Journal of Southeast University (English Edition) Vol. 29, No. 2, pp. 158 − 161 June 2013 ISSN 1003—7985

Effective thermal and electrical conductivity of graphite nanoplatelet composites

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Abstract: The relationship between the thermal/electrical conductivity enhancement in graphite nanoplatelets (GNPs) composites and the properties of filling graphite nanoplatelets is studied. The effective thermal and electrical conductivity enhancements of GNP-oil nanofluids and GNP-polyimide composites are measured. By taking into account the particle shape, the volume fraction, the thermal conductivity of filling particles and the base fluids, the thermal and electrical conductivity enhancements of GNP nanofluids are theoretically predicted by the generalized effective medium theory. Both the nonlinear dependence of effective thermal conductivity on the GNP volume fraction in nanofluids and the very low percolation threshold for GNP-polyimide composites are well predicted. The theoretical predications are found to be in reasonably good agreement with the experimental data. The generalized effective medium theory can be used for predicting the thermal and electrical properties of GNP composites and it is still available for most of the thermal/electrical modifications in two-phase composites.

Key words: graphite nanoplatelet; nanofluids; thermal conductivity; electrical conductivity; percolation threshold
doi: 10.3969/j. issn. 1003 − 7985. 2013. 02. 009

Recently, there has been an increasing interest in graphite nanoplatelet composites because of their unique electrical, thermal and optical properties[1-6]. The thermal conductivity of large enough graphite nanoplatelets (GNPs) should be higher than that of bulk graphite[7]. Recent experimental studies have also shown that the thermal conductivity of few-layer GNPs is of a similar aspect ratio to that of single-wall nanotubes (SWNTs) but with twice the increase in the thermal conductivity when embedded in epoxy composites[8]. Moreover, the nonlinear dependence of the effective thermal conductivity on the volume fraction of GNPs has been reported[7]. In the case of the electrical conductivity, very low percolation thresholds in carbon nanotube composites have also been reported[9-10]. Surprisingly, the thermal transport measurements on GNP-oil nanofluids bear no signature of the percolation threshold. The contrasting behavior should be carefully examined since both the thermal and electrical transport processes are described by the same continuum equation[11-12]. The relationship between the thermal/electrical conductivity enhancement and the properties of filling GNP particles requires quantitative study of the thermal/electrical transfer processes in GNP composites.

1 Experiment

In this paper, we study the thermal conductivity enhancement of GNP-oil nanofluids and the low percolation threshold of GNP-polyimide composites. Considering the shape and the volume fraction of graphite nanoplatelet particles, we would like to generalize the effective medium theory to investigate the effective thermal and electrical properties of GNP composites. Our theoretical prediction on the effective thermal and electric conductivity of GNP composites is in good agreement with the experimental results.

Graphite nanoplatelet particles are grown on substrates by the ultrasonic spry pyrolysis method and under the typical controlled exfoliation and dispersion process[8, 13]. In order to investigate the effective thermal conductivity enhancement of GNP-oil nanofluids, the GNP particles of 0.05%, 0.1%, 0.2%, 0.4%, 0.6%, 1.0% in volume fraction are chosen. Then the chosen amount of GNP particles are filled into the base oil liquid and operated under a high rotation speed for about 100 min to ensure a good dispersion of additional filling GNP particles in oil. The thermal conductivity of GNP-oil nanofluids is measured by a thermal testing device (ZKY-BRDR). The effective electric conductivity enhancement of GNP-polyimide composites is measured by a resistivity test fixture (Keithley8009) and an electrometer (Keithley6517).

Received 2012-11-29.
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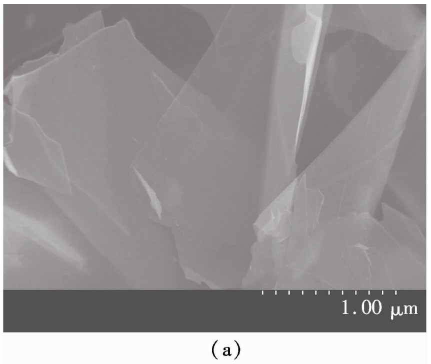
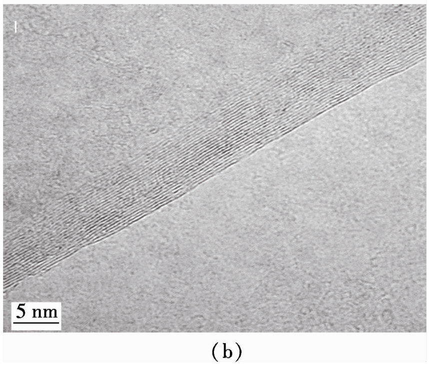
Foundation items: The National Natural Science Foundation of China (No. 50906073, 31070517), China Postdoctoral Science Foundation (No. 20110491332), Jiangsu Planned Projects for Postdoctoral Research Funds (No. 1101009B), the Science and Technology Development Plan of North Jiangsu (No. BC2012444).
Citation: Zhou Xiaofeng, Zhang Xiaosong, Zhou Jiancheng. Effective thermal and electrical conductivity of graphite nanoplatelet composites [J]. Journal of Southeast University (English Edition), 2013, 29(2): 158 − 161. [doi: 10.3969/j. issn. 1003 − 7985. 2013. 02. 009]

2 Results and Discussion

To observe details of the as-prepared graphite nanoplatelet particles, a scanning electron microscope (SEM) and high resolution transmission electron microscopy (TEM) are employed. The results are shown in Fig. 1. From Figs. 1(a) and (b), it can be seen that the average lateral dimensions and the average thicknesses of GNPs are 0.5 to 3 μm and 10 to 20 nm, respectively. The SEM and TEM images show that the as-prepared products consist of nanoplatelets with irregular shape and thickness distribution.



(a)



(b)

Fig. 1 Micrographs of GNPs. (a) SEM; (b) TEM

In the course of understanding the transport behavior of the GNP mixture, we would like to generalize the effective medium theory[14]. We consider a graphite nanoplatelet composite in which the graphite nanoplatelet particles with the volume fraction f and matrix particles with conductivity K_m are randomly mixed. For simplicity, we assume that matrix particles are spherical and graphite nanoplatelet particles are spheroidal in shape with the half radii a, b, c, and $b = c$. Since graphite nanoplatelet particles are randomly oriented, the effective conductivity K_e is isotropic[15-16]. For such a composite, the effective medium theory gives[17] that

$$f\sum_{j=x,y,z}\frac{K_e - K_{c,j}}{K_e + L_j(K_{c,j} - K_e)} + 9(1-f)\frac{K_e - K_m}{2K_e + K_m} = 0 \tag{1}$$

where $K_{c,j}$ is the equivalent thermal (or electrical) conductivity along the j-axis, and the depolarization factor L_j depends on the GNP aspect ratio $P = a/c$, which is expressed as

$$L_z = \begin{cases} \dfrac{1}{2p^3}\left(-2p + e\ln\dfrac{e-p}{e+p}\right) & e<1 \\ \dfrac{1}{2q^3}\left(2q - e\pi + 2e\arctan\dfrac{e}{q}\right) & e>1 \end{cases} \tag{2}$$

For the thermal transportation, due to the large interfacial thermal resistance, graphite nanoplatelet particles are physically anisotropic. In order to take such an effect into account, one often assumes that spheroidal particles are coated with a layer of material with thickness d and conductivity K_s. The interfacial thermal resistance is concentrated on a surface of zero thickness, which is defined as

$$R_{Bd} = \lim_{\delta\to 0,\, K_s\to 0}(\delta/K_s)$$

Hence, one has $K_{c,j} = K_p/(1 + QR_{Bd}L_jK_p)$ with $Q = (2a + c)/(ac)$. As a result, Eq. (1) is simplified as

$$f\left[\frac{K_e - K_p}{K_e + L_z(K_p - K_e)} + 4\frac{K_e - K_p}{2K_e + (1-L_z)(K_p - K_e)}\right] + 9(1-f)\frac{K_e - K_m}{2K_e + K_m} = 0 \tag{3}$$

For the electrical transport, the interfacial electrical resistance is so small that it can be ignored, i.e., $R_{Bd} = 0$. Therefore, we calculate the effective electrical conductivity by substituting $K_{c,j} = K_p$ into Eq. (3). Note that Eq. (3) can predict the non-zero percolation threshold.

Fig. 2 shows a comparison between Eq. (3) and our measured effective thermal conductivity of GNP-oil nanofluids. In the calculation, the thermal conductivities of the oil and the GNPs are taken as 0.144 8 and 1 000 W/mK, respectively[8], and $R_{Bd} = 8\times10^{-8}$ m^2 · K/W. In addition, the depolarization factor tends to be zero for graphite nanoplatelet particles with a large aspect ratio. Fig. 2 shows the thermal conductivity enhancement as a function of the filler loading. The enhancement (as high as 50%) in the thermal conductivity is observed for the GNP-oil nanofluids with a volume fraction of only 1% GNPs. At the same time, our theoretical results are found

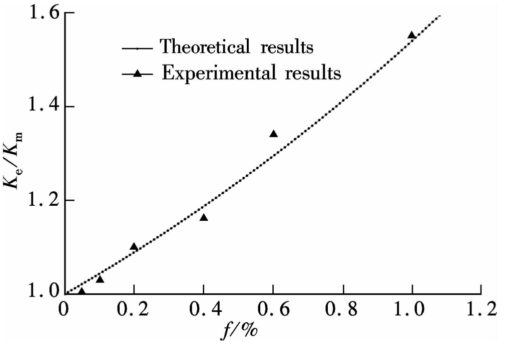


Fig. 2 Effective thermal conductivity enhancement of GNP-oil nanofluids compared with the theoretical results

to be in reasonably good agreement with the measured experimental data.

Fig. 3 shows the enhancement of the effective electrical conductivity vs. the volume fraction in GNP-polyimide composites. We can see that the theoretical results are in good agreement with the measured experimental data. For GNPs in polyimide composites, the depolarization factor is estimated as 0.000 07; hence, $f_c = 0.0004$. Such a percolation value is of the same order as the data reported in Ref. [11]. When the experimental data is compared with our theoretical results, we find that a very low percolation threshold for GNP-polyimide composites is well predicted.

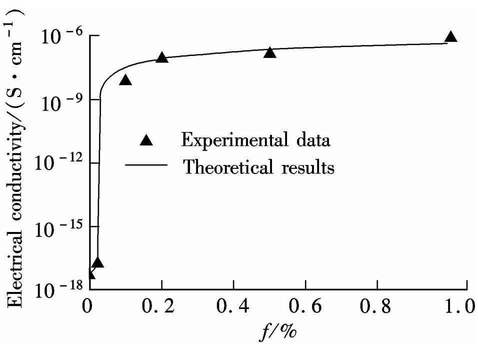


Fig. 3 Effective electrical conductivity vs. volume fraction in GNP-polyimide composites

3 Conclusion

The effective thermal and electrical conductivity enhancements of GNP-oil nanofluids and GNP-polyimide composites are measured. As high as 50% enhancement in the thermal conductivity is observed for GNP-oil nanofluids with a volume fraction of only 1%. For the GNP-polyimide composites, the non-zero percolation threshold is well investigated. Both the nonlinear dependence of the effective thermal conductivity on the GNP volume fraction in nanofluids and the very low percolation threshold for GNP-polyimide composites are well predicted. Our theoretical predications are in good agreement with the experimental data. Our model can be applied for predicting the thermal and electrical properties of GNP composites, which is still available for most of the thermal/electrical modification in two-phase composites.

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石墨纳米片复合物的有效热导率和电导率

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摘要:研究了石墨纳米片复合物的电、热传输性质的增强与添加的石墨纳米片热导率、电导率之间的关系.测量了石墨纳米片/油纳米流体、石墨纳米片/聚酰亚胺复合物的有效热导率和电导率增强.通过考虑颗粒形状、体积分数、添加颗粒的热导率和基质性质,依据发展的有效媒质理论,理论预测了石墨片纳米流体的电导率、热导率增强.解释了石墨纳米片的添加量和增强之间的非线性关系,同时还阐明了小体积份数下石墨纳米片复合物的渗流阈值.理论计算结果与实验结果吻合.发展的有效媒质理论不仅适用于预测石墨纳米片复合物的电导率、热导率性质,而且还适用于所有两相复合体系的电导率、热导率改性.

关键词:石墨纳米片;纳米流体;热导率;电导率;渗流阈值

中图分类号:O472.2