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ABSTRACT

The influence of CuO nanoparticles on the flow-boiling of R-134a/polyolester mixtures in a horizontal tube is quantified. A lubricant-based nanofluid is prepared with a synthetic ester and 30-nm diameter CuO particles stably suspended in the mixture to a 4% volume fraction. For a 0.5% nanolubricant mass fraction in a mixture with R-134a, the nanoparticles have no apparent effect on the two-phase boiling heat transfer coefficient. However, for a nanolubricant mass fraction of 1%, an enhancement between 42% and 82% in the heat transfer coefficient is manifest, over that of the same refrigerant-oil mixture without nanoparticles. A further increase in the nanolubricant mass fraction to 2% results in a still larger improvement in heat transfer coefficient of between 50% and 101%. In addition to heat transfer enhancement, it is found that the presence of nanoparticles has an insignificant effect on the system pressure drop (within the experimental uncertainty) when compared to baseline data.

1. INTRODUCTION

Solid particles of the nominal size 1-100 nm are called nanoparticles (Figure 1). Nanofluids are produced by dispersing metallic, ceramic, or carbon nanoparticles into base fluids such as water and ethylene glycol. Early studies of nanofluids have mainly focused on thermal conductivity enhancements which are often far beyond the predictions of classical models for larger-sized particle suspensions. Recently, Hwang *et al.* (2006) showed that multi-walled carbon nanotubes (MWCNT) in oil have a higher thermal conductivity enhancement than that of MWCNT in water, and that an ethylene-glycol-copper-oxide nanofluid has a higher thermal conductivity enhancement than that of copper oxide (CuO) in water. These results imply that higher thermal conductivity enhancements can be obtained for base fluids of lower thermal conductivity. In addition, Eastman *et al.* (1997) reported copper nanoparticles in pump oil resulted in an improvement in thermal conductivity similar to copper oxide in water, but required half the nanoparticle concentration.

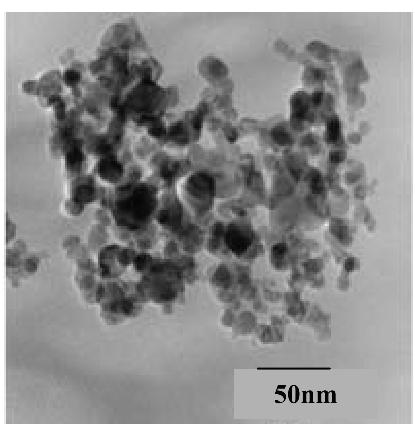


Figure 1: Copper (II) oxide nanoparticles (average size ~ 30 nm). Image from Nanophase Technologies Corp.

For single-phase convective heat transfer of nanofluids, most experimental investigations have found that the heat transfer coefficient increases with higher particle concentrations and increasing Reynolds number (e.g. Ding *et al.*, 2006). Furthermore, pressure drop measurements of pipe flows indicate an insignificant penalty for both laminar and turbulent flows for particle concentrations of 1-3% by volume in water.

In recent years, some studies on phase-change heat transfer of nanofluids have been reported; however, these studies are limited in their relevance to practical applications because they were mainly focused on pool boiling heat transfer of aqueous nanofluids. Kim *et al.* (2006b) found that nanofluids significantly increase the critical heat flux over that of a pure base fluid, and they attributed this enhancement predominantly to nanoparticle deposition on the heater and consequent changes of the surface microstructure. On the other hand, an independent study by Kim *et al.* (2006b) noted an increased wettability due to nanoparticle deposition on boiling surfaces. Lee and Mudawar (2007) studied aqueous Al_2O_3 nanofluids of 1% and 2% volume concentration in micro-channels and found that the single-phase heat transfer enhancement was more pronounced in the entrance region of the micro-channel under laminar flow conditions. Under evaporative two-phase flow conditions, however, large clusters of nanoparticles were formed near the exit of the channel causing clogging, and a steady state was never reached.

Despite the recent activity in nanofluid research for heat transfer enhancement and the continuously expanding scope, our current understanding of nanofluids is limited with respect to most two-phase flow conditions, especially those used in vapor compression systems. Previous experiments for single- and two-phase convective heat transfer predominantly use aqueous suspensions of nanoparticles. While water is widely used in heat transfer applications and generally produces stable nanoparticle suspensions, the feasibility of creating refrigerant-based nanofluids as well as characterizing their thermal effects must be explored if the potential heat transfer enhancements are to be realized in air-conditioning and refrigeration applications.

While screening fluid-particle combinations for this research, a rapid agglomeration and settling of numerous uncoated metal, ceramic, and carbon nanoparticles was observed in refrigerants. A slower rate of sedimentation was observed with hydrophobically treated Silica nanoparticles. Preliminary experiments were conducted with 0.5% volume concentration of these treated silica nanoparticles seeded directly into the refrigerant. Very little agitation of the mixture could be performed due to the nature of the base fluid and ultimately a 30% reduction in the two-phase heat transfer coefficient was found. Upon inspection of the experimental system, accumulated silica particles were found lining the walls of the tubing, thereby increasing the resistance to heat transfer within the test section.

Recently, Kedzierski and Gong (2007) studied the effect of a "nanolubricant," a polyolester oil with dispersed copper (II) oxide nanoparticles mixed with R-134a in pool boiling experiments (instead of direct particle-refrigerant mixing). As Kedzierski (2003) reported in earlier work, the mechanism of the boiling heat transfer of refrigerant/lubricant mixtures is strongly governed by the lubricant excess layer near the boiling surface. Kedzierski and Gong observed that the boiling process can drive the nanoparticles to the heat transfer surface where a more stable dispersion can be formed within the lubricant excess layer. Some of the particles will also be entrained in the vigorous boiling of the fluid. If the nanoparticles significantly change the thermal conductivity of the lubricant excess layer, that may cause an enhancement or a degradation in heat transfer depending on whether the increased conduction causes a reduced available superheat or whether it increases the thermal boundary layer thickness.

Notably, Kedzierski and Gong found significant enhancement of nucleate boiling heat transfer (between 50% and 245% for a nanolubricant mass fraction of 0.5% at 4% volume CuO nanoparticles) due to the presence of the nanoparticles in the oil. They also reported an unprecedented stability of the R-134a/nanolubricant mixture. On the basis of this recent finding, and in view of our early fluid-particle screening, we set out to determine the effect of dilute concentrations of nanoparticles on the two-phase flow boiling and pressure drop of halocarbon refrigerant/lubricant mixtures.

2. EXPERIMENTAL APPROACH

In order to investigate the influence of nanoparticles on refrigerant/lubricant two-phase heat transfer, the heat transfer coefficients for three R-134a/nanolubricant mixtures flowing through a horizontal tube at mass fluxes between 100 and 400 kg/m^2-s were measured. A commercial polyolester lubricant (RL68H) with a nominal kinematic viscosity of 72.3 μm^2/s at 313.15K was the base lubricant that was mixed with copper (II) oxide nanoparticles (79.55 g/mol) with a nominal size of 30 nm. Initially, a high-concentration RL68H/CuO mixture (nanolubricant) was purchased in which CuO particles constituted 40% of the volume. The manufacturer used a proprietary surfactant at a mass between 5% and 15% of the mass of the CuO particles to improve dispersion. The mixture was then diluted in our laboratory to a 4% CuO volume fraction by adding neat RL68H and ultrasonically agitating the solution for 24 hours. The nanolubricant, i.e. RL68H/CuO (96/4) volume fraction blend, was mixed with pure R-134a to obtain three R-134a/nanolubricant mixtures at nominally 0.5%, 1% and 2% mass fraction for two-phase flow boiling tests. In addition, the two-phase heat transfer for three R-134a/RL68H mixtures (0.5%, 1% and 2% mass fraction), without nanoparticles, were measured to serve as baseline for comparison to the nanofluids.

Evaporative heat transfer and pressure drop data, with and without nanoparticle seeding was obtained using a two-phase flow loop (Figure 2). The liquid refrigerant is pumped with a gear pump that is driven by a variable-frequency drive through a Coriolis-effect mass flow meter (±0.1%), through a preheater used to reach the desired vapor quality and to the test section inlet. The preheater consists of a copper tube section wrapped with electric strip heaters. The gauge pressure before the inlet of the preheater is measured using a pressure transducer with an accuracy of ±3.5 kPa. The temperatures before the inlet of the preheater and within the test section are measured with type-T thermocouples (±0.1ºC). These pressures and temperatures are used to determine the thermodynamic states necessary to compute the test section inlet quality. Downstream of the test section, the refrigerant is condensed in a water-cooled brazed-plate heat exchanger and is delivered to the receiver tank. A water-cooled brazed-plate subcooler is used upstream of the pump in order to avoid cavitation. Pressure, temperature, and quality in the test section are controlled by using the two throttling valves, preheater and by varying the temperature and flow rate of the cold water entering the condenser.

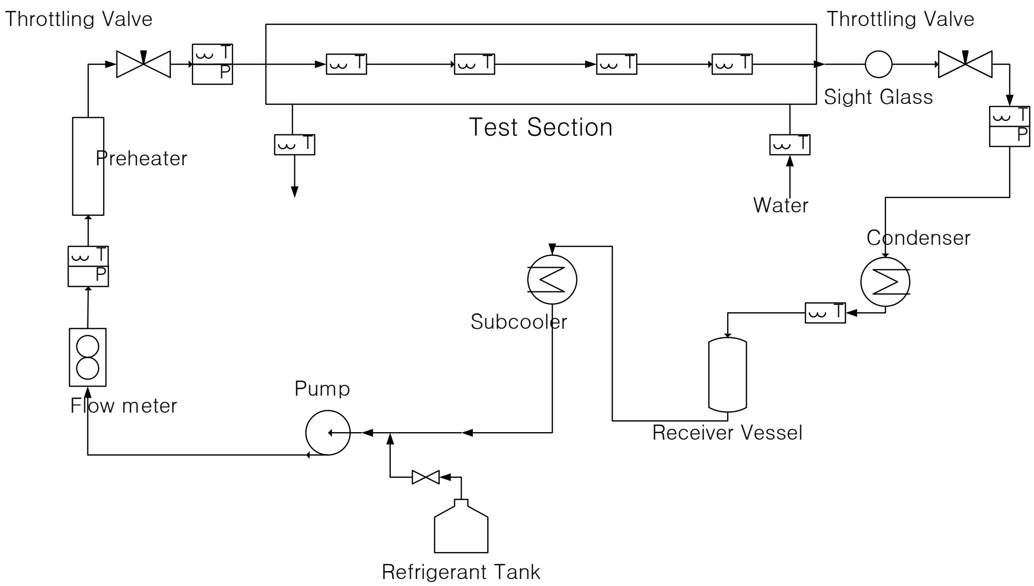


Figure 2: Schematic of the experimental apparatus

The 2-meter-long test section allows for various values of mass flux and heat flux over a range of inlet qualities with only small quality changes; thus, quasi-local values of the heat transfer coefficient can be obtained. The test section is a concentric counter flow heat exchanger, where the refrigerant flows through the smooth, copper inner tube (7.9 mm, ID). Evaporation within the test section is driven by a counter flow of hot water in the outer tube with the temperature and pressure measured at stations both in the flow and along the refrigerant tube surface. The temperature of the heating water is controlled using a thermostatic bath and the mass flow rate of water is measured using a vortex flow meter with an uncertainty of ±3.0%. To test the accuracy and reliability of the system, experiments were conducted using pure R-134a and the results were compared to correlations from the literature.

Once validation was complete, baseline and enhancement experiments were conducted by injecting a measured mass of lubricant or nanolubricant with a syringe into the receiver vessel. The experimental system was then evacuated and charged with pure R-134a to a predetermined mass. The refrigerant/lubricant solution was mixed by flushing high velocity refrigerant through the system for approximately 3 hours. All compositions were determined from masses of the charged components and are given on a mass fraction basis.

3. DATA REDUCTION AND ANALYSIS

The following calculation was employed to determine the vapor quality of the refrigerant at the inlet and outlet of the test section as well as the evaporative heat transfer coefficient, from the steady-state data recorded during two-phase heat transfer experiments. All of the thermophysical and transport properties were evaluated using commercial software (Engineering Equation Solver, *EES*) to determine pure R-134a properties. At the oil concentrations used in this work, the effect of the oil on the saturation state and enthalpy is within the experimental uncertainty.

The vapor quality at the inlet of the test section (x_{in}) was determined from the measured pressure and the enthalpy as given by:

$$i_{in} = i_{sub} + Q_{heater} / m_{ref} \quad , \tag{1}$$

where i_{sub} is the enthalpy of subcooled liquid based on the pressure, P_{pre} and the temperature, T_{pre} measured before the preheater, Q_{heater} is the heat transfer rate into the refrigerant from the preheater, and m_{ref} is the mass flow rate of the refrigerant. Similarly, the vapor quality at the exit of the test section (x_{out}) is determined from the measured pressure and the exit enthalpy given by,

$$i_{exit} = i_{in} + Q_w / m_{ref} \quad , \tag{2}$$

where i_{exit} is the refrigerant enthalpy at the exit of the test section and Q_w is the heat transfer rate into the refrigerant from the circulating hot water. The heat transfer, Q_w is found from the measured water temperatures and flow rate from:

$$Q_w = m_w C_{pw} (T_{w,in} - T_{w,out}) \quad . \tag{3}$$

Experiments were conducted such that quality changes were always less than 7.5%, allowing a quasi-local, average evaporative heat transfer coefficient ($\overline{h}$) to be determined from:

$$Q_w = \left(T_{wall,avg} - T_{ref,avg}\right)\left[\frac{1}{\overline{h}2\pi r_i L} + \frac{\ln(r_o / r_i)}{2\pi k L}\right]^{-1} \tag{4}$$

or, rearranging,

$$\overline{h} = kQ_w / \left[2\pi r_i kL(T_{wall,avg} - T_{ref,avg}) - Q_w r_i \ln(r_o / r_i)\right] \tag{5}$$

In Equation (5) k is the thermal conductivity of the tube wall (copper alloy); r_i and r_o are the inside and outside diameters of the test section tube, respectively; L is the length of the test section; $T_{wall,\ avg}$ is the tube wall temperature averaged from thermocouple readings at the outer surface; and $T_{ref,avg}$ is the saturation temperature of the refrigerant in the test section which is evaluated at $P=(P_{in}+P_{out})/2$.

Two existing correlations were used to validate the baseline data. Equation (6) was proposed by Gnielinski (1976) for fully-developed single-phase turbulent flow in a smooth circular tube and Equation (7) was suggested by Panek *et al.* (1992) for convective boiling of pure R-134a.

$$h = \frac{k(f/8)(\mathrm{Re}_D - 1000)\,\mathrm{Pr}}{2r_i(1+12.7(f/8)^{1/2}(\mathrm{Pr}^{2/3}-1))} \tag{6a}$$

where f, the friction factor is given by:

$$f = (0.790 \ln \mathrm{Re}_D - 1.64)^{-2} \tag{6b}$$

$$\frac{h_{tp}}{h_{lo}} = \frac{3.686}{X_{tt}^{0.563}} \tag{7a}$$

where

$$h_{lo} = \frac{0.023k_l}{2r_{in}} \mathrm{Re}_{lo}^{0.8} \mathrm{Pr}_l^{0.4} \tag{7b}$$

and

$$X_{tt} = \left(\frac{1-x}{x}\right)^{0.9} \left(\frac{\rho_g}{\rho_l}\right)^{0.5} \left(\frac{\mu_l}{\mu_g}\right)^{0.1} \tag{7c}$$

In Equation (7) h_{lo} is the liquid-only heat transfer coefficient and Re_{lo} is the liquid-only Reynolds number. Properties with subscript l refer to liquid properties and subscript g refers to the vapor phase.

Table 1 summarizes the range in saturation temperature, heat flux, mass flux and average vapor quality achieved within the test section during both baseline and enhancement experiments. The average uncertainty of the reported evaporative heat transfer coefficients, calculated using EES, is 22.1%, 24.5% and 13.8% for the 0.5%, 1% and 2% nanolubricant mixtures, respectively. For the 1% and 2% nanolubricant mixtures, the enhancement as reported in the results section of this paper is significantly greater than the aforementioned uncertainty. This establishes the validity of the heat transfer enhancement found with the 1% and 2% R-134a/nanolubricant mixtures.

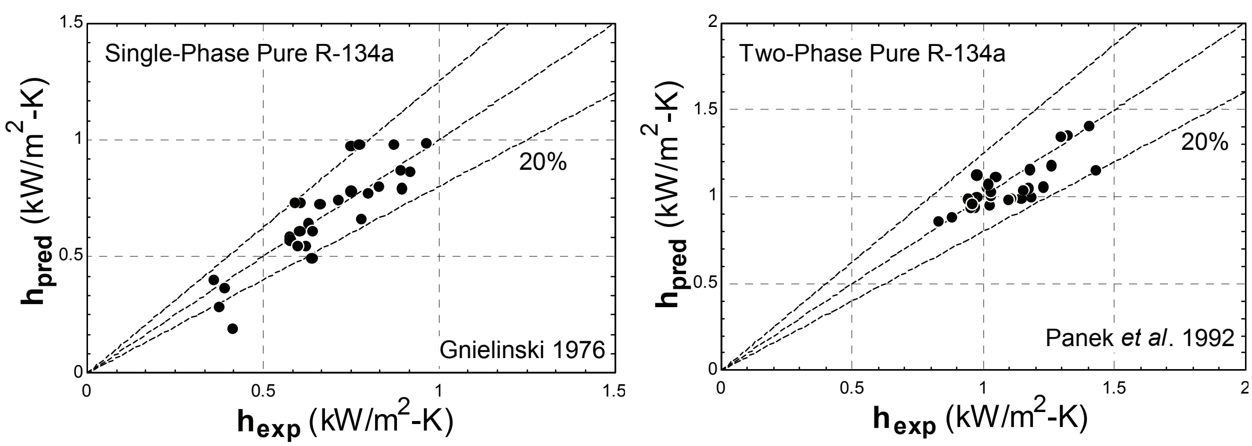
Table 1: Range of experimental parameters

Saturation Temperature	**Heat Flux**	**Mass Flux**	**Average Vapor Quality**
3.6 - 14.6* [°C]	0.69 - 3.06 [kW/m^2]	125 – 390 [kg/m^2-s]	2.44-6.33 [%]
*For the 2% nanolubricant mixture the saturation temperature ranged from 37.9 - 41.0°C			

4. RESULTS

4.1 Apparatus Validation with Pure R-134a

Before measuring the two-phase heat transfer coefficient and pressure drop of a refrigerant/nanolubricant mixture, the reliability and accuracy of the experimental system were assessed by measuring the single-phase and two-phase heat transfer coefficient of pure R-134a. Single-phase heat transfer data were collected and compared to predictions by Gnielinski (1976) as shown in Figure 3(a). For two-phase heat transfer measurements using pure R-134a, results were compared to Panek *et al*. (1992) and are shown in Figure 3(b). The good agreement between the measured data and well-established predictions by Gnielinski and Panek serves to validate the apparatus and data reduction methods.



(a) (b)

Figure 3: Comparison of experimental data to (a) Gnielinski's correlation for the single-phase-flow data, and (b) Panek's model for the flow-boiling data

4.2 Evaporative heat transfer and pressure drop with R-134a/POE/CuO nanofluids

Experiments were conducted with and without CuO nanoparticle seeding in order to allow baseline and nanofluid comparisons at the same refrigerant mass flux and inlet vapor quality. For nanofluid experiments conducted at mass fractions of 0.5% and 1% nanolubricant, the saturation temperature was controlled to within ±2ºC of the relevant baseline condition for the majority of experiments. Saturation temperatures were found to be higher with the nanolubricant mixtures relative to the observed baseline saturation temperatures and increased with increasing nanolubricant mass fraction. The throttling valves, preheat, and temperature and flow rate of the cold water entering the condenser were adjusted to fit the enhancement test parameters to those of the corresponding baseline conditions. Even with these controls, the saturation temperatures for the 2% nanolubricant enhancement tests were between 25ºC and 31ºC higher than baseline tests with corresponding refrigerant mass flux and inlet vapor quality.

As reported in most prior work, an insignificant pressure drop penalty (within the experimental uncertainty of the pressure transducers) was found with the CuO nanolubricant for all three nanofluids of this study. Seemingly, nanofluids cause little or no penalty to pumping power because at these very low concentrations the particles do not substantially affect viscosity.

The heat transfer coefficient for the R-134a/POE/CuO nanofluid, 0.5% nanolubricant by mass, is compared to that of the baseline R-134a/POE mixture in Figure 4. The nanoparticles had no apparent effect on the heat transfer coefficient with enhancement test data varying within ±10% of the baseline.

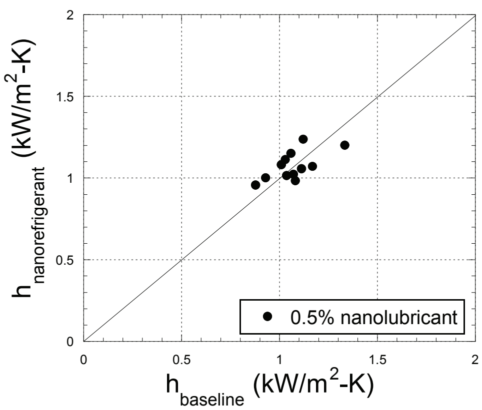


Figure 4: A mass fraction of 0.5% nanolubricant has no apparent effect on the evaporative heat transfer coefficient of R-134a

The effect of nanolubricant at mass fractions of 1% and 2% is shown in Figure 5. The R-134a/POE/CuO nanofluid, 1% nanolubricant by mass, shows a 42-82% enhancement in the heat transfer coefficient relative to that of a "pure" R-134a/POE mixture (99/1 by mass). A further increase in the nanolubricant mass fraction to 2% resulted in a still larger heat transfer enhancement of 50-101% over baseline data collected with pure R-134a/POE (98/2 by mass).

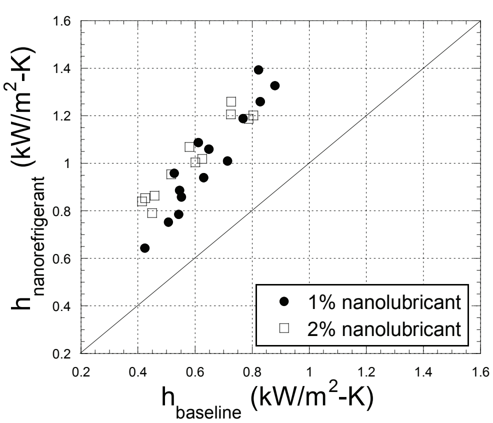
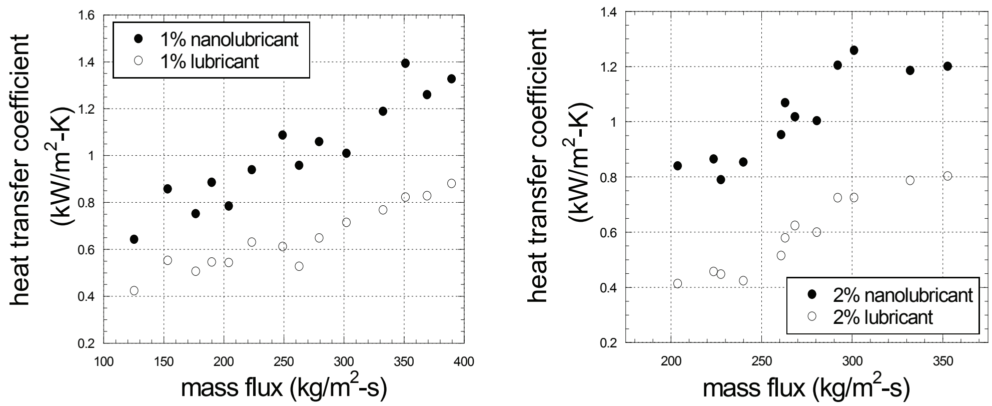


Figure 5: Mass fractions of 1% and 2% nanolubricant increase the evaporative heat transfer coefficient of R-134a

These baseline and nanofluid data are presented in Figure 6 as a function of mass flux, where again the dramatic enhancement is apparent. During these experiments, the average quality ranged from 2.44% to 6.33%, and the heat flux ranged from 0.69 to 3.06 W/m^2. While the experimental range is limited, the results are compelling.



(a) (b)

Figure 6: Heat transfer enhancement as a function of mass flux for (a) 1% nanolubricant and (b) 2% nanolubricant

The heat transfer results show that nanofluids have significant potential for improving the flow-boiling heat transfer of refrigerant/lubricant mixtures. However, the reasons behind this marked improvement with nanolubricant mass fractions of 1% and 2% are not clearly understood. It is known that the suspended nanoparticles increase the thermal conductivity of the base lubricant; however, the thermal conductivity of CuO (20 W/m-K) is just two orders of magnitude larger than that of RL68H (0.132 W/m-K). Therefore, as reported by Kedzierski and Gong (2007), the measured thermal conductivity of the 4% volume CuO nanolubricant (0.139 W/m-K) is just 5% higher than that of neat RL68H. The mechanism responsible for the heat transfer enhancement seems to go beyond what would be expected from the increase in thermal conductivity, unless a particle-rich layer forms near the heat transfer surface. It might be that the particles induce secondary nucleation near the heat transfer surface. Moreover, chaotic movement of the ultrafine CuO particles might increase the thermal mixing.

5. CONCLUSIONS

The effect of CuO nanoparticles on the flow boiling of R-134a/POE mixtures in a horizontal tube was investigated. For a nanolubricant mass fraction of 0.5%, no effect on the heat transfer coefficient was apparent. However, for nanolubricant mass fractions of 1% and 2%, the heat transfer coefficient of the R-134a/POE/CuO nanofluid increased in comparison to baseline experiments with corresponding R-134a/POE mixtures. For a 1% nanolubricant mass fraction, the nanoparticles caused a heat transfer enhancement between 42% and 82%, and for a 2% nanolubricant mass fraction an enhancement between 50% and 101% was measured. It was also noted that nanoparticle presence had an insignificant effect on pressure drop. It is unclear why the large increase in heat transfer with an insignificant pressure increase is realized. Moreover, obvious challenges with particle circulation and unknown effects on the compressor of an air-conditioning or refrigeration system have not been addressed. Nevertheless, these findings are compelling and further research should be undertaken.

ACKNOWLEDGEMENT

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NOMENCLATURE

C_p	specific heat at constant pressure	(kJ/kg·K)
f	friction factor	(-)
h	heat transfer coefficient	(kW/m^2·K)
i	enthalpy	(kJ/kg)
k	thermal conductivity	(W/m·K)
L	length of test section	(m)
m	mass flow rate	(kg/s)
P	pressure	(kPa)
Pr	Prandtl number	(-)
Q	heat transfer rate	(kW)
Re	Reynolds number	(-)
r_i	Tube inner radius at test section	(m)
r_o	Tube outer radius at test section	(m)
T	temperature	(°C)
x	vapor quality	(-)
X_{tt}	Lockhart-Martinelli parameter	(-)

Greek Symbols

μ	dynamic viscosity	(Pa·s)
ρ	density	(kg/m^3)

Subscripts

avg	average
exit	test section exit
g	vapor
in	test section inlet
l	liquid
lo	liquid only
ref	refrigerant
sat	saturation condition
sub	subcooled
tp	two-phase
w	water
wall	tube wall surface

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