

# Effects of roughness and size ratio on alumina fluid deposition patterns

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**Abstract.** In this paper, the deposition patterns of alumina nanofluids with different particle size ratios (10 nm:30 nm = 0:1, 1:9, 3:7, 5:5 and 1:0) evaporated on different roughness (0.1, 0.15 and 0.2 μm) substrates were studied. It was found that the surface roughness of substrates and particle size ratio in solution were important factors affecting the height of deposition rings and the morphology of deposition patterns. A dimensionless criterion number  $A_{jw}$  was defined to represent the deposition patterns. The  $A_{jw}$  number increases with the increase of the roughness of the floor and the proportion of small particles in the solution.

## 1 Introduction

Droplet evaporation is a complex thermodynamic behavior involving heat transfer and evaporation processes at three interfaces: solid-liquid, solid-gas and liquid-gas. Its evaporation characteristics and deposition patterns after evaporation have been extensively studied. The study of droplet evaporation characteristics has been widely applied in spray cooling and ink jet printing fields [1-3]. The study of deposition patterns has been widely applied in thin film coating and biomedical diagnosis. The research of droplet evaporation characteristics has been carried out in the field of spray cooling and ink jet printing. The research of There are also important applications in the field of fault diagnosis [4,5].

Deegan et al [6,7] first put forward the concept of "coffee ring effect" and studied the evaporation process and deposition pattern of nanoparticle solution, which proved that the solution inside the droplet moved from inside to outside. With in-depth study, it is necessary to consider many factors affecting droplet evaporation, such as temperature, humidity, magnetic field and roughness, and droplet size, type, concentration, acidity and alkalinity. Yahui Wang et al. [8] carried out an experimental study on the factor of magnetic field. The experimental results show that the deposition pattern of particles is closely related to the applied direction of external magnetic field. Liu et al. [9] studied the crack pattern formed by evaporation of dried colloidal suspension droplets on different surface roughness boards. The experimental results show that the roughness of boards affects the distance or number of cracks; Yongjian Zhang, et al. [10] studied the deposition patterns of silica nanoparticles suspension solution on different roughness surfaces were studied and compared, and the results showed that roughness

inhibited coffee ring effect and Marangoni effect to some extent. NUNZI et al. [11] experimentally studied the deposition patterns of Al<sub>2</sub>O<sub>3</sub>-H<sub>2</sub>O with different mass concentration evaporated on different temperature substrates, and obtained concentric results. In addition to the three typical patterns of circular, peripheral and thin homogeneous layers with thicker rings, concentric polycyclic structures [12], dendritic structures [13] and spiral structures [14] were also found in other experimental studies. ZHONG et al. [15] studied the deposition patterns of suspension mixtures of small and large particles of aluminum oxide nanoparticles, and found that the concentric polycyclic structure, dendritic structure and spiral structure were evaporated. Deposition patterns containing two mixtures of nanoparticles either exhibit the characteristics of a single pattern or are similar to only one of them. AMJAD et al. [16] studied the evaporation rate and deposition mode of mixed droplets containing silicon and silver nanoparticles, and found that the presence of mixed nanoparticles did have a great influence on the evaporation rate and drying mode of the droplets.

At present, the research on the deposition pattern of evaporation by roughness and multiparticle size has been relatively mature, but the research on the effect of both of them is not enough. Therefore, this paper studies the effect of the two on the deposition pattern by changing the roughness of the bottom plate and the particle size ratio of Al<sub>2</sub>O<sub>3</sub>-H<sub>2</sub>O solution. The shape and size of the deposition pattern are observed by microscopy and three-dimensional surface measurement instrument, and the specific effects of the roughness and particle size ratio are discussed.

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## 2 Materials and methods

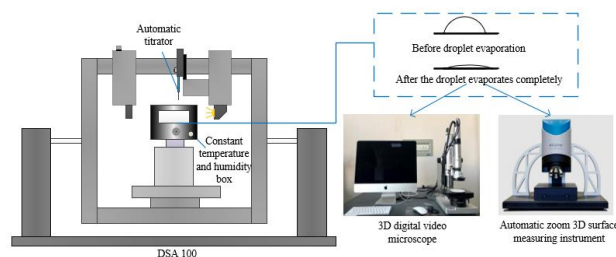
### 2.1 Experimental materials

The nanoparticles used in the experiment are 20% nano-alumina dispersion slurry with uniform particle size distribution and high purity (produced by Aladdin Reagent Co., Ltd.). The solvent is deionized water, which is colourless, clear and transparent (produced in College of Biological Food, Tianjin Commercial University). In order to make the nanoparticles disperse evenly and avoid agglomeration, the anionic surfactant sodium dodecylbenzene sulfonate (produced by China Pharmaceutical Group Chemical Reagent Co., Ltd.) was added to the nanofluid solution. The surface roughness of solid bottom plate is 40mm×30mm×8mm and the resistivity is infinite. The roughness is 0.1, 0.15 and 0.2 $\mu$ m, respectively.

### 2.2 Experimental procedure

Shimadzu analytical balance(model:AUW220D, accuracy: 0.0001g) was used to weigh 20wt% nano-alumina dispersion slurry and deionized water, and sodium dodecylbenzenesulfonate, respectively. Then the weighted nano-alumina dispersion slurry, deionized water and surfactant are mixed in the required proportion, and the ultrasonic cell crusher (model: XO-150) is used to stir for half an hour, then the weighted nano-alumina dispersion slurry is poured into the solution, and the ultrasonic cell crusher is used to stir for 2 hours, so that the nano-particles in the solution are evenly dispersed and seven different particles are formed. Al<sub>2</sub>O<sub>3</sub>-H<sub>2</sub>O nanosuspensions with diameter ratios (10nm:30nm = 0:1, 1:9, 3:7, 5:5 and 1:0) are relatively stable.

The experimental device is mainly composed of an optical measuring instrument(model:DSA100, accuracy:  $\pm 0.3^\circ$ ), a 3D digital video microscope (model: RH-2000) and an automatic zoom 3D surface measuring instrument (model: IFM G5), as depicted in Fig.1.



**Fig. 1.** Schematic diagram of experimental system.

In the whole evaporation process, the temperature and humidity of the chamber are controlled to be  $22\pm 0.5$  and  $30\pm 1\%$ . After evaporation, the solution with different particle size ratios is dripped on the glass substrates of 0.1, 0.15 and 0.2 $\mu$ m respectively with the automatic titrator of optical measuring instrument. The 3D digital video microscope is used to control the temperature and humidity of the chamber. and the automatic zoom three-dimensional surface measuring instrument observed the deposition pattern after droplet evaporation and measured the deposition area, pattern

spacing and the height and depth of the deposition patterns.

## 3 Results and discussion

### 3.1 Experimental results

The evaporation patterns of Al<sub>2</sub>O<sub>3</sub>-H<sub>2</sub>O nanofluids with different particle sizes and roughness ratios can be observed by 3D video microscopy (model: RH-2000), as shown in Table 1. From the experimental results, it can be seen that there are obvious coffee ring patterns at the edge of the three-phase line, and there are different sizes of ring patterns in the deposition pattern. At the same time, the two-ring structure appears in the deposition pattern containing two different particle sizes of nano-solution. On the same roughness of the substrate, when the particle size ratio of nanofluids increases gradually, the annular structure size in the deposition pattern decreases significantly and shifts to the center position. When the droplet size ratio is constant, the size of deposition patterns on different roughness boards changes visually.

On three different roughness boards, when droplets only contain particles with a single particle size of 10nm, there is a smaller inner ring in the center of the deposition pattern, while when droplets only contain particles with a single particle size of 30nm, there is a larger inner ring near the coffee ring of the deposition pattern. Both of the two different depositional patterns have uniform particle deposition near the inner ring, and obvious particle gap appears at the inner ring.

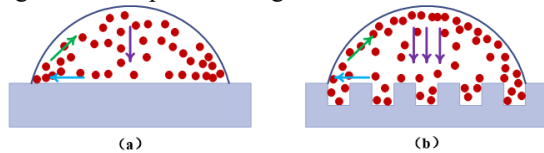
When there are 10nm and 30nm particles in the solution, the deposition pattern is completely different. When the particle size ratio of droplets is 10nm:30nm=0:1, the edges of internal sediments on each rough floor are clearly distinguishable; when the particle size ratio of droplets is 10nm:30nm=5:5, the embedded double-ring structure is blurred, and only one ring structure remains in the deposition pattern on the 0.15 and 0.2 micron floor; when the particle size ratio of droplets is 10nm:30nm=1:0, the roughness on the 0.15 micron floor is 0.15 micron. The annular pattern will appear inside the depositional pattern.

**Table 1.** Deposition patterns of alumina nanofluids with different size ratios on bottom plates with roughness of 0.1, 0.15 and 0.2 $\mu$ m.

Particle size ratio	Roughness		
	0.1 $\mu$ m	0.15 $\mu$ m	0.2 $\mu$ m
10nm:30nm=0:1			
10nm:30nm=1:9			
10nm:30nm=3:7			
10nm:30nm=5:5			
10nm:30nm=1:0			

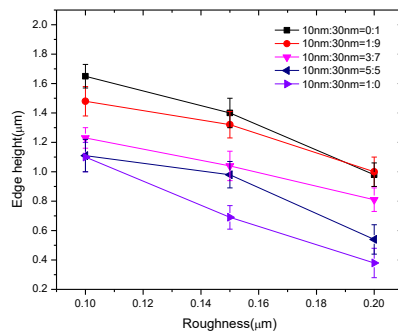
### 3.2 Nanoparticles distribution and deposition

Because of the difference of the bottom plate of droplet evaporation, with the increase of the roughness of the bottom plate, the flow resistance inside the droplet increases, which affects the circulation flow inside the droplet, and the droplet spread more on the solid surface, droplet height decreases, temperature gradient decreases, Marangoni effect decreases, which makes the particles stay uniformly on the surface of the droplet to form a film, and the formation of particle film hinders the droplet evaporation. The droplet evaporation rate is slowed down, the particles have enough time to deposit, and the area of uniform deposition pattern becomes larger and larger, eventually covering the contact area completely. Moreover, due to the increase of the roughness of the floor, some particles are easy to deposit in low-lying areas and cannot be moved. As shown in Fig.2, combined with the influence of capillary flow and Marangoni flow, it is easier for particles to deposit uniformly on the solid surface, thereby affecting the height of the deposition ring.



**Fig.2.** Schematic diagram of deposition pattern formation during droplet evaporation (a) Smooth bottom plate; (b) Rough bottom plate.

Fig.3. shows the height of the outermost coffee ring of the sedimentary pattern. It can be seen that for the same nanofluids, the height of the outer coffee ring decreases with the increase of roughness, and is less than that of roughness from 0.1 to 0.15 $\mu\text{m}$ . When the roughness obtained under the same experimental conditions is constant, the height of the outer coffee ring in the deposition pattern decreases gradually with the increase of the proportion of small and medium size particles in the nanofluids, but the height is always higher than the diameter of 10 and 30nm in the solution, which indicates that the particles accumulate and form a multi-layer ring stacking structure during the deposition process.

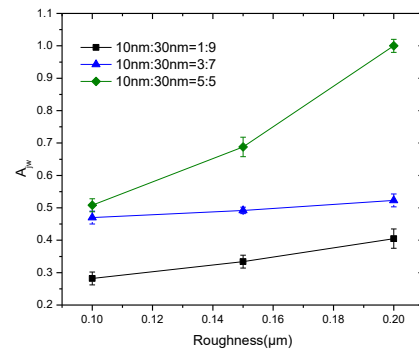


**Fig.3.** Schematic diagram of coffee circle height on mixed surface with nano-alumina mixed solution containing multiple particle size ratios.

In order to further explore the influence of particle size ratio and roughness on the distribution of sedimentary particles, it is necessary to quantitatively analyse the area of sedimentary patterns containing two kinds of particle solutions. However, due to the different morphologies of sedimentary patterns after evaporation, in order to avoid the errors caused by the difference in the area of sedimentary patterns, a dimensionless criterion number  $A$  is defined which can change the area of uniformly distributed areas of sedimentary patterns. it is defined as:

$$A_{jw} = \frac{A_j}{A_w} \quad (1)$$

Among them,  $A_j$  is the area of uniform distribution of particles within the sedimentary pattern, in units of  $\mu\text{m}^2$ ;  $A_w$  is the area of the droplet rigid droplet on the solid surface, in units of  $\mu\text{m}^2$ .



**Fig.4.** Dimensionless number  $A_{jw}$  in a deposition pattern with two particle droplets.

The calculated results are shown in Fig.4. The data shows that the overall deposition patterns of two different droplets behaved regular changes, with the increase of the particle ratio and roughness at 10nm.

Dimensionless  $A_{jw}$  shows an upward trend, The reason for this phenomenon may be that a small number of droplets are deposited in the bottom groove during the spreading process. With the increase of roughness, the number of particles deposited in the groove increases. Because of the difference of roughness and particle size by three orders of magnitude, the particles deposited in the groove cannot easily move with the flow of droplets, so they are rough. Degree first affects the distribution area of particles and the pattern of deposition patterns.

Because of the influence of roughness and particle size ratio, the particle velocity is relatively slow during the whole evaporation process. In order to describe the settling speed of particles more concretely, Stokes law can be used to estimate the settling speed of particles  $U_s$ [17], as shown in formula (2):

$$U_s = \frac{d^2(\rho_p - \rho)g}{18\mu} \quad (2)$$

Among them,  $d$  is particle size, unit m;  $\rho_p$  is particle density, unit  $\text{g}/\text{cm}^3$ , value  $1.06\text{g}/\text{cm}^3$ ;  $\rho$  is the density of deionized water in  $\text{g}/\text{cm}^3$  with a value of  $1\text{g}/\text{cm}^3$ ;  $g$  is the acceleration of gravity in  $\text{m}/\text{s}^2$  with a value of  $9.8\text{m}/\text{s}^2$ , and  $\mu$  is the viscosity in  $\text{Pa}\cdot\text{s}$ . References for the results are given[18,19].

Since the suspended particle size in droplets is only 10nm and 30nm, the  $U_s$  values of the two sizes can be calculated. Among them, the  $U_s$  values of the 10nm particles are  $2.85 \times 10^{-12}$ m/s and the  $U_s$  values of the 30nm particles are  $3.33 \times 10^{-12}$ m/s.

Therefore, when the droplet evaporates, the 30nm particles are deposited on the bottom plate at a relatively fast rate before they move to the droplet surface or when the initial film is formed, which results in the confusion of the distribution of the 30nm particles in the deposition pattern. In contrast, the deposition rate of the 10nm particles is slower and there is enough time to form a thin film, and the distribution of the particles is gradually uniform, which makes the deposition pattern of the 10nm particles more orderly and orderly. Similarly, the deposition patterns of alumina nanofluids with different particle size ratios on different roughness substrates are different. This is because the particles with larger particle size first deposit under the action of gravity, while some particles with smaller particle size move to the edge or center under the action of capillary force and Marangoni effect, and finally form different deposition patterns.

## 4 Conclusions

(1) Solutions consisting of a single particle form a double-ring pattern, while those containing two different particle sizes present a three-ring pattern.

(2) Surface roughness of floor and particle size ratio in solution are important factors affecting the height of deposition ring. The height of the outer coffee ring in the deposit pattern decreases gradually with the increase of the proportion of the roughness particles, but the height is always higher than the diameter of 10 and 30nm in the solution.

(3) The surface roughness of the bottom plate and the particle size ratio in solution also play an important role in the distribution of droplet deposition particles. The increase of roughness can inhibit the internal circulation flow, the Marangoni effect is weakened and the evaporation rate of droplets is slowed down, which makes the particles inside droplets distribute evenly and deposit slowly. Moreover, because the sedimentation velocity of 10nm particles is less than that of 30nm particles, the distribution of 10nm particles is more uniform. Therefore, with the increase of surface roughness and the proportion of 10nm particles in solution,  $A_{jw}$  increases gradually and the area of uniform deposition of particles increases.

## ACKNOWLEDGMENTS

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## NOMENCLATURE

$A_{jw}$  Dimensionless parameter of area change of uniform distribution area of sedimentary pattern particles

$A_j$	The area of uniform distribution of particles in the depositional pattern[ $\mu\text{m}^2$ ]
$A_w$	The area of a droplet just dropped on the surface of a solid[ $\mu\text{m}^2$ ]
$U_s$	Settling velocity of particles[m/s]
$d$	Particle size[m]
$\rho$	Density of deionized water[g/cm <sup>3</sup> ]
$\rho_p$	Particle density[g/cm <sup>3</sup> ]
$g$	Acceleration of gravity[m/s <sup>2</sup> ]
$\mu$	Viscosity[Pa•s]

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