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Amplifying and attenuating the coffee-ring effect in drying sessile nanofluid droplets

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Experiments and simulations to promote or attenuate the “coffee-ring effect” for pinned sessile nanofluid droplets are presented. The addition of surfactant inside a water suspension of aluminum oxide nanoparticles results in coffee-ring formation after the pinned sessile droplets are fully dried on a substrate, while droplets of the same suspension without the surfactant produce a fine uniform coverage. A mathematical model based on diffusion-limited cluster-cluster aggregation has been developed to explain the observed difference in the experiments. The simulations show that the particle sticking probability is a crucial factor on the morphology of finally dried structures.

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I. INTRODUCTION

Evaporation of a colloidal sessile droplet leads to the self-assembly of a finally dried particle structure. The phenomena can be observed in nature and many industrial applications. One of the most commonly observed drying patterns is a coffee-ring effect reported by Deegan *et al.* [1,2]. Possible simple ways to alter or eliminate the coffee-ring effect were investigated recently [3–8]. For example, Yunker *et al.* showed that the effect can be eliminated by changing the shape of polystyrene microparticles from spherical to elliptical [8]. It was indicated that the increased long-ranged particle interaction resulted in the formation of large arrested clusters and finally led to the uniform substrate coverage. In the present work a surfactant is used to control the nanoparticle interactions. Experimentally, the addition of hexadecyltrimethylammonium bromide (CTAB) as a surfactant to a water-based suspension of aluminum oxide nanoparticles significantly reduces the particle aggregation into clusters in the drying process of droplets and results in coffee-ring formation. Alternatively, in nanofluids without a surfactant, the suspended particles tend to aggregate into clusters and deposit into a uniform layer after the sessile nanofluid droplet is totally dried. A mathematical model implementing the diffusion-limited cluster-cluster aggregation (DLCA) approach [9] with a linearized outward capillary flow in a pinned drying droplet has been developed to explain the dependence between the final deposition pattern and the particle aggregation rate on the sticking parameter.

II. EXPERIMENTS

In the experiments, deionized water was used as the base fluid for preparing nanofluids with 25-nm aluminum oxide nanoparticles (Nanostructured and Amorphous Materials, Inc.) with concentrations of 2 and 5 g/L. A solution of 0.1% CTAB (by weight) as a surfactant was added to some nanofluid samples but not to other fluid samples. Mass preparation was carried out by using a sensitive mass balance with an accuracy of 0.1 mg. The surfactant was used to prevent the agglomeration of nanoparticles initially in the base fluids [10]. The cationic dispersant kept the aluminum oxide particles

well dispersed in the based fluids and decreased the particle sticking probability, while the nanofluids free of the surfactant are considered to have a higher sticking probability. The prepared nanofluids were well stirred and then maintained in an ultrasonication bath (Fisher Scientific model 500) for more than 2 h to ensure even dispersion of nanoparticles in the base fluids initially. A silicon wafer was used as the substrate for the nanofluid droplet drying. Right after the suspensions were prepared, a tiny sessile droplet of 1 mm or less in diameter was placed on a clean substrate to dry naturally. The drying process was performed in open conditions at 25°C and 40% relative humidity. The morphology of drying structures was observed under an optical microscope (Zeiss).

III. MODEL

The mathematical model to explain the experimental trend is based on the DLCA model [9] with linearized outward capillary flows in a sessile droplet. Initially, all the solid nanoparticles with the assumed shape are uniformly distributed inside the circular domain on the square lattice shown in Fig. 1. The domain shape mimics the top view of the pinned sessile droplet. At each step of the simulation, each particle performs a random move within the circular domain with probability distribution p_{move} for left, right, up, and down directions. We use the following linear distribution function for p_{move} in this approach:

$$p_{\text{move}} = \left\{ \frac{1}{2} - \frac{x}{4R}, \frac{x}{4R}, \frac{y}{4R}, \frac{1}{2} - \frac{y}{4R} \right\}, \quad (1)$$

where $0 \leq x \leq 2R$ and $0 \leq y \leq 2R$ are the Cartesian particle coordinates and R is the domain radius. The p_{move} distribution implements the outward capillary flow with an increasing velocity toward the edge of the droplet, which might be induced by an uneven evaporation rate with a maximum value at the edge [1,11–15]. After each random move the particles occupying the adjacent cells stick to each other and aggregate into a cluster with probability p_{stick} .

The value of p_{stick} is fixed throughout the single simulation run. If any particles aggregate into a cluster, on the next Monte Carlo step (MCS) the cluster is treated as a single particle and assumed to perform random moves with the same value of p_{move} . The cluster coordinates, $\langle x \rangle$ and $\langle y \rangle$, are averaged for the correspondent coordinates of the contained particles. All

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FIG. 1. Schematic representation of the capillary flow implementation. The circular domain of the square lattice is filled with liquid and randomly distributed particles. Particles are allowed to randomly move inside the liquid but the probability distribution promotes motion in the outward radial direction.

clusters can progressively stick to each other and form ever larger aggregates. Moreover, we assume that more particles start outward motion at the thicker part of the sessile droplet in the vicinity of the axis of symmetry. Therefore, every 10 MCS we randomly place $N_{\text{new}}(r)$ additional particles inside the domain, and $N_{\text{new}}(r) \propto h(r)$, in which $h(r)$ is the thickness profile of a spherical cap and r is the distance from the center of symmetry with a value of $\sqrt{(x-R)^2 + (y-R)^2}$. Since the entire simulation is performed in a two-dimensional (2D) domain, some necessary assumptions, which might look artificial, have to be applied to the model in order to simulate the actual capillary flow inside a three-dimensional (3D) spherical cap. The necessity of additional particles arises from the fact that the real 3D droplet has multiple layers; and while the outward flow is strong near the surface, the bottom particle layer might be slowly moving or not moving at all, until the droplet shrinks further down. In this case, the lower droplet layers serve as additional sources of particles for the outward flow. The experimental observations show that particles at different depth move with different velocity, and, moreover, this velocity profile rapidly changes as the droplet shrinks. Since the presented simple 2D model cannot include all the sophisticated dynamics, an alternative configuration with a distributed particle source is proposed. The new particles are added according to the law that new particles are more likely to appear at places where the shrinking layer is thicker, i.e., near the axis of symmetry (circular domain center). In the model it is assumed that the outward flow is simply restricted to a single 2D layer (simulation domain), while the additional deeper (or “hidden”) layers merge with the main layer as the droplet shrinks. All the particles are then transferred to the main layer and start their outward motion. The particle sticking probability, p_{stick} , is used to explain the main observed trend in the experimental results. The proposed model does not imply that the sticking values used

exactly match with the outcome of the real particle collisions; however, the main goal of the work is to show the trend in the pattern transition as a result of the changed particle interaction. The simulation results show a mechanism by which a real dried pattern might be changed from uniform coverage to a coffee-ring deposition (or vice versa), and the experimental results provide an example of how it might be achieved in reality by changing the particle agglomeration rates via the addition of CTAB surfactant. Note that the individual particle and cluster diffusivity are actually not the same. Their motion is indeed governed by the same p_{move} distribution, but there is an exclusion condition, stating that two different particles are not allowed to occupy the same lattice position at once. (This rule is necessary for the coffee-ring simulation and is reasonably dictated by reality.) Therefore, the large cluster has more “rejected” moves, and after the rejection it holds the position until the next cycle. Therefore, this rule lowers the diffusivity of the larger aggregates (especially for the branched aggregates).

IV. RESULTS AND DISCUSSION

Figure 2 displays a series of images captured from the recording of the experimental drying process of a nanofluid droplet without surfactant. The result represents the last stage of the process, when the remaining liquid layer is thin and the outward capillary flow has the highest velocity. As seen from Fig. 2(a), a thin coffee ring has already been present due to the accumulation of particles at earlier stages. Also, a small number of immobile particles (illustrated as separate black dots) are seen on the substrate. However, the ratio of particles attached onto the substrate is not significant, thus proving that the attraction forces between the silicon substrate and the dispersed aluminum oxide particle do not play a crucial role in this case; the mechanism of deposition driven by particle-substrate interactions has been previously described [7,16]. Figures 2(b)–2(d) show that sparse particle aggregates, characterized by a semitransparent brown color and carried by the capillary flow, start to accumulate behind the outer ring. Consequently, Fig. 2(e) illustrates the formation of a thin layer from sparse particle clusters during the last drying stage. The resulting totally dried pattern is shown in Fig. 2(f), displaying a fine uniform coverage of the substrate area inside the outer ring. The appearance of the finally dried structure allows us to distinguish initially deposited dense particle clusters, which are displayed as small dark spots and cover a small proportion of the substrate area. Structures formed from sparser colloidal aggregation inside the liquid phase are present and uniformly cover most of the substrate inside the initial coffee ring.

The experimental process of the surfactant-induced coffee-ring deposition is displayed in Fig. 3 for the 2 g/L nanofluids. The process starts similarly [see Fig. 3(a)] to the one observed without surfactant [see Fig. 2(a)], since a thin initial coffee ring and negligible initial sedimentation are observed. However, the last stage of the drying process in the presence of CTAB surfactant is qualitatively different. Figures 3(b)–3(d) show that particles do not form visible aggregates and accumulate into a second layer of the coffee ring. The final structure, shown in Fig. 3(f), shows a thicker than initial coffee ring and some particles attached to the substrate left inside the ring. After the

FIG. 2. (Color online) Experimental images of the uniform coverage formation evolution process. The concentration of aluminum oxide particles in the nanofluid is 5 g/L. Images (a)–(f) show the last stage of the droplet drying process, focusing on the droplet edge, with (f) displaying the fully dried structure. The scale bar on the images indicates a length of 25 μm . The real-time video is available as Supplementary Video-1 [17].

droplet is totally dried, most of the substrate inside the ring is left free of particles; however, some spots are still covered by the small particle aggregates, initially sedimentated. This observation indicates that particle attraction to the substrate in the presence and the absence of surfactant is comparable,

but the difference in the particle agglomeration process is responsible for the difference in final pattern morphology. We have to mention that the pinned three-phase line of the sessile nanofluid droplets was fixed in all the present experiments. In the drying process, no contact line shrinking was observed.

FIG. 3. (Color online) Experimental images of the coffee-ring development evolution process. The concentration of aluminum oxide particles in the nanofluid is 2 g/L. Images (a)–(f) show the last stage of the droplet drying process, focusing on the droplet edge, with (f) displaying the fully dried structure. The scale bar on the images indicates a length of 25 μm . The real-time video is available as Supplementary Video-2 [17].

FIG. 4. (Color online) Experimental images of the fully dried aluminum oxide water-based nanofluid droplets: (a)–(d) without surfactant addition; (e)–(h) with 0.1% CTAB surfactant addition. The concentration of 25-nm aluminum oxide nanoparticles is 2 g/L [(a), (b), (e), and (f)] and 5 g/L [(c), (d), (g), and (h)].

This suggests that the contact angle changed continually from an initial angle of about 30° and approached 0° finally as the droplets fully dried.

The images of finally dried nanoparticle structures are summarized in Fig. 4. As we can see from the results in Figs. 4(a) and 4(b), the dried spot is evenly covered with the remaining particle aggregates for the 2 g/L nanofluid samples. The presence of the white spots inside the disk is associated with the loose packing of particles, and the thin outer ring indicates the ringlike particle deposition at earlier stages of the drying. Increasing the mass particle concentration to 5 g/L does not change the pattern of the dried structure [see Figs. 4(c) and 4(d)]. However, the resulting structure has a higher density, as the spots are darker for the almost same diameter disk. The outer ring is also more visible, since a greater number of particles reach the three-phase line at early stages. Furthermore, the addition of surfactant was tested to affect the dried patterns. The resulting patterns have changed significantly for both 2 g/L [Figs. 4(e) and 4(f)] and 5 g/L [Figs. 4(g) and 4(h)] particle concentrations after the 0.1% CTAB surfactant was added to the prepared nanofluids. The evident coffee ring was formed, and the thickness of the ring became more significant with an increase of nanoparticle concentration. The drying process of the 5 g/L aluminum oxide–water nanofluids with the surfactant can be seen in Supplementary Video-3 [17], where a much thicker coffee ring is shown to form there. In these cases only a small fraction of particles is left on the substrate inside the outer ring. It has been suggested that the difference in the drying patterns arises from the effect of surfactant on the nanoparticle aggregation [10,18,19]. The effect might be characterized by the various sticking probabilities among the nanoparticles. It is interesting to mention that in a previous work Still *et al.* reported the opposite effect from surfactant addition to a polystyrene particle suspension [20]. The work focused on coffee ring suppression by an inward Marangoni flow, caused by the presence of surfactant. The addition of CTAB surfactant

to the aluminum oxide nanoparticle suspensions in our cases triggered the qualitatively different mechanisms and led to the opposite phenomena. Another interesting effect of the colloidal suspension salinity was investigated previously [21]. The study involved suspensions with a significantly higher volume concentration and showed that the addition of salt changed the resulting crack patterns in the deposit. Therefore, it might be also possible to affect the particle interactions in other ways in conjunction with the performed CTAB addition.

Simulation runs for different values of p_{stick} were performed. For the assumed initial particle coverage of 5%, which is not a critical parameter in the simulation, the self-assembly process throughout 1800 MCS is compared. Figure 5 shows the progression of the coffee-ring formation in the simulation with a low sticking probability value, $p_{\text{stick}} = 0.5\%$. The simulation run starts with a uniform distribution; afterward, the outward capillary flow pushes all particles toward the edge of the circular domain. The average velocity of outward particle motion linearly increases toward the edge of the domain. Eventually, a coffee ring starts to form and progressively grows as the added new particles join the outward motion. In the simulation, a low sticking probability value allows the particles to reach the boundary of the ring almost independently, and the number of aggregation events in the bulk of the liquid is relatively small. After reaching a densely packed boundary area, the particles become immobile and finally aggregate into a coffee ring.

Figure 6 illustrates the progression of uniform coverage formation process for the highest value of sticking probability, p_{stick} , at 100%. In this case, the process dynamics is completely different from the observation in Fig. 5. The simulations start from the same initial state shown in Fig. 6(a); however, a high p_{stick} value leads to rapid agglomeration of particles in the liquid bulk. As a result, a lot of large branched clusters are formed far inside the domain edge [Fig. 6(c)]. After reaching the droplet boundary, the clusters continue to aggregate with

FIG. 5. Progression of the coffee-ring formation during a single DLCA simulation run with $p_{\text{stick}} = 0.5\%$. Images are continuously captured from 0 MCS (a) to 1800 MCS (j); the time gap between any two consecutive images is 200 MCS in a circular domain ($2R = 300$). The animation of the coffee-ring formation is available as Supplementary Video-4 [17].

each other, forming the loosely packed structures as illustrated in Fig. 6(d). These structures rapidly grow from the boundary toward the domain center and finally jam the outward flow [Fig. 6(f)]. During the last stages of the process, the remaining gaps are slowly filled in with the newly placed particles [Figs. 6(i) and 6(j)], and finally the whole domain is covered by the loosely packed, but uniform branched clusters in Fig. 6(j). The total number of particles in the structures shown in Figs. 5(j) and 6(j) is almost the same, but a densely packed

coffee ring occupies only a thin region near the droplet edge in Fig. 5(j), while a branched DLCA cluster spreads over the whole circle in Fig. 6(j).

In the further parameter study, we have continuously changed the p_{stick} value from 0% to 100% and observed a continuous transformation from a coffee-ring pattern to uniform domain coverage. Initially, the coffee-ring pattern is still preserved at $p_{\text{stick}} = 1\%$ [Fig. 7(c)], but the gaps between particles inside the ring grow and the looser ring

FIG. 6. Progression of the uniform coverage formation during a single DLCA simulation run with $p_{\text{stick}} = 100\%$. Images are continuously captured from 0 MCS (a) to 1800 MCS (j) with a step size of 200 MCS in a circular domain ($2R = 300$). The animation of the uniform coverage formation is available as Supplementary Video-5 [17].

FIG. 7. (Color online) The variety of particle patterns depending on the sticking probability value. The results are shown for p_{stick} values of (a) 0%, (b) 0.1%, (c) 1%, (d) 5%, (e) 10%, (f) 20%, (g) 25%, (h) 50%, and (i) 100%. Images captured after 2000 MCS are with a lattice size of 300×300 . The total number of particle clusters during the simulation runs is shown in (j), the sticking probability increases from the top curve to the bottom one. Diagrams (k) and (l) show the dependence of the final particle distribution on the sticking probability parameter. ϕ in (k) is the local particle coverage, and ρ in (l) is the cumulative particle coverage.

becomes thicker. However, for the case with $p_{\text{stick}} = 5\%$, a clear transition is already seen; the branched boundary clusters spread over a large area, and only one large gap is left in the domain center [Fig. 7(d)]. A further increase of p_{stick} leads to a steady switch toward uniform coverage [Figs. 7(d)–7(g)]. The central gap finally vanishes at p_{stick} at approximately 25% [Fig. 7(g)]. The dynamics of the total cluster number in Fig. 7(j) is significantly different for low and high sticking values. The low p_{stick} corresponding to the coffee-ring-like structures produces an initial growth of the total cluster number. After that, the number starts to fall slowly. High values of p_{stick} produce an immediate drop in the cluster number, and the stronger stickiness apparently results in a sharper drop. Therefore, we can make an observation that the final particle patterns can be directly coupled with the cluster number dynamics. For a given model, having known only the trend of the total cluster number, we could estimate the resulting particle distribution inside the domain. Figure 7(k) shows plots of local particle density, ϕ , as a function of a given radius, while Fig. 7(l) reflects the “cumulative” density, i.e., particle coverage ρ within a disk of $r < R$. The sharp rise in the curves indicates the correspondent coffee rings. Both diagrams clearly prove the continuous dependence on the sticking probability and the transition from the coffee ring to uniform coverage.

The general self-assembly comparison between the simulations and experiments suggests that the coffee-ring and uniform self-assembly patterns can be manipulated and transformed into each other by changing a nanofluid property and the particle aggregation chance as a result. It is noted that the experimental structures with surfactant [Figs. 8(a) and 8(b)] show a clear coffee-ring pattern quite similar to the simulation result for the low sticking probability, p_{stick} [Figs. 8(c) and 8(d)]. Moreover, we also note that the experimental results for 5 g/L concentration with surfactant [Fig. 8(b)] is similar to the simulation with 1% sticking [Fig. 8(d)], whereas the experiments with a lower concentration at 2 g/L [Fig. 8(a)] are in better agreement with the simulations for $p_{\text{stick}} = 0.1\%$ [Fig. 8(c)]. This might be understood since an increase in particle number for the low p_{stick} values leads to an increase in the actual agglomeration events due to a smaller distance between the particles and consequently a higher chance to encounter each other. That is the same situation that occurs when we slightly increase the sticking probability itself, resulting in more intensive agglomeration as well. The fact explains that a thicker particle suspension with the surfactant has a slightly looser packed coffee ring due to the higher number of agglomerated particles.

A quantitative analysis of the particle density distributions within a circular domain was also performed. Figures 8(a)–8(e)

FIG. 8. (Color online) Quantitative analysis of the particle density distributions in the simulation and experimental results. (a)–(e) Comparison of coffee-ring particle distribution profiles between the finally dried experimental structures [at particle concentration of 2 g/L (a) and 5 g/L (b)] and the final simulated patterns captured after 2000 MCS [(c) and (d)]. (f)–(j) The correspondent comparison of the uniform particle deposition patterns between the finally dried experimental structures [at particle concentration of 2 g/L (f) and 5 g/L (g)] and the final simulated patterns captured after 2000 MCS [(h) and (i)]. Local particle coverage in (e) and (j) is plotted for each image and ranges from 0 (local empty surface) to 1 (local fully covered surface).

show good agreement between the particle distribution in the experimentally observed structures with the CTAB addition [Figs. 8(a) and 8(b)] and simulations with a low sticking probability [Figs. 8(c) and 8(d)]. The sharp rise in the curves of Fig. 8(e) indicates the inner radius of the correspondent thick coffee ring. Moreover, the simulated results with a high sticking probability [Figs. 8(h) and 8(i)] show a good statistical agreement with the experimentally captured dried pattern from the nanofluids without the CTAB addition [Figs. 8(f) and 8(g)]. A thin coffee ring might also be seen on the experimental images without the surfactant. The observations demonstrate that it forms during the early stages of droplet evaporation, before the main phase of the outward flow and cluster agglomeration starts. The width of the outer ring in this case is significantly smaller than in the presence of surfactant. A statistical image analysis shows that the average ratios of

the coffee ring width to the droplet base radius are 0.04 ± 0.01 and 0.08 ± 0.01 for 2 and 5 g/L samples without surfactant, respectively, but increase to 0.15 ± 0.01 and 0.24 ± 0.01 for 2 and 5 g/L samples with the surfactant, correspondingly. Based on the qualitative and statistical comparisons, it is seen that the model manages to explain the observed transition from the coffee ring toward uniform coverage in drying the pinned sessile nanofluid droplets. The p_{stick} parameter is coupled to the presence of CTAB surfactant in the nanofluid suspension. The alternation of simulation parameter reflects the correspondent changes in the experimental conditions.

V. CONCLUSIONS

The experimental results indicate that the addition of surfactant into a nanofluid can be used to control the final

nanoparticle self-assembly patterns after the full evaporation of a pinned sessile nanofluid droplet. The DLCA-based mathematical model can be used to explain the observed experimental difference by changing the interparticle sticking probability. In this case, high particle sticking probability values are assigned for the samples without the surfactant. Oppositely, it is assumed that the modeled droplets with the added CTAB surfactant contain particles with low sticking values. The simulation results show the transition from the coffee-ring pattern for low sticking probability to uniform deposition for high sticking probability. This qualitative agreement between the experiments and simulations indicates that the control of interparticle aggregation inside the nanofluid is crucial to promote or suppress the coffee-ring effect. The

applied addition of surfactant is only one possible and simple way to make the transition, although the surfactant effects on the fluid thermal properties including surface tension and viscosity have not been discussed here. However, the modeled mechanism is shown to be more universal and other ways to affect particle sticking can be considered in future experiments.

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