ON POOL BOILING OF NANOFLUIDS

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ABSTRACT

Past research has shown contradicting trends in Pool Boiling Heat Transfer Coefficient (PBHTC) of Nanofluids. An experimental study has been conducted to investigate the effect of the preparation techniques on the PBHTC. Nanofluids made of water and alumina nanoparticles have been used. The effects of electrostatic stabilization (ES) and preparation of nanofluids from dry particles or from ready-made suspensions have been investigated. It is found that ES has a significant effect on the PBHTC of nanofluids prepared from dry particles.

INTRODUCTION

During the past decade there has been an exponential interest in nanofluids boiling. Particle suspensions made of micro- and millimeter size particles resulted in erosion, sedimentation and fouling problems. The emergence of nanotechnology has made synthesis of particles of nanometer scale possible and affordable. With smaller particles the drawbacks of particle-suspensions become less significant.

Most working fluids used in industry have a thermal conductivity an order of magnitude lower than the metals used in heat exchange equipment. That is why working fluids are considered the bottleneck in heat exchanges effectiveness. The key property that makes nanofluids attractive is their enhanced thermal conductivity, as compared with their base fluids. It is believed that nanofluids could be the next generation of working heat transfer fluids. Figure 1 shows the rate of increase in research in nanofluids over the past few years.

Xuan et al. [1] measured the thermal conductivity of nanofluids prepared from copper nanoparticles dispersed in water and transformer oil. A 75% enhancement in the thermal conductivity was recorded for 7.5 % vol. particle concentration of the copper-water nanofluids. Moreover, nanofluids have a temperature dependant thermal conductivity, which increases with temperature. Das et al. [2] measured the thermal conductivity of nanofluids at room temperature and at 51°C.
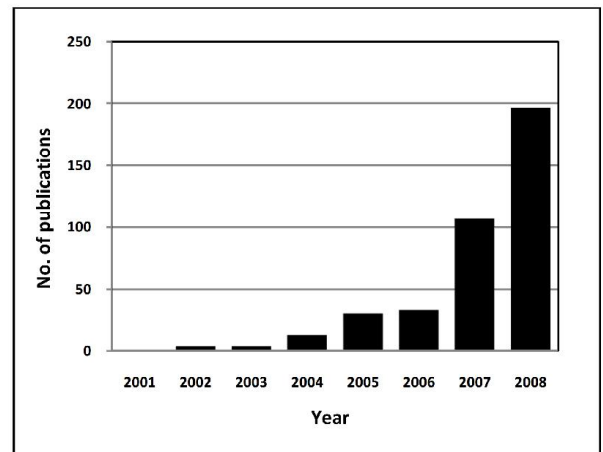
Figure 1: Number of publications in nanofluid research over the last eight years.

NOMENCLATURE

Symbol	Units	Description
$\emptyset_v$	[-]	Volumetric concentration
$\emptyset_m$	[-]	Mass concentration
ρ	$[kg/m^3]$	Density
q''	$[W/m^2]$	Heat Flux
μ_l	$[Ns/m^2]$	Liquid viscosity
h_{lv}	$[J/kg]$	Latent heat of vaporisation
σ	$[N/m]$	Surface tension
g	$[m/s^2]$	Gravitational acceleration
Pr_l	[-]	Liquid Prandtl number
C_{pl}	$[J/kg°C]$	Specific heat
C_{sf}	[-]	Surface factor
$T_s - T_{sat}$	$[°C]$	Surface superheat
R_a	$[m]$	Average surface roughness
d_p	$[m]$	Particle size

Subscripts

p	Particle
f	Fluid
l	Liquid
v	Vapour

Nanofluids prepared from alumina (38.4nm) and copper oxide (28.6nm) nanoparticles dispersed in water were used. They found the thermal conductivity of the 4 vol. % alumina nanofluids to increase from 9.4% to 24.3% when the temperature was raised from 21°C to 51°C. The thermal conductivity of the copper oxide nanofluids at the same concentration increased from 14% to 36% for the same temperature rise. They attributed the thermal conductivity enhancement to the Brownian motion of the particles, which is intensified at higher temperatures. This temperature-dependant thermal conductivity makes nanofluids more attractive for thermal applications operating at high temperatures.

Nanofluids have been prepared using several techniques by researchers. One of the most important issues when using nanofluids is the "stability" of the nanoparticles in the fluid. The nanoparticles have a tendency to form larger aggregates as well as settle or "crash-out" of the fluid. Surfactants such as laureate salts may be added to enhance the stability of the nanofluids, as well as changing the pH value.

Research in pool boiling of nanofluids has shown contradicting trends. The first work on pool boiling of nanofluids was carried out in 1984 by Yang and Maa [3]. They used a horizontal 3.2 mm diameter cylindrical rod as their heater element and employed a magnetic stirrer to mix the solution and prevent the particles from sedimentation. Alumina nanoparticles of 50, 300 and 1000 nm in size were used at concentrations of 0.1 – 0.5 wt. %. (equivalent to 0.03 – 0.14 vol. %). Their results show an increase in the Pool Boiling Heat Transfer Coefficient (PBHTC) of nanofluids with respect to water. The PBHTC increased with particle concentration to a maximum enhancement of 4 times that of water. They also showed that the PBHTC was enhanced with decreasing particle size. They did not provide a detailed discussion of this trend; however, they attributed the enhancement in the PBHTC to the effect of the nanoparticles on the thermal boundary layer. There was no mention of how Yang and Maa prepared their nanofluids or the heater surface properties.

Das et al. [4] carried out a similar experimental investigation using tubular heaters of 20 mm diameter, and different surface roughness, of 400 nm and 1150 nm Ra. Alumina nanofluids of 0.1 – 4 vol. % concentrations were used. The nominal particle size was 38 nm. Because surfactants change the surface tension of nanofluids, which alters their boiling characteristics, surfactants were not used as a stabilizing agent in this study. Dry nanoparticles were mixed with water and ultrasonic vibration was used for four hours prior to each experiment to break down particle agglomerates. Their results showed that the PBHTC was reduced, varying from 0.4 to 0.8 that of pure water. A considerable deterioration of PBHTC was found with concentrations as low as 0.1 vol. %. This deterioration increased with increasing the concentration, however, not proportionally. The smoother surface (Ra = 400 nm) showed a higher deterioration in the heat transfer than did the rougher surface (Ra = 1150 nm). A layer of nanoparticles was found on the heater surface. Measurements of surface roughness after the experiments revealed that surface roughness decreased because of the nanoparticles deposition. The deterioration of the heat transfer was attributed to the particles size being one or two orders of magnitude smaller than that of

the surface roughness, which caused the particles to settle in the surface voids; making the heater surface smoother. These results were opposite to the expected trend that the PBHTC would increase due to the enhanced thermal conductivity of the nanofluids and its effect on the bubble sliding mechanism where the fluid thermal conductivity plays a major role. This expected effect was apparently overshadowed by the effect of particles deposition on the heater, which changed the surface characteristics.

Bang and Chang [5] investigated the pool boiling phenomenon of nanofluids on horizontal and vertical flat surfaces. Their surfaces were very smooth having a surface roughness of 37 nm Ra, which is almost equal to the size of their nanoparticles of 47 nm on average. Alumina nanoparticles and water were mixed to concentrations of 0.5 – 4 vol. %. Ultrasonic vibration was used for eight hours to break down the nanoparticles agglomerates. An electrical mixer was used to enhance the distribution of the nanofluids; however, its use did not show any significant effect on the results. Their results showed deterioration in the PBHTC ranging from 0.25 to 0.5 that of pure water. This deterioration was found to increase with increasing nanofluid concentration. They also found that nucleate boiling regime started at wall temperatures higher than that in the case of pure water. Bang and Chang compared their results to the Rohsenow correlation [9] to validate their experimental setup. The Rohsenow correlation is widely used among boiling researchers to validate nucleate boiling results. The correlation accounts for liquid and boiling surface properties to predict the boiling heat flux for a given surface temperature. Their pure water boiling results were found to agree with the Rohsenow correlation whereas the nanofluids boiling results deviated from the correlation.

Similar to the findings reported in [4], Bang and Chang found a layer of nanoparticles deposited on the boiling surface after the experiment, which was easily but not completely removable using a water jet. They argued that the settling of nanoparticles altered the surface characteristics, which explains the deviation of their results from the Rohsenow correlation. Their experimental setup included an observation window, which allowed them to observe less bubble generation with nanofluids than with pure water. These observations indicated that the number of nucleation sites decreased with nanofluids.

Liu and Liao [6] investigated the effect of surfactants to stabilise nanofluid solutions on their boiling characteristics. They used water and alcohol based fluids with 50 nm copper oxide (CuO) particles and 35 nm silica particles, respectively. Concentrations from 0.07 – 0.7 vol. % were used. Sodium dodecyl benzene sulphate (SDBS) was used as the surfactant. The nanofluids were sonicated in an ultrasonic bath for twelve hours before every experiment. A smooth horizontal surface was used as the heater surface with Ra value about 200 nm.

When the surfactant was used with the water based nanofluids a layer of nanoparticles was found to form and adhere to the surface which was not removable by a water jet. The sorption layer caused a surface temperature fluctuation of ±2°C at surface temperatures above 112°C indicating that the surfactant broke down and the stability of the nanofluids was severely reduced. When no surfactant was used, the boiling performance was found to be close to that of pure water. A

minor enhancement in the nucleate boiling regime was recorded.

The first study that employed the electrostatic stabilisation (ES) method was the study carried out by Wen and Ding [7]. The main concept of the ES method is to adjust the pH value of the nanofluid away from the iso-electric point of the nanoparticles. An electric double layer forms around the nanoparticles which creates repulsion forces between the particles and maintain the stability of the nanofluid. The iso-electric point of alumina is pH = 9.1, so they maintained a pH of 7 during their experimental investigation. A horizontal heater surface of about 3000 nm Ra surface roughness was used as the heater surface. Alumina nanoparticles, 10 – 50 nm in size, were mixed with water to concentrations of 0.1 – 0.35 vol. %. Their results showed an increase in the PBHTC with higher concentrations. A maximum enhancement of 40% was recorded at the highest concentration of 0.35 vol. %. They observed almost no particle deposition on the heater surface after the experiments.

Das et al. [8] investigated the enhancement and deterioration of the PBHTC of nanofluids. Alumina nanoparticles of 47 and 150 nm in size were used at concentrations of 0.15 - 0.57 vol. % with water as the base fluid. A vertical tubular heater was used with a surface roughness of 48, 98 and 150 nm. The nanofluids were electrostatically stabilised by adjusting the pH value to 5.5. Ultrasonic vibration was also used to break down particle agglomerates. They found that the largest deterioration in heat transfer was achieved when the particle size was close to that of the surface roughness. They concluded that when the size of the particles and the size of the surface roughness are comparable, the particles settle in the voids of the surface and block the nucleation sites. The PBHTC was enhanced when the particles were either much larger or much smaller than the size of the heater surface roughness. Das et al. argued that when the heater surface roughness is much larger than the particles, the particles would settle in the voids and multiply the nucleation sites. They introduced a "Surface Interaction Parameter" defined as

$$R_a / d_p$$

to compare the size of the particles and the surface roughness. They found a maximum deterioration at $R_a/d_p=1$ and a concentration of about 0.57 vol. %, giving a PBHTC 90% that of water. They also recorded a maximum enhancement of heat flux at $R_a/d_p=11$ and a concentration of 0.57 vol. % of about 180 % that of pure water.

Considering the above literature review, researchers have reported contradicting trends of enhancement and deterioration in the PBHTC of nanofluids with respect to pure water. The deterioration was attributed to particles settling on the surface, creating an insulation layer and blocking the nucleation sites. The enhancement was attributed to the enhanced thermal conductivity of the nanofluids, to the method of stabilisation of the suspension, which prevented the particles from settling on the heater surface, and to the fact that particles settle in larger voids tend to multiply the number of nucleation sites.

The use of surfactants has proven to be impractical in stabilising the nanofluids as reported by Liu and Liao [6]. As mentioned before, some investigations attributed the enhancement in the PBHTC to the improved stability of the nanofluids. To the best of our knowledge, no study has investigated the effects of the pH value or the preparation method of nanofluids from dry or ready-made suspensions on their boiling performance. This paper provides some preliminary results of an experimental study carried out at the Thermal Processing Laboratory considering these effects.

EXERIMENTAL APPARATUS

The boiling apparatus used in this investigation is shown in Figure 2. The boiling vessel consists of a large stainless steel
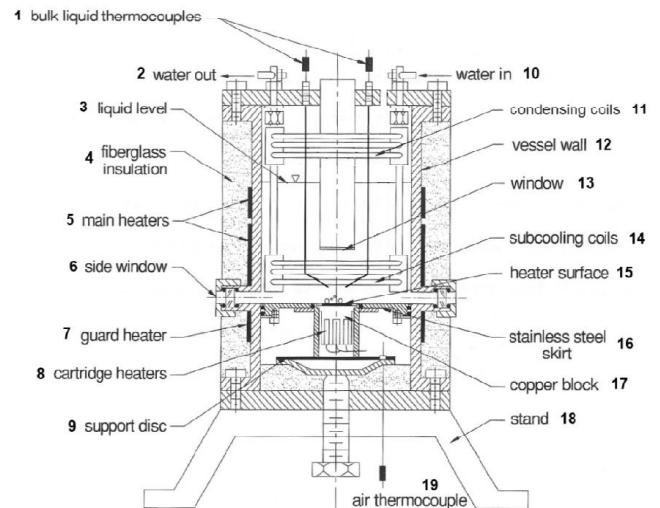

Figure 2: Experimental apparatus

pipe of 20 cm diameter (12). A stainless steel skirt (16) was installed to support the fluid. Two main heaters (5) were used to heat the working fluid to the saturation temperature. A copper block (17) of 25.4 mm diameter was fixed to the stainless steel skirt and served as the heating surface. Three high-power-density heaters (8) were installed in the copper block which could provide a maximum heat flux of 1480 [KW/m²]. Three E-type thermocouples were installed in the copper block at different axial positions beneath the copper block surface to determine the axial temperature distribution. Linear extrapolation was used to determine the temperature gradient in the copper block as well as its surface temperature. Error analysis has been carried out showing an estimated error of 4.3 % in the heat flux measurement and a 0.9°C error in the surface temperature.

A guard heater (7) beneath the stainless steel skirt (16) was installed to heat the air around the copper to reduce radial heat losses from the block. A support disk (9) was used to trap the hot air around the copper block. A thermocouple (19) was installed to monitor the air gap temperature around the copper block. Two bulk liquid thermocouples (1) were inserted from the top of the vessel to record the bulk liquid temperature. Two cooling coils (11, 14) were installed; one for liquid subcooling (which was not used in this study) and another for vapour condensation. Vapour condensation was critical so as to not change the concentration of the nanofluids with too much evaporation taking place during the experiments. Small

openings in the top disk maintained atmospheric conditions inside the vessel. The thermocouples were connected to a Kiethley Data Acquisition System Model 2700 which was connected to a personal computer, and temperature readings were sampled at 5 second intervals.

NANOFLUIDS PREPARATION

Alumina nanoparticles of 45 nm nominal particle size were acquired in dry form and concentrated ready-made suspension in water from Nanophase Technologies Incorporation. The ready-made suspensions were prepared by the manufacturer months before the experiments were carried out. The conversion from volumetric to mass concentrations was carried out using equation (1).

$$\emptyset_V = \frac{1}{\left(\frac{1-\emptyset_m}{\emptyset_m}\right)\frac{\rho_p}{\rho_f}+1} \tag{1}$$

Dry nanoparticles or ready-made suspension were added to water to obtain the required concentration. Hydrochloric acid was added to the suspension to adjust the pH of the nanofluid. The nanofluids were then placed in an ultrasonic bath at 40KHz for five hours prior to each experiment. The ultrasonic vibration was used to break down particle agglomerate as nano-sized particles have a high tendency to agglomerate due to strong Van der Waals forces [7].

EXPERIMENTAL PROCEDURE

The boiling vessel was filled with five liters of fluid. The main and guard heaters were switched on to heat the water/nanofluids to the saturation temperature. The main heaters would then be controlled to maintain the bulk liquid temperature at the saturation temperature. The cartridge heaters in the copper block would then be switched on and controlled at different levels of heat flux and the entire setup would be allowed to reach steady-state. The guard heaters were controlled to maintain the air temperature around the copper block close to the block average temperature in order to minimize radial heat losses.

Prior to every experiment, the boiling surface was polished to ensure that no nanoparticles were sticking to the boiling surface which may alter the surface characteristics from one experiment to the other. Surface roughness measurements were taken before and after each experiment using a Zygo white light interferometer. This method of surface roughness measurement provides non-contact surface roughness measurement. Conventional contact surface roughness measurement techniques have the disadvantage of potentially scraping through the deposited nanoparticles on the surface and giving inaccurate measurements. Therefore, non-contact surface roughness measurement is advantageous for this study. Pre- and post-boiling surface roughness measurements were used as an indication to the degree, if any, of particle deposition on the surface after the experiments.

Nanofluids at 0.1 vol. % concentration were prepared from dry particles as well as ready-made suspensions. Acid was added to the nanofluids to electrostatically stabilize them by adjusting the pH to 5. Water boiling experiments at both a neutral pH of 6.5 and an acidic pH of 5 were conducted to relate the boiling performance of the nanofluids. Table 1 provides details of the experiments reported in this paper.

RESULTS AND DISCUSSION

Validation of the Experimental Setup

Results of boiling heat flux (BHF) of pure water have been compared to the BHF calculated using Rohsenow's correlation [9] shown in Equation (2).

$$q'' = \mu_l h_{lv} \left[\frac{\sigma}{g(\rho_l - \rho_v)}\right]^{-1/2} Pr_l^{-s} \left[\frac{c_{pl}}{h_{lv}c_{sf}}(T_s - T_{sat})\right]^{1/r} \tag{2}$$
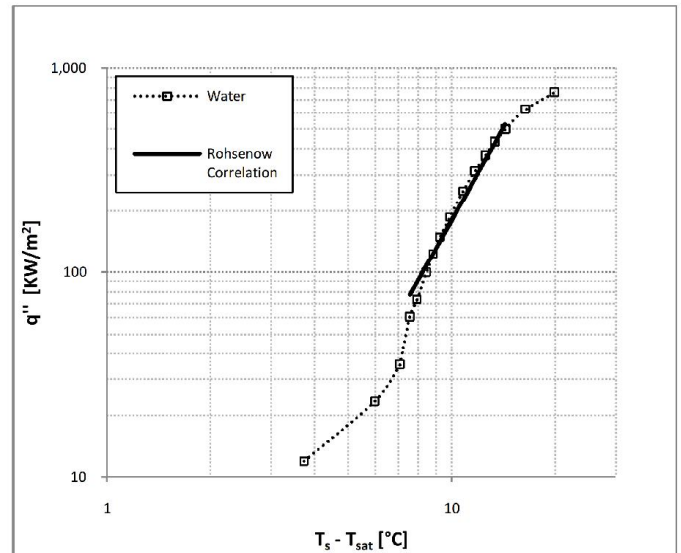

Figure 3: Comparison of present boiling heat flux (BHF) of pure water and BHF calculated using equation (2)

Rohsenow suggested s = 1 and r = 0.33 for water and C_{sf} = 0.0128 for emery polished copper. It can be seen in Figure 3 that the data fits the correlation well in the range of 8 to 15 °C surface superheat. The value for C_{sf} in this work that best fits the data has been found to be 0.0177. The present data deviates from the correlation at surface temperatures below the incipience of boiling. The deviation from the correlation at surface temperatures larger than 15 °C is due to the boiling becoming very vigorous and slugs and columns of vapor form and give lower heat fluxes. Pure water boiling experiments were carried out at a heat flux ≤ 800 KW/m² as CHF was found to occur above that range.

Effect of the Electrostatic Stabilization Method

Four cases of nanofluids have been investigated in this study, as shown in Table 1. The boiling performance of nanofluids is shown in Figure 4. The graph indicates that boiling nanofluids resulted in deterioration of the PBHTC of nanofluids with respect to pure water. The deterioration in the PBHTC for the nanofluids compared with pure water is expected. This is because the size of surface cavities is very close to the particle size. Das et al. [8] suggested that deterioration in the PBHTC is expected when the "surface interaction parameter" is close to unity. The value of R_a/d_p in the present study is close to 1.

The percentage deterioration in the PBHTC evaluated at a surface superheat of 15 °C is shown in Table 1. A deterioration in the PBHTC of about 50% was found for the cases of

Table 1: Nanofluids cases and the corresponding experimental results

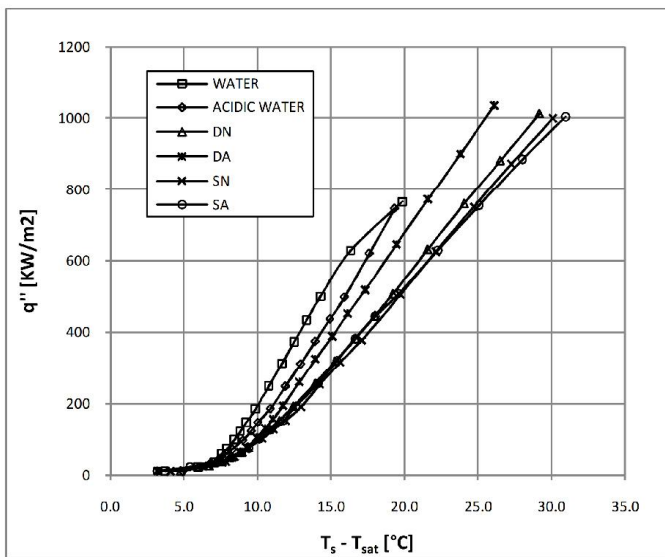| Case Designation	Nanofluid Prepared from	pH	PBHTC % of Water @ 15°C	Post-Boiling Surface Roughness	Effective Particle size
DN	Dry	Neutral (6.5)	55	6000-7500	215
DA	Dry	Acidic (5)	69	400-500	180
SN	Suspension	Neutral (6.5)	51	2500-3500	170
SA	Suspension	Acidic (5)	54	300-500	150

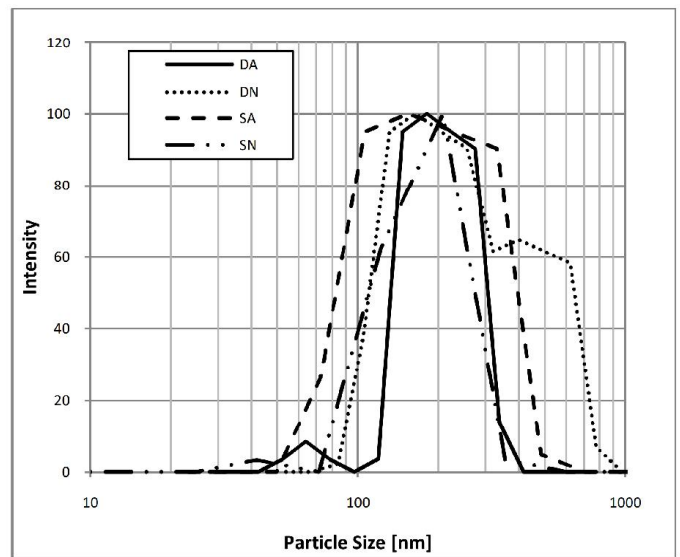**Figure 4:** Water and nanofluid boiling curves

Figure 5: Particle size distributions

nanofluids prepared from ready-made suspensions and dry particles at pH = 6.5 (SN, SA, and DN). The case of nanofluids prepared from dry particles with an acidic pH (DA) resulted in only 31% deterioration, i.e., the heat flux of this case is 25 - 35 % higher than that of the other three cases.

Experiments using water at an acidic pH of 5 were also carried out in order to investigate the effect of the pH value on the boiling performance of water. The PBHTC of acidic water was 80 % that of pure water. This reduction in PBHTC is because the surface tension of hydrochloric acid is higher than that of water. A higher surface tension will increase the vapor bubble diameter and therefore reduce the bubble departure frequency, which in turn reduces the heat flux at a given surface superheat.

Examining the surface roughness after each experiment has revealed some interesting results. A layer of white nanoparticles was seen by the naked eye on the surface after the experiments of the neutral nanofluids (DN and SN). This layer was attached to the surface as it was not possible to wash it away with a water jet. The nanoparticles could be removed upon rinsing the surface by hand under a water jet. Much less particle deposition was visually observed to settle on the surface after boiling the acidic nanofluids (DA and SA).

The value of the surface roughness in terms of Ra before all experiments was found in the range between 100 and 150 nm. Surface roughness measurements after pure water experiments did not show any appreciable change, regardless of the pH value. Post-boiling surface roughness measurements showed that particle deposition in the cases of neutral pH nanofluids (DN and SN) is far more significant than the cases of acidic nanofluids (DA and SA). As shown in Table 1, the surface roughness was found in the rage of 6000 - 7500 nm and 2500 - 3500 nm for cases DN and SN, respectively. The value of post-boiling surface roughness was reduced to 400 - 500 nm and 300 - 500 nm for cases DA and SA, respectively.

Particle size analysis using Dynamic Light Scattering has been carried out and the results are also displayed in Table 1. The electrostatically stabilized nanofluids had a smaller effective particle size than the non-stabilized nanofluids.

The effect of the acidity seems to have a significant effect on the stability of nanofluids as reflected in the post-boiling surface roughness measurements. As shown in Table 1, the surface roughness in the case of the acidic nanofluids is almost an order of magnitude lower than the case of neutral nanofluids. This can be attributed to the fact that the acidity creates a charge on the particles which cause them to repel one another

and enhance their stability [7]. Although a rougher surface is expected to result into a higher PBHTC, however, in this case, the post-boiling surface roughness indicates settling of nanoparticles on the surface, which creates an insulating layer on the boiling surface, resulting into a lower PBHTC. The effect of the acidity is also reflected in the particle size. The acidic nanofluids have a smaller effective particle size. The charge on the particles reduces the chance for them to agglomerate due to the Van der Waals forces [7].

The acidity was found to have a noticeable effect on the PBHTC for the nanofluids prepared from dry particles. The PBHTC has increased by 25 % upon addition of the acid. This effect is expected as the nanofluids become more stable and less particles deposit on the surface. The more stable nanofluids are also expected to have a higher thermal conductivity which may provide another explanation for the enhancement in the PBHTC. The effect of the acidity alone on the water was seen to reduce the PBHTC. Therefore, the enhancement in nanofluid stability seems to overshadow the surface tension effect on the PBHTC due to the addition of the acid.

Effect of Preparation Method

The nanofluids prepared from ready-made suspensions appear to be already stable. The addition of the acid causes a much lesser change in particle deposition on the boiling surface for the nanofluids prepared from suspensions compared to the nanofluids prepared from dry particles. About 7000 nm increase in surface roughness for the nanofluids prepared from dry particles, as opposed to only about 3000 nm for those prepared from ready-made suspensions. Almost no effect was found on the PBHTC for the nanofluids prepared from a ready-made suspension upon addition of the acid. This was contrary to expectations being that the acidity will enhance the stability of the nanofluids. Also the acid causes a smaller reduction in the effective particle size for the nanofluids prepared from a ready-made suspension (20nm reduction) than those prepared from dry particles (35 nm reduction). This may explain why the PBHTC did not change when stabilizing the nanofluids prepared from a ready-made suspension.

However, the reason why the PBHTC of the nanofluids prepared from dry particles and ready-made suspensions are different is unknown. The length of time of suspending the particles in the water seems to have an effect on the performance of the nanofluids under pool boiling.

More work is needed to provide explanations for the observed difference in boiling behavior between nanofluids prepared from dry nanoparticles and ready-made suspensions. Nanofluids properties such as surface tension, thermal conductivity and viscosity need to be measured to give a better understanding of the nanofluid behavior during pool boiling.

CONCLUSION

The addition of acid has been confirmed to improve the PBHTC of nanofluids by making them more stable when prepared from dry nanoparticles. The acidity has no effect on the PBHTC when the nanofluids are prepared from a ready-made suspension. Deposition of nanoparticles on the heater surface during pool boiling is greatly reduced when nanofluids are slightly acidic. Nanoparticles have a strong tendency to agglomerate and it is very difficult to break them down to their individual particle size. Effective particle size is usually 3-4 times larger than the nominal particle size. More research is needed to understand the dynamics of nanofluids boiling.

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