



Local aggregation characteristics of a nanofluid droplet during evaporation



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ABSTRACT

This study experimentally investigates non-uniform particle distributions and evaporation characteristics of nanofluid droplets containing 50 nm average diameter alumina (Al_2O_3) particles, on a hydrophilic glass surface. Using an inverted microscope, the size distribution of aggregated nanoparticles was visualized and analyzed at different sight-of-view locations. From the digital images captured using CMOS cameras and a magnifying lens, the effect of particle concentrations on droplet evaporation rates was examined. In particular, in order to understand the significance of the early stage of droplet evaporation, the dynamics of a corresponding triple line were visualized using a high-speed imaging technique. From the results, it was found that as the volume fraction of nanoparticles in nanofluids increased the total evaporation time and the initial contact angle decreased, while the corresponding perimeter of the droplet increased. Local aggregation was observed when a nanofluid droplet was in contact on the surface, suggesting that the non-homogeneous characteristics should be considered in estimating thermal conductivity of a nanofluid droplet.

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1. Introduction

Many researchers have tried to improve the thermal conductivity of base fluids by suspending metal or ceramic solid particles since the thermal conductivity of solids is generally higher than that of liquids. Recently, nanofluids (NF) have attracted great interest because of their potential in exhibiting much better thermal performance [1–4]. Their enhanced thermal performance makes nanofluids promising for various applications related to thermal management. The excellent potential of nanofluids as the next generation of cooling fluids for automobiles and electronic devices has recently led to increased research. In particular, it is generally accepted that well-dispersed nanofluids could be easily produced with nanoparticles smaller than 50 nm thereby reducing sedimentation, pipe erosion, and agglomeration [5]. Moreover, wetting characteristics between nanofluids and solid surfaces have been recognized as an important phenomenon in multiphase flows and interfacial transport applications. After Deegan et al. [6,7] reported the coffee ring effect of the colloid suspended sessile droplet, numerous experimental and theoretical studies on heterogeneity at the interface among solid, liquid, and vapor phases have been studied [8–12]. The characteristics of droplet

evaporation have also been studied with nanofluid droplets [13,14]. With the evaporation of a nanofluid droplet, micro/nano-patterning can be obtained by using self-assembly and deposition of nanoparticle patterns. This may be a very easy and inexpensive way as compared to existing methods used in traditional micro-electronics industries [13,15–17]. In the nanofluid droplet case especially, there are important factors that influence the thermal efficiency, viscosity, wetting, and evaporation characteristics of the droplet such as the types of nanoparticles and base fluids, particle size, and concentration.

In the nano-scale, unexpected phenomenon can occur compared to the micron- or macro-scale. Shin et al. [12] studied the evaporation characteristics on submicron-scale patterned silicon hydrophobic surfaces. The well-known regimes (constant contact area mode, constant contact angle mode, and mixed mode) were not distinctively observed during the corresponding evaporation process. Vafaei et al. [8] conducted an experiment on the contact angle variation depending on the nanoparticle concentration and particle size. The role of the nanoparticles inside in droplet-based micro/nano-fluidics is important. Also, the nanoparticles inside the droplet can be acting as a surfactant. They might be involved in the stabilization of droplet interfaces [18]. Upon evaporation, nanofluid droplets show strong pinning along the initial droplet contact line [13]. Shin et al. [19] proposed that the droplet edge shrinks quickly near the last stage of evaporation; one characteristic that is a key in understanding the flow inside the droplet

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during evaporation. Furthermore, the evaporation rate accelerated inside the nanofluid droplet due to nanoparticles with high thermal conductivity. These results can be helpful in the paint industry e.g. to improve the pigment dispersion and to prohibit unwanted sedimentation phenomenon, as well as in future academic studies.

There are many theoretical and experimental studies for thermal conductivity enhancement using nano-sized particle suspensions that have been conducted since Maxwell's theoretical approach [20]. Many theoretical relations were proposed to estimate effective thermal conductivity of nanofluids because the conventional understanding of mixtures based on continuum formulations could not be applied [21–31]. Effective thermal conductivity is an important factor in predicting the total evaporation time of the nanofluid droplets. Hu and Larson [32] and Popov [33] proposed relations for predicting the total evaporation time. However, those models lack consideration of the effective thermal conductivity of the droplet. In fact, many researchers have attempted to measure and predict the effective thermal conductivity of nanofluids considering the particle size effect [34–40]. Nevertheless, these approaches can be accepted only if the nanoparticles stay in a homogeneous distribution. The aggregation phenomenon could largely decrease the effect of thermal conductivity which is directly opposed to the purpose of the nanofluids [21,41,42]. As a result, a dissimilar evaporation rate would be induced and this could affect the total evaporation time of the nanofluid droplet. Recently, Kondaraju et al. [43] conducted a numerical study for the changes of the effective thermal conductivity of nanofluids containing multiple nanoparticle sizes by mimicking an analogous inhomogeneous agglomeration condition. To date, (to the authors' knowledge), however, there has been no relevant experimental examination of non-homogeneous distribution of nanoparticles in nanofluid droplets. In this study, we experimentally observed that the nanoparticles are quickly aggregated right after the drop was dispensed even though they were dispersed well before dispensing. We also observed that there was spatial non-uniformity due to the agglomeration inside the droplet.

The main objective of the present study is to experimentally investigate the aggregation effect and wetting and evaporation characteristics of Al_2O_3 nanofluid droplets on glass surfaces, particularly the total evaporation time. First, this study observed the spatial non-uniformity of the nanoparticles due to the aggregation inside the nanofluid droplets. Second, the total evaporation time was examined for various volume fractions of nanoparticles in nanofluid droplets. Third, we analyzed the factors that could affect the total evaporation time such as the measured apparent initial equilibrium contact angles (ECAs) and the perimeters of nanofluid droplets with different volume fractions.

2. Experimental method

Four different volume fractions of 0.01 vol%, 0.05 vol%, 0.1 vol%, and 0.5 vol% were prepared to examine evaporation of 2 μl nanofluid droplets deposited on a slide glass surface. The alumina (Al_2O_3) nanoparticles (Sigma Aldrich Co.) with an average diameter of 50 nm were suspended in de-ionized (DI) water as a working base fluid. The nanofluids were dispersed for 5 h in each case at room temperature and atmospheric pressure by using an ultrasonic disruptor. Each experiment was repeated ten times and the environmental temperature and humidity were maintained at 24 ± 0.5 °C and $40 \pm 1\%$, respectively.

As seen in Fig. 1, the experimental system consisted of two CMOS cameras (ARTRAY Co., ARTCAM-300MI, 46 fps, and Xi, one Mega-pixel, 30 fps), micropipettes (Gilson, P2), a telecentric lens

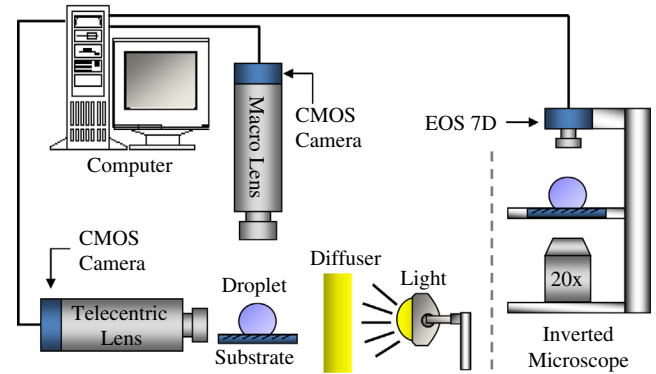


Fig. 1. Schematic of the experimental setup to visualize top, side, and transparent views.

(Computer, TEC-M55), a halogen lamp (400 W, 3 M Overhead Project), and a soda-lime slide glass (MARRIENFELD, 76×26 mm). For consistent experiments, well-cleaned target surfaces should be prepared. In this study, all the glass surfaces were sufficiently cleaned by the air-blower which removed any dust deposited on the surface. This hydrophilic substrate glass showed the initial ECA of a water droplet to be about 34° . The droplets were gently dispensed onto the glass slide, and the front and top sides of the droplets were dynamically recorded every 5 s during evaporation via the CMOS cameras. In addition, the inverted microscope (Olympus, CKX41) with a digital camera (Cannon, EOS 7D, 5184×3456) was used in order to investigate the spatial non-uniformity generated by nanoparticle aggregation. The built-in illumination system applied a 30 W halogen lamp, which spectrum has more intensity at around 650 nm to about 950 nm. The 20X objective lens (Olympus, LCAChN 20X PhP, $\text{NA} = 0.4$) was used and the field of view, which is the ratio of field number ($\text{FN} = 22$) and total magnification, was 1.1 mm through a current system. The microscopic images at every focal plane in every diametral region inside the droplet were taken after the drop dispensed on the slide glass.

With the captured images, as shown in Fig. 2, the dynamic contact angles, the contact radii, and the perimeters of droplets were obtained using Image-J software. The LB-ADSA (Low-Bond Axisymmetric Drop Shape Analysis) [44] method was employed to measure the ECAs. In order to count particles, the MaxEntropy method, proposed by Kapur et al. [45], was used to determine the threshold gray-levels for the imaging process. The posteriori entropy of the gray-level histogram in Pun's algorithm [46] was reformulated by Kapur et al. [45] to correct Pun's errors. The MaxEntropy method uses the calculation of the probability distribution of gray-levels. This allowed for automatically or interactively setting lower and upper threshold values, then segmenting the grayscale gradient of the image into target particles and background. The measurement uncertainty including ECAs, contact radii, and perimeters of droplets was estimated as 2.9% for ten data sets in each experimental condition (95% confidence level). The experimental uncertainty coming from some of environmental sources including temperature, humidity, and the equipment such as micropipette used was estimated to be 12.1%.

3. Results and discussion

3.1. Local aggregation phenomenon inside the nanofluid droplets

The 50 nm Al_2O_3 nanoparticles were split into 4 groups of 0.01 vol%, 0.05 vol%, 0.1 vol%, and 0.5 vol%. The corresponding nanofluid evaporation experiments were performed in ambient

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