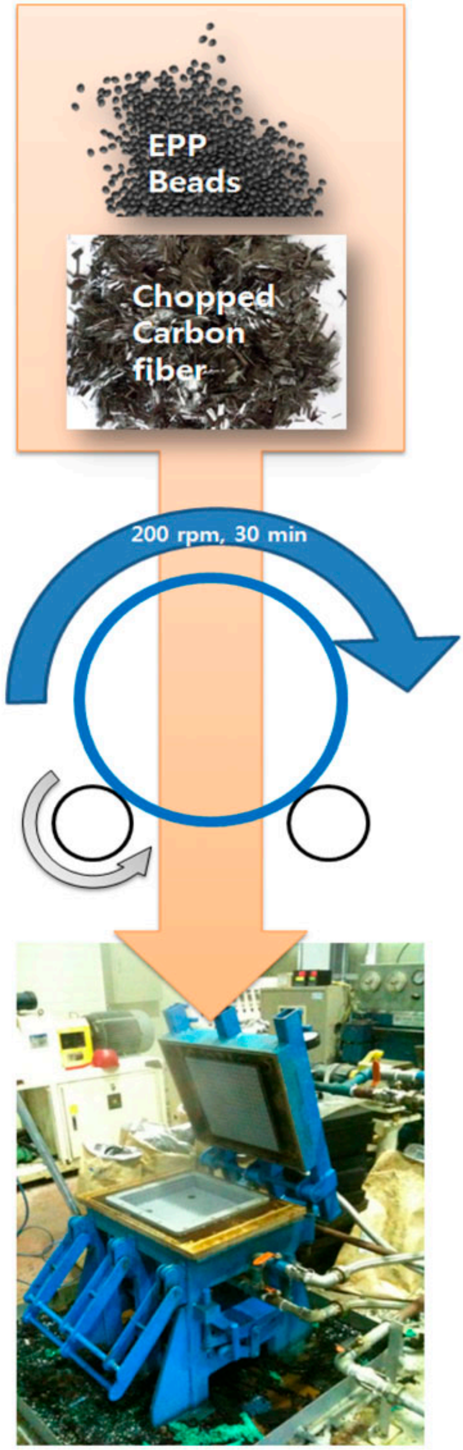

Figure 2. A schematic of manufacturing process.

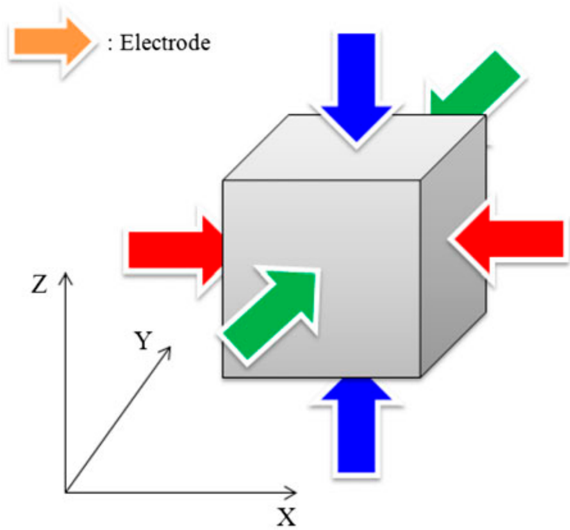

Figure 3. Illustration of measurement of electrical conductivity of the specimen.

The measurements were done in each direction for five specimens for each data point.

Measurement of thermal conductivity

The thermal conductivity of the carbon fiber/EPP composite was measured by a thermal conductivity analyzer (Mathis TCi, C-Therm Technologies Ltd) which employed the modified transient plane source technique.[23,24] Specimens for measuring the thermal conductivity were machined to a dimension of 10 cm (length) × 10 cm (width) × 5 (height) cm with a water jet cutter. As illustrated in Figure 4, three-dimensional thermal conductivities of the specimen were measured at the room temperature. The

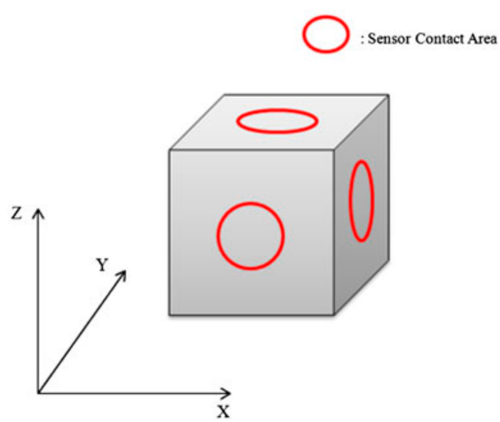

Figure 4. Illustration of measured area on the specimen.

measurements were done in each direction for three specimens for each data point. Figure 5 shows the specimen being installed in the analyzer.

Analysis of the morphology

In order to assess the orientation of the carbon fibers along the gaps between the beads, the morphologies of the cross-sections of carbon fiber/EPP composite were observed by FE-SEM (JSM-6700F, JEOL).

Results and discussion

Electrical conductivity

The volume electrical conductivity of the specimens was measured and the results are shown in Figure 6. In this composite, formation of electrical networks via the contact between the fibers may be considered as a major mechanism of electrical conductivity. At low fiber volume fraction of less than 5.2%, the electrical conductivities in both directions are low because of scarce chances of contact between fibers. However, the chances of contact between fibers and the aggregates of fibers in the composite become larger as the fiber volume fraction increases. Furthermore, the EPP expansion and the orientation of the carbon fibers were promoted slightly more in the thickness direction during the sample processing. For these reasons, the average volume conductivity of the specimen tends to increase with an increase in the amount of carbon fibers. Also, the volume electrical conductivity of the out-of-plane (thickness) direction was generally higher than that of the in-plane directions and the variation of the data becomes

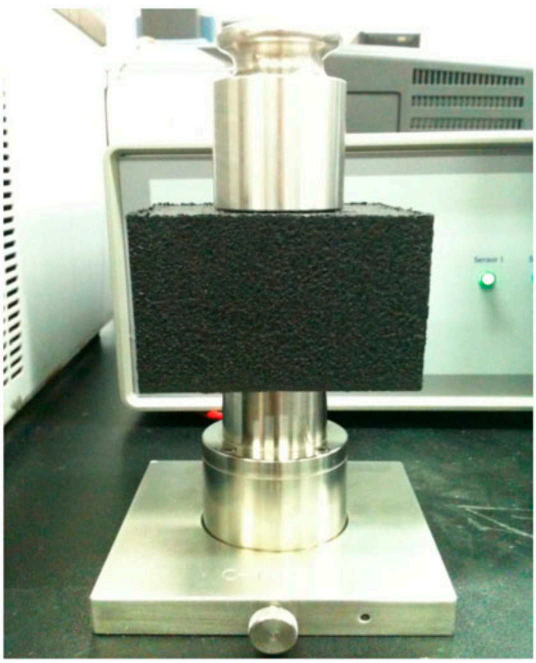

Figure 5. Installation view for measurement of thermal conductivity.

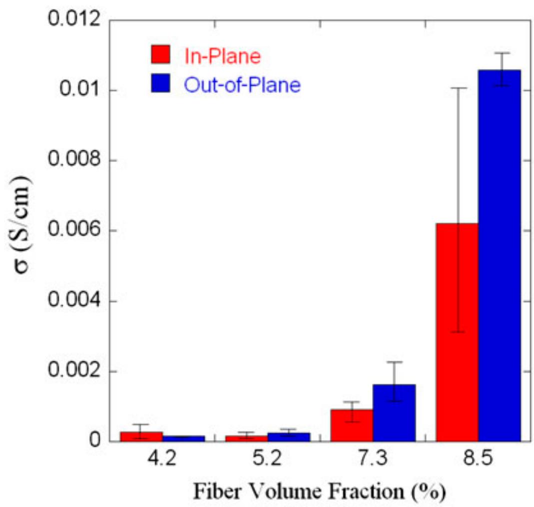

Figure 6. Electrical conductivity of carbon fiber/EPP composite.

greater as the fiber volume fraction increases. Volume electrical conductivity of 0.0062 and 0.0106 S/cm for the in-plane and out-of-plane directions, respectively, were obtained for carbon fiber volume fraction of 8.5% and it may be plausible to say that the electrical percolation threshold lies between 7.3 and 8.5% in this study.

Without secondary expansion of EPP, as shown in Figure 7, the sample with a fiber volume fraction of 8.5% shows anisotropic volume electrical conductivities (0.0014 and 0.0007 S/cm for the in-plane and out-of-plane directions, respectively).

The above electrical conductivity results were compared with the theoretical values obtained by Kirkpatrick's model given by the following [25,26]:

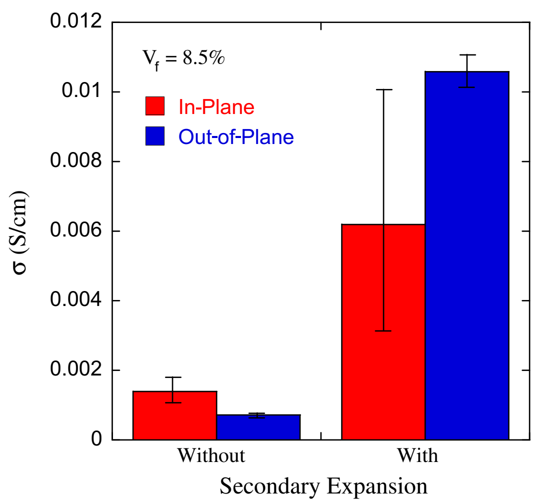

Figure 7. Electrical conductivity of carbon fiber/EPP composite ($V_f = 8.5\%$) with and without secondary expansion.

$$\sigma_c = \sigma_f(\phi - \phi_{\text{crit}})^t$$

where σ_c is the electrical conductivity of the composite in S/cm, σ_f for the electrical conductivity of filler in S/cm, ϕ for the fiber volume fraction, ϕ_{crit} for the percolation threshold, and t for the critical exponent.

The critical exponents of 2.61 and 2.49 for the in-plane and the out-of-plane directions, respectively, were calculated by the model with an assumption of the percolation threshold of 7.3%.

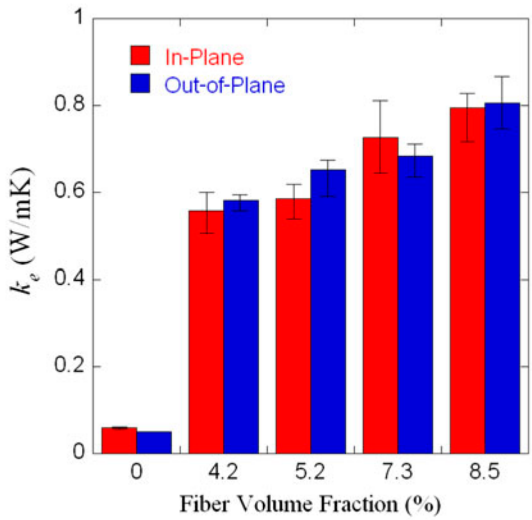

Figure 8. Thermal conductivities of the carbon fiber/EPP composite through the directions of in-plane and out-of-plane.

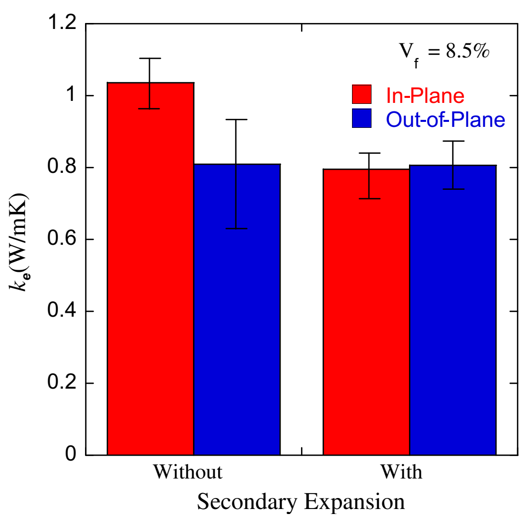

Figure 9. Thermal conductivity of carbon fiber/EPP composite ($V_f = 8.5\%$) with and without existence of secondary expansion.

Thermal conductivity

The mechanisms of electrical and thermal conduction in the composite are different qualitatively.[15] The thermal conductivity is attributed to the phonon conduction. By adding carbon fibers in the composites, the thermal conductivities were enhanced generally. As shown in Figure 8, conductivity values along each plane were similar to each other. The thermal conductivity of the composite containing a fiber volume fraction of 4.2% was increased by 9.4 times in the in-plane direction compared to that of the neat EPP. In the out-of-plane direction, the thermal conductivity was increased by 11.5 times. By increasing the volume amount of carbon fibers in the composites, the thermal conductivities were increased accordingly. At a fiber volume fraction of 8.5%, the thermal conductivities were enhanced by 13.3 times and 15.9 times along the in-plane and

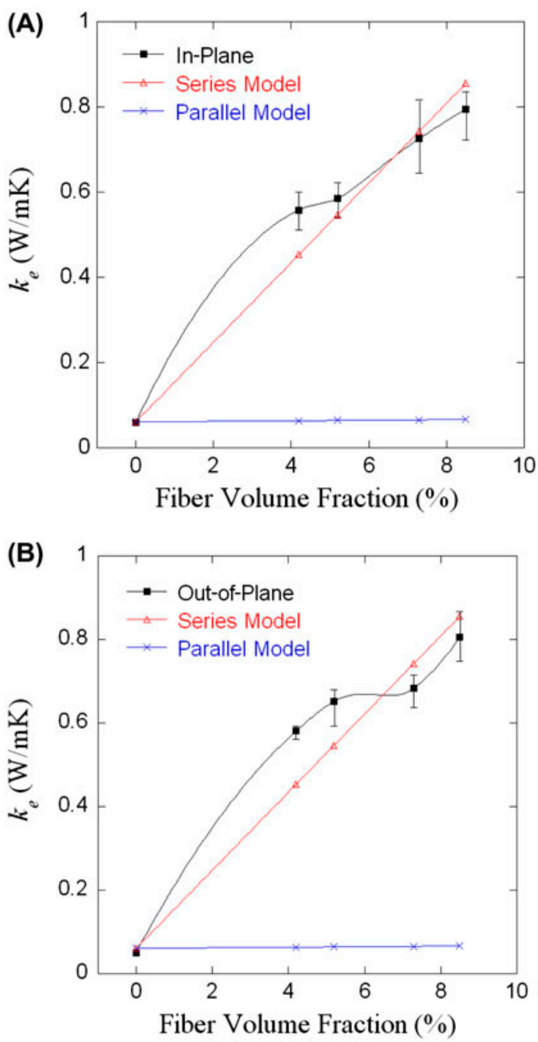

Figure 10. Comparison of the experimental and theoretical values for the thermal conductivity; (A) thermal conductivity of the in-plane, (B) thermal conductivity of the out-of-plane.

out-of-plane directions, respectively. Consequently, the thermal conductivities of the carbon fiber/EPP composites were enhanced by about 16 times using 8.5 vol.% of the carbon fibers and the isotropy of the thermal conductivity was realized within the deviation range, though the slight differences of thermal conductivities between the in-plane and the out-of-plane directions are shown.

However, without secondary expansion of EPP, the sample with a fiber volume fraction of 8.5% shows thermal conductivities of 1.04 and 0.81 W/mK for the in-plane and out-of-plane directions, respectively (see Figure 9).

Unlike the electrical conductivities which are attributed to the percolation mechanism, the thermal conductivities of the composite are similar between in-plane and out-of-plane directions because of high thermal contact resistances between the fiber and the polymer and between the fibers. Therefore, the increase of the thermal conductivities is more difficult, and the variation of data may become smaller. For these reasons, the fiber volume fraction of the sample can be considered as a main factor of the thermal conductivity of the composite as shown below equations.

The above thermal conductivity results were compared with the theoretical values obtained using the series model and the parallel model in Figure 10. The theoretical values from the series model and parallel model were estimated using the following equations [27]:

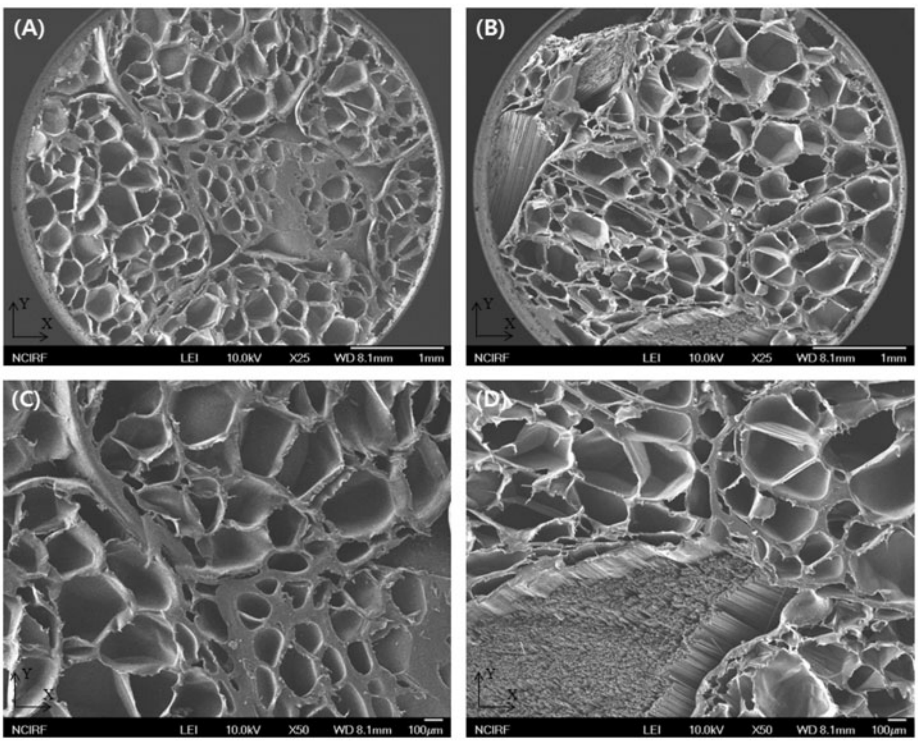

Figure 11. The surface morphologies of the cross section of the EPP and the carbon fiber/EPP composite; (A) EPP, (B) carbon fiber/EPP composite, (C) magnification of (A), (D) magnification of (B).

$$k_e = (1 - \phi)k_m + \phi\, k_f; \quad \text{Series model}$$

$$\frac{1}{k_e} = \frac{(1 - \phi)}{k_m} + \frac{\phi}{k_f}; \quad \text{Parallel model}$$

where k_e is the thermal conductivity of the composite in W/mK, ϕ for the fiber volume fraction, k_m for the thermal conductivity of the matrix in W/mK, and k_f for the thermal conductivity of the fiber in W/mK.

The measured values of the three-dimensional thermal conductivity were close to the theoretical values from the series model as can be seen in Figure 10.

Surface morphology

Numerous cells are observed on the cross-section of the neat EPP as shown in Figure 11(A). Low thermal conductivity of the neat EPP is attributed to the insulation effect by cell walls. By utilizing this characteristic, the carbon fiber could be oriented along the gaps between the bead walls along the out-of-plane. The cross-sectional view (in-plane) of the carbon fiber/EPP composite with a fiber volume fraction of 8.5% shows oriented carbon fibers along the gaps between the beads as shown in Figure 11(B). This could be interpreted as a reason for the increased electrical conductivity in the thickness direction of the specimen (out-of-plane).

Conclusion

The manufacturing process of carbon fiber/EPP composites was introduced and their quasi-isotropic electrical and thermal conductivities were observed. A random mixture of EPP having a spherical shape and carbon fibers having a cylindrical rod shape could make the carbon fibers positioned along the gap between the expanding EPP beads. Therefore, the secondary expansion which occurs during the fusion-bonding process of EPP could be an effective way to realize the isotropic conductivities. Through the addition of carbon fibers, the average volume electrical conductivity could be increased to 0.0062 and 0.0106 S/cm for the in-plane and out-of-plane directions, respectively, and the thermal conductivity was enhanced isotropically.

Funding

This work was supported by the Basic Science Research Program through the National Research Foundation (NRF) of Korea funded by the Ministry of Science, ICT, and Future Planning, Korea under [grant number NRF-2012R1A1A1044253].

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