

## Enhanced thermal conductivity and viscosity of copper nanoparticles in ethylene glycol nanofluid

J. Garg,<sup>1,a)</sup> B. Poudel,<sup>2</sup> M. Chiesa,<sup>1</sup> J. B. Gordon,<sup>1</sup> J. J. Ma,<sup>1</sup> J. B. Wang,<sup>4</sup> Z. F. Ren,<sup>2</sup> Y. T. Kang,<sup>5</sup> H. Ohtani,<sup>3</sup> J. Nanda,<sup>3</sup> G. H. McKinley,<sup>1</sup> and G. Chen<sup>1</sup>

<sup>1</sup>*Department of Mechanical Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA*

<sup>2</sup>*Department of Physics, Boston College, Chestnut Hill, Massachusetts 02467, USA*

<sup>3</sup>*Materials and Nanotechnology Department, Research and Advanced Engineering, Ford Motor Company, Dearborn, Michigan 48121, USA*

<sup>4</sup>*School of Environmental Science and Engineering, Huazhong University of Science and Technology, People's Republic of China*

<sup>5</sup>*School of Mechanical and Industrial Engineering, Kyung Hee University, South Korea*

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This study investigates the thermal conductivity and viscosity of copper nanoparticles in ethylene glycol. The nanofluid was prepared by synthesizing copper nanoparticles using a chemical reduction method, with water as the solvent, and then dispersing them in ethylene glycol using a sonicator. Volume loadings of up to 2% were prepared. The measured increase in thermal conductivity was twice the value predicted by the Maxwell effective medium theory. The increase in viscosity was about four times of that predicted by the Einstein law of viscosity. Analytical calculations suggest that this nanofluid would not be beneficial as a coolant in heat exchangers without changing the tube diameter. However, increasing the tube diameter to exploit the increased thermal conductivity of the nanofluid can lead to better thermal performance. © 2008 American Institute of Physics. [DOI: 10.1063/1.2902483]

### I. INTRODUCTION

Properties that mainly determine the thermal performance of a liquid for heat transfer applications are the thermal conductivity, viscosity, specific heat, and density. Fluids such as air, water, ethylene glycol, and mineral oils are typically used as heat transfer media in applications such as power generation, chemical production, automobiles, computing processes, air conditioning, and refrigeration. However, their heat transfer capability is limited by their very low thermal conductivity. These fluids have almost two orders of magnitude lower thermal conductivity compared to metals, resulting in low heat removal efficiencies.

First attempts to improve the thermal conductivity involved dispersing micron-sized particles in these liquids. Ahuja<sup>1</sup> and Liu *et al.*<sup>2</sup> studied the heat transfer augmentation and rheology of the slurries. One of the drawbacks associated with the use of slurries is the abrasive action of the particles causing erosion of the components. Secondly, micron-sized particles tend to rapidly fall out of the suspension due to their large mass and, thus, can cause fouling of the components thereby clogging the flow path and increasing the pressure drop.

Decreasing the particle size to the nanometer range offers the potential to overcome these drawbacks. Masuda *et al.*<sup>3</sup> first reported the thermal conductivity and viscosity of liquids containing nanometer sized particles. Choi<sup>4</sup> also investigated the thermal conductivity enhancement of such colloidal suspensions and coined the term “nanofluids.” In addition to overcoming the drawbacks associated with the use of micron-sized suspensions, nanofluids have often exhibited

thermal conductivity enhancement substantially higher than that predicted by the Maxwell effective medium theory.<sup>5</sup> Over the past decade, a significant amount of data has been gathered on the thermal conductivity of nanofluids. Typical materials used for nanoparticles include metals such as copper, silver, and gold and metal oxides such as alumina, titania, and iron oxide. Carbon nanotubes have also been used to enhance the thermal conductivity of liquids. Experimental data on the thermal conductivity of nanofluids widely varies and mechanisms responsible for the thermal conductivity enhancement are under debate, as summarized in recent reviews.<sup>6,7</sup> Mechanisms proposed to explain thermal conductivity enhancement include Brownian motion of nanoparticles,<sup>8–10</sup> layering of fluid around nanoparticles,<sup>11,12</sup> near field radiative heat transfer,<sup>13</sup> and nanoparticle aggregation.<sup>14–16</sup> There is clearly a need to resolve the differences and confirm the repeatability of data obtained. This can be achieved by reporting the exact components of the nanofluid tested such as surfactants, particle material, base fluid, method of synthesis of nanoparticles, and any possible contamination with other agents. Also, the data obtained by different groups need to be verified for repeatability. Reaching a consensus in regards to the experimental data is critically important for an accurate physical model to be developed to explain the anomalous enhancement.

There is limited data available on the thermal conductivity enhancement of copper in ethylene glycol nanofluids. Eastman *et al.*<sup>17</sup> first reported an almost 40% improvement in thermal conductivity through the dispersion of 0.3 vol % Cu nanoparticles in ethylene glycol. They used direct condensation of metallic vapor into nanoparticles by contact with a flowing low vapor pressure liquid. Zhu *et al.*<sup>18</sup> produced cop-

<sup>a)</sup>Electronic mail: jivtेश@mit.edu.

per in ethylene glycol nanofluid by reducing a mixture of copper sulfate pentahydrate in ethylene glycol with sodium hypophosphite monohydrate. They used polyvinylpyrrolidone as a surfactant and reported an almost 9% increase in thermal conductivity with 0.1 vol % loading of copper nanoparticles in ethylene glycol. The thermal conductivity enhancement was reported for only 1 vol % loading and the effect of polyvinylpyrrolidone on the thermal conductivity of ethylene glycol was not reported. Liu *et al.*<sup>19</sup> synthesized copper nanoparticles in water using the chemical reduction method. Samples of volume fractions ranging from 0.05% to 0.2% of copper nanoparticles in water were prepared. They reported a maximum increase in thermal conductivity of water of about 23.8% with a volume fraction of 0.1 vol % copper nanoparticles in water. However, the reported thermal conductivity increase was found to be a strong function of time after sonication and decayed to nearly zero after about 10 min of sonication. It was not clear if the measured enhancement was due to the instability of the nanofluid. The instability of the nanofluid also affects the accuracy of the measurement and it was not clear what the accuracy of the thermal conductivity measurement was.

In this article, we present an approach to synthesize copper in ethylene glycol nanofluid using chemical synthesis. We measure both thermal and rheological properties of the nanofluid and discuss cases in which this nanofluid could be beneficial as a coolant. The experimental data show that the increase in thermal conductivity is almost twice of that predicted by the Maxwell effective medium model<sup>5</sup> and the increase in viscosity is almost four times of that predicted by the Einstein law of viscosity.<sup>20</sup> The measured enhancement in thermal conductivity was much lower than that reported by Eastman *et al.*<sup>17</sup>

## II. EXPERIMENT

In order to prepare the copper in ethylene glycol nanofluid, the copper nanoparticles were first synthesized using a chemical synthesis process and then dispersed in ethylene glycol.

In the present work, a chemical reduction method was used to synthesize copper nanoparticles with water as the solvent. The method used was similar to the one adopted by Liu<sup>19</sup> *et al.* except for the addition of sodium hydroxide as one of the reactants. In a typical synthesis, 2 g of copper acetate was dissolved in 50 ml of distilled water using a magnetic stirrer. Thereafter, 0.5 g of sodium hydroxide was added to the solution and dissolved. The solution was then cooled down to approximately 5 °C. At this stage, 3 ml of hydrazine was added, dropwise, to the solution while keeping the stirrer on. The solution was stirred at room temperature for 12 h to allow for the reduction reaction to complete. After this step, copper nanoparticles were obtained. To separate the particles from the solution, the reaction solution was centrifuged. The particles thus obtained were washed with acetone twice before being vacuum dried at 50 °C for 12 h to remove water. Removal of water from the particles is necessary as water has higher thermal conductivity than ethylene glycol and the presence of water can lead to erroneous

thermal conductivity data. The nanoparticles thus obtained were finally dispersed in ethylene glycol under an argon environment in a glovebox. To prepare the copper in ethylene glycol nanofluid of a certain volume fraction, the mass of the nanoparticles required was calculated using the bulk density of copper and added to ethylene glycol. The nanoparticles were dispersed in ethylene glycol using Sonics Vibra Cell VCX 750 sonicator. No significant sedimentation was observed in the nanofluid thus prepared for about a day.

Structural characterization on the as synthesized Cu nanoparticles to establish its structural phase and crystallinity was performed. Figure 1(a) shows the x-ray diffraction (XRD) of the copper nanoparticles showing characteristic peaks at  $2\theta$  of 43.2° and 50.4°, corresponding to the fcc phase of copper nanoparticles. Figure 1(b) shows a typical transmission electron micrograph (TEM) of a very diluted sample of the nanofluid. The TEM image of particles shown in Fig. 1(b) indicates an average primary particle size of about 200 nm. Particles with much smaller dimension can be seen as well. The Cu nanoparticles were highly crystalline as evident from the high magnification picture [Fig. 1(c)] showing atomically resolved planes of Cu and also from the corresponding selected area diffraction micrograph (inset).

## III. THERMAL CONDUCTIVITY MEASUREMENT

Thermal conductivity of the nanofluid was measured using the transient hot wire method. In order to measure the thermal conductivity of electrically conducting fluids, the method developed by Nagasaka and Nagashima<sup>21</sup> was used. The experimental setup consists of a thin platinum wire (25  $\mu\text{m}$ ) coated with an electrically insulating layer (1.5  $\mu\text{m}$  thick isonel layer). The wire is immersed in the nanofluid and a constant current is passed through it. The temperature rise of the wire is measured as a function of time. The temperature rise of the wire was measured for a time of 2 s. The thermal conductivity of the nanofluid can be calculated from the obtained data by using

$$k = \frac{Q}{4\pi L \frac{dT}{d \ln t}}. \quad (1)$$

Here,  $k$  is the thermal conductivity of the nanofluid,  $Q$  is the total power dissipated in the wire,  $L$  is the length of the wire,  $T$  is the wire temperature, and  $t$  is the time. The data between 0.1 and 1 s were used to calculate  $dT/d(\ln t)$  used in the thermal conductivity calculation. It was observed that by using the data between 0.1 and 1 s, the effects associated with the thermal capacitance of the wire and the natural convection heat transfer could be avoided. Typical temperature rise of the wire in de-ionized (DI) water is plotted in Fig. 2.

The constant current used in the measurement served two purposes; one to act as a heat source and second to enable the temperature measurement of the wire. In order to measure the temperature rise, the hot wire was made part of a Wheatstone bridge. Before starting the experiment, the bridge was balanced. During the experiment, the change in wire temperature caused a change in wire resistance leading to an imbalance in the bridge. The change in wire resistance

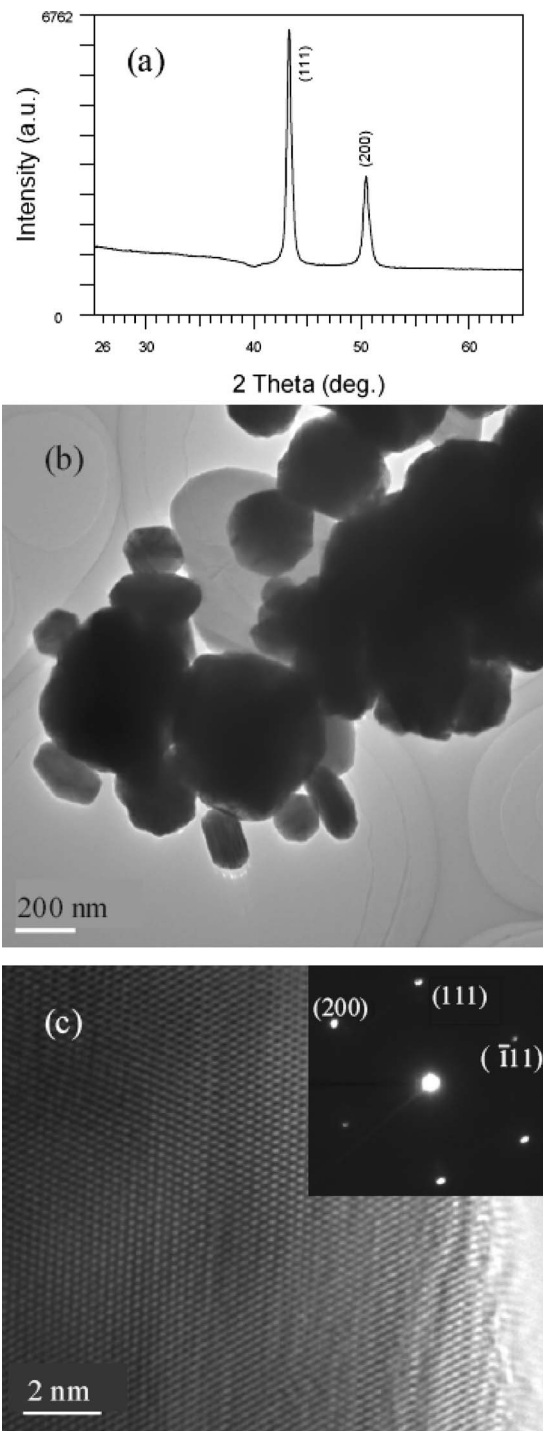


FIG. 1. (a) XRD, (b) TEM, and (c) high resolution TEM (HRTEM) images of Cu nanoparticles (HRTEM shows high crystallinity, the selected area electron diffraction in inset shows the Cu diffraction pattern).

was measured by measuring the voltage imbalance in the bridge. The temperature coefficient of resistance of the wire was accurately determined by measuring the resistance as a function of the temperature of the wire. The temperature rise of the wire was calculated from the change in wire resistance data and using the determined temperature coefficient of resistance. By measuring the slope of the temperature rise versus log of time curve and using Eq. (1), the thermal conductivity of the nanofluid was measured.

The experimental setup was calibrated by comparing the

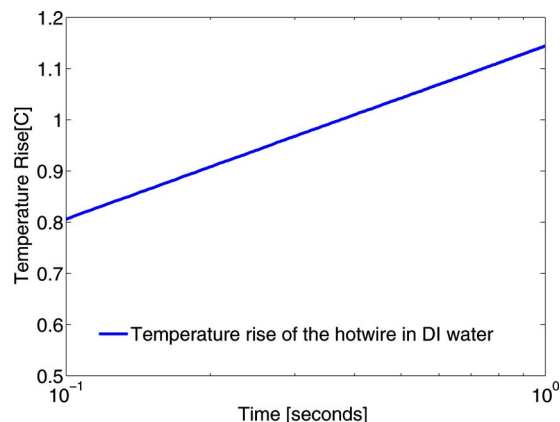


FIG. 2. (Color online) Experimental temperature rise of the hotwire in DI water vs time with time plotted on log scale.

measured value of thermal conductivity for ethylene glycol against the literature value. The literature value was reproduced with an uncertainty of 0.5%. The thermal conductivity for each volume fraction was measured 15 times over a period of about 30 min after sonication and the average of the 15 data points is reported in this article. The thermal conductivity measurement did not appreciably vary over the 30 min interval and the typical standard deviation involved was 0.2%.

It is known that ethylene glycol absorbs water when exposed to atmosphere. Since water has a higher thermal conductivity than ethylene glycol, any presence of water in ethylene glycol would lead to an increase in the effective thermal conductivity. Therefore, presence of water in ethylene glycol based nanofluids could lead one to erroneously conclude that the measured enhancement is due to the presence of nanoparticles. In order to ensure that measured enhancement was not due to the presence of water, several steps were taken. A sample of ethylene glycol was exposed to atmosphere and the thermal conductivity change was measured as a function of exposure time. It was found that the thermal conductivity increased by 2% after 12 days and 4% after 33 days of exposure to atmosphere.<sup>22</sup> The thermal conductivity of the copper in ethylene glycol nanofluid was measured in less than a day after dispersing the nanoparticles in ethylene glycol. Any exposure of the nanofluid to atmosphere during this period of one day would have resulted in water absorption that would have caused a change in thermal conductivity of much less than 2%. Thus, it was concluded that the measured change in thermal conductivity could not be due to the presence of water.

Figure 3 shows the thermal conductivity of the copper in ethylene glycol nanofluid for volume fractions ranging from 0.4% to 2%. The thermal conductivity ratio predicted by the Maxwell effective medium theory<sup>5</sup> is given by

$$\frac{k_{nf}}{k_f} = \frac{k_p + 2k_f - 2\phi(k_f - k_p)}{k_p + 2k_f + \phi(k_f - k_p)}. \quad (2)$$

Here,  $k_f$ ,  $k_p$ , and  $k_{nf}$  are the thermal conductivity of the base fluid, particle, and nanofluid, respectively, and  $\phi$  is the volume fraction of the nanoparticles. The above expression for effective thermal conductivity does not take into account the

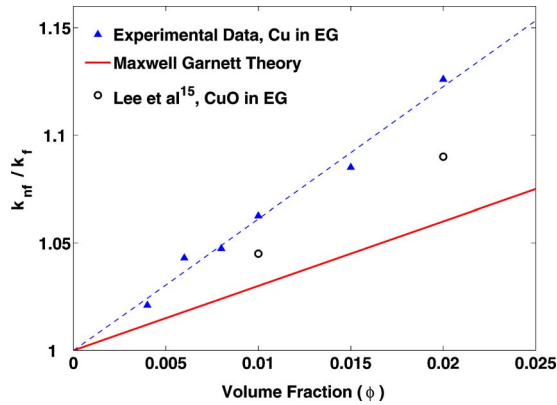


FIG. 3. (Color online) Effective thermal conductivity of copper in ethylene glycol nanofluid as a function of volume fraction. Dots are experimental data and dashed line is a linear fit to the experimental data. Solid line is based on effective medium theory.

thermal interface resistance between the particle and the fluid, the reduced thermal conductivity of the particles due to size effects, and is applicable only for spherical particles. Interfacial thermal resistance and particle size effects can be included while computing the effective thermal conductivity.<sup>23,24</sup> However, their inclusion will only reduce the predicted effective thermal conductivity. If the particle thermal conductivity is much higher than the base fluid thermal conductivity, then the ratio  $k_f/k_p$  can be approximated as zero and the above equation simplifies to

$$\frac{k_{nf}}{k_f} = \frac{1 + 2\phi}{1 - \phi} \approx 1 + 3\phi. \quad (3)$$

The thermal conductivity ratio prediction based on the above equation is also plotted in Fig. 3 and it can be seen that the relative increase in thermal conductivity was almost twice of that predicted by the Maxwell effective medium theory for all volume fractions studied. The experimental thermal conductivity ratio can be linearly fitted to

$$\frac{k_{nf}}{k_f} = 1 + C_k\phi \quad \text{where } C_k \approx 6. \quad (4)$$

The thermal conductivity ratio for 2 vol % fraction copper in ethylene glycol was measured to be about 1.12 and compares well with Lee *et al.*<sup>25</sup> who measured a ratio of 1.09 with a volume fraction of 2% CuO in ethylene glycol.

#### IV. VISCOSITY MEASUREMENT

The viscosity of the nanofluid was measured using a TA instruments AR-G2 rheometer equipped with a 6 cm 1° cone rheometer. The measured viscosity was compared against the Einstein law of viscosity<sup>20</sup> which predicts the effective viscosity of a fluid according to the following equation:

$$\frac{\mu_{nf}}{\mu_f} = 1 + 2.5\phi. \quad (5)$$

In the above equation,  $\mu_f$  and  $\mu_{nf}$  are the viscosity of the base fluid and the nanofluid, respectively. Both the measured and predicted viscosity ratios are plotted in Fig. 4. The experimental viscosity ratio can be linearly fitted to

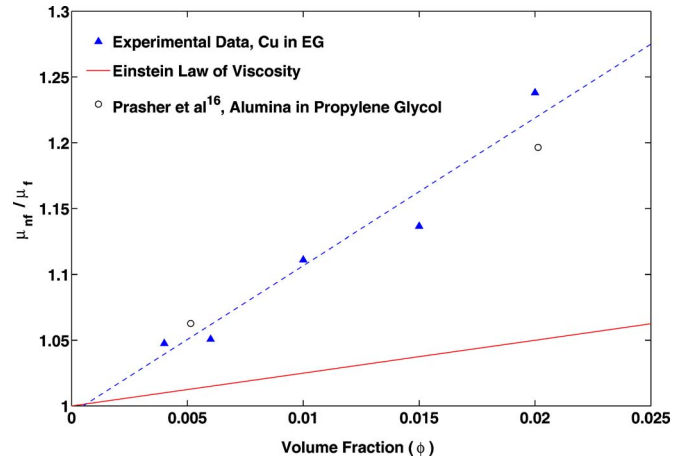


FIG. 4. (Color online) Viscosity of copper in ethylene glycol nanofluid. Dots are experimental data measured at a shear rate of  $3000 \text{ s}^{-1}$  and solid line is based on effective medium theory. Dashed line is a linear fit.

$$\frac{\mu_{nf}}{\mu_f} = 1 + C_\mu\phi \quad \text{where } C_\mu \approx 11. \quad (6)$$

The measured viscosity ratio compares well with the data obtained by Prasher *et al.*<sup>26</sup> who found that the experimental data matched well with the linear fit for a value of 10 for  $C_\mu$ . The viscosity of the nanofluid was also measured as a function of the shear rate ( $3\text{--}3000 \text{ s}^{-1}$ ) and is presented in Fig. 5. The viscosity did not appreciably vary, indicating Newtonian behavior, over the shear rate studied.

From the data for thermal conductivity and viscosity, it can be seen that the viscosity increase was higher than the thermal conductivity enhancement. It can be shown, following the analysis done by Prasher *et al.*,<sup>26</sup> that if the heat transfer hardware is redesigned, nanofluids could still be beneficial as coolants despite their higher viscosity increase, as compared to thermal conductivity increase. We present the analysis in the following section.

#### V. HEAT TRANSFER CONSIDERATIONS

From the above data, it is clear that the increase in viscosity is higher than the increase in thermal conductivity

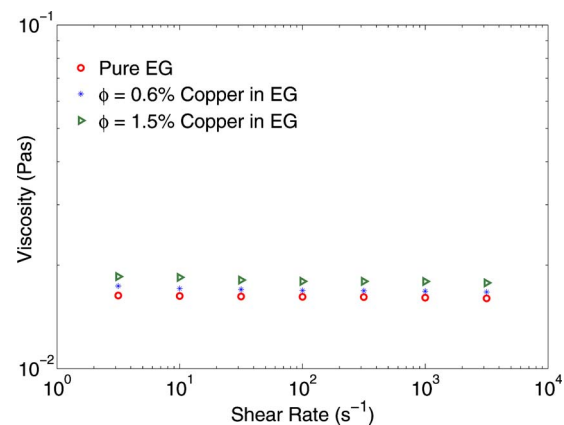


FIG. 5. (Color online) Viscosity of copper in ethylene glycol nanofluid as a function of shear rate ( $3.0\text{--}3000 \text{ s}^{-1}$ ) for three different volume fractions (0%, 0.6%, and 1.5%) at  $25^\circ \text{C}$ .

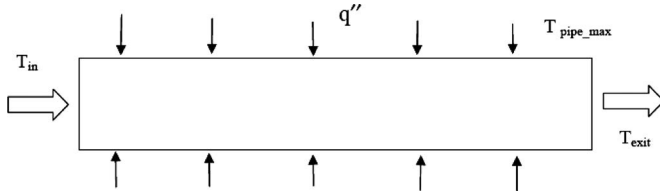


FIG. 6. Schematic of a duct being cooled by a liquid flowing through it.

when copper nanoparticles are added to ethylene glycol. In order to compare the cooling performance of a nanofluid with the corresponding base fluid, let us consider a pipe being cooled by a fluid flowing through it, as shown in Fig. 6. Let the uniform heat flux on the outside of the tube be  $q''$ . If the bulk temperature rise of the fluid is negligible, the maximum wall temperature of the pipe  $T_{\text{pipe\_max}}$  occurs at the exit end and is given by<sup>27</sup>

$$T_{\text{pipe\_max}} = \frac{q''d}{\text{Nu}k} + T_{\text{in}}, \quad (7)$$

where  $T_{\text{in}}$  is the bulk inlet fluid temperature,  $q''$  is the heat flux on the outside surface of the pipe,  $\text{Nu}$  is the Nusselt number,  $d$  and  $L$  are the pipe diameter and length, respectively,  $\dot{m}$  is the mass flow rate,  $c_p$  is the fluid specific heat, and  $T_{\text{pipe\_max}}$  is the maximum pipe temperature.

One can define a figure of merit to compare the performance of the two fluids (base fluid and the nanofluid) as

$$\eta = \frac{(\text{heat removed})_{nf}/(\text{pumping power})_{nf}}{(\text{heat removed})_f/(\text{pumping power})_f}. \quad (8)$$

In order to compare the figure of merit for the base fluid and the nanofluid, it is assumed that the volumetric flow rate is the same for both. If it is assumed that the Nusselt number stays constant for the base fluid and the nanofluid, it can be shown following the analysis done by Prasher *et al.*,<sup>26</sup> that for the case where the pipe diameter is not changed, and for the same maximum wall temperature,

$$\eta = \frac{k_{nf}/\mu_{nf}}{k_f/\mu_f} = \frac{1 + C_k\phi}{1 + C_\mu\phi} \approx 1 - (C_\mu - C_k)\phi = 1 - 5\phi. \quad (9)$$

Clearly for this case, the figure of merit is lower than one, indicating that nanofluid leads to poorer thermal performance compared to the base fluid. It is assumed in the above analysis that the heat transfer coefficient increases in proportion to the thermal conductivity. However, the increased thermal conductivity of the nanofluid may not directly translate into enhanced heat transfer and the mechanisms behind the heat transfer coefficient enhancement are still being investigated.

For the case where the pipe diameter is varied to take advantage of the higher thermal conductivity of the nanofluid and the heat flux is maintained constant, it can be shown that

$$\begin{aligned} \eta &= \left(\frac{k_{nf}}{k_f}\right)^5 \bigg/ \left(\frac{\mu_{nf}}{\mu_f}\right) = \frac{(1 + C_k\phi)^5}{1 + C_\mu\phi} \approx 1 + (5C_k - C_\mu)\phi \\ &= 1 + 19\phi. \end{aligned} \quad (10)$$

Thus, for the case where the pipe diameter is increased in proportion to the thermal conductivity of the nanofluid, the nanofluid is a better coolant compared to the base fluid as

long as  $C_\mu < 5C_k$ . This is because of the fact that the pumping power is inversely proportional to the fourth power of the diameter and increasing the diameter significantly lowers the pumping power required to maintain the same maximum wall temperature. Prasher *et al.*<sup>26</sup> compared the pressure drops while maintaining the same Nusselt number and showed that nanofluids were beneficial as coolants for  $C_\mu < 4C_k$ . By defining a different figure of merit which compares the total amount of heat removed while maintaining the same pressure drop, we have extended the range of applicability of nanofluids to higher values of  $C_\mu$ , i.e.,  $C_\mu < 5C_k$ . Equation (10) shows that a 2 vol % fraction of copper in ethylene glycol nanofluid would lead to a 38% better performance as a coolant compared to pure ethylene glycol.

## VI. CONCLUSION

Limited data exist for the thermal conductivity of copper in ethylene glycol nanofluid. In this article, we developed a technique for chemical synthesis of copper in ethylene glycol nanofluid and measured its thermal conductivity and viscosity. Particle size was measured to be about 200 nm using TEM. No surfactants were used to stabilize the nanoparticles. Thermal conductivity of the nanofluid was measured using a transient hot wire method for volume fractions ranging from 0.4% to 2%. The data show that the thermal conductivity enhancement is almost twice of that predicted by the Maxwell model. The measured increase in thermal conductivity was found to be much lower than that reported by Eastman *et al.*<sup>17</sup> The viscosity increase was almost four times of that predicted by the Einstein law of viscosity. Due to the larger increase in viscosity than the increase in thermal conductivity, nanofluids would be poorer coolants as compared to base fluids if they are used in the existing heat transfer hardware. However, if the pipe diameter is increased to take advantage of the higher thermal conductivity of the nanofluid, then nanofluids can prove beneficial as coolants.

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