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Thermal- and Phase-Change Characteristics of Self-Assembled Ethanol/Polyalphaolefin Nanoemulsion Fluids

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The strategy of adding solid particles to fluids for improving thermal conductivity has been pursued for more than one century. Here, a novel concept of using liquid nanodroplets for enhancing thermal performance has been developed and demonstrated in polyalphaolefin nanoemulsion fluids with dispersed ethanol nanodroplets. The ethanol/polyalphaolefin nanoemulsion fluids are spontaneously generated by self-assembly and are thermodynamically stable. Their thermophysical properties, including thermal conductivity and viscosity, and impact on convective heat transfer are investigated experimentally. The thermal conductivity enhancement in these fluids is found to be moderate but increases rapidly with increasing temperature in the measured temperature range of 35–75°C. A very remarkable increase in the convective heat transfer coefficient, by a factor of up to 2.2, occurs in the nanoemulsion fluids due to the explosive vaporization of the ethanol nanodroplets at the superheat limit (i.e., spinodal states, about 122°C higher than the atmospheric boiling point for ethanol). Such an explosive liquid–vapor phase transition might augment the fluid heat transfer through the heat of vaporization (which intuitively raises the base fluid specific heat capacity) and the fluid mixing induced by the sound waves. The development of such phase-changeable nanoemulsion fluids would open a new direction for thermal fluids studies.

Nomenclature

C = heat capacity, $J/m^3\,K$
H = heat of vaporization, J/m^3
h = convective heat transfer coefficient, $W/m^2\,K$
k = thermal conductivity, $W/m\,K$
P = pressure, N/m^2
q = heat flux, W/m^2
T = temperature, °C
V = volume, m^3
ϕ = volume fraction
μ = fluid viscosity, $N \cdot s/m^2$
ν = velocity, m/s
ρ = mass density, kg/m^3

I. Introduction

COOLING is one of the most limiting technical challenges faced by a multitude of diverse industry and military groups [1–3,32,5–20]. The coolants, lubricants, oils, and other heat transfer fluids used in today's thermal systems typically have inherently poor heat transfer properties. Hence, there is an urgent need for innovative heat transfer fluids with improved thermal properties over those currently available. The idea of adding solid particles to improve thermal conductivity has been pursued for more than 100 years since Maxwell's theoretical work [4]. Early studies were confined to large particles (millimeter to micron sized). About a decade ago, nanofluids, heat transfer fluids containing suspensions of solid nanoparticles, were proposed to improve the thermal performance of the base fluids [1–3]. Materials commonly used for nanoparticles include oxides, such as alumina, silica, and titania; metals, such as copper and gold; and carbon nanotubes.

Recently, the authors have proposed a radically new design for thermal fluids, "nanoemulsion fluids," that completely eliminate solid particles and, instead, use liquid nanostructures [12,21,22]. Nanoemulsion fluids are suspensions of liquid nanodroplets (e.g., spherical droplets) in conventional thermal fluids, which are part of a broad class of multiphase colloidal dispersions. The present work focuses on an experimental study of convective heat transfer of self-assembled ethanol-in-polyalphaolefin (PAO) nanoemulsion fluids. The thermophysical properties, including thermal conductivity and viscosity, and their temperature dependence are also investigated.

II. Experimental Results and Discussion

A. Nanoemulsion Fluids

The ethanol/PAO nanoemulsion fluids are spontaneously generated by self-assembly. These ethanol nanodroplets are, in fact, reverse micelles swollen with ethanol and stabilized by the surfactant molecules, sodium bis(2-ethylhexyl) sullfosuccinate (Sigma Aldrich), which have hydrophilic heads facing inward and hydrophobic tails facing outward into the base fluid PAO (Chevron Phillips Chemical Company). Figure 1 shows the prepared ethanol/PAO nanoemulsion fluids and the pure PAO. The ethanol/PAO nanoemulsion fluid is transparent, but scatters light due to the Tyndall effect. The self-assembled ethanol/PAO nanoemulsion fluids are thermodynamically stable; their free energy is even lower than in the unmixed system [23].

The size distribution of the ethanol droplets in the self-assembled nanoemulsion fluids were determined by dynamic light scattering (DLS) with a Zetasizer Nano series (Malvern Instruments). All measurements were conducted at 20°C. The nanodroplet size was found to be less than 100 nm in diameter, as shown in Fig. 2. These results are consistent with the literature data [24].

PAO has been widely used as a heat transfer fluid and lubricant. It is able to remain oily in a wide temperature range due to the flexible alkyl branching groups on the C–C backbone chain. Ethanol is chosen as the dispersed phase mainly because it has a much lower atmospheric boiling point (78°C) than PAO [25,26].

B. Thermal Conductivity Characterization

Thermal conductivity and viscosity are macroscopically observable parameters that affect the thermal performance of the fluids, and their characteristics are investigated in the ethanol/PAO nanoemulsion fluids. The thermal conductivity of the pure PAO and ethanol/PAO nanoemulsion fluids (4 and 8 vol %) was measured in the temperature range of 35–75°C using a 3ω-wire method. Unlike the conventional hot-wire method [27], the 3ω-wire method

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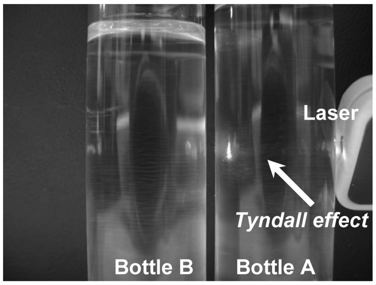


Fig. 1 Ethanol/PAO nanoemulsion fluids (bottle A) and pure PAO (bottle B). Liquids in both bottles are transparent. The Tyndall effect (i.e., a light beam can be seen when viewed from the side) can be observed only in bottle A when a laser beam is passed through bottles A and B. Picture taken by a Canon PowerShot digital camera.

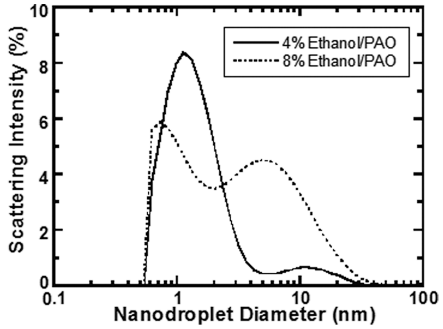


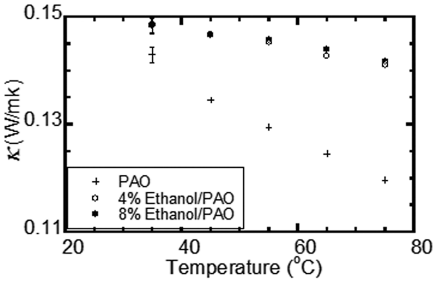
Fig. 2 Size distribution of ethanol nanodroplets in the ethanol/PAO nanoemulsion fluids. DLS measurements were conducted at 20°C.

determines the fluid conductivity by detecting the frequency dependence of temperature oscillation in a metal wire [11–13,22,28–31]. One advantage of this 3ω-wire method is that the temperature oscillation can be kept small enough (below 1 K, compared with about 5 K for the hot-wire method) within the test liquid. Calibration experiments were performed for oil and water at atmospheric pressure. Literature values were reproduced with an error of $<1\%$.

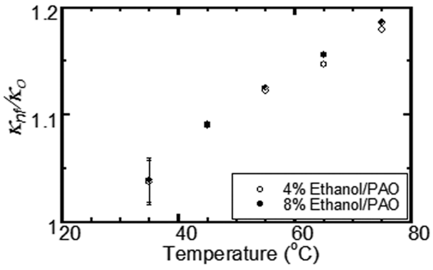
The thermal conductivity of the pure PAO and ethanol/PAO nanoemulsion fluids, as well as the relative thermal conductivity, are plotted in Fig. 3. The relative thermal conductivity is defined as k_{nf}/k_o, where k_o and k_{nf} are the thermal conductivities of the base fluid and nanoemulsion fluids, respectively. The PAO thermal conductivity is experimentally found to be 0.143 W/m K at room temperature, which compares well with the literature values [26]. As seen in Fig. 3, the relative thermal conductivity, or the thermal conductivity enhancement, increases rapidly with increasing temperature in the measured temperature range, although the thermal conductivity of both the pure PAO and ethanol/PAO nanoemulsion fluids goes down with temperature. The magnitude of the conductivity increase is rather moderate in the ethanol/PAO nanoemulsion fluids, for example, an 18% increase at 75°C for 4 vol %. These experimental results are within the range predicted by the traditional effective medium theory [32,4,33,34]. However, the dependence of the relative thermal conductivity on the ethanol concentration is found to be pretty weak in the ethanol/PAO nanoemulsion fluids, and the reason is still under investigation. It is noteworthy that ethanol has a thermal conductivity only slightly higher than PAO, $k_{\mathrm{PAO}} = 0.143$ W/mK and $k_{\mathrm{ethanol}} = 0.158$ W/mK, at room temperature [25,26].

C. Viscosity Characterization

The dynamic viscosity of the pure PAO and ethanol/PAO nanoemulsion fluids was measured using a commercial viscometer (Brookfield DV-I Prime). Results for the viscosity are shown in Fig. 4. The viscosity of the pure PAO and the ethanol/PAO nanoemulsion fluids was measured at a spindle rotational speed from 6 to 30 rpm and exhibited a shear-independent characteristic of



a)



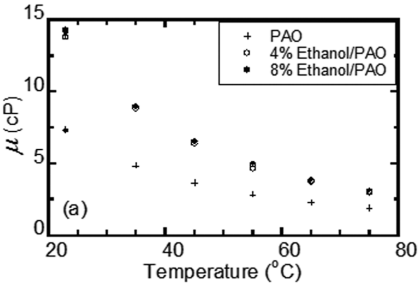
b)

Fig. 3 Shown are the following: a) thermal conductivity of the pure PAO and ethanol/PAO nanoemulsion fluids vs temperature, and b) relative conductivity of the ethanol/PAO nanoemulsion fluids vs temperature.

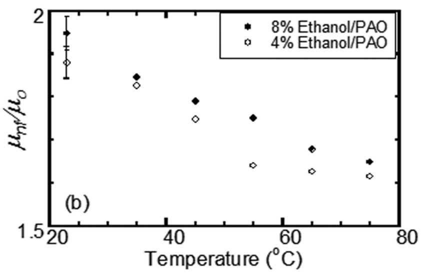
Newtonian fluids. Unlike the relative thermal conductivity, the relative viscosity is found to decrease with increasing temperature in the ethanol/PAO nanoemulsion fluids, for example, a decrease from 1.88 to 1.61 for 4 vol % when the temperature rises from 23 to 75°C. The similar trend of the relative viscosity has also been observed in many other nanofluid systems [31,35,36].

D. Convective Heat Transfer

The convective heat transfer experiments were conducted on a platinum wire 25 μm in diameter and 3 cm long, subjected to a crossflow of the fluids. This platinum wire serves as both the heater



a)



b)

Fig. 4 Shown are the following: a) dynamic viscosity of the pure PAO and ethanol/PAO nanoemulsion fluids vs temperature, and b) relative viscosity of the ethanol/PAO nanoemulsion fluids vs temperature.

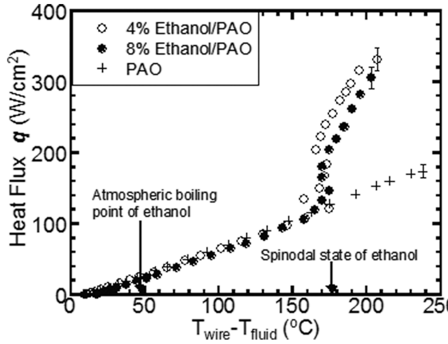


Fig. 5 Convective heat transfer curves for the pure PAO and the ethanol/PAO nanoemulsion fluids. Note that $T_{\mathbf{fluid}} = 30°\mathrm{C}$ and that the atmospheric boiling point of ethanol is 78°C.

and the thermometer. It is heated electrically by use of a dc power supply, and so the dissipated heat can be calculated directly from its voltage and resistance. The wire temperature can also be determined from its electrical resistance. The electric resistance of the platinum wire has a linear relationship with its temperature. The temperature coefficient of the platinum wire is found to be $0.00399\ \mathrm{C}^{-1}$. A thermocouple is used to measure the bulk temperature of the fluids, which is kept at 30°C. The flow rate is maintained at $3\ \mathrm{cm/s}$ for the fluids, monitored by a flow meter (King Instrument Company).

The convective heat transfer curves are plotted in Fig. 5 for the pure PAO and nanoemulsion fluids containing 4 and 8 vol % ethanol nanodroplets. The curve for the PAO is classified as being in the liquid forced convection regime, as the measured temperature range is far below its atmospheric boiling point [37]. When $\Delta T < 170°\mathrm{C}$ (i.e., $T_{\mathrm{wire}} < 200°\mathrm{C}$), the heat transfer coefficient values of all fluids including the pure PAO appear to be the same. This indicates that these ethanol nanodroplets have little effect on the fluid heat transfer efficiency without undergoing phase transition. When the heater temperature is further increased, however, an abrupt increase in the convective heat transfer coefficient, by a factor of up to 2.2, is observed in the ethanol/PAO nanoemulsion fluids, compared with the pure PAO case. For example, the dissipated heat flux q is found to be 125 and 275 $\mathrm{W/cm^2}$ at $T_{\mathrm{wire}} - T_{\mathrm{fluid}} = 180°\mathrm{C}$ for the pure PAO and the 4 vol % ethanol/PAO nanoemulsion fluid, respectively.

The causes of the observed abrupt increase in heat transfer coefficient can be examined by first evaluating the Hilpert correlation that works for laminar flow over a cylinder, [37] $h = Ak^{2/3}(\rho C_p)^{1/3}\nu^{0.0033}$, where A is a constant, ρ is the fluid mass density, C_p is the fluid heat capacity, k is the fluid thermal conductivity, and ν is the fluid kinematic viscosity. The mass density ρ of the tested nanoemulsion fluids remains nearly constant with the value of pure PAO and, therefore, has little effect on the increase of the heat transfer coefficient in the Hilpert correlation. The measured conductivity and viscosity increase is rather moderate in the ethanol/PAO nanoemulsion fluids. According to the Hilpert correlation, the net effects on heat transfer induced by the increased thermal conductivity and viscosity would be negligible in the ethanol/PAO nanoemulsion fluids, which is consistent with the fact that, when $T_{\mathrm{wire}} < 200°\mathrm{C}$, the heat transfer coefficient of the ethanol/PAO nanoemulsion fluids is almost same as that of the pure PAO.

Therefore, the abrupt increase in heat transfer coefficient, by a factor of up to 2.2, would be mainly accredited to the vaporization of those ethanol nanodroplets dispersed in the base fluid PAO. A direct impact of the nanodroplet vaporization is that the heat of vaporization could significantly enhance the effective heat capacity of the fluid. The effective heat capacity can be evaluated using the following formula: $C_{\mathrm{eff}} = C_0 + \phi \cdot H_{\mathrm{droplet}}/\Delta T$, where ϕ is the volume fraction of the phase-changeable nanodroplets, H_{droplet} is the heat of vaporization of the nanodroplets per unit volume, and ΔT is the temperature difference between the heat transfer surface and the bulk fluid [38]. In this experiment, if $\Delta T = 10°\mathrm{C}$ is assumed, the effective volumetric specific heat can be increased by up to 162% for the 4 vol % nanoemulsion fluid when the ethanol nanodroplets undergo liquid-vapor phase transition. This would provide as much as a 38% enhancement of the heat transfer coefficient according to the Hilpert correlation.

In addition, the nanodroplet vaporization can enhance heat transfer through inducing drastic fluid motion within the thermal boundary layer around the heat transfer surface. The ethanol/PAO interface constitutes a hypothetically ideal smooth surface, free of any solid motes or trapped gases, so that the heterogeneous nucleation and ordinary boiling are suppressed. In this case, the ethanol nanodroplets can be heated to a temperature of about 120°C above their normal atmospheric boiling point (78°C). Such a temperature is very close to thermodynamic limit of superheat or the spinodal state of ethanol and is only about 10% below ethanol's critical point. The spinodal states, defined by states for which $\frac{\partial P}{\partial V}|_{T,n} = 0$ [39], represent the deepest possible penetration of a liquid in the domain of metastable states. When those ethanol nanodroplets vaporize after reaching their limit of superheat, the energy released could create a sound shock wave, a so-called vapor explosion [40–42]. This sound wave would lead to strong fluid mixing within the thermal boundary layer, therefore enhancing the fluid heat transfer.

It is also observed in Fig. 5 that the heat flux appears to vary only slightly in the cases of 4 and 8 vol % ethanol/PAO nanoemulsion fluids. No satisfactory explanation is currently available. One possible explanation is that the vapor explosion effects on heat transfer might be mainly related to the droplet number density, rather than the droplet size, in the self-assembled ethanol/PAO nanoemulsion fluids.

III. Conclusions

In summary, the thermal and phase-change characteristics of the ethanol/polyalphaolefin nanoemulsion fluids have been investigated experimentally. These fluids are spontaneously generated by self-assembly, are thermodynamically stable, and can be mass produced. A remarkable increase in the convective heat transfer coefficient, by a factor of up to 2.2, is observed experimentally in the 4 vol % ethanol/PAO nanoemulsion fluids. Such an increase in the heat transfer coefficient is not related to the increased thermal conductivity and viscosity of the fluids. The ethanol nanodroplets vaporize explosively after reaching their limit of superheat, thereupon enhancing the fluid heat transfer through the heat of vaporization (which intuitively raises the base fluid specific heat capacity) and the fluid mixing induced by the sound waves. The ethanol/PAO nanoemulsion fluids are self-recoverable after the vaporization/condensation of the dispersed ethanol nanodroplets.

Acknowledgments

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