On the Effect of Graphene Nanoplatelets on Water–Graphene Nanofluid Thermal Conductivity, Viscosity, and Heat Transfer Under Laminar External Flow Conditions

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Experiments were conducted with graphene nanoplatelets (GNP) to investigate the relative benefit of the thermal conductivity increase in relationship to the potential detriment of increased viscosity. The maximum enhancement ratio for GNP nanofluid thermal conductivity over water was determined to be 1.43 at a volume fraction of 0.014. Based on GNP aspect ratios, the differential effective medium model is shown to describe the experimental results of this study when using a fitted interfacial resistance value of 6×10^{-8} m^2 *K* W^{-1}. *The viscosity model of Einstein provided close agreement between measured and predicted values when the effects of temperature were included and the intrinsic viscosity model term was adjusted to a value of 2151 representative for GNP. Heat transfer in external flows in laminar regime is predicted to decrease for GNP nanofluids when compared to water alone.* [DOI: 10.1115/1.4038835]

Introduction

Graphene is a monolayer of graphite with reported thermal conductivities in the range of 2000 W m^{-1} K^{-1} to 5350 W m^{-1} K^{-1} [1–3], and a graphene–water nanofluid is expected to have a greater thermal conductivity than water alone. The magnitude of overall convective heat transfer when using graphene nanoplatelets (GNP) nanofluids is a function of the thermal conductivity as well as the viscosity of the nanofluid. There are no explicit studies in the literature for GNP nanofluids for external flow configurations under laminar flow regimes over flat plates, which is of interest in applications of film cooling of surfaces.

Description of the Thermal Conductivity Prediction Model. A differential effective medium model was presented by Chu et al. [1]

$$9(1-f)\frac{K_e-K_m}{2K_e+K_m}=f\left[\frac{2K_x}{K_e\left(\frac{2R_kK_x}{L}+1\right)}-\frac{K_e\left(\frac{2R_kK_z}{t}+1\right)}{K_z}-1\right] \quad (1)$$

where K_e is the effective thermal conductivity of the nanofluid, K_x is the thermal conductivity along the transverse x-axis, K_z is the thermal conductivity along the longitudinal z-axis, K_m is the thermal conductivity of the base fluid, and R_k is the interfacial thermal resistance. Differential effective medium models have been shown to be in good agreement with nanofluid thermal conductivity experimental data [4]. Chu et al. [1] showed the importance of the interfacial thermal resistance yet stated that it is theoretically uncertain and its value ranges between 3 and 9×10^{-8} m^2 K W^{-1}. Also, Murshed et al. [5] assumed an interfacial thermal conductivity to be 1.25 to 3 times that of the base fluid because of their uncertainty about the exact thermal conductivity of the interfacial layer. f is the volumetric fractional composition of the graphene in solution. This model was chosen, because unlike other models, it accounts for the interfacial resistance and also takes into account the effects of morphology of the dispersed particles by its inclusion of dimension L (length of the particles) and t (thickness of the particles).

Model of Viscosity

The Einstein [6] model for prediction of viscosity as modified by Anoop et al. [7] as the power series model is used in this study for prediction of nanofluid viscosity

$$\text{relative viscosity}=\frac{v_{nf}}{v_b}=1+[\eta]f+\mathrm{K}[\eta]^2f^2+Of^3+\cdots \quad (2)$$

The intrinsic viscosity constant of the nanofluid is $[\eta]$, and the Huggins' constant is K. The volumetric fraction, f, accounts for disturbances in flow due the presence of solid particles. Anoop et al. [7] further describe that these disturbances in flow give rise to an increase in dissipations energy, and f^2 accounts for the effect of pair interactions between suspended particles. The intrinsic viscosity constant value is determined by geometry of the suspended particles. For spheres, $[\eta]=2.5$ and with simplifications neglecting higher order terms; the model becomes

$$\text{relative viscosity}=\frac{v_{nf}}{v_b}=1+2.5f \quad (3)$$

The Influence of Thermal Conductivity and Viscosity on Heat Transfer. The configuration of interest for this study is the laminar external flow of GNP nanofluids over a flat plate. Heat transfer through the nanofluid is predominantly due to convection

$$\dot{Q}=\bar{h}A\Delta T \quad (4)$$

where $\bar{h}$ is the coefficient of heat transfer and ΔT is the change in temperature.

The convection heat transfer coefficient can be obtained from the dimensionless Nusselt number term as shown in the following equation:

$$\bar{h}=\frac{K}{D}\mathrm{Nu} \quad (5)$$

where D is the length of the flat plate and Nu is the well-known Nusselt number. Hence, with substitution, Eq. (5) becomes

$$\dot{Q}=\left(\frac{K}{D}\mathrm{Nu}\right)A\Delta T \quad (6)$$

For laminar flow of a fluid over a flat plate, Nusselt number can be predicted based as shown in Eq. (7) for $\mathrm{Pr}>0.6$

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$$\mathrm{Nu} = 0.664\mathrm{Re}^{0.5}\mathrm{Pr}^{\frac{1}{3}} \quad (7)$$

where Re is the Reynolds number and Pr is the Prandtl number

$$\mathrm{Re} = \frac{VD}{\nu} \quad (8)$$

where V is the velocity of flow of the fluid over the flat plate and ν is the kinematic viscosity of the fluid. The Prandtl number is

$$\mathrm{Pr} = \frac{C_p \mu}{K} \quad (9)$$

where C_p is the heat capacity of the fluid, K is the thermal conductivity, and μ is the absolute viscosity of the fluid. Consequently, a proportionality relation for heat transfer is derived for water as follows:

$$\dot{Q}_w \propto \frac{K_w^{\frac{2}{3}} C_{pw}^{\frac{1}{3}} \rho_w^{\frac{1}{3}}}{\nu_w^{0.167}} \quad (10)$$

where $\dot{Q}_w$ is the water heat transfer, K_w is the water thermal conductivity,

$C_{p,w}$ is the water heat capacity, ρ_w is water density, and ν_w is the water kinematic viscosity.

The analogous expression for a heat transfer in a nanofluid (nf) is

$$\dot{Q}_{\mathrm{nf}} \propto \frac{K_{\mathrm{nf}}^{\frac{2}{3}} C_{p,\mathrm{nf}}^{\frac{1}{3}} \rho_{\mathrm{nf}}^{\frac{1}{3}}}{\nu_{\mathrm{nf}}^{0.167}} \quad (11)$$

Finally, the expected relative ratio of nanofluid heat transfer to water based on differences in properties associated with differing thermal conductivities and viscosities becomes

$$\text{relative heat transfer} = \frac{\dot{Q}_{\mathrm{nf}}}{\dot{Q}_w} = \left(\frac{K_{\mathrm{nf}}}{K_w}\right)^{\frac{2}{3}} \left(\frac{C_{p,\mathrm{nf}}}{C_{p,w}}\right)^{\frac{1}{3}} \left(\frac{\rho_{\mathrm{nf}}}{\rho_w}\right)^{\frac{1}{3}} \left(\frac{\nu_w}{\nu_{\mathrm{nf}}}\right)^{\frac{1}{6}} \quad (12)$$

This formulation requires the heat capacity of the nanofluid, which can be estimated from the model as described by O'Hanley et al. [8] as shown in the following equation:

$$C_{p,\mathrm{nf}} = \frac{f \rho_n C_{p,n} + (1-f)\rho_w C_{p,w}}{f \rho_n + (1-f)\rho_w} \quad (13)$$

where $C_{p,\mathrm{nf}}$ is nanofluid heat capacity, $C_{p,n}$ is nanoplatelet heat capacity (2.1 J g^{-1} K^{-1}), and ρ_n is the density of the nanoplatelet (300 mg mL^{-1}).

Experimental Methods

Nanofluids for thermal conductivity measurement samples were prepared with 0.42 g of gelatin dissolved in 15 mL of de-ionized water at 99 °C. 15 mL of cold (10 °C) additional de-ionized water was then added as well as a known mass (0.1 g to 1.0 g) of carbon GNP (CAS 7782-42-5). The suspension was sonicated for 1 h. Nanofluids of high volume fraction were used and gelatin prevented sedimentation during thermal conductivity measurements. Nanofluid samples for viscosity and sizing were prepared in similar manner as the thermal conductivity samples but without gelatin. Based on manufacturer's data, the graphene used has a nominal size of 5 μm in diameter and 3 nm thickness. Scanning electron microscope measurements for diameter and thickness and dynamic light scattering measurements for diameter showed that platelet dimensions were monodisperse and unchanged after the sonication; thus, the manufacturer's recommended dimensions were adopted and used in model calculations. A hot wire instrument was used to measure the thermal conductivity. A temperature-controlled rheometer fitted with an ultralow-viscosity adapter was used to measure the viscosity. Temperatures tested were 277, 298, and 333 K. The rotational speed used was 60 rpm with a shear rate of 29 s^{-1}.

Results and Discussion

Shown in Fig. 1 are the relative thermal conductivity data obtained in this study for various GNP volume fractions. Thermal conductivity data from other studies are limited to relative low GNP volume fractions of less than 0.002 [9] with the exception of results for functionalized GNP from Kole and Dey [10] who studied GNP volume fractions as high as 0.0047, and these values included for comparison in Fig. 1 show close agreement with the results of this study.

The maximum relative thermal conductivity found was 1.47 at a volume faction of 0.014 and represents the largest reported GNP nanofluid thermal conductivity enhancement known to the authors. Shown in Fig. 2 are how the experimental thermal conductivity enhancement data compare with the predicted thermal conductivity enhancement from Eq. (1). For the predicted thermal conductivity, the limits of R_k, as suggested by Chu et al. [1], from 3×10^{-8} to 9×10^{-8} m^2 K W^{-1} are shown. The experimental results obtained in this study are between these limits and an R_k value of 6×10^{-8} m^2 K W^{-1} was fitted such that the experimental results matched the predicted thermal conductivity enhancement. GNP-reported thermal conductivities are in the range of 2000 W m^{-1} K^{-1} to 5350 W m^{-1} K^{-1} [1–3]. Within this range, the calculated relative thermal conductivity values in Fig. 2 are essentially independent of the graphene thermal conductivity with the system dominated by the interfacial thermal resistance term (R_k) for the particle aspect ratios used. The approach here used to determine R_k differs from other studies [10,11] where R_k was not accounted for and instead a much lower effective GNP

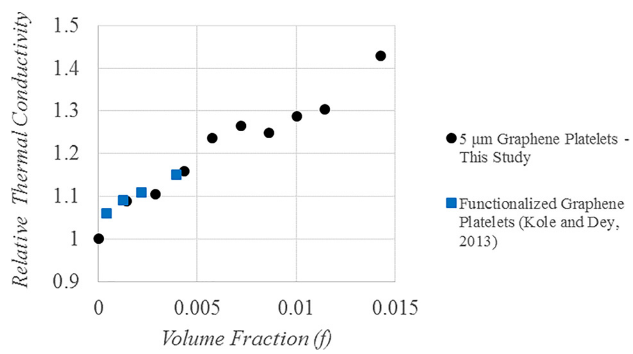

Fig. 1 Relative thermal conductivity for differing GNP volume faction

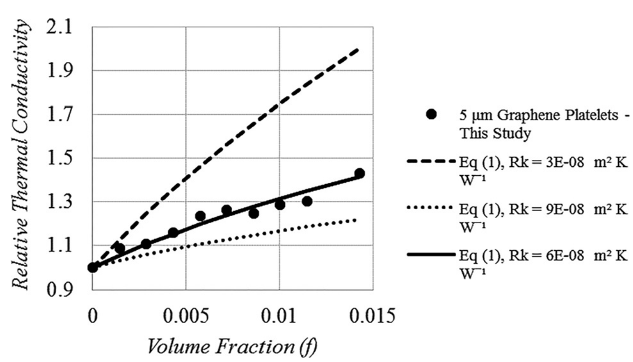

Fig. 2 Experimental thermal conductivity enhancement compared with the thermal conductivity enhancement predicted from the differential effective medium model presented by Chu et al. [1] for different R_k

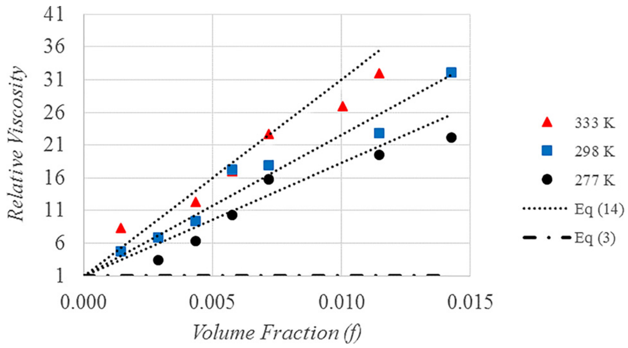

Fig. 3 Experimental relative viscosity compared to those predicted by Eqs. (3) and (14) for differing temperatures and GNP volume fraction

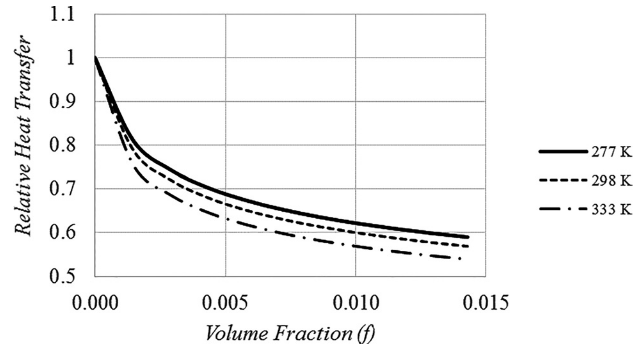

Fig. 4 Predicted heat transfer through a nanofluid with GNP volumetric fraction

thermal conductivity, on the order of 10 W m^{-1} K^{-1}, was used to predict thermal conductivity data.

Displayed in Fig. 3 are experimental relative viscosity data at 277 K, 298 K, and 333 K. The relative viscosity increases in an approximate linear relation with the volumetric fraction and also increases with temperature. The Einstein [6] classical model underpredicts the experimental data. A similar observation was made by Anoop et al. [7] who attributed particle agglomeration the cause for observed viscosity increases beyond conventional theoretical predictions and noted that the intrinsic viscosity is shape dependent. An intrinsic value has not been found for platelets. Nevertheless, Douglas and Garboczi [12] suggest values as high as 204.9 for oblate spheroids for aspect ratio of 300. In this study, GNP aspect ratio was nearly 17,000 and based on a sum of least squared difference analysis, a shape-dependent value of 2151 for all the experimental data can be seen to closely match the experimental data when an empirical temperature relationship is incorporated as shown in Eq. (14). Where T is nanofluid temperature in degrees Kelvin

$$\text{relative viscosity} = \frac{v_{\text{nf}}}{v_b} = 1 + 2151 f \frac{T}{298}^{3} \quad (14)$$

The temperature effects upon relative viscosity have been reported by others studying GNP nanofluids [9,13] but theoretical formulations for this temperature dependence are lacking. Relative viscosity predictions without temperature effects neglect the higher order effects of particle–particle interactions. It is plausible that observed nanofluid temperature dependence may be attributable to ignoring these particle–particle interactions that occur to greater extent in higher temperature solutions. Regardless, it should be noted that the viscosity enhancement found for dispersion of GNP in water in this study is much greater than any other GNP nanofluid. A viscosity increase of over 30 times at 0.015 GNP volume fraction is found in this study. This high viscosity of the GNP nanofluid is attributed to the high aspect ratio shape of these nanoparticles, the high volume fractions used, and the relatively low viscosity of the water base fluid.

Application of Results

Given that both nanofluid thermal conductivity and viscosity play important roles in determining nanofluid heat transfer, the relative benefit of GNP nanofluids for heat transfer applications can be considered by using the proportionality relationship of Eq. (12). Literature values for thermal conductivity, viscosity, and specific heat capacity were used for water. Equation (14) was used for relative viscosity. Equation (13) was used for GNP nanofluid heat capacity and Eq. (1) with $R_k = 6 \times 10^{-8}$ m^2 K W^{-1} was used for relative thermal conductivity. Figure 4 shows change in heat transfer through a nanofluid with GNP volumetric fraction relative to the base fluid. The downward trend shows that heat transfer with GNP dispersed in water decreases with increasing volumetric fraction of the nanoparticles in the base fluid at all temperatures. This effect is attributed to detrimental viscosity effects dominating the process more so than the beneficial enhancements in thermal conductivities for GNP nanofluids. Few studies have directly measured heat transfer rates for GNP under laminar flow conditions but our results agree with the findings of Tharayil et al. [14] who studied the heat transfer performance of miniature loop heat pipe with graphene–water nanofluid experimentally. They found heat transfer performance to decrease at GNP concentrations above 0.002 volume fraction, although they attributed this effect to graphene particles sticking to the evaporator and not to detrimental viscosity effects.

Conclusions

A maximum enhancement ratio for GNP nanofluid over water was 1.43 at a volume fraction of 0.014. However, adding GNP to water also increases solution viscosities with a viscosity enhancement ratio of 33.1 for GNP nanofluids at a volume fraction of 0.014.

The differential effective media model presented by Chu et al. [1] describes the experimental results of this study using a fitted R_k value of 6×10^{-8} m^2 K W^{-1}. This value falls within the range suggested in the literature as reasonable for R_k and is the first experimental measurement obtained for R_k of GNP known to the authors. Adjusting the intrinsic viscosity term to a fitted value of 2151 representative for the GNP provided much close agreement between measured and predicted viscosity values when an empirical temperature correction was applied to the data.

In this study, the highest enhancement of thermal conductivity is less than the highest corresponding enhancement of viscosity at the corresponding volume fraction. Heat transfer is a nonlinear function of these parameters and the magnitude of heat transfer is predicted to decrease for GNP nanofluids when compared to water alone for applications of external laminar flow over flat plates. The use of nanofluids to enhance heat transfer processes, as proposed by other investigators studying thermal conductivity enhancements alone, appears not to be a viable approach for external laminar flow over flat plate configurations when detrimental effects associated with viscosity increases are considered as in this study.

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