FUNDAMENAL STUDY OF THE EFFECT OF NANOPARTICLES ON BOILING HEAT TRANSFER

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Abstract. Two sets of experiments are performed in this work to reveal the potential effect of nanoparticles on boiling heat transfer: i) pool boiling of nanofluids on two well-defined boiling surfaces and ii) bubble formation in a quiescent pool of nanofluids under adiabatic conditions. Different to conventional thoughts that the modification of boiling heat transfer is the result of solid surface modification by particle sedimentations forming a porous or coating layer, the result suggests that both the particle deposition effect, and the particles suspended in liquid could affect the boiling heat transfer significantly. The particle deposition effect is affected by the relative size between particles suspended in the liquid medium and the surface geometry, and their interactions. Nanoparticles suspended in the liquid can affect bubble dynamics significantly by modifying pinning behaviour of the triple line when forming bubbles and subsequent bubble dynamics. Both roles are likely co-existent in a typical boiling system. Depending on different applications, properly surface engineered nanoparticles could minimize the particle deposition effect but still contribute to the modification in heat transfer through the second mechanism.

Keywords: Boiling, bubble, nanoparticles, nanofluids, wetting

1. INTRODUCTION

Nanofluids are functional liquid suspensions containing particles that are smaller than 100nm. With some enhanced thermophysical properties, they could be used to intensify heat and mass transfer in a variety of systems including microfluidic system. Over a decade's research in the nanofluids technology has advanced our understanding on the subject, especially on their thermophysical properties, but with many questions remaining. While the original idea of using thermal nanofluids was to enhance thermal conductivities of some typical heat transfer fluids including water, mineral oil and ethylene glycol, the inclusion of nanoparticles has been found more profound than the mean properties effect. As reviewed recently, our current understanding on the subject is still very limited especially on the interactions between particles, liquid and heating surfaces (Wen et al. 2009). Many conflict experimental results have been reported, from mean properties measurement to different applications. Recent international benchmark study has revealed that the enhancement in thermal conductivities is mainly due to the increase in heat transfer surface area associated with large specific surface area of nanoparticles, and the enhancement could be predicted by the effective medium theory (Buongiorno et al. 2009). For nanofluids applications especially under pool boiling conditions, some significant boiling heat transfer enhancement has been reported (Wen and Ding 2005, Wen et al., 2006, Park and Jung 2007, Liu et al. 2007, Kedzierski 2009); this however contrasts significantly to others where large heat transfer deterioration was reported (Das et al, 2003, Bang and Chang, 2005, Kim and Kim 2007, Trisaksri and Wongwises, 2009). The results were very inconsistent even for the same nanoparticles under similar experimental conditions. Not like the thermophysical properties such that the thermal conductivity is always increased with particle concentrations, conflicting trends have been extensively reported on the influence of particle concentrations on the boiling heat transfer (Park et al. 2009, Kathiravan et al.2009). Meanwhile, a large number of studies reported different levels of enhancement, from 10% to 400%, in the critical heat flux (CHF) (You et al. 2003, Vassallo et al. 2004, Kumar and Milanova 2009, Park et al. 2009, Kathiravan et al.2009).

There are a few possible parameters affecting boiling heat transfer with nanofluids such as the morphological and thermophysical properties of nanoparticles and nanofluids, the stability of nanofluids, the content of nanofluids such as surfactants and ions, and the deposition and interactions of nanoparticles with heating surfaces (Kim et al. 2006). A few studies have suggested that different boiling heat transfer results are related to the deposition of nanoparticles on the heating surface (Kim and Kim 2007, Narayan et al. 2007, Part et al. 2009). As boiling heat transfer is very sensitive to surface characteristics especially the number and shape of potential nucleate sites, any change in the surface would probably result in different boiling behaviors. However the surface effect alone should not be the whole picture; otherwise we should revive old studies on boiling enhancement by surface treatment rather than using uncontrolled nanoparticle deposition. Is there any roles played by nanoparticles suspended in the liquid phase? One aspect is obvious; the inclusion of nanoparticles would modify the mean properties of base fluids. Of boiling considerations, the effect due to thermal conductivity or viscosity modification would be small under dilute conditions. The key parameters would be surface tension and wettability as they affect bubble formation and dryout significantly. Some studies have indeed showed that small concentrations of nanoparticles could affect these parameters to a great extend (Sefiane 2006, Vafaei and Wen, 2010a, 2010b). Apart from the mean property effect, some theoretical studies have shown that well

suspended nanoparticles in the liquid phase alone could modify triple line dynamics and bubble formation significantly, for a given ideal surface (Wen 2008, Chengara et al. 2004, Wasan and Nikolov 2003).

This work aims to identify the roles of nanoparticles in boiling heat transfer with a particular focus on the possible dual roles played by nanoparticles: i) modification of the heating surface through particle deposition and ii) modification of bubble dynamics through particles suspended in the liquid phase. The role i) will be identified by conducting pool boiling experiments on two carefully fabricated plate heaters with quantitative characterization of boiling surfaces before and after boiling through an atomic force microscopy (AFM), energy dispersive X-ray spectrometry (EDS) and scanning electronic microscopy (SEM). A particular focus is laid on the modification of surface properties and their subsequence influence on nucleate boiling heat transfer. To illustrate the role ii), i.e. the effect of particles suspended in liquid, separate experiments will be performed on the formation of a single bubble in alumina nanofluids on top of a stainless steel substrate, as well as the discussion of its implication to boiling heat transfer.

2. THE EFFECT OF PARTICLE DEPOSITION

To avoid the influence of surfactants or dispersants, which alone would have a strong effect on the boiling process (Wasekar and Manglik, 2002, Wen and Wang 2002), Al2O3 nanofluids are made freshly by dispersing dried α - alumina nanopowers into de-ionized water under ultrasonic agitation conditions. Three low concentrations of alumina nanofluids are formulated at 0.001%, 0.01% and 0.1% concentration by volume. The nanofluids formulated are not permanently stable but can last for a couple of hours without visible sedimentations, which resembles a large body of nanofluids studies reported in literature (Das et al. 2003, Bang and Chang 2005 You et al. 2003, Narayan et al, 2007, 2008, Kedzierski 2009) . Figure 1a shows a SEM image of the alumina nanoparticles, where the primary particles are spherical and have a wide size distribution in the range of 20 ~ 150nm. Figure 1b shows an in-situ size distribution of alumina particles in the suspending liquid measured by the Malver nanosizer. It clearly illustrates a much larger size profile than that of dried particles as captured by SEM. The particle size lies within 50~900nm with an average diameter of 405 nm, a first peak at 370 nm, and a second peak at ~ 5 micrometers. The second peak area accounts for ~6% of all light scattered and will be the first batch of particles deposited onto the surface that interacts with potential nucleate sites. We emphasis here the large differences in particle size profiles between those observed by SEM and those suspended in a liquid medium. Numerous experiments and evidence have shown that such a difference is common for all nanofluids formulated without proper stabilization and surface modification. Such a difference is understandable as dried nanoparticles frequently appear in the form of agglomerates, some of which, particularly those formed due to strong agglomerations or sintering during the particle production process, are very difficult to break through mechanical forces such as ultrasonification, milling and high-shearing. As a consequence, the in-situ particle size in a liquid suspension will be much larger than the size of primary particles as shown in Figure 1a. For any nanofluids applications, it is believed that an in-situ particle size profile in its host medium will be a more proper parameter to reflect the particle size, rather than the popularly-used primary particle size as characterized by electronic microscopes.
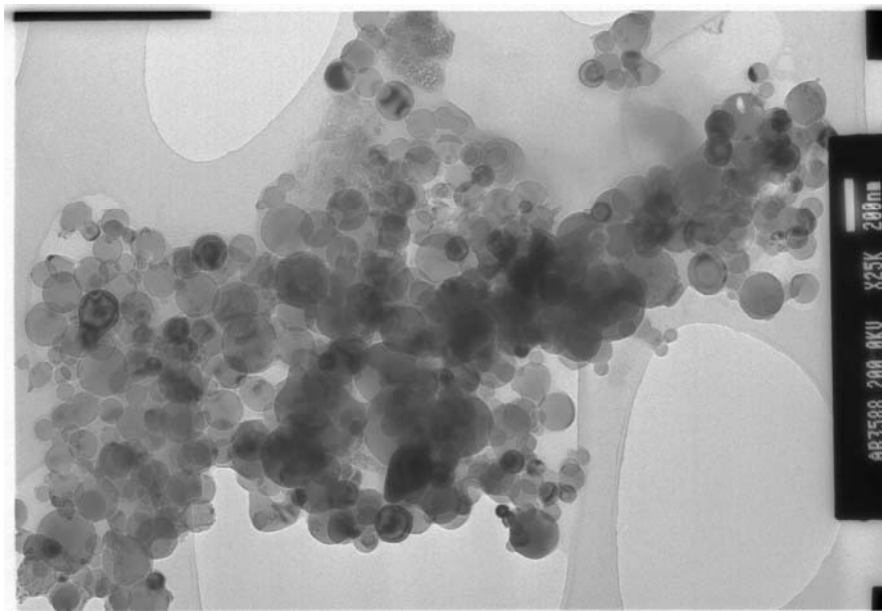
(a) SEM image of alumina nanoparitcles

Size Distribution by Intensity
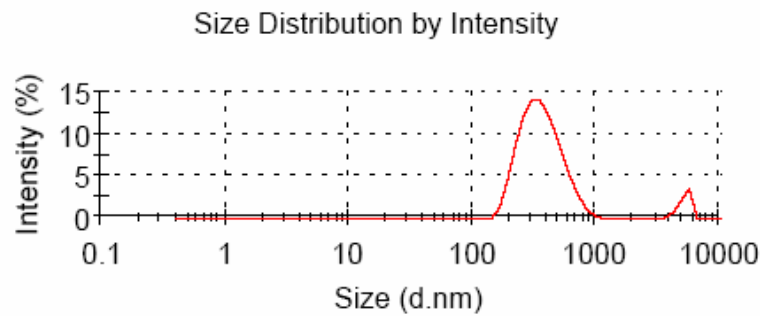
(b) In-situ paritlce size distribution measured by Nanosizer,Malvern

Figure 1: Characterisation of particles through SEM and in-situ particle profile measured by nanosizers

Pool boiling experiments are conducted on two plate boiling surfaces, which are made of copper having a rectangular shape of 20 mm by 20mm and a thickness of 10 mm. The heaters are cleaned thoroughly in an ultrasonic bath and bonded carefully to a transparent high temperature Pyrex housing through a water proof, high temperature resistant epoxy mould. The center of one side of the boiling plate accommodates a ϕ=6.7 mm cylindrical cartridge heater (18 mm in length), which is controlled by a variable DC power supply to provide heat. Three type-K thermocouples are located 0.2 mm below the boiling surface, separated by a distance of 2mm starting from the center of the heater, and one is immersed in the liquid pool to monitor the liquid temperature. The whole system is well insulated except the top of the housing that is vented to the ambient. All thermocouple readings are collected by a data acquisition system through a multichannel amplifier. The uncertainty of temperature measurement is controlled at 0.2K and the heat loss to the surroundings is estimated as ~5%. An overall uncertainty of 7% in the heat flux is achieved in the experiments.

Two boiling plates are manufactured with different surface finishes, termed as rough and smooth surface respectively. The average roughness values of the rough and smooth surfaces as characterized by AFM are 420 nm and 25 nm respectively. Of particular note though, the averaged roughness only gives an arithmetic average of peak-to-peak values, which is not a proper parameter to describe a boiling surface. Only some specially shaped cavities with gas entrapment capability can be acted as active nucleate sites that control different boiling behaviors. However without delving too much detail here, the convention is adopted here and the average roughness is used as the parameter to reflect the nucleation site distribution.

In a typical experiment, nanofluids are prepared freshly before each experiment. Here we will focus on two well-controlled experiments To avoid nanoparticles contamination and establish a reliable comparison, the experiments for DI water and 0.001% concentration of nanofluids are conducted under exactly the same experimental conditions on the two surfaces. DI water is boiled first and then the 0.001% nanofluids. Example results are shown in Figure 2 and Figure 3. It is apparent that the 0.001% nanofluids increase the boiling heat transfer rate for the smooth surface, i.e. a two-fold increase in the boiling heat transfer coefficient is observed under low heat flux conditions. However this is not the case for the rough surface, which produces nearly identical results for both 0.001% nanofluids and DI water.
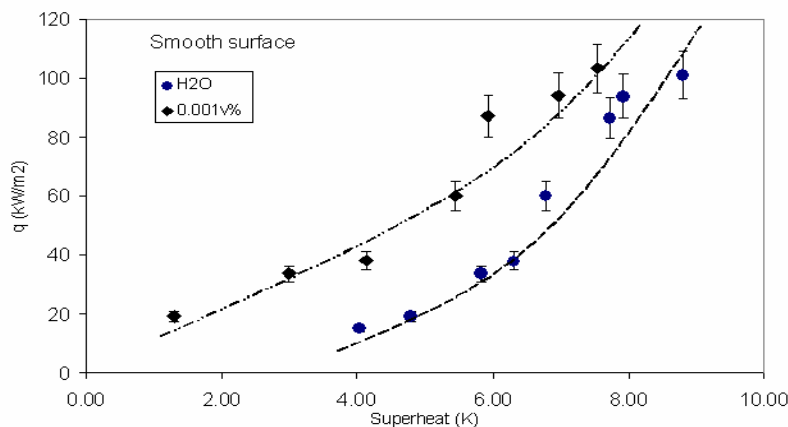
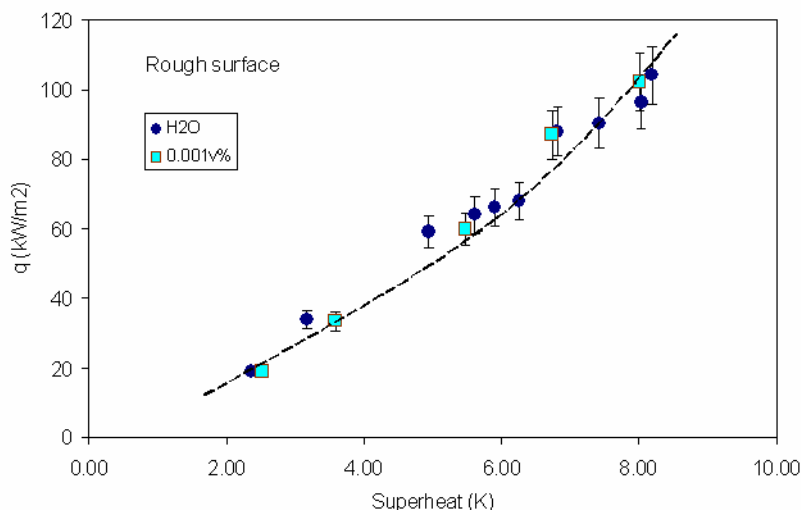Figure 2:Comparative boiling experiments on the smooth surface

Figure 3 Comparative boiling experiments on the rough surface

The boiling surface is then removed, washed by DI water and examined in details by the AFM. Example AFM images before and after the first boiling experiments are illustrated in Figure 4. It is surprise to notice that layers of nanoparticles are deposited onto the surface after boiling even under such low particle concentrations and low heat flux conditions. As the surface after boiling was carefully washed before the AFM examination, these nanoparticles appear to become an inherent part of the boiling surface and would certainly modify subsequent boiling processes. The instability of the nanofluids, possible fouling and even sintering of nanoparticles onto the surface during boiling should be responsible. We would like to emphasis here that similar to most of other reported experiments, the nanofluids after boiling still look the same as before, without any visible sedimentation. The AFM study here clearly reveals that such surface modification must have been occurred in nearly all boiling experiments with nanofluids reported by different research groups.

The difference in boiling behaviour can be further revealed by examining microscopic details of the surface geometry before and after boiling. For the rough surface, the surface geometry profile maintains nearly the same after nanofluids boiling, with similar peak-to-peak values. However for the smooth surface, the peak-to-peak values and the average height increase nearly three folds, and the averaged roughness increases over 2 folds for both sampling locations after the first time boiling with nanofluids. These results again are understandable by comparing the relative in-situ particle size with the surface geometry. For geometry with an average roughness of ~ 25 nm having peak-to-peak values of ~ 300 nm, the nanofluids with peaks in 370 nm and 5 microns will no double have an overall increase in the roughness after boiling. Although not a perfect parameter into the boiling mechanism, such increased roughness will contribute to the enhancement of boiling heat transfer as shown in Figure 3.   It is apparent that the relative size between nanoparticles in a liquid medium and boiling surface geometry plays a crucial role in determining the heat transfer effect. Narayan et al. (2008) proposed a parameter, the ratio of primary particle size to the average roughness of the surface, to differentiate boiling with nanofluids where they claimed that boiling heat transfer coefficient was decreased for smooth surfaces, which is direct contradictory to our findings. Rather than the primary particle size, the in-situ particle size in the liquid medium should be used to accurately reflect the interactions between particles and boiling surface. Beside the size effect, the thermophysical properties of particles and the heating surface will reflect how strongly the particles will be bonded to the heating surface. Such a surface modification would certainly affect the number of active nucleation sites, solid surface energy and the wettablity of liquids, all of which have important roles in boiling heat transfer.

It is believed that the change of surface geometry is an inherent feature of nanofluids even for stable ones. The retaining and trapping of nanoparticles will occur when nanofluids come in touch with surfaces, irrespective of the surface orientation. The increase in surface temperature by heating will increase the Van Der Waals force that would accelerate particle entrapments. The situation becomes intensified under boiling conditions where the rise of bubbles and rewetting by surrounding fluids after bubble departure bring a constant supply of nanoparticles to the surface that intensifies nanoparticle deposition. For unstable nanofluids as chosen in this work, a macroscopic sedimentation would accelerate the deposition process. Large particles especially those shown in the second peak areas (Figure 1b) will deposit first. The relatively high temperature of the heating surface makes strong adherence and even sintering of nanoparticles onto the surface that modifies subsequent boiling processes. For single phase flow in relative large channels, however, the heat transfer rate will not be affected significantly as the heat transfer is not sensitive to surface

geometries, which has been evidenced by many observation of enhanced convective heat transfer coefficient by nanofluids. For single phase flow in microchannels where the surface roughness becomes comparable to that of the channel dimension, we expect that a strong surface effect will emerge.
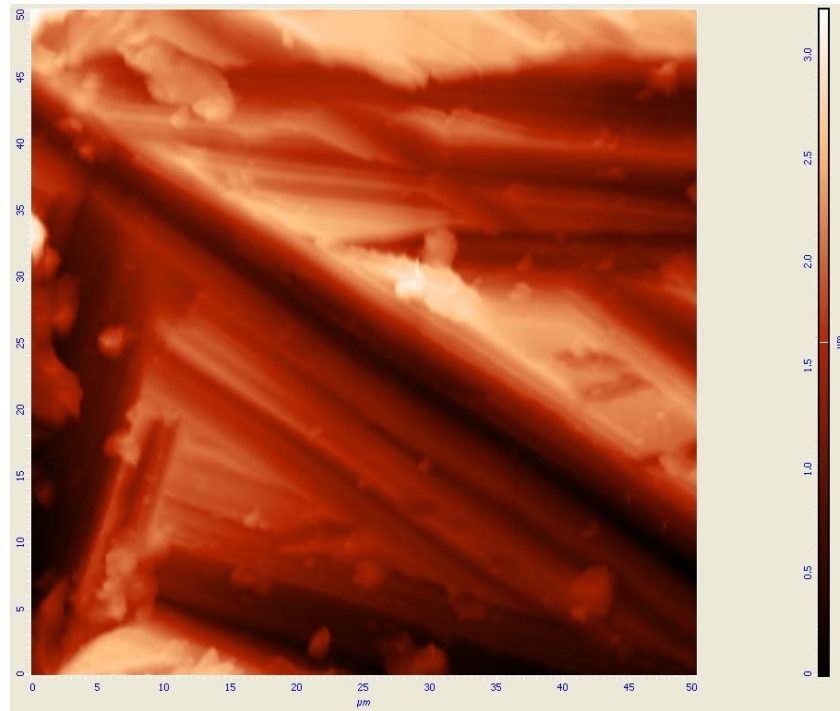
(a) before boiling
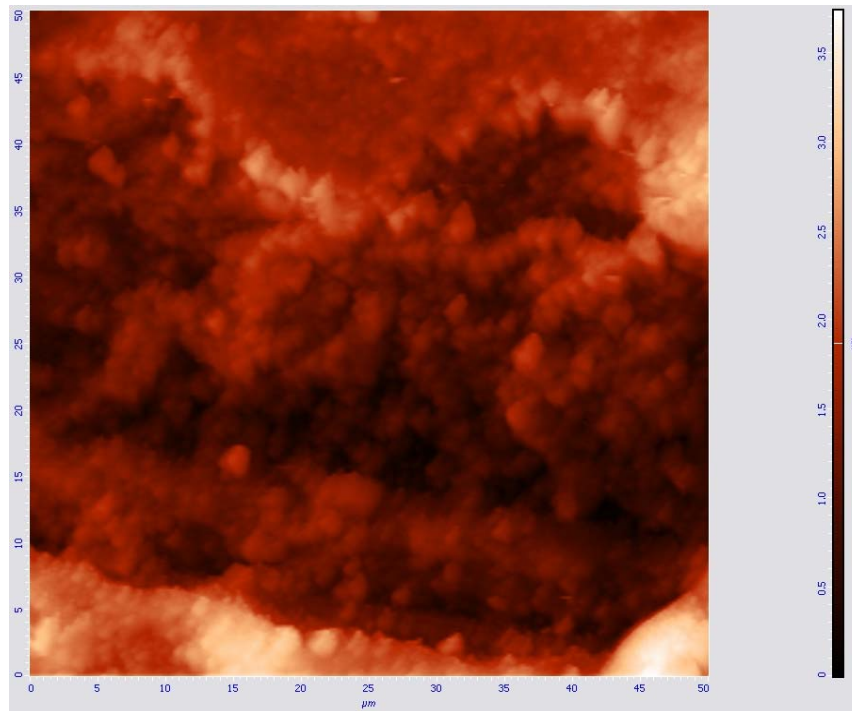
(b) after boiling

Figure 4: AFM image of the rough surface before and after boiling with nanofluids

3. THE EFFECT OF PARTICLES SUSPENDED IN FLUIDS

To reveal the effect of nanoparticles suspended in the liquid, it is essential to seclude the effect of particle deposition and observe bubble dynamics under well controlled conditions. As various experiments have shown that the particle deposition is mainly caused by heating, here single bubble will be formed under adiabatic condition, i.e formation of gas bubbles in nanofluids, to avoid the heating influence and minimize the particle deposition effect. An cylindrical artificial cavity of 0.4 mm in diameter is fabricated on a stainless steel substrate, the same material as that in flow boiling experiment, and polished afterwards (Ra 0.021 with Rz of 0.03 μm). The experimental system for bubble formation is detailed in Vafaei and Wen (2010b). For brief, it includes a gas supply system, camera and microscope, the stainless steel substrate and a measurement system. The substrate is submerged into a transparent square-sized glass container of the size of 20 by 20 mm and a height of 72 mm. The glass container is filled with quiescent deionized water or nanofluids to a height of 20 mm and open to the atmosphere under ambient conditions. The air flow is supplied from a pressurized air cylinder through a pressure reduction valve and flows vertically into the orifice. The flow rate is controlled by a flow controller in the range of 0.015-0.83 ml/min. The flow controller has a specified accuracy of $\pm 0.5\%$ of the nominal reading. A high speed camera (1200 frame/sec) and an optical microscope head are used to capture the images of formation of bubble. The images are stored in a computer for later processing.

During the experiments, the high speed optical camera captured details of the bubble formation process. Late image processing provided accurate measurements of some bubble parameters such as the radius of bubble contact line and the bubble height. Figure 5 shows the variation of bubble height and radius of contact line with time over formation of bubble on top of a stainless steel substrate nozzle inside water and alumina nanofluids (0.001 v%), where significant different behavior is observed. In general, bubbles form and develop earlier in alumina nanofluids, the triple line is pinned at larger radius for given bubble volumes. The bubble departure volume is larger and takes relatively longer time to depart. It is of interest to note that such a phenomenon is different to nanofluids made of smaller cylindrical-shaped gold nanoparticles (~10 nm) where a reverse trend is observed (Vafaei and Wen,2010). It is apparent that bubble dynamics is affected significantly by the nanoparticle materials, the size and shape. Such an observation is understandable from molecular kinetic aspect as fundamentally, the presence of nanoparticles at the vicinity of a substrate will modify the interactions among liquid-liquid molecules and liquid-solid moleculars, which would modify both the driving force and resistance force at the meniscus, changing the dynamics of the triple line. Different nanoparticle materials, size and shape will have different effects on these interactions that would result in different triple line dynamcis, where detailed understanding requires further studies.
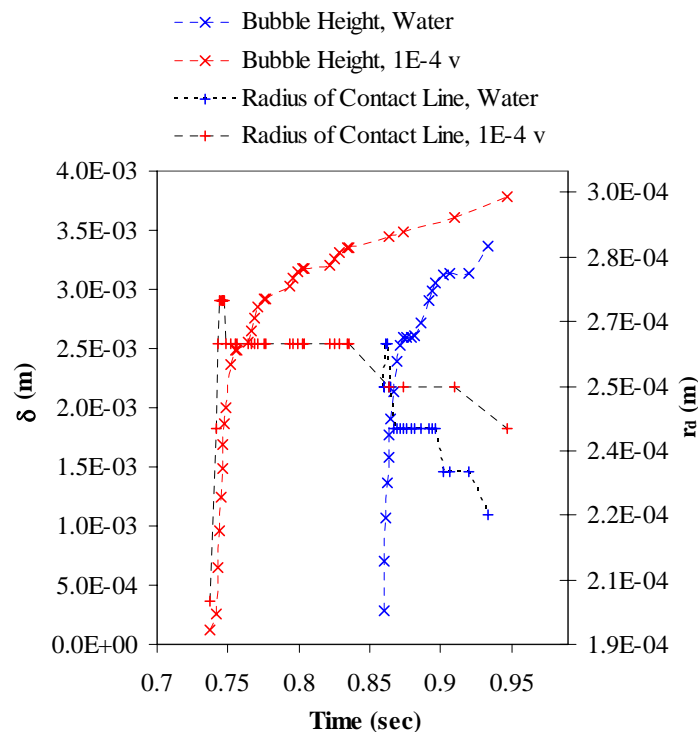

Figure 5. Variation of bubble height and radius of contact line with time inside water and alumina nanofluids (0.001 v%). The nominal gas flow rate is 0.5 ml/min.
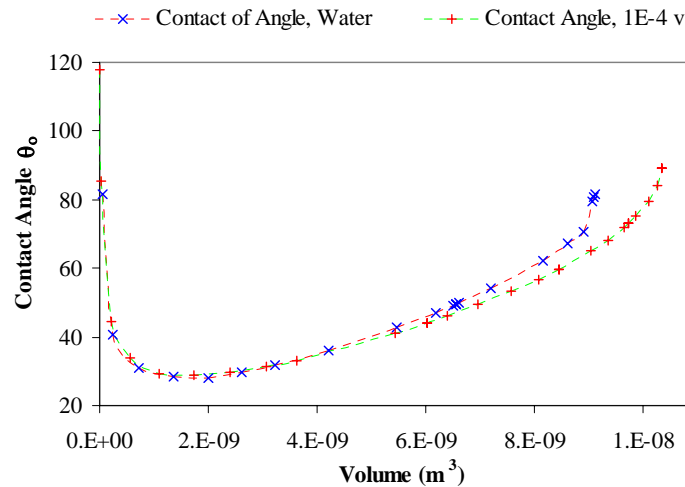
Figure 6. Variation of bubble contact angle with volume on top of stainless steel substrate plate inside water and 1E-4 v alumina nanofluids. The nominal gas flow rate is 0.5 ml/min.

Other bubble parameters that are difficult to determine accurately by image processing are obtained through Young-Laplace equation. The Young-Laplace equation represents a mechanical equilibrium condition between two fluids separated by an interface, which can be solved by knowing two bubble parameters among the radius of contact line, the bubble height, the radius of curvature at the bubble apex and bubble contact angle in a running angle and curvature coordinate. Using the bubble height and the radius of contact line as the only two inputs, other bubble parameters are obtained by solving Young-Laplace equation in Matlab using the 4th order Runge-Kutta method. Detailed description of the numerical process and discussion on the applicability of Young-Laplace under similar conditions can be found in reference (Vafaei and Wen,2010a). One example on the variation of contact angle of bubble on top of a stainless steel substrate nozzle inside water and alumina nanofluids (0.001 v%) is shown in Figure 6. The bubble contact angle is quite similar at early stages of bubble formation when the bubble volume is small; however a lower value is observed at large bubble volumes. As the variation of liquid-gas surface tension due to alumina particles inside nanofluids is observed to be small (Kim et al. 2007), the modification of solid surface tensions on the force balance at the triple line, together with modified triple line and bubble dynamics, could contribute the reduction of the bubble contact angle.

4. DISCUSSION: THE DUAL ROLES OF NANOPARTICLES

Depending on different boiling fluids and surface properties, boiling heat transfer could be convection dominated nucleation dominant or both modes are important. Not only modifying the fluids properties such as thermal conductivity, the nanoparticles could affect both surface topography of the heating surface and bubble dynamics by modifying the contact angle, the bubble volume and the bubble departure frequency, which will affect different contributions from evaporation and convection heat transfer. In general, the investigation of nucleate boiling heat transfer is semi-empirical that based on a number of correlations. The total heat transfer is dependent on the active nucleate site density, superheat, bubble departure frequency and departure diameter, which can be summarized as the following form:

$$q \sim (n_a)^{a1}(T_w - T_s)^{a2} f^{a3} D_d^{a4}$$

(1)

where a1,a2, a3,a4 are experimental constants, a1 ranges from 1/3 to 4/3, a2 ranges from 1 to 1.5, a3 =1, a4 is between 2~3. f is the departure frequency, Dd is the bubble departure diameter, na is the active nucleate site density. The role of nanoparticle will manifest itself by the modification of these controlling parameters, especially n_a, f and D_d.

Particle deposition and subsequent modification of surface morphology could have different effects on nucleate site density, n_a, which have been long speculated as the main reasons for different boiling heat transfer coefficients of nanofluids. It is generally observed that particle deposition effect is small under stationary or low heating conditions, i.e under single phase convective heat transfer. However significant building up of porous layer of nanoparticles on the heating surface was found under boiling conditions. It is suggested that evaporation of such a microlayer with subsequent deposition of the nanoparticles is the main reason for the formation of the porous layer of nanoparticles on top of heating surface (Kim et al. 2007). We have showed that as nanofluids concentration increases, more particles appeared onto the heating surface, forming a layer of porous structures, being thicker and more condensed for higher concentrations, as shown in Figure 4. Detailed AFM results suggest that the influence of particle deposition on the nucleate site density is not so straightforward; it is dependent upon the relative size between the in-situ particle size and

the average roughness of the heating surface. Careful analysis of surface topography before and after boiling with nanofluids is needed.

The surface wettabililty can affect the nucleation sites a lot. To characterize surface wettability change after boiling with nanofluids, many studies have used equilibrium contact angle of static droplets. For instance, Kim et al [29] measured the droplet contact angle and surface roughness inside the stainless steel pipe after testing flow boiling heat transfer with water and different concentrations of alumina nanofluids, and observed that the droplet contact angle was reduced from 83 to 20 degree, and the average surface roughness, R_a, was changed from 2.15 to 1.72 μm after experiments. It appeared that the variation of surface roughness and consequently nucleate site density was not much affected, and the main reason for the critical heat flux enhancement was the improvement of surface wettability. It was also claimed by the authors that the use of nanofluids mitigated the propagation of hot spots that could explain why burnout tends to be more localized compared to water, which might be caused by high surface wettability as a result of the deposition of nanoparticles on the heating surface (Kim and Kim, 2007). However the bubble contact angle would be more appropriate to use, since it is more properly for boiling conditions. As shown in Figure 5, the presence of alumina nanoparticles could reduce bubble contact angle for a given bubble volume, an implication of an improved wettability.

Using bubble formation in a quiescent pool of alumina nanofluids as an example, we show that the modification of bubble dynamics is not due to the modification of solid surface by nanoparticles deposition. Under these heating conditions, the substrate surface wettability modification is a direct result of particle deposition and formation of porous layer. Here we demonstrate that modification of bubble dynamics is mainly due to the actions of nanoparticles suspended in the liquid. To ascertain that such variation in bubble dynamics is not caused by particle deposition, bubbles are formed again on surfaces that have been used in alumina nanofluids. Similar bubble dynamics as that of on surfaces inside pure water are observed. Not only affecting contact angles, these nanoparticles alone influence the triple line development, bubble volume and bubble frequency as well, which would affect subsequent boiling heat transfer. Its potential influence in boiling heat transfer is two-folded, as shown in Eq (1): the increase in bubble volume will contribute to boiling heat transfer, but the decrease in departure frequency will make the boiling heat transfer less effective. The total effect is dependent on the relative importance of these two effects, whose details require further detailed studies.

For nanofluids application under boiling conditions, our current experiments suggest that nanoparticles will have two main roles to play: i) modification of the heating surface through nanoparticle deposition and ii) modification of force balance at the triple line due to variation of solid surface tensions through suspended nanoparticles in the base liquid. The first role is apparent, which have been reported by many prior studies. The bubble formation experiment suggests that the second role is also universal. In a typical boiling system, it is believed that both roles would be manifesting themselves. With properly surface engineering, an improvement in heat transfer could be achieved even without the influence of particle deposition, which would open a promising window for its future applications. For nanofluids application in microchannel heat sinks, many prior studies has shown that nanoparticle deposition and subsequent increase in flow resistance or even channel blockage is one of the main constraints (Lee and Mudawar, 2007). One possible way out may be through proper engineering the surface properties of nanoparticles, such as with a hydrophobic coating layer, which would minimize the particle deposition effect but in the same time may still modify bubble dynamics and heat transfer through the second mechanism.

However it needs to be cautious to extend or apply directly gas bubble results to boiling conditions. As shown by Smith et al (2002), there are many similarities, as well as differences, between gas bubbles and vapor bubbles. Pertinent to current work, different flow conditions, different cavity sizes and different surroundings should be considered. The discussions based on homogeneous assumption, Eq. (1), is also debatable as the distribution of nanoparticles in the liquid is likely heterogeneous where the two-phase nature of the fluids should be considered. Nevertheless, as an initial analysis, this work illustrates that both particle deposition and nanoparticles in the liquid phase could affect bubble formation significantly. Further work to identify the exact contribution from each part is required, which is currently undergoing.

5. CONCLUSION

This work conducted a fundamental study of the roles of nanoparticles under boiling conditions through two well-controlled experiments. Apart from the particle deposition effect, which has been reported previously and further proved in this study, this study shows that the role of nanoparticles suspended in the liquid phase is also important. The boiling mechanism is affected by both factors, i.e. i) modification of the heating surface through particle deposition and ii) modification of bubble dynamics through particles suspended in the liquid phase. More specific, this study shows that:

- The enhancement or deterioration of boiling heat transfer is affected by the relative size between particles suspended in the liquid medium and the surface geometry, and their interactions.

- The surface modification by nanoparticles is found to be an inherent feature of nanofluids that occurs each time after boiling. The experimental results are affected by the number and frequency of the usage of boiling surface.
- Nanoparticle suspended in the liquid phase can modify bubble dynamics significantly by varying contact angles, departure bubble volume and frequency. The bubble contact angle is found to be smaller for bubbles forming in aluminum nanofluids for a given volume.
- Both roles are likely co-existent in a typical boiling system, whereas properly surface engineered nanoparticles can minimize the particle deposition effect but still contribute to the modification in heat transfer through the second mechanism, which is promising for microchannel applications.

6. ACKNOWLEDGEMENT

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