

# Effect of Diamond Nanolubricant on R134a Pool Boiling Heat Transfer

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*This paper quantifies the influence of diamond nanoparticles on the pool boiling performance of R134a/polyolester mixtures on a roughened, horizontal, and flat surface. Nanofluids are liquids that contain dispersed nanosize particles. A lubricant based nanofluid (nanolubricant) was made by suspending 10 nm diameter diamond particles in a synthetic ester to roughly a 2.6% volume fraction. For the 0.5% nanolubricant mass fraction, the nanoparticles caused a heat transfer enhancement relative to the heat transfer of pure R134a/polyolester (99.5/0.5) up to 129%. A similar enhancement was observed for the R134a/nanolubricant (99/1) mixture, which had a heat flux that was on average 91% larger than that of the R134a/polyolester (99/1) mixture. Further increase in the nanolubricant mass fraction to 2% resulted in boiling heat transfer degradation of approximately 19% for the best performing tests. It was speculated that the poor quality of the nanolubricant suspension caused the performance of the (99.5/0.5), and the (98/2) nanolubricant mixtures to decay over time to, on average, 36% and 76% of the of pure R134a/polyolester performance, respectively. Thermal conductivity and viscosity measurements and a refrigerant/lubricant mixture pool-boiling model were used to suggest that increases in thermal conductivity and lubricant viscosity are mainly responsible for the heat transfer enhancement due to nanoparticles. Particle size measurements were used to suggest that particle agglomeration induced a lack of performance repeatability for the (99.5/0.5) and the (98/2) mixtures. From the results of the present study, it is speculated that if a good dispersion of nanoparticles in the lubricant is not obtained, then the agglomerated nanoparticles will not provide interaction with bubbles, which is favorable for heat transfer. Further research with nanolubricants and refrigerants are required to establish a fundamental understanding of the mechanisms that control nanofluid heat transfer. [DOI: 10.1115/1.4005631]*

*Keywords: additives, boiling, diamond, enhanced heat transfer, nanotechnology, refrigerants, refrigerant/lubricant mixtures*

## Introduction

Energy efficiency is a primary component for net zero energy, high performance green building-design [1,2]. Chillers that provide air conditioning for buildings consume nearly 13% of total building electric expenditures [3]. Consequently, a cost-effective means for improving the efficiency of chillers would facilitate meeting green building goals. In recent years, nanofluids, i.e., liquids with dispersed nanosize particles, have been shown to be a potential means for enhancing the performance of chillers. For example, Liu et al. [4] have demonstrated that a water-based nanofluid in the evaporator improved the efficiency of a laboratory chiller by more than 5%. Given that nanoparticles are not likely to be trapped by filters or increase abrasion because of their small size [5], they appear to be a viable option for improving chiller performance.

In addition to water-based nanofluids, lubricant-based nanofluids, i.e., nanolubricants, have been used to improve refrigerant boiling heat transfer that will, likewise, benefit refrigerant cycle performance. For example, recent studies by Kedzierski [6] and Bi et al. [7] have recommended the use of nanolubricants as a means for improving efficiencies of chillers and refrigerators, respectively. Peng et al. [8] have shown that diamond nanolubricants can significantly improve the nucleate pool boiling of R113 at atmospheric pressure. In addition, Marquis and Chibante [9] discuss the improved lubricating qualities of nanolubricants for engines. In a similar manner for engines, improved lubricity is

also expected to benefit the performance of the chiller compressor by reducing power requirements.

Nanoparticle properties are crucial for determining the performance characteristics of nanolubricants. According to Bobbo et al. [10], the way in which nanoparticle material, dimension, shape, and concentration affect nanolubricant properties is complex and not well understood. Marquis and Chibante [9] point out that nanoparticle size is more influential in determining thermal conductivity than is the shape of the nanoparticle. Kedzierski [6] has shown that the concentration of CuO nanoparticles may determine whether an enhancement or a degradation in refrigerant/lubricant boiling heat transfer is obtained. The same study also showed that the improvement in thermal conductivity was not the governing factor in determining the magnitude of the enhancement despite CuO having nearly 2 orders-of-magnitude greater thermal conductivity than the base lubricant. Clearly, knowledge of how the properties of nanoparticles influence the heat transfer behavior of nanolubricants must be obtained before the performance can be optimized.

In order to further investigate the influence of nanoparticle thermal conductivity on refrigerant/lubricant pool boiling, the boiling heat transfer of three R134a/nanolubricant mixtures on a roughened, horizontal, flat (plain), and copper surface was measured. A commercial polyolester lubricant (RL68H)<sup>1</sup> with a nominal kinematic viscosity of 72.3  $\mu\text{m}^2/\text{s}$  at 313.15 K was the base lubricant that was

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<sup>1</sup>Certain commercial equipment, instruments, or materials are identified in this paper in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.

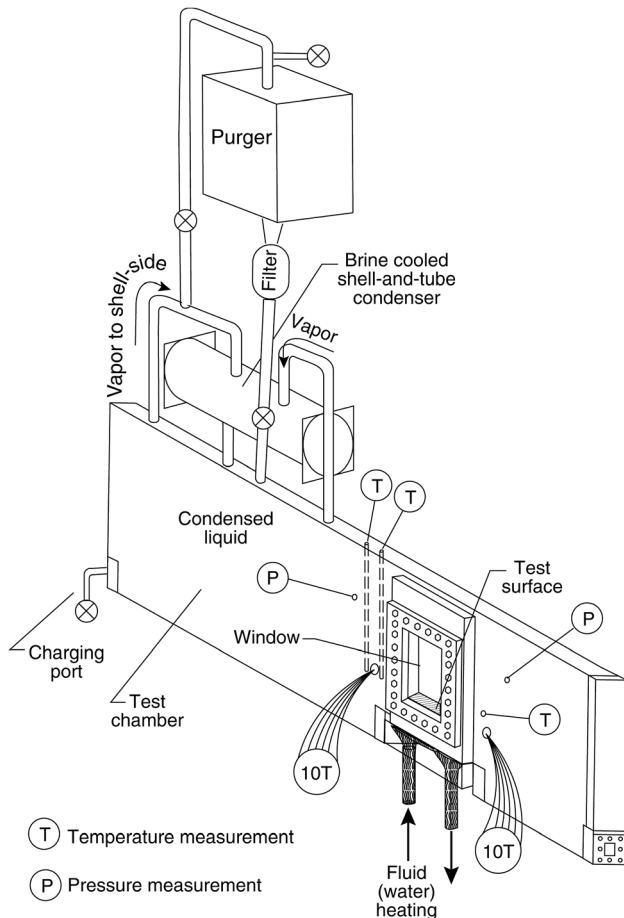


Fig. 1 Schematic of test apparatus

mixed with nominally 10 nm diameter, dry diamond nanoparticles. Diamond nanoparticles have many commercial applications including use in polishing, tooling, and die work. The RL68H/diamond mixture (nanolubricant) was made by ultrasonically mixing commercial diamond nanoparticles without a surfactant with pure RL68HA over a 5 month period. The mixture was made such that 2.6% of the volume was diamond particles. The volume fraction was chosen so that it was large enough to give a large thermal conductivity, while small enough so that the viscosity would be acceptable for compressor applications. The particle size and dispersion were measured with a light scattering technique and a sieving technique and were found to have a wide range of agglomerated particles between approximately 10 nm and 250  $\mu\text{m}$ . The RL68H/diamond (97.4/2.6)<sup>2</sup> volume fraction mixture, a.k.a. RL68H2C, was mixed with pure R134a to obtain three R134a/RL68H2C mixtures at nominally 0.5%, 1%, and 2% RL68H2C mass fractions for the boiling tests. In addition, the boiling heat transfer of three R134a/RL68H mixtures (0.5%, 1%, and 2% mass fractions), without nanoparticles, was measured to serve as a baseline for comparison to the RL68H2C mixtures.

### Apparatus

Figure 1 shows a schematic of the apparatus that was used to measure the pool boiling data of this study. More specifically, the apparatus was used to measure the liquid saturation temperature ( $T_s$ ), the average pool-boiling heat flux ( $q''$ ), and the wall temperature ( $T_w$ ) of the test surface. The three principal components of the apparatus were the test chamber, the condenser, and the purger. The internal dimensions of the test chamber were 25.4 mm  $\times$  257

<sup>2</sup>The equivalent mixture is RL68H/diamond (91.3/8.7) in terms of mass.

mm  $\times$  1.54 m. The test chamber was charged with approximately 7 kg of refrigerant, giving a liquid height of approximately 80 mm above the test surface. As shown in Fig. 1, the test section was visible through two opposing, flat 150 mm  $\times$  200 mm quartz windows. The bottom of the test surface was heated with high velocity (2.5 m/s) water flow. The vapor produced by liquid boiling on the test surface was condensed by the brine-cooled, shell-and-tube condenser, and returned as liquid to the pool by gravity. Further details of the test apparatus can be found in Kedzierski [11,12].

### Test Surface

Figure 2 shows the oxygen-free high-conductivity (OFHC) copper flat test plate used in this study. The test plate was machined out of a single piece of OFHC copper by electric discharge machining. A tub grinder was used to finish the heat transfer surface of the test plate with a crosshatch pattern. Average roughness measurements were used to estimate the range of average cavity radii for the surface to be between 12  $\mu\text{m}$  and 35  $\mu\text{m}$ . The relative standard uncertainty of the cavity measurements was approximately  $\pm 12\%$ . Further information on the surface characterization can be found in Kedzierski [12].

### Measurements and Uncertainties

The standard uncertainty ( $u_i$ ) is the positive square root of the estimated variance  $u_i^2$ . The individual standard uncertainties are combined to obtain the expanded uncertainty ( $U$ ), which is calculated from the law of propagation of uncertainty with a coverage factor. All measurement uncertainties are reported at the 95% confidence level except where specified otherwise. For the sake of brevity, only an outline of the basic measurements and uncertainties is given below. Complete detail on the heat transfer measurement techniques and uncertainties can be found in Kedzierski [13,14], respectively.

All of the copper-constantan thermocouples and the data acquisition system were calibrated against a glass-rod standard platinum resistance thermometer and a reference voltage to a residual standard deviation of 0.005 K. Considering the fluctuations in the saturation temperature during the test and the standard uncertainties in the calibration, the expanded uncertainty of the average saturation temperature was no greater than 0.04 K. Consequently, it is believed that the expanded uncertainty of the temperature measurements was less than 0.1 K.

Twenty 0.5 mm diameter thermocouples were force fitted into the wells of the side of the test plate shown in Fig. 2. The heat flux and the wall temperature were obtained by regressing the measured temperature distribution of the block to the governing

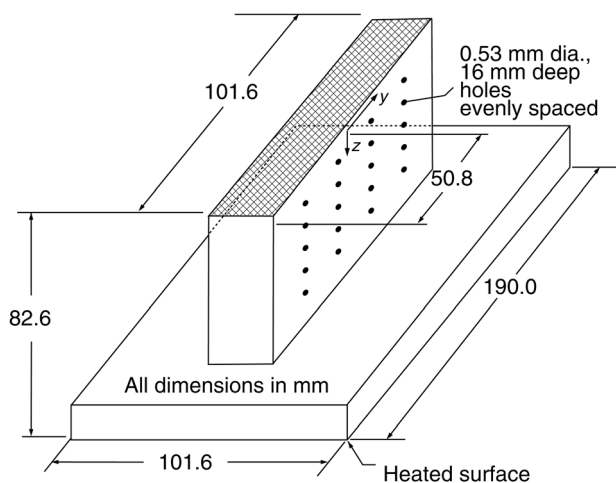


Fig. 2 OFHC copper flat test plate with cross-hatched surface and thermocouple coordinate system

two-dimensional conduction equation (Laplace equation). In other words, rather than using the boundary conditions to solve for the interior temperatures, the interior temperatures were used to solve for the boundary conditions following a backward stepwise procedure given in Kedzierski [15]<sup>3</sup>. The origin of the coordinate system was centered on the surface with respect to the  $y$ -direction at the heat transfer surface. Centering the origin in the  $y$ -direction reduced the uncertainty of the wall heat flux and temperature calculations by reducing the number of fitted constants involved in these calculations.

Fourier's law and the fitted constants from the Laplace equation were used to calculate the average heat flux ( $q''$ ) normal to and evaluated at the heat transfer surface based on its projected area. The average wall temperature ( $T_w$ ) was calculated by integrating the local wall temperature ( $T$ ). The wall superheat was calculated from  $T_w$  and the measured temperature of the saturated liquid ( $T_s$ ). Considering this, the relative expanded uncertainty in the heat flux ( $U_{q''}$ ) was greatest at the lowest heat fluxes, approaching 10% of the measurement near 20 kW/m<sup>2</sup>. In general, the  $U_{q''}$  remained approximately between 3% and 6% for heat fluxes greater than 50 kW/m<sup>2</sup>. The average random error in the wall superheat ( $U_{T_w}$ ) remained between 0.06 K and 0.14 K. Plots of  $U_{q''}$  and  $U_{T_w}$  versus heat flux can be found in Kedzierski [14].

## Experimental Results

**Heat Transfer.** The heat flux was varied approximately between 7 kW/m<sup>2</sup> and 130 kW/m<sup>2</sup> to simulate a range of possible operating conditions for R134a chillers. All pool-boiling tests were taken at 277.6 K saturated conditions. The data were recorded consecutively starting at the largest heat flux and descending in intervals of approximately 4 kW/m<sup>2</sup>. The descending heat flux procedure minimized the possibility of any hysteresis effects on the data, which would have made the data sensitive to the initial operating conditions. Kedzierski [14] presents the measured heat flux and wall superheat for all the data of this study.

The mixtures were prepared by charging the test chamber (see Fig. 1) with pure R134a to a known mass. Next, a measured mass of nanolubricant or lubricant was injected with a syringe through a port in the test chamber. The refrigerant/lubricant solution was mixed by flushing pure refrigerant through the same port where the lubricant was injected. All compositions were determined from the masses of the charged components and are given on a mass fraction basis. The maximum uncertainty of the composition measurement is approximately 0.02%, e.g., the range of a 2.0% composition is between 1.98% and 2.02%. Nominal or target mass compositions are used in the discussion. For example, the "actual" mass composition of the RL68H in the R134a/RL68H (99.5/0.5) mixture was 0.51% ± 0.02%. Likewise, the RL68H mass fractions for R134a/RL68H (99/1) and the R134a/RL68H (98/2) mixtures were 0.98% ± 0.02% and 2.00% ± 0.02%, respectively. Using the same uncertainties, the nanolubricant mass fractions as tested with R134a were 0.49%, 1.02%, and 1.97%.

Figure 3 is a plot of the measured heat flux ( $q''$ ) versus the measured wall superheat ( $T_w - T_s$ ) for the baseline measurements of the refrigerant/pure-lubricant mixtures at a saturation temperature of 277.6 K. The boiling curve for pure R134a, taken from Kedzierski and Gong [16], is shown as the leftmost solid line. The open circles, squares, and stars represent the measured heat flux ( $q''$ ) versus the measured wall superheat ( $T_w - T_s$ ) at a saturation temperature of 277.6 K for the R134a/RL68H (99.5/0.5), R134a/RL68H (99/1), and R134a/RL68H (98/2) mixtures, respectively. Five days of boiling the R134a/RL68H (99.5/0.5) mixture produced 140 measurements over a period of approximately 1 week. The solid lines shown in Fig. 3 are cubic best-fit regressions or estimated means of the data. Four of the 140 R134a/RL68H (99.5/0.5) measurements

<sup>3</sup>For the record, Table 1 provides functional forms of the Laplace equation that were used in this study in the same way as was done in Kedzierski [15] and in similar studies by this author.

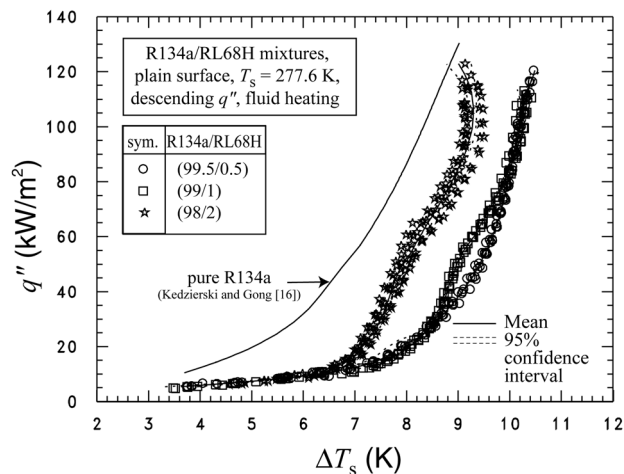


Fig. 3 R134a/RL68H mixtures boiling curves

were removed before fitting because they were identified as "outliers" based on having both high influence and high leverage [17]. Each of the data sets presented here exhibited a similar number of outliers and was regressed in the same manner. Kedzierski [14] gives the constants for the cubic regression of the superheat versus the heat flux for all of the fluids tested here. The residual standard deviation of the regressions—representing the proximity of the data to the mean—is given in Kedzierski [14]. The dashed lines to either side of the mean represent the lower and upper 95% simultaneous (multiple-use) confidence intervals for the mean. From the confidence intervals, the expanded uncertainty of the estimated mean wall superheat was, on average, 0.11 K and 0.05 K for superheats less than and greater than 8 K, respectively. Kedzierski [14] provides the average magnitude of 95% multi-use confidence interval for the fitted wall superheat for all of the test data.

A general overview of the effect that the variation in the pure lubricant mass fraction has on R134a/lubricant pool boiling can be obtained from Fig. 3. Comparison of the three mean boiling curves shows that the superheats are within approximately 1.4 K of each other for the entire tested heat flux range. For the most part, the superheat for the refrigerant/lubricant mixtures is 1 K–3 K greater than that for pure R134a indicating a heat transfer degradation with respect to pure R134a. Kedzierski [18] has shown that, in general, degradations associated with increased lubricant mass fractions occur when the concentration induced bubble size reduction, and its accompanying loss of vapor generation per bubble, is not compensated by an increase in site density. Typically, heat transfer degradations have been observed to increase with respect to increasing lubricant mass fraction. The present measurements are inconsistent with this trend in that the heat transfer performance of the (99.5/0.5) mixture and that of the (99/1) mixture are, for the most part, the same. To add to the inconsistency, the (98/2) mixture exhibits a heat transfer enhancement compared with the (99.5/0.5) and the (99/1) mixtures for heat fluxes greater than 10 kW/m<sup>2</sup>. Although the measurements are inconsistent with typical refrigerant/lubricant data, they are fairly repeatable within this study and with respect to the previous study by Kedzierski and Gong [16]. The boiling curves for the present (99.5/0.5) and the (99/1) mixtures agree with those given by Kedzierski and Gong [16] for the same mixtures on the same surface. However, the superheat for the present (98/2) mixture is approximately 1.5 K less than what was measured in the previous study for the same conditions. The reason for why only two of the three data sets are repeatable between this study and the Kedzierski and Gong [16] study is not known. Kedzierski [18] has shown that refrigerant/lubricant pool boiling with larger lubricant concentrations typically exhibit greater between-run variability. Also, Kedzierski [19] has shown that, on occasion, a boiling surface can perform as if it has two different sets of active cavities on a given



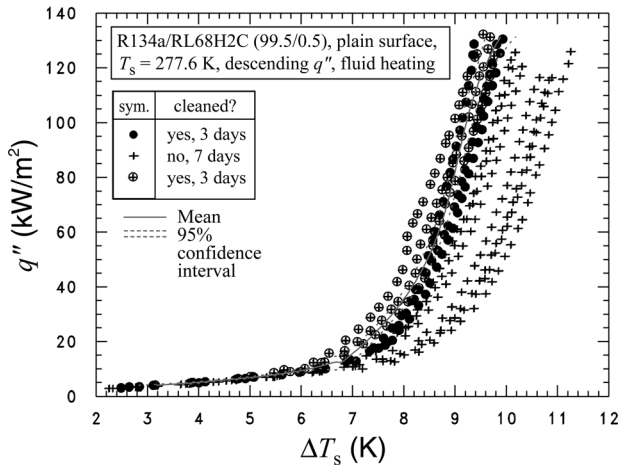


Fig. 4 R134a/RL68H2C (99.5/0.5) mixture boiling curves

day with one being more favorable for boiling than the other. Considering the above, and without a better explanation, the discrepancy between the two (98/2) data sets over roughly a year between measurements can be explained as a potential characteristic of refrigerant/lubricant boiling with greater lubricant concentrations.

Figures 4–6 present the boiling heat transfer measurements for the three refrigerant/nanolubricant mixtures of R134a and RL68H2C. Figures 4 and 6 show that the R134a/RL68H2C (99.5/0.5) mixture and the R134a/RL68H2C (98/2) mixture both exhibit a significant between-run variation manifested in a stratification of the daily boiling curves. In general, the performance for the (99.5/0.5) and the (98/2) refrigerant/nanolubricant mixtures was stable and repeatable for the first few days; following the stable period, the performance degraded for successive test days. However, the R134a/RL68H2C (99/1) does not exhibit this behavior, i.e., the (99/1) daily boiling curves are random with respect to one another and the data scatter are typical of refrigerant/lubricant boiling. A more detailed discussion of these measurements is given in the following.

Figure 4 shows the measured heat flux ( $q''$ ) versus the measured wall superheat ( $T_w - T_s$ ) for the R134a/RL68H2C (99.5/0.5) mixture at a saturation temperature of 277.6 K. Thirteen boiling curves (one for each test day) were measured over the span of approximately a month. The measurements are separated into three groups as represented by the three different symbols used in the plot. The close circles represent measurement made over the first 3 days where the surface was initially exposed to diamond nanoparticles.

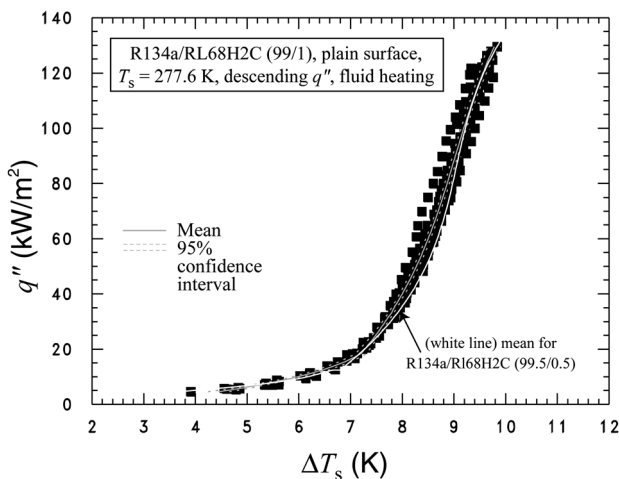


Fig. 5 R134a/RL68H2C (99/1) mixture boiling curves

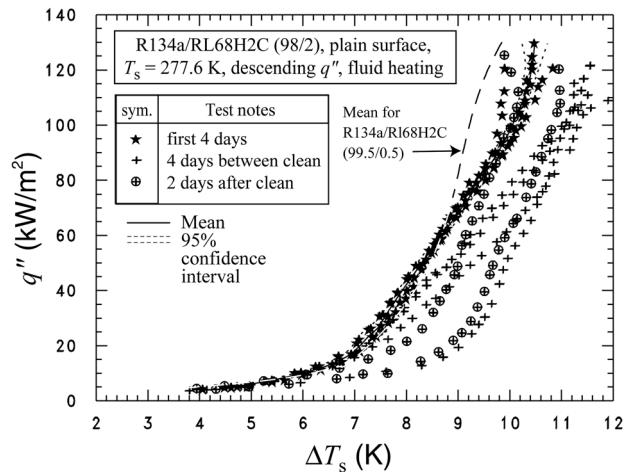
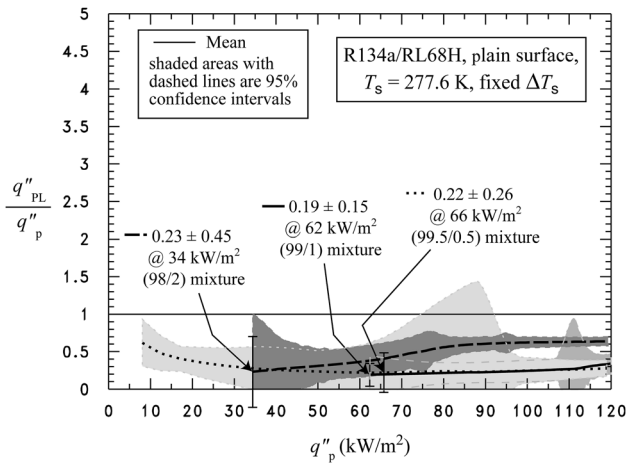


Fig. 6 R134a/RL68H2C (98/2) mixture boiling curves

As represented by the plus symbol, successive measurements resulted in an average gain of approximately 0.2 K in superheat relative to the previous day's measurements, i.e., an increasing degradation in heat transfer performance as the surface experienced more boiling hours. The deterioration of the heat transfer performance with increased usage was believed to be caused by the accumulation of diamond nanoparticles on the surface. To test this hypothesis, the boiling surface was cleaned with acetone and a commercial copper cleaner in an attempt to remove the nanoparticles from the surface. The open circles centered with plus signs represent the boiling measurements that were made over the next 3 days after the surface was cleaned. The measurements that were made after cleaning agree well with those taken after the first 3 days of test for heat fluxes greater than roughly 80 kW/m<sup>2</sup>. For measurements between approximately 10 kW/m<sup>2</sup> and 80 kW/m<sup>2</sup>, the superheat is roughly 0.2 K less for the cleaned surface than for the first 3 days of test. All in all, cleaning has restored the heat transfer performance of the boiling surface. The solid line along with confidence intervals documents the fit of the six best test runs, i.e., three boiling curves after the initial run plus three boiling curves after cleaning the surface. The average expanded uncertainty of the estimated mean wall superheat for the fitted refrigerant/nanolubricant mixture was 0.23 K and 0.13 K for superheat greater than and less than 6 K, respectively.

Figure 5 shows the measured heat flux ( $q''$ ) versus the measured wall superheat ( $T_w - T_s$ ) for the R134a/RL68H2C (99/1) mixture at a saturation temperature of 277.6 K. Eight boiling curves were measured over the span of approximately 2 weeks. Unlike the R134a/RL68H2C (99.5/0.5) measurements, the measurements for the (99/1) mixture exhibited a modest random variation in performance rather than a continuous degradation with respect to test day. For most heat fluxes, the R134a/RL68H2C (99/1) superheat measurements, represented by the closed squares, agree well with the six best measurement days for the R134a/RL68H2C (99.5/0.5) mixture. In fact, the mean boiling curve for the (99.5/0.5) mixture differs from the mean for the (99/1) mixture by no more than approximately 0.2 K. The average expanded uncertainty of the estimated mean wall superheat for the fitted refrigerant/nanolubricant mixture was 0.12 K and 0.06 K for superheat greater than and less than 8 K, respectively.

Figure 6 shows the measured heat flux ( $q''$ ) versus the measured wall superheat ( $T_w - T_s$ ) for the R134a/RL68H2C (98/2) mixture at a saturation temperature of 277.6 K. Ten boiling curves were measured over the span of approximately 3 weeks. The measurements for the (98/2) refrigerant/nanolubricant mixture exhibit a behavior similar to that of the measurements for the (99.5/0.5) refrigerant/nanolubricant mixture. The first 4 days of measurement for the (98/2) mixture, as represented by the closed stars, are fairly repeatable and random between runs. Consequently, the first 4



**Fig. 7** Boiling heat flux of R134a/RL68H mixture relative to that of pure R134a

days were regressed together and are shown as the solid and dashed lines representing the mean and 95% confidence interval, respectively. The average expanded uncertainty of the estimated mean wall superheat for the fitted refrigerant/nanolubricant mixture was 0.17 K and 0.10 K for superheat greater than and less than 7 K, respectively. The plus symbols show measurements taken after the first 4 days at a degraded performance. The open circles centered with plus signs represent the boiling measurements that were made after cleaning the surface. Cleaning restored the performance of the surface for higher heat fluxes for one run but had essentially no effect on the performance for another run.

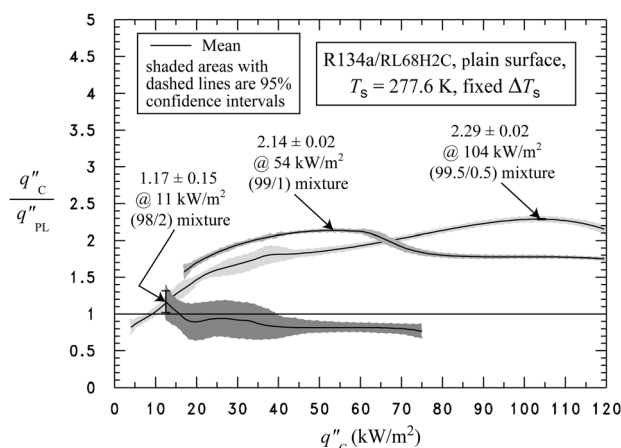
A more precise comparison of the R134a/RL68H and the R134a/RL68H2C heat transfer performances relative to R134a and R134a/RL68H, respectively, is given in Figs. 7 and 8. Figure 7 plots the ratio of the R134a/RL68H mixture heat flux to the pure R134a heat flux ( $q''_{PL}/q''_P$ ) versus the pure R134a heat flux ( $q''_P$ ) at the same wall superheat. Figure 7 illustrates the influence of lubricant mass composition on the R134a/RL68H boiling curve with solid lines representing the mean heat flux ratios for each mixture and shaded regions showing the 95% confidence level for the mean. Overall, lubricant for all compositions has caused a heat transfer degradation relative to the heat transfer of pure R134a for all measured  $q''_P$ . The refrigerant/lubricant pool boiling heat flux is shown to be between roughly 20% and 60% of that of the pure refrigerant. The minimum performance for each refrigerant/lubricant mixture is approximately 20% of the performance of

pure R134a. For example, at roughly  $64 \text{ kW/m}^2$ , the minimum heat flux ratio for the R134a/RL68H (99.5/0.5), and the R134a/RL68H (99/1) mixture was  $0.22 \pm 0.26$ , and  $0.19 \pm 0.15$ , respectively. The minimum heat flux ratio for the R134a/RL68H (98/2) mixture was  $0.23 \pm 0.45$  at approximately  $34 \text{ kW/m}^2$ . Overall, the average heat flux ratio for the R134a/RL68H (99.5/0.5), the R134a/RL68H (99/1), and the R134a/RL68H (98/2) mixture for the heat flux range shown in Fig. 7 for each fluid was 0.27, 0.24, and 0.48, respectively.

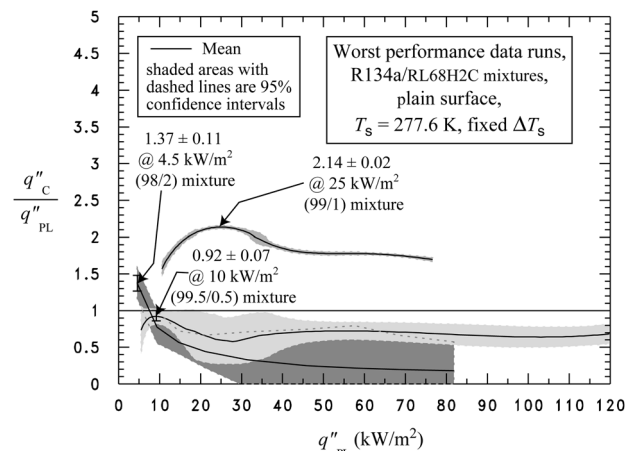
Figure 8 details the effect that the diamond nanoparticles had on the R134a/RL68H boiling curves for the repeatable, best performance runs as described in the discussion of Figs. 4 and 6. The figure plots the ratio of the R134a/RL68H2C heat flux to the R134a/RL68H heat flux ( $q''_C/q''_{PL}$ ) versus the R134a/RL68H2C mixture heat flux ( $q''_C$ ) at the same wall superheat. The three different compositions are represented by three different lines where each R134a/nanolubricant mixture is compared with the R134a/pure-lubricant mixture at the same mass fraction. A heat transfer enhancement exists where the heat flux ratio is greater than one and the 95% simultaneous confidence intervals (depicted by the shaded regions) do not include the value one. Figure 8 shows that the R134a/RL68H2C (99.5/0.5) and (99/1) mixtures exhibit a significant boiling heat transfer enhancement over that of the R134a/RL68H (99.5/0.5) and the R134a/RL68H (99/1) mixtures (without nanoparticles), respectively. The heat flux ratio varies between roughly 0.8 and 2.29 for the R134a/RL68H2C (99.5/0.5) mixture for heat fluxes between  $4 \text{ kW/m}^2$  and  $120 \text{ kW/m}^2$ . The R134a/RL68H2C (99/1) mixture shows a maximum heat flux ratio of approximately 2.14 at approximately  $54 \text{ kW/m}^2$ . The R134a/RL68H2C (98/2) mixture shows a maximum heat flux ratio of approximately 1.17 and a region between  $11 \text{ kW/m}^2$  and  $40 \text{ kW/m}^2$  where no difference can be established between the two fluids because the confidence intervals include the value of one. Overall, the average heat flux ratio for the R134a/RL68H2C (99.5/0.5) mixture and the R134a/RL68H2C (99/1) mixture from approximately  $15 \text{ kW/m}^2$  to  $120 \text{ kW/m}^2$  was 1.98, and 1.91, respectively. The average heat flux ratio for the R134a/RL68H2C (98/2) mixture from approximately  $11 \text{ kW/m}^2$  to  $75 \text{ kW/m}^2$  was 0.81.

The enhancements obtained by Peng et al. [8] of between 1.17 and 1.63 for their diamond nanolubricant boiling with R113 are consistent with the ones presented here. Comparisons of their regressed model with the present measurements were not possible due to an error that was made in the presentation of the coefficients of the model in their paper [20].

Given that the boiling curves showed a surface aging effect for the (99.5/0.5) and the (98.2) mixtures with nanoparticles, Fig. 9 provides the heat flux ratios, as done in Fig. 8, but for the two



**Fig. 8** Boiling heat flux of R134a/RL68H2C mixtures relative to that of R134a/RL68H without nanoparticles for best performance data runs



**Fig. 9** Boiling heat flux of R134a/RL68H2C mixtures relative to that of R134a/RL68H without nanoparticles for worst performance data runs

worst performance runs shown in Figs. 4 and 6. In this way, Figs. 8 and 9 can be used to bracket the observed performance for tests conducted with nanoparticles in this study.<sup>4</sup> The heat flux ratio for the R134a/RL68H2C (99/1) mixture is the same as was presented in Fig. 8 with the exception that it is plotted against the refrigerant/pure-lubricant heat flux rather than the refrigerant/nanolubricant heat flux. The maximum heat flux ratio for worst performing R134a/RL68H2C mixtures was  $0.92 \pm 0.07$  and  $1.37 \pm 0.11$  for the (99.5/0.5) and the (98/2) mixture, respectively. Averaged for heat fluxes between approximately  $5 \text{ kW/m}^2$  and  $80 \text{ kW/m}^2$ , the heat flux ratio for the worst performing R134a/RL68H2C runs for the (98/2) and the (99.5/0.5) mixtures was 0.36 and 0.76, respectively.

**Particle Size.** The size of the diamond nanoparticles in the nanolubricant was measured with a dynamic light scattering (DLS) technique using a 633 nm wavelength laser and a sieving technique using a syringe filter and an optical microscope. The two methods were required because the diamond nanolubricant was very polydispersed and contained particles that were larger than what could be measured with the DLS technique.

The DLS technique was used to measure particles sizes less than 1000 nm. Approximately 0.01 g of the nanolubricant was mixed with approximately 3 g of toluene and pushed through a  $1 \mu\text{m}$  syringe filter into the sample cuvette that was analyzed with the DLS system. An index of refraction of 2.42 for diamond was used in the Brownian motion based calculation that was done internally by the DLS instrument for the particle size. The uncertainty of the packaged DLS instrumentation was confirmed with a National Institute of Standards and Technology (NIST) traceable 60 nm nanofluid standard. The measured diameter of the standard with the DLS system was within 5 nm of range of uncertainty of the standard. For the diamond nanolubricant, for particles smaller than  $1 \mu\text{m}$ , the 10 nm diameter<sup>5</sup> diamond nanoparticles were agglomerated primarily into two clumps of 30 nm and 400 nm with a bandwidth of approximately  $\pm 60\%$  for each of approximately equal intensities. Consequently, for particles smaller than  $1 \mu\text{m}$ , the nanolubricant had nearly an equal number of nearly discrete diamond nanoparticles (single or clumps of 2–5 particles), and agglomerated clumps of approximately 40 particles.

The  $1 \mu\text{m}$  syringe filter material used above was examined with an optical microscope. Figure 10 shows the diamond particles that can be seen in the filter are between roughly  $10 \mu\text{m}$  and  $50 \mu\text{m}$ . However, particle agglomeration as large as  $250 \mu\text{m}$  was observed when the mixture was left to settle over several weeks. Jillavenkatesa et al. [21] show that it would require thousands of size measurements to obtain an uncertainty in the size distribution for a 95% confidence level. Consequently, only an approximate range for the particle size is given here.

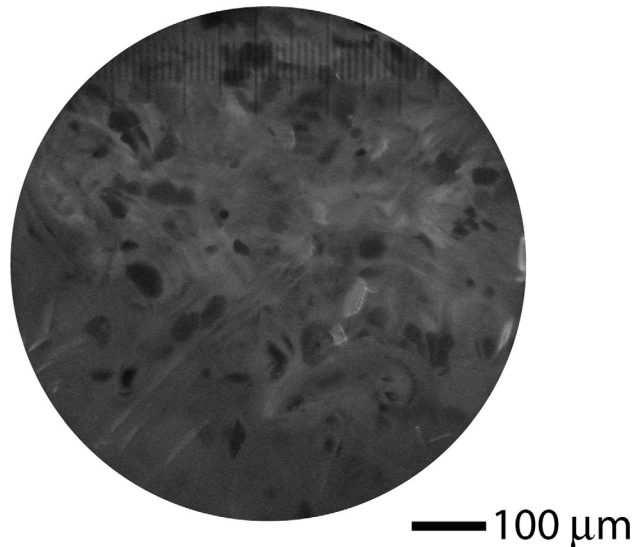
Combining the results of the two measurement methods shows that the nanolubricant is not a good dispersion. The particles are dispersed from single 10 nm diameter particles to agglomerations of particles as large as  $50 \mu\text{m}$ . The large clumps of particles are expected to settle and further agglomerate as demonstrated by the  $250 \mu\text{m}$  agglomerations observed in the aged sample. In other words, the diamond nanolubricant dispersion made in this study was not stable and experienced settling over time.

## Discussion

As far as refrigerant boiling is concerned, there are two direct consequences that an agglomerated nanolubricant-dispersion has on heat transfer. The first is that the polydispersed nanolubricant has a very large viscosity: approximately  $450 \mu\text{m}^2/\text{s}$  at 313.15 K, which is nearly a 550% increase over the viscosity of the pure

<sup>4</sup>Because of the consistent trend of the stratified data, it is likely that a larger degradation would have been exhibited by the worst performing mixtures had more data runs been taken.

<sup>5</sup>The 10 nm particle diameter was specified by the manufacturer of the nanopowder that was used to make the nanolubricant.



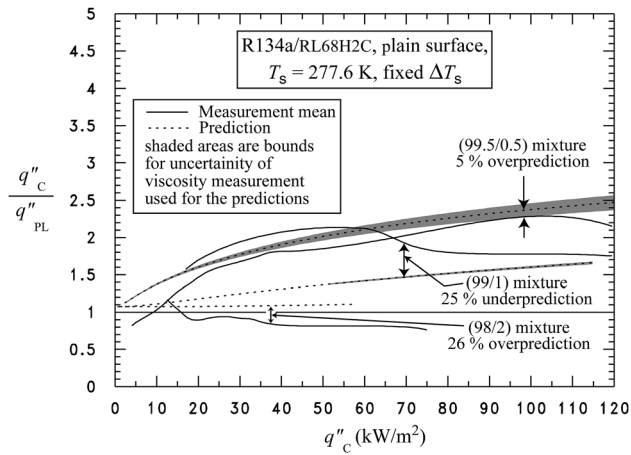
**Fig. 10 Agglomerated diamond nanoparticles in syringe filter material**

lubricant [14]. Kedzierski [18] showed that increases in lubricant viscosity resulted in improvements in boiling heat transfer. From this, it is shown below that the viscosity increase of the diamond nanolubricant is largely responsible for the boiling heat transfer enhancement associated with this particular refrigerant/nanolubricant mixture. The second consequence of a poor dispersion is that it can cause a heat transfer instability if the particles are large enough to settle over time despite the vigorous mixing as caused by the boiling.

A previous study [6] suggested that nanoparticle interaction with bubbles was the main boiling enhancement mechanism of boiling refrigerant/CuO-nanolubricant mixtures. In order for the interaction to occur, the nanoparticles must remain suspended in the lubricant excess layer that resides on the boiling surface. Nanoparticles that become lodged in surface cavities are likely to degrade heat transfer performance via loss of bubble nucleation sites. Particles that are not governed by Brownian motion, i.e., particles that are too large, are not likely to provide favorable and stable interaction with bubbles. Over time, the larger particles are likely to agglomerate and settle onto the boiling surface. The worsening performance over time exhibited by the (99.5/0.5)-nanolubricant mixture and the (98/2)-nanolubricant mixture is speculated to be a direct result of particle settling over time.

The above argument suggests that nanoparticle interaction with bubbles is not likely to be responsible for the heat transfer enhancements that were observed for the refrigerant/diamond-nanolubricant mixtures. Considering that diamond-nanoparticle interaction is not the governing heat transfer enhancement mechanism, then is it possible for increased nanolubricant viscosity to be the cause? In an attempt to demonstrate that it is, Fig. 11 compares the enhancement ratio for the R134a/nanolubricant mixtures to those predicted by the refrigerant/lubricant mixture, pool-boiling model given in Ref. [22]. The dashed lines of Fig. 11 are predictions from the refrigerant/lubricant pool boiling model while using the properties of the refrigerant and the measured fluid properties of the nanolubricant [14] evaluated at the 2.6% diamond volume fraction. Overall, the model predicts the relative stratification of the boiling measurements for the three mixtures with the smaller nanolubricant mass fractions having the best improvement. In addition, fairly good absolute agreement was achieved between measurements and predictions for each nanolubricant mass fraction. For example, the heat flux ratios for the (99.5/0.5) mixture and the (98/2) mixture were overpredicted, on average, by roughly 5% and 26%, respectively. Similarly, the (99/1) mixture is underpredicted by about 25% on average. Given that the





**Fig. 11 Predicted heat flux ratio for RL68H2C (99.5/0.5) mixture using Kedzierski [22] model compared with measurement means**

model cannot account for nanoparticle interaction with bubbles, the agreement suggests that the boiling heat transfer enhancement with the nanolubricant of this study was due mainly to favorable changes in fluid properties, i.e., increases in liquid viscosity and thermal conductivity<sup>6</sup> with respect to the pure lubricant.

At this point, it is appropriate to address the stable performance of the R134a/RL68H2C (99/1) mixture and how it relates to the above discussion. It was noted by Jillavenkatesa et al. [21] that the particle distribution in a nanofluid has the potential for significant variation. Considering this and the wide range of particle size of the diamond nanolubricant of this study, it may have been possible that the nanolubricant that was used to make the R134a/RL68H2C (99/1) mixture had less particle agglomeration than what was used to make the other two mixtures even though it was taken from the same batch. If this were the case, there would be less particle settling, which would lead to greater measurement repeatability between runs. In addition, the boiling heat transfer performance was on average 25% greater than what was predicted by the model based on refrigerant/lubricant properties. Potentially, that 25% difference, if it is not due to prediction errors, could be due to particle interaction as caused by a better nanofluid suspension.

Future research is required to investigate the influence of the particle material, its shape, size, distribution, and concentration on refrigerant boiling performance. In particular, the importance of having a good dispersion in promoting nanoparticle-bubble interaction should be verified with further refrigerant boiling studies with well-dispersed diamond nanolubricants. Sarkas [23] suggests that this may be more easily said than done because the surface chemistry of diamond may be well suited for encouraging particle agglomeration. As a result, the surface chemistry of diamond nanoparticles must be altered (e.g., hydrogen-atom terminated bonds [23]) before it can become a viable material for chiller nanolubricants. Further investigation in this area and with other nanoparticle materials may lead to a theory that can be used to develop nanolubricants that improve boiling heat transfer for the benefit of the refrigeration and air-conditioning industry.

## Conclusions

The effect of diamond nanoparticles on the boiling performance of R134a/polyolester mixtures on a roughened, horizontal flat surface was investigated. A nanolubricant containing diamond nanoparticles at 2.6% volume fraction with a polyolester lubricant was mixed with R134a at three different mass fractions. Because the nanolubricant was polydispersed with relatively large agglomer-

<sup>6</sup>As shown in Ref. [14], the nanoparticles resulted in an approximate 7% increase in the thermal conductivity of that of the pure lubricant.

ated particles, the boiling heat transfer performance was shown to degrade with time for two out of three of the test mixtures and was likely due to particles filling nucleation sites. As a result, measurement comparisons were made for both the worst and the best-case heat transfer performances. For the best case, the 0.5% nanolubricant mass fraction, the diamond nanoparticles caused a heat transfer enhancement relative to the heat transfer of pure R134a/polyolester (99.5/0.5) on average of 98%. A similar enhancement was observed for the R134a/nanolubricant (99/1) mixture, which had a heat flux that was on average 91% larger than that of the R134a/polyolester (99/1) mixture. Further increase in the nanolubricant mass fraction to 2% resulted in a boiling heat transfer degradation of approximately 19% on average for the R134a/nanolubricant (98/2) mixture. For the worst case, the R134a/nanolubricant (98/2) mixture and the R134a/nanolubricant (99.5/0.5) mixture exhibited a 64% and a 24% degradation, respectively. The R134a/nanolubricant (99/1) mixture performance did not degrade over time, and as a result exhibited a consistent 91% enhancement.

From the results of the present study, it is speculated that if a good dispersion of nanoparticles in the lubricant is not obtained, then the agglomerated nanoparticles will not provide interaction with bubbles, which is favorable for heat transfer. The clumps of nanoparticles can provide heat transfer enhancement via purely fluid property effects, i.e., increases in liquid viscosity and thermal conductivity. These property improvements persist only as long as the particles remain suspended in the lubricant excess layer. Reductions in boiling heat transfer performance increase as the agglomerated nanoparticles settle out of the excess layer and into the cavities of the boiling surface. For this reason, unless the surface chemistry of the diamond nanoparticles can be altered, diamond-nanolubricants do not appear to be a viable material for chiller applications.

The present study demonstrates the necessity of having the particles well dispersed in the nanolubricant so that the particles remain suspended to provide a favorable performance and induce interaction with bubbles for further boiling heat transfer enhancement.

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## Nomenclature

### Symbols

$q''$  = average wall heat flux,  $W m^{-2}$

$T$  = temperature, K

$T_w$  = temperature at roughened surface, K

$U$  = expanded uncertainty

$u_i$  = standard uncertainty

### Greek Symbols

$\Delta T$  = temperature difference, K

$\Delta T_s$  = wall superheat:  $T_w - T_s$ , K

### Subscripts

C = diamond nanoparticles

p = pure R134a

PL = R134a/RL68H mixture

$q''$  = heat flux  
 $s$  = saturated state  
 $T_w$  = wall temperature  
 $w$  = wall

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