International Journal of Heat and Mass Transfer 105 (2017) 230–236

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Study of residue patterns of aqueous nanofluid droplets with different particle sizes and concentrations on different substrates

H.H. Lee [a], S.C. Fu [a], C.Y. Tso [a,b], Christopher Y.H. Chao [a,*]

[a] Department of Mechanical and Aerospace Engineering, The Hong Kong University of Science and Technology (HKUST), Hong Kong
[b] HKUST Jockey Club Institute for Advanced Study, The Hong Kong University of Science and Technology (HKUST), Hong Kong

ARTICLE INFO

Article history:
Received 12 April 2016
Received in revised form 19 September 2016
Accepted 19 September 2016

Keywords:
Droplet
Evaporation
Nanofluid
Residue pattern

ABSTRACT

Nanofluid droplet evaporation has attracted great interest due to its applications such as in painting, coating and patterning. In most studies, either the particle size or the concentration of nanofluid is considered as a factor in the formation of the residue pattern. This work aims to investigate the effect of both particle size and concentration on the residue pattern. A comprehensive study was made of the residue patterns of Al_2O_3 and TiO_2 aqueous nanofluid droplets on different substrates (i.e. glass, stainless steel and Teflon). It was found that a ring-shaped pattern was formed at low concentrations and small particle sizes, while a uniform pattern was formed at high concentrations and large particle sizes for Al_2O_3 nanofluids. In addition, only ring-shaped residue patterns were observed for all concentrations of TiO_2 nanofluids. In the case of different substrates, on a material with a high contact angle with water, it was difficult to form a ring-shaped pattern. The widths of the ring-shaped pattern were analyzed as well. The results showed that the width of the ring-shaped pattern was larger for small particles. The materials of substrate and nanoparticle also influenced the width.

1. Introduction

The evaporation of a colloidal droplet and the formation of the residue pattern are crucial to many applications as such inkjet printing [1], virus detection [2], DNA stretching [3–5] and colloidal self-assembly [6–8]. The size of particle can alter its movement inside the droplet [2,9,10]. Small particles tend to approach the three-phase contact line of the droplet and form ring-shaped patterns while large particles appear closer to the center in the residue. Hence, this results in different residue patterns for different particle sizes. This influences the performance of the applications. For example, the size of pigment particles inside a commercial ink product for printing ranges from tens to hundreds of nanometers [1]. Bermel et al. [1] found that using a smaller particle size pigment could improve the printed image quality without losing the lightfastness by reducing the particle size less than 50 nm. However, it is easier for small particles in a droplet to move to the contact line. This causes poor performance due to the uneven distribution of pigment and formation of ring-shaped patterns. Therefore, the suppression of the ring-shaped residue pattern is necessary in inkjet printing. The residue pattern depends on the particle size, evaporation dynamics, concentration of fluid, material of substrate and material of nanoparticle, etc.

Some factors of drying patterns of a colloidal droplet have been investigated. For instance, Monteux and Lequeux [11] found that nanoparticles segregated at the edge of the drop of a polydispersed nanometric and micrometric particles colloidal suspension. They suggested nanoparticles had a different packing behavior to form patterns from microparticles. Adding surfactant into nanofluid is a common practice for the stability. However, adding surfactant influences the residue pattern. Crivoi and Duan [12] investigated the effect of surfactant on drying patterns. Adding Cetyltrimethylammonium Bromide (CTAB) surfactant into graphite nanofluid can help the formation of ring-shaped pattern instead of uniform pattern. The effect of different surfactant, sodium dodecyl sulfate (SDS), was investigated by Still et al. [13] and they found addition of SDS caused a significantly more uniform pattern from drying of polystyrene colloidal suspension. Yunker et al. [14] studied the shaped of the particle in colloid to control the deposition pattern. Ellipsoidal particles uniformly deposited during evaporation. However, Dugyala and Basavaraj [15] investigated the patterns with different shapes of the particle and pH value of the fluid. The result revealed that the particle–particle and particle–surface interaction was more important than the aspect ratio of ellipsoidal particle to control the ring-shaped pattern. Lin et al. [16] found the effect of

---

[*] Corresponding author at: Department of Mechanical and Aerospace Engineering, Main Academic Building, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong.

E-mail address: meyhchao@ust.hk (C.Y.H. Chao).

http://dx.doi.org/10.1016/j.ijheatmasstransfer.2016.09.093
0017-9310/© 2016 Published by Elsevier Ltd.

Nomenclature

c_1	empirical parameter
c_2	empirical parameter
CCA	constant contact angle [–]
CCR	constant contact radius [–]
r	radius of the ring-shaped patterns [m]
w	width of ring-shaped pattern [m]
Greek symbol	
ϕ	concentration of nanofluid [vol%]

the surface hydrophobicity on the patterns of silica nanofluids. The surface with higher surface energy could result in smaller width of ring because the nanoparticles were more readily to move to the contact line.

Some researchers have studied the influence of the nanoparticles size and concentration in nanofluids on droplet residue patterns. With regard to particle size, Chon et al. [17] studied the effect of nanoparticle size and number density on the evaporation and dry out characteristics of strongly pinned nanofluid droplets. Four types of nanofluids were tested experimentally, namely 2 nm Au, 30 nm CuO, 11 nm Al_2O_3, and 47 nm Al_2O_3 at 0.5 vol%. A nanofluid droplet was put on a micro-heater array under a constant voltage mode. They found that smaller nanoparticles had a wider edge and more central deposition while larger nanoparticles resulted in narrower stains at the edge with a less central deposition. In other words, larger particles were likely to form a ring-shaped pattern because larger particles have a higher number density of particles, leading to a lower viscosity of the fluid. Wong et al. [2] investigated the sizes of particle separation, which is caused by different traveled distances of the various particle sizes during droplet evaporation. Carboxylate-modified negatively charged-stabilized fluorescent polystyrene spheres with diameters of 40 nm, 1 μm and 2 μm prepared in deionized water were used in their experiment. Suspension droplets on a precleaned glass substrate underwent evaporation under ambient conditions. Three well-separated rings were observed on the substrate after evaporation. The outermost rings and the innermost rings were formed by 40 nm and 2 μm particle suspensions, respectively. Similar results were found for using amine-modified positively charge-stabilized particles. The smaller particles moved closer to the contact line whereas the larger particles moved closer to the center. The result is not the same as that of Chon et al. [17] and it is believed that apart from particle size, other factors may also affect the residue pattern.

Regarding the concentrations, a series of experiments were conducted by Sefiane [18]. Aqueous nanofluids with Al_2O_3 nanoparticles with diameters smaller than 100 nm were examined and a Teflon surface was used as the substrate. The effect of concentration (0.1%, 0.5%, 1% and 2%) on the residue patterns was studied. It was found that all nanofluids formed ring-shaped patterns and the thickness of the rings increased with the concentration so that the increase of concentration was approximately proportional to the increase of the thickness of the ring. Jing and Ma [19] studied a 75 nm silica aqueous nanofluid from 20 wt% to 53 wt% evaporations and patterns experimentally. The result showed that the average radius of the circular cracks, which are cracks inside the residue pattern, decreased with increasing the concentration of nanofluid. While the average distance between two adjacent circular cracks increased with the increasing concentration of nanofluid. Therefore, the patterns are dependent on the concentration of the nanofluid. Brutin [20] studied the patterns of a nanofluid with 24 nm diameter particles whose density was equal to water, with 0.01−5.7% concentrations. It was observed that from 0.01% to 0.47%, ring-shaped patterns were formed and the width of the ring increased with the concentration. For the concentrations of 1.15% and above, axisymmetric flower petal patterns were discovered and the thicknesses increased with concentration. Also, the width of the ring and the concentration were correlated in the power law. Lebovka et al. [21] studied the effect of the concentration of Laponite-based aqueous nanofluids. It was found the coffee-ring effect only for the initial Laponite concentration less than 1 wt% while dome-like deposition for the Laponite concentration exceeding 1 wt%.

The works mentioned above either focused on the particle concentration or the particle size, but there has been a lack of studies to investigate the combined effect of both. Deegan [22] investigated a 0.1 and 1 μm sulfate-terminated polystyrene microsphere suspension with 0.063−2 vol% on a mica substrate. It was reported that the 0.1 μm suspension formed four well-defined ring patterns for the 1 vol% and less organized rings for lower concentrations. For 1 μm suspension, ring patterns were formed with many arches for all concentrations and the distribution function of the arches shifted to a larger value with decreasing concentrations. In addition, the relation of the width of the ring and the nanofluid concentration was shown in a power law equation. These results show that the residue pattern depends not only on the particle size, but also the concentration. However, only two sizes were considered in Deegan's experiment and the smallest size was 100 nm which cannot represent the case of nanofluids.

The objective of this paper is to systematically investigate the residue patterns of different particle sizes and concentrations of nanofluid and different substrate materials. Nanofluids with Al_2O_3 nanoparticle sizes of 9 nm, 13 nm, 20 nm, 80 nm and 135 nm and with TiO_2 nanoparticle sizes of 21 nm in diameter and 0.01 vol%, 0.05 vol%, 0.1 vol%, 0.5 vol%, 1 vol%, 2 vol%, 3 vol% and 4 vol% on glass, stainless steel and Teflon, respectively, are examined in this work. After the evaporation of the nanofluid droplets, the patterns of the deposited particles were captured by a camera. Since the patterns are influenced by the evaporation dynamics [23], the contact angle and the contact diameter were measured by a goniometer during evaporation. The formation of different residue patterns will be discussed and the patterns will be characterized, including the analysis of the width of the ring-shaped patterns.

2. Materials and experimental set-up

A two-step approach [24–27] was used to prepare the nanofluids. The first step was to synthesize the nanoparticles, and the second step was to disperse the nanoparticles in the base fluid. The benefits of this process are that they are easy to perform and applicable to different types of nanoparticles. However, the nanoparticles may easily become agglomerated with each other, leading to instability of the nanofluids. If the effective size of the particles becomes larger with agglomeration, the characteristics of the nanofluids, e.g. their thermophysical properties are affected. To prevent agglomeration, the nanofluids were put in an ultrasonic bath (Branson 3800, Branson Ultrasonics, US) in the preparation process.

Fig. 1 shows a schematic diagram of the experimental setup for investigating the evaporation kinetics of nanofluids. A droplet, whose volume was controlled at 2.5 μL by a micro-pipette, was