Effect of Particle Size on Thermal Conductivity of Nanofluid

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Nanofluids, containing nanometric metallic or oxide particles, exhibit extraordinarily high thermal conductivity. It is reported that the identity (composition), amount (volume percent), size, and shape of nanoparticles largely determine the extent of this enhancement. In the present study, we have experimentally investigated the impact of Al_2Cu and Ag_2Al nanoparticle size and volume fraction on the effective thermal conductivity of water and ethylene glycol based nanofluid prepared by a two-stage process comprising mechanical alloying of appropriate Al-Cu and Al-Ag elemental powder blend followed by dispersing these nanoparticles (1 to 2 vol pct) in water and ethylene glycol with different particle sizes. The thermal conductivity ratio of nanofluid, measured using an indigenously developed thermal comparator device, shows a significant increase of up to 100 pct with only 1.5 vol pct nanoparticles of 30- to 40-nm average diameter. Furthermore, an analytical model shows that the interfacial layer significantly influences the effective thermal conductivity ratio of nanofluid for the comparable amount of nanoparticles.

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I. INTRODUCTION

THE need for transportation of sensible heat over a finite distance down the thermal gradient using an appropriate fluid is quite common in engineering devices, machines, and plants producing energy, work, or functions. Heat transfer through a fluid mostly occurs by convection that can be enhanced by appropriate flow geometry, boundary conditions, or increased thermal conductivity of the fluid. It is known that the suspension of solid particles in a fluid, as originally proposed by Maxwell,[1] may increase the thermal conductivity of the base fluid, because thermal conductivity of solids is orders of magnitude higher than that of liquids. However, coarse particles, due to their larger size and greater mass than that of finer ones, are prone to sedimentation, develop resistance to fluid flow, and cause erosion to conduits. On the other hand, fluids with suspensions of ultrafine or nanoparticle in very low quantity, called nanofluid (say by Choi[2]), offer exciting possibilities to enhance heat-transfer performance of a given fluid for exceeding the level predicted by conventional models based on the rule of average for fluid with suspension of microsized particles. Larger relative surface area of nanoparticles, compared to that of coarse particles, should significantly enhance heat-transfer capabilities, improve stability of the suspensions, and reduce erosion of channels or conduits. Thus, nanofluids can offer a significant advantage in the thermal management of both large installations such as heat exchangers, evaporators, or radiators as well as miniature or microelectronics devices. Keblinski et al.[3] have made a useful review of the properties of the nanofluid. The performance of nanofluid critically depends upon the size, quantity (volume percentage), shape, and distribution of dispersoids and their ability to remain suspended and chemically unreacted in the fluid. Despite the exciting opportunities, lack of agreement among experimental results, poor characterization of suspension, and inadequate theoretical understanding of the mechanisms of heat transfer by nanofluid are serious impediments against large-scale commercial exploitation of nanofluids for thermal management in important fields such as electronics, transportation, medicine, and thermal engineering.[4] In the past, experimental studies with metallic and ceramic nanoparticles by several researchers including Choi,[2] Das et al.,[5] Xuan et al.,[6] Eastman et al.,[7,8] and Lee et al.[9] have reported 5 to 60 pct enhancement of thermal conductivity containing barely 0.1 to 5 vol pct nanoparticles. It has recently been shown that a two-stage approach of synthesizing nanometric powders by mechanical alloying and subsequent dispersion of the same in a given fluid could be a more flexible method of producing nanofluid with a greater scope of scaling up the process of synthesis.[10,11] However, a detailed effort to study the influence of particle size on the degree of enhancements of thermal conductivity ratio has not been reported.

In the present work, a systematic effort has been made to synthesize Al-rich Al-Cu and Al-Ag alloy nanoparticles of different sizes from the appropriate elemental blends by means of mechanical alloying. These particles were dispersed in ethylene glycol and water in very low volume fraction following a special routine to prepare a new type of nanofluid and carry out characterization and thermal conductivity measurement of this nanofluid

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using an indigenously developed thermal comparator device. An attempt has also been made to improvise an existing model to predict the thermal conductivity ratio as a function of size and amount of suspended nanoparticles and to compare the same with experimental data obtained in this study.

II. EXPERIMENTAL

Nanofluid is a uniform and stable colloidal suspension and precludes the tendency for agglomeration or chemical change of the particles or fluid. In this study, Al_2Cu and Ag_2Al dispersed nanofluids were prepared by a two-step method, in which Al_2Cu and Ag_2Al nanoparticles were first produced by mechanical alloying using a high energy planetary ball mill followed by dispersing these nanometric solid particles into ethylene glycol and water to produce nanofluids with different volume fractions and particle sizes. Elemental powder blends of Al-30 at. pct Cu and Al-30 at. pct Ag were subjected to mechanical alloying at room temperature using a high energy planetary ball mill with WC media (vials and balls) at 300 rpm and 10:1 ball-to-powder weight ratio. Milling was carried out in wet condition (using about 50 mL of toluene) to prevent undue oxidation, agglomeration of powders, changing the milling dynamics by smearing, coating the vials and ball surfaces with Al/Cu/Ag powders, and ensuring sufficient output (alloyed powder) from the milling operation. Solid particles were deagglomerated and homogenized by intensive ultrasonic vibration and magnetic stirring, respectively. Use of 1 vol pct oleic acid as the surfactant and appropriate stirring were effective to ensure uniform and proper dispersion of particles and stability against the sedimentation without affecting the thermophysical properties of the nanofluid. The particle and grain size of Al_2Cu and Ag_2Al powders were varied by milling up to different hours. Milling was carried out in a wet condition in toluene to prevent undue oxidation, cold welding, and agglomeration. Identity and grain size of phases were determined by X-ray diffraction (XRD). Average crystallite size and lattice strain were measured by separating the XRD peak broadening contributions due to the Gaussian and Cauchian factors after subtracting the broadening due to instrumental errors.[12]

It is known that the flow pattern of a liquid-solid mixture depends on the hydrodynamic force acting upon the surface of solid particles. Therefore, the volume fraction of the solution is considered a more important factor than mass fraction. Volume fraction was calculated using the following conversion formula, because it is very difficult to measure the precise volume of nanoparticles:

$$\phi_v = \frac{1}{\left(\frac{1-\phi_m}{\phi_m}\right)\frac{\rho_p}{\rho_f} + 1} \quad [1]$$

where ϕ and ρ represent concentration and density with m and v as subscripts representing mass and volume and with p and f as subscripts representing solid particles

and base fluid, respectively. This equation leads to the density expression for a dilute mixture of liquid-solid as follows:

$$\rho = \rho_f(1 - \phi_v) + \rho_p\phi_v \quad [2]$$

III. RESULTS AND DISCUSSION

A. Measurement of Thermal Conductivity of Nanofluids

To measure thermal conductivity of nanofluids, we used an indigenously developed thermal comparator, based on the original concept of Powell,[13–15] that works on the principle that total heat transferred from a hot probe is a function of the thermal conductivity of both the fluid and solid particles (the sample fluid/nanofluid). For the proper operation of the thermal comparator, heat transfer between the sample and environment must be constant (or negligibly small) for all samples. This unit is easy to fabricate, economical, portable, and can be a complementary method to measure thermal conductivity by the more common but expensive transient hot wire technique. Figure 1(a) shows the schematic view of this setup that consists of a metallic copper probe, a temperature-controlled heating coil, a direct current microvoltmeter, and a voltage stabilizer.[10] The key to precise measurement in this setup lies in ensuring a point contact between the liquid surface and copper probe (to ensure one-dimensional heat flow).

When an electrically heated source is brought in contact with the sample (fluid) surface, the thermocouple attached to the probe tip senses the temperature through the thermo-emf generated and records the latter through the circuit covering the probe assembly, sample (nanofluid), and base. The thermo-emf is proportional to the temperature difference between the thermocouple probe tip and reference.

Figure 1(b) shows the calibration curve obtained by recording the thermal conductivity of various standard and synthetic fluids using the thermal comparator. Regression analysis of this variation of thermal conductivity (k) as a function of comparator reading (x) yields a polynomial to correlate x (in volt) with k (in W/m K) as follows:

$$k = \alpha + \beta x + \gamma x^2 \quad [3]$$

Here, α, β, and γ are the regression coefficients with the values $\alpha = 2.9 \times 10^{-1}$ W/mK, $\beta = -2.5 \times 10^{-3}$ W/mK/V, and $\gamma = 8.1 \times 10^{-6}$ W/mK/V^2, respectively.

Initial measurements were carried out using water, ethylene glycol, liquid paraffin, and carbon tetrachloride to obtain the calibration curve. Subsequently, the calibration was validated through measurements using two more standard liquids, namely, toluene and benzene. The comparator readings for toluene (218.2 μV) and benzene (221.1 μV), when converted into the corresponding thermal conductivity using Eq. [3], yielded values (0.137 and 0.140 W/mK) that were within ± 3 pct deviation from the corresponding standard k values of toluene (0.133 W/mK) and benzene
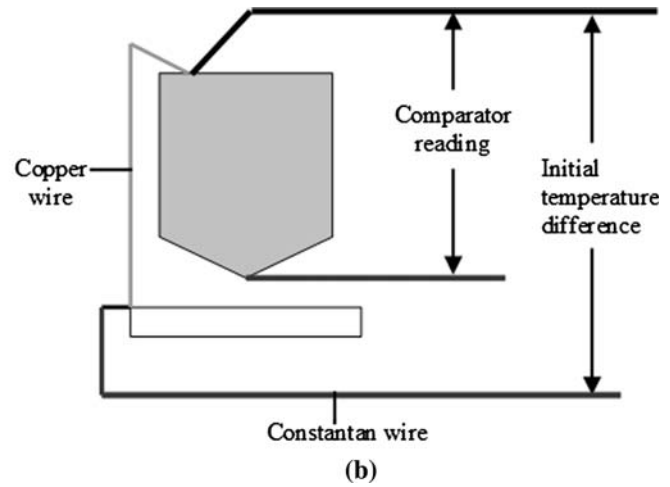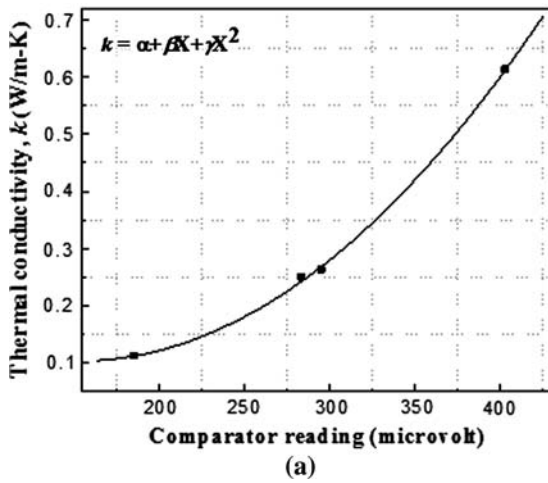
Fig. 1—(*a*) A schematic setup for recording differential thermo-emf based on the modified thermal comparator method. (*b*) Calibration curve to convert comparator reading (*x*) into thermal conductivity (*k*) of fluid/nanofluid.

(0.144 W/mK) at comparable temperature (300 K), respectively.[16] This small deviation may arise due to the impurity (water) present in the fluids. Thus, Figure 1(b) provides a reasonably accurate calibration to convert the experimental values of *x* (comparator reading) into corresponding *k* (thermal conductivity) of the concerned fluid or nanofluid.

B. *Phase Identification and Crystallite/Grain Size Measurement of Al₂Cu and Ag₂Al*

As mentioned earlier, the metallic powder used as colloidal dispersion in nanofluid was prepared by mechanical alloying. Figure 2(a) records the XRD profiles obtained from mechanically alloyed $Al_{70}Cu_{30}$

elemental powder blend at different hours of cumulative milling time. The elemental peaks tend to diminish or disappear by 10 hours of milling due to the formation of a new phase, identified as the equilibrium Al_2Cu or θ phase that stabilizes and remains in the microstructure until the end of milling. Thus, the final milling product of $Al_{70}Cu_{30}$ blend is single-phase nanocrystalline Al_2Cu. Figure 2(b) shows the compilation of XRD profiles obtained from the Al-30 at. pct Ag elemental powder blend subjected to mechanical alloying for different lengths of cumulative milling time. It is apparent that the sharp peaks of elemental Ag and Al disappear within 10 hours of milling due to formation of a new transition phase that finally transforms into a face-centered-cubic (fcc) phase beyond 30 hours of milling. Upon careful comparison with the relevant JCDPS data [14-0647], the milling product is identified as single-phase (metastable) Ag_2Al. Because mechanical alloying is a nonequilibrium processing route, formation of a transition metastable
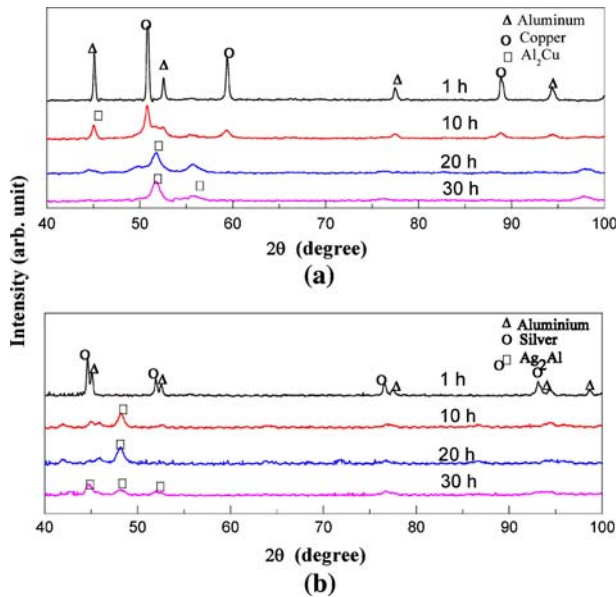

Fig. 2—XRD patterns of (*a*) $Al_{70}Cu_{30}$ and (*b*) $Al_{70}Ag_{30}$ elemental powder blends at different stages of mechanical alloying. Note that the final product is single-phase nanocrystalline Al_2Cu and Ag_2Al in the $Al_{70}Cu_{30}$ and $Al_{70}Ag_{30}$ blends, respectively.
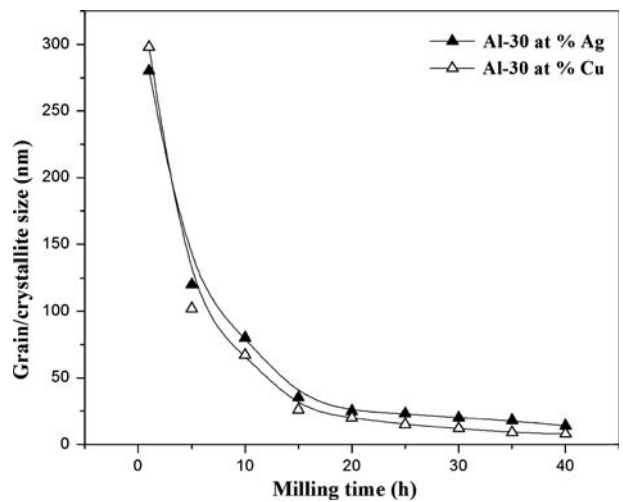

Fig. 3—Variation of crystallite size of mechanically alloyed of Al-30 at. pct Cu (Δ) and of Al-30 at. pct Ag ($\blacktriangle$) powder blends as a function of milling time.

phase is not unusual. Careful measurement of the peak-width at half-maximum intensity enables calculation of the crystallite/grain size of Al_2Cu and Ag_2Al through the Scherrer equation with an appropriate correction[12] to eliminate the influence of strain and instrumental error. It should be noted that the actual particle size may be larger than that calculated from the Scherrer formula due to agglomeration, because the particles are essentially poly-nanocrystalline. Figure 3 shows the variation of the grain/crystallite size of Al-30 at. pct Cu and Al-30 at. pct Ag alloys as a function of milling time. It appears that grain size reduction is significant within the first 10 hours of milling and tends to saturate by 30 hours of milling.

C. Thermal Conductivity of Al_2Cu-Dispersed Water and Ethylene Glycol Based Nanofluid

Thermal conductivity measurements were carried out at room temperature (300 K). Figure 4 shows the variation of the ratio of effective thermal conductivity of nanofluids (k_e) to that of the base fluid (k_f) as a function of nanoparticle size. It is evident that dispersoid size strongly influences the conductivity ratio. The smaller the dispersoid size, the greater is the enhancement in the thermal conductivity of nanofluids. This enhancement can be as high as 80 to 100 pct with approximately 30-nm particles and can be attributed to the greater specific surface area of nano compared to coarse particles. Figure 4 also reveals that the higher the volume percent of nanoparticles, the greater is the effective thermal conductivity ratio of water and ethylene glycol based nanofluid at comparable particle sizes of Al_2Cu and Ag_2Al.

It should be noted that the small size of particles not only enhances the thermal conductivity due to the larger surface areas (relative to that of coarse particles) but also increases the stability and homogeneity of suspensions. For nanofluids using the same amount of nanoparticles (Al_2Cu), the conductivity ratio (k_e/k_f) of the water based nanofluid is always slightly higher than that of the ethylene glycol based nanofluid. This is possibly because ethylene glycol is more viscous than water and the particles are more stable in ethylene glycol per Stoke's law.[2] Thus, both size/density of solid dispersoid and thermal/physical property of base fluid directly influence the thermal conductivity of nanofluid.

The variation of the thermal conductivity ratio with the volume percent of nanoparticles seems to follow a steady increase between 1.0 and 2.0 vol pct of particles. In terms of stability of dispersion (resistance to sedimentation and clogging), ethylene glycol based nanofluid is better than the water based one (because the former is more viscous).

Figures 5(a) and (b) show the TEM images of Al_2Cu dispersed ethylene glycol and water based nanofluid, respectively. The particles are truly nanometric (20 to 30 nm) albeit with some tendency of agglomeration.

D. Thermal Conductivity of Ag_2Al-Dispersed Water and Ethylene Glycol Based Nanofluid

Figure 6 shows that ethylene glycol and water based nanofluids containing a small volume percent (< 2 vol pct) of Ag_2Al nanoparticles induce a significantly higher thermal conductivity than that of the base liquid without nanoparticles, respectively. Comparison between Figures 4 and 6 reveals that the conductivity ratio of the Ag_2Al containing nanofluid is slightly higher than that of the Al_2Cu containing nanofluid. This is perhaps due to the higher thermal conductivity of silver (418.68 W/mK at 273 K) than that of copper (386.11 W/mK at 273 K).[16] Furthermore, Figure 6 suggests that enhancement in thermal conductivity follows a linear relationship both with the particle size and volume percent within the present range of investigation.

The results show that the factors that primarily affect the thermal conductivity of nanofluid are the size and amount (volume percent) of nanoparticles. The effective thermal conductivity ratio of the nanofluid increases with the decrease in size and increase in volume fraction of Ag_2Al or Al_2Cu nanoparticles. Comparison between the experimental results and preliminary experimental results of Eastman et al. (1997)[7] shows that the present (Ag_2Al or Al_2Cu dispersed) nanofluids exhibit superior thermal conduction properties. This is attributed to the narrow size distribution range of nanoparticles used in the present study (within 10 to 20 nm) than the wider size range (~30 to 100 nm) at CuO or Cu nanoparticles used by Eastman et al.[7] Because the surface area–to–volume ratio for particles with 20-nm diameter is 5 times higher than that for particles with 100-nm diameter, a significant improvement in effective thermal conductivity of nanofluids is expected if the average particle size is smaller and narrower in size distribution.

Figures 7(a) and (b) show that the Ag_2Al nanoparticles are near spherical in shape and show some tendency for agglomeration. A close scrutiny suggests that the particles tend to adhere together and form a chain structure. According to Hamilton and Crosser,[17] heat
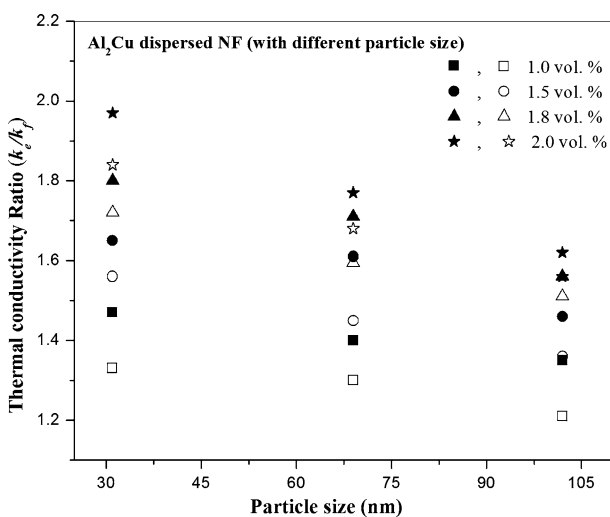

Fig. 4—Variation of thermal conductivity ratio (k_e/k_f) as a function of nanoparticle size of Al_2Cu dispersed ethylene glycol (open symbol) and water (filled symbol) based nanofluid.
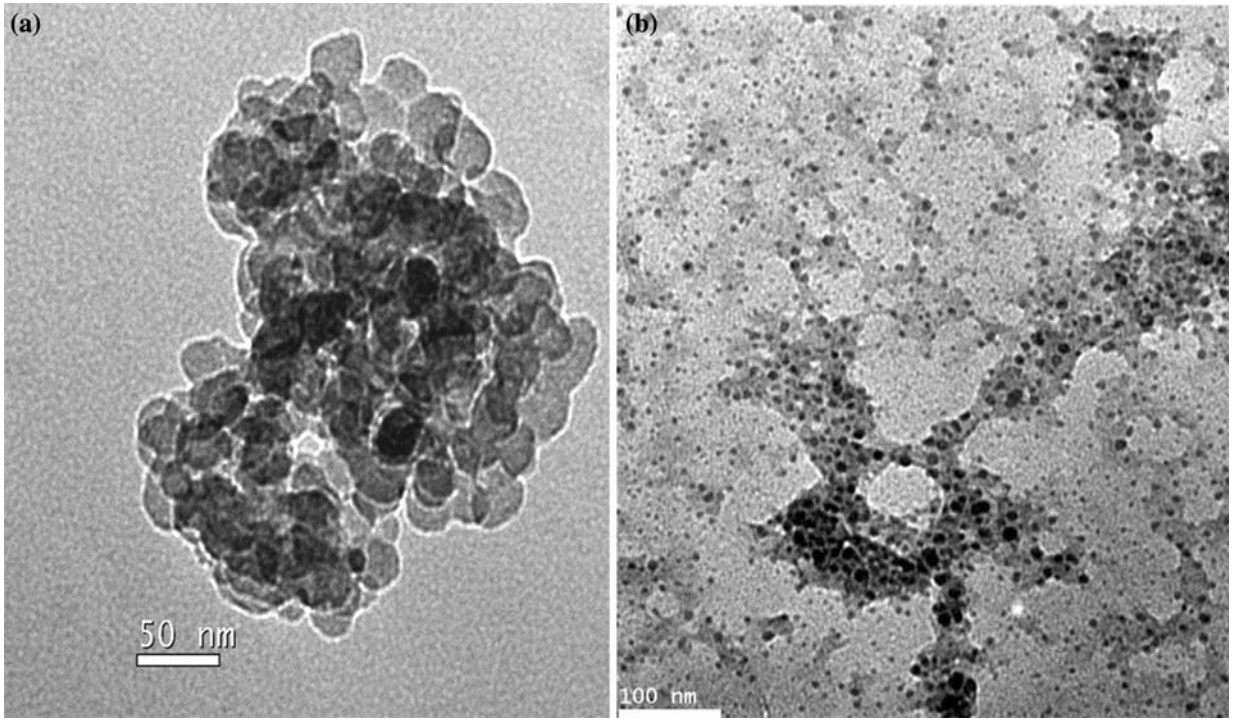
Fig. 5—TEM images showing nanometric Al_2Cu powder particles suspended in (*a*) ethylene glycol and (*b*) water.
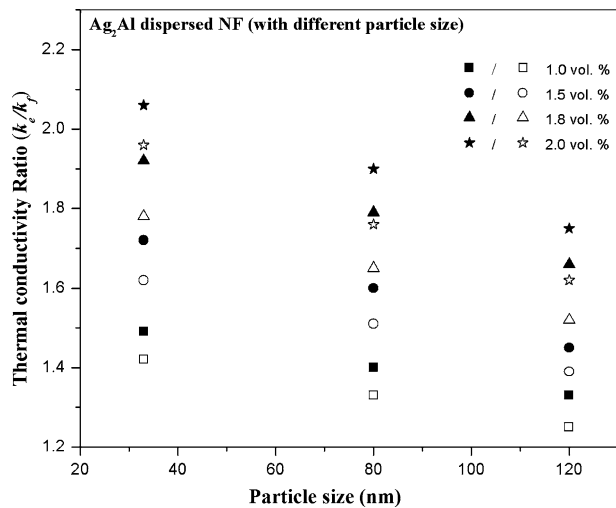

Fig. 6—Variation of thermal conductivity ratio (k_e/k_f) as a function of nanoparticle size of Ag_2Al dispersed in ethylene glycol (open symbol) and water (filled symbol) based nanofluid.

transfer could be enhanced if the particles form chain structures that may aid conduction of heat along those chains aligned in the direction of heat flux. Therefore, it is possible that such a chain structure contributes to an increase in thermal conductivity of nanofluid. Because a loosely arranged periodic aggregate of nanoparticles occupies more volume than the algebraic sum of the volume of individual nanoparticles that make up the aggregate, the effective volume of such loose ensemble is larger than the actual volume of nanoparticles.[3]

Though we do not have direct evidence of such macroensemble of loosely packed nanoparticles, we believe this could be one of the possible reasons for "extraordinary" thermal conductivity of nanofluid. It should be pointed out that the average particle size of Ag_2Al, 35 nm, after 25 hours of mechanical alloying calculated from the Scherer equation[12] is substantiated by TEM evidence in Figure 7. Figure 7 also reveals that Ag_2Al is within the narrow size range of 30 to 40 nm.

E. Analytical Model and Comparison between Predicted and Experimental Results

In the recent past, several attempts have been made to model the mechanism of heat transfer by nanofluid, although no unified theory has emerged to date. According to Keblinski et al.[3] and Eastman et al.,[7] the possible mechanisms of superior heat conduction by nanofluid can be explained on the basis of the (a) Brownian motion of the nanoparticles, (b) molecular-level layering of the liquid at the liquid/particle interface, (c) phonon-assisted ballistic heat transfer, or (d) formation of microensemble comprising clusters of nanoparticles. Wang et al.[18] have argued that the thermal conductivity of nanofluids should depend on distribution and the microscopic movement (Brownian motion and interparticle forces) of nanoparticle. Alternatively, formation of molecular level ordered or periodic arrangements of liquid/solid interfacial layer is believed to be one of the possible reasons for significant improvement of thermal conductivity in nanofluid. Recently, Koo and Kleinstreuer[19] proposed a new model for nanofluid that considers the effect of particle
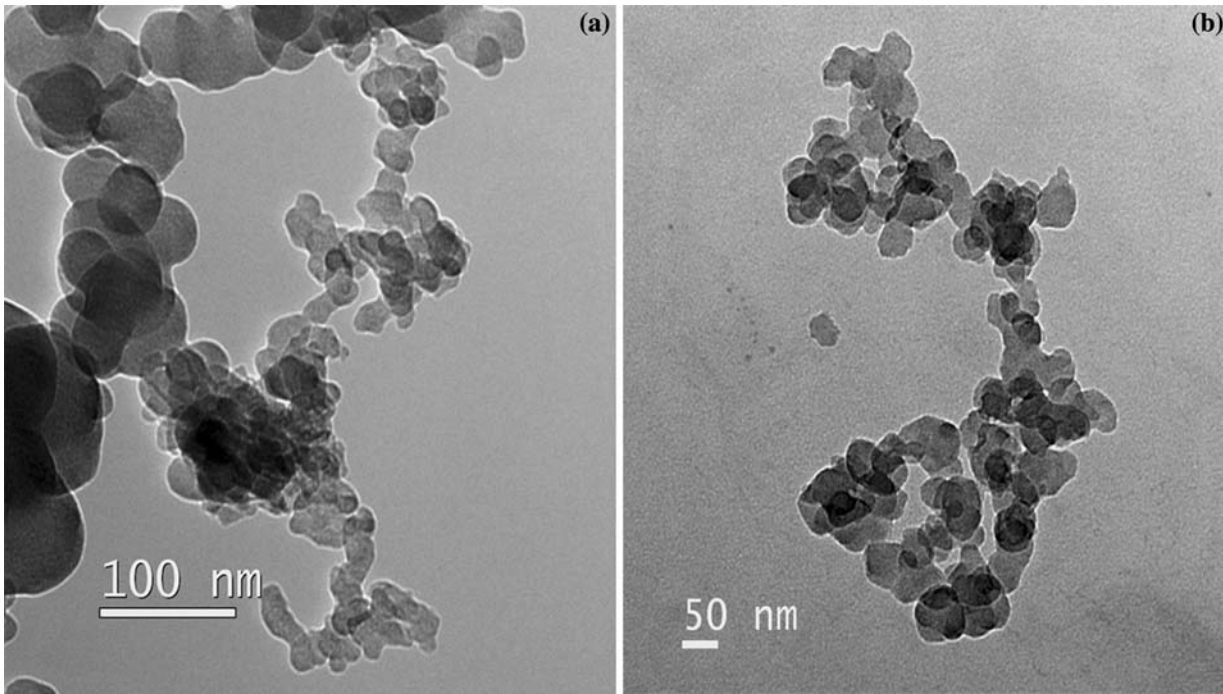
Fig. 7—TEM images showing nanometric Ag$_2$Al powder particles suspended in (a) ethylene glycol and (b) water.

size and volume fraction and the temperature dependence of the properties of the base fluid and nanoparticles subjected to Brownian motion. However, Evans et al.[20] have opined that the contribution of Brownian motion to the heat transfer by nanofluid is rather small and cannot account for the extraordinary thermal transport properties of nanofluid in stationary condition. Yu and Choi,[21] based on the so-called effective medium theory, have proposed a modified Maxwell model that considers a very thin liquid molecular layer around the nanoparticles in nanofluid having a thermal conductivity equivalent to that of the solid particles.

In one of the later developed models by Xue,[22] it has been suggested that an interfacial shell exists between the nanoparticles and liquid matrix with more ordered arrangement of atoms/molecules than that in the bulk liquid. If nanoparticles with interfacial shell are regarded as complex nanoparticles, then the nanofluid itself can be considered as a quasi-single-phase fluid with complex nanoparticles dispersed in it. Drawing parallels between complex nanoparticles and a conducting ellipsoid and using average polarization theory, it has been proposed that the effective thermal conductivity (k_e) of the complex nanoparticle-fluid system is a function of the depolarization factor ($B_{2,j}$) that depends entirely on the shape of complex nanoparticles.[22]

The rather complex expression for effective thermal conductivity of nanofluid presented by Xue[22] has been simplified by us as follows:

$$2A^3\Gamma - A^2\delta + A\chi + \theta = 0 \qquad [4]$$

where A is the thermal conductivity ratio (k_e/k_f); k_f is the thermal conductivity of pure fluid; and Γ, δ, χ, and θ are

constants that depend on the volume percent (v) of nanoparticles, thermal conductivity of the elliptical nanoparticles (k_2), concentric shell around it (k_1) with a finite thickness (t), and $B_{2,j}$, as follows:

$$\Gamma = 1 + B_{2,x} + 4C \qquad [5]$$

$$\delta = 2Y - 4C(2C - 2B_{2,x} - 2Y + 1) \\ - B_{2,x}(2Y - 1) - 1 + 2Z \qquad [6]$$

$$\chi = 2Y^2 - B_{2,x}(2Y^2 - 2Y + 4C) \\ - Y(2 - 2C - Z) - 2Z + 4C^2 \qquad [7]$$

$$\theta = Y^2(1 + B_{2,x}) - Y \qquad [8]$$

$$Y = k_{c,x}/k_f, \quad C = k_{c,y}/k_f, \quad Z = 9(1 - v/\lambda) \qquad [9]$$

$$\lambda = abc/[(a + t)(b + t)(c + t)] \qquad [10]$$

Here, a, b, and c are the half-radii of the elliptical nanoparticle. Thus,

$$k_{c,x} \text{ or } k_{c,j} = k_1 \frac{(1 - B_{2j})k_1 + B_{2j}k_2 + (1 - B_{2j})\lambda(k_2 - k_1)}{(1 - B_{2j})k_1 + B_{2j}k_2 - B_{2j}\lambda(k_2 - k_1)} \qquad [11]$$

According to Eq. [4], the effective thermal conductivity of nanofluid depends on the shape and surface area to volume of the complex nanoparticle. Figures 8 and 9
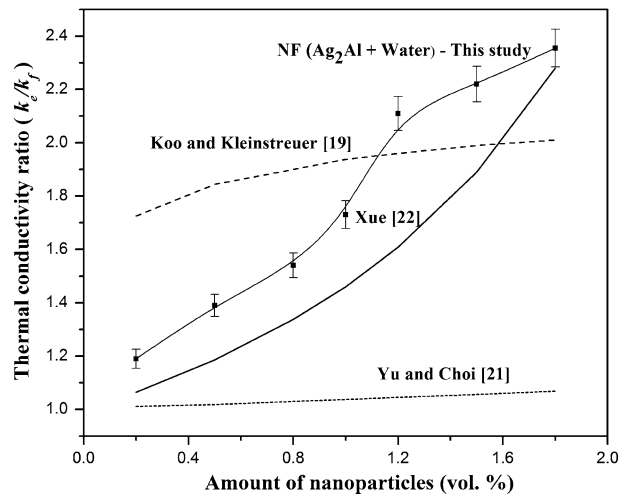
Fig. 8—Comparison between experimental results as a function of Ag$_2$Al nanoparticle volume percent in water-based nanofluid[23] and that predicted by modified Xue,[22] Koo and Kleinstreuer,[19] and Yu and Choi[21] model.
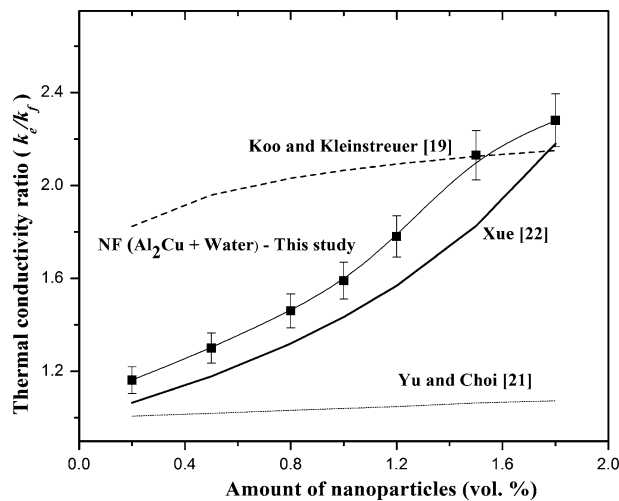

Fig. 9—Comparison between experimental results as a function of Al$_2$Cu nanoparticle volume percent in water-based nanofluid[23] and that predicted by the modified Xue,[22] Koo and Kleinstreuer,[19] and Yu and Choi[21] model.

compare the thermal conductivity ratio as a function of nanoparticle volume percent of Al$_2$Cu and Ag$_2$Al dispersed in water based nanofluid *vis-à-vis* that predicted by the modified model of Xue,[22] Koo and Kleinstreuer,[19] and Yu and Choi.[21] More detailed experimental results on the same nanofluid system have recently been reported elsewhere.[23] For calculation of effective thermal conductivity, the following data have been used.[16,24] Thermal conductivity of Al$_2$Cu nanoparticles = 319 W/mK, Ag$_2$Al nanoparticles = 358 W/mK, thickness of interfacial layer = 1 nm, nanoparticle diameters = 30 nm, density of Al$_2$Cu particles = 5.83 Mg/m^3, density of Ag$_2$Al particles = 7.56 Mg/m^3, specific heat of Al$_2$Cu particles = 0.643 kJ/kgK,

and specific heat of Ag$_2$Al particles = 0.48022 kJ/kgK.

It is obvious that the results predicted by the existing models fail to match the same obtained from the relevant experimental studies. Both the modified Maxwell model by Yu and Choi[21] and the model by Koo and Kleinstreuer[19] fail to predict the plausible value of thermal conductivity for the present nanofluids. The predicted results from the modified Xue model[22] appear somewhat closer to our experimental results but still are lower by about 10 pct from the corresponding experimental data. Thus, there is a need to develop more of a comprehensive theory to understand the behavior and mechanism of heat conduction in nanofluids. In this regard, Prasher[25] opines that the complex Brownian motion dominated convection is primarily responsible for the significant enhancement of the conductivity ratio in the nanofluid. Perhaps, additional macroscale effects such as heat conduction, particle-driven natural convection, or forces of electrophoresis or thermophoresis should be considered while considering the overall heat transfer through a nanofluid. On the other hand, microscopic thermal conductance modeling by molecular dynamic simulation between fluctuating dipoles by Domingues *et al.*[26] suggests that Coulombic interaction and polariton resonance within a few atomic diameter distances may account for the extraordinary thermal property of the nanofluid. In any case, interaction in the length scale extending to only a few particle diameters or lesser than that (shell around the nanoparticle) seems crucial and different than the existing macroscopic conduction, and radiation models and physical collision or contact among nanoparticles may not be necessary.[26]

IV. CONCLUSIONS

The effective thermal conductivities of Al$_2$Cu and Ag$_2$Al nanoparticle dispersed water and ethylene glycol based nanofluid have been measured using a thermal comparator. The experimental results show that the thermal conductivity ratio, relative to that of base fluid, increases nonlinearly with the increase in volume fraction and the decrease in the size/diameter of nanoparticles. Existence of an interfacial shell around nanoparticles having similar ordered or periodic arrangement of fluid molecules as that in solids seems plausible, because the results predicted by the model based on such an assumption lie reasonably close to the corresponding experimental data.

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