Numerical Simulation of Colloidal Drop Deposition Dynamics on Patterned Substrates for Printable Electronics Fabrication

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(Invited Paper)

***Abstract*—One of the major challenges in printable electronics fabrication is the print resolution and accuracy for high precision applications such as printable displays. In this paper, the lattice Boltzmann method (LBM) is used for the direct numerical simulation of an inkjet-printed colloidal drop wetting on a patterned substrate for confined deposition. The two-dimensional multiphase particle suspension LBM model previously developed by the authors is extended to three-dimensional with the addition of the particle rotational dynamics and an improved treatment of particle–particle forces. The model is used to study the contact angle hysteresis and the stick–slip behavior of the contact line motion as a liquid drop wetting and evaporating on a patterned substrate. Finally, the dynamics of a colloidal drop containing many suspended particles are examined with and without evaporation. Results show that colloidal jamming occurs at the liquid-vapor interface as the drop preferentially wets and evaporates on a patterned substrate. This model development is an important first step towards understanding the complex transport phenomena present in an inkjet-printed evaporating drop for printable electronics fabrication.**

***Index Terms*—Inkjet printing, lattice Boltzmann method (LBM), particle suspensions, patterned substrates, printable electronics.**

I. Introduction

ENVIRONMENTALLY, benign roll-to-roll (R2R) electronics fabrication using inkjet printing and direct laser patterning on flexible substrates is an enabling technology that will provide desired high-volume, low-cost production of flexible electronic devices, ranging from displays, solar cells, low-power lighting, to highly specialized small scale sensors and healthcare devices [1]. An important advantage of this technology over conventional silicon devices is its ability to create micron-sized electronic functions like transistors on flexible substrates such as plastic films and textiles. However, the intrinsic limits on the spatial accuracy of inkjetting devices, wetting, de-wetting, contact line pinning, interfacial instabilities, microflows within the deposited drop, and the self-assembly of particulate matter during drop evaporation all contribute to the lack of precise control of deposited electronic materials. This has created a critical need to better understand how the dynamics of specific transport processes impact the final microstructure and properties of deposited materials on flexible substrates.

The process of deposition by drop-on-demand (DOD) inkjet printing essentially comprises two separate physical stages—drop impingement on the substrate and evaporation of the carrier liquid that leads to the final deposition of the particulate phase. For micron-scale drops produced by inkjet printing, the impact stage is characterized by rapid drop deformation and spreading to a maximum diameter followed by either minor oscillations or complete rebound, depending on the surface energy of the substrate [2]. For wettable substrates, drop oscillations viscously dissipate until equilibrium is reached, which is characterized by a stable spherical cap shape. It has been observed [3] that the equilibrium profile of a jetted drop may also be characterized by a contact angle that is different than the equilibrium static contact angle because of the contact angle hysteresis.

The impact phase is followed by evaporation of solvent where either the drop contact angle or the imprint radius decreases with time. As reported by Deegan *et al.* [4], [5], as long as the contact line of the drop is pinned, the higher evaporative flux at the contact line will drive a replenishing radial flow from the center. This flow is responsible for the so-called “coffee-ring” pattern left by evaporating drops carrying solute, as depicted in Fig. 1(a). The contact line pinning that is essential to this process may initially occur due to substrate heterogeneities but subsequently be reinforced as particles “jam” the contact line during the radial flow [6]. However, if no initial pinning of contact line occurs, the colloidal drop will undergo self-similar evaporation, i.e., the drop imprint radius decreases but the contact angle remains roughly the same [7] as shown in Fig. 1(b) until a mixed-stage of drop evaporation is reached where both the contact angle and contact line diameter decrease with time [7], [8]. For a case dominated by self-similar evaporation, more uniform particle deposition morphology is expected. It is important to note here that the relation between the contact line pinning and

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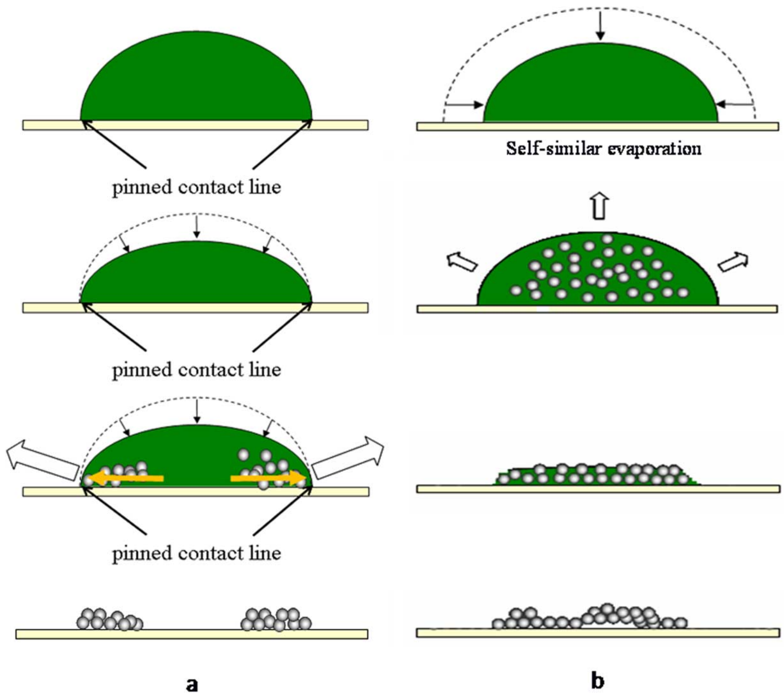

Fig. 1. Colloidal drop deposition for different evaporation mechanisms. (a) Pinned contact line leading to coffee ring deposits. (b) Constant contact angle and mixed mode leading to relatively more uniform deposits.

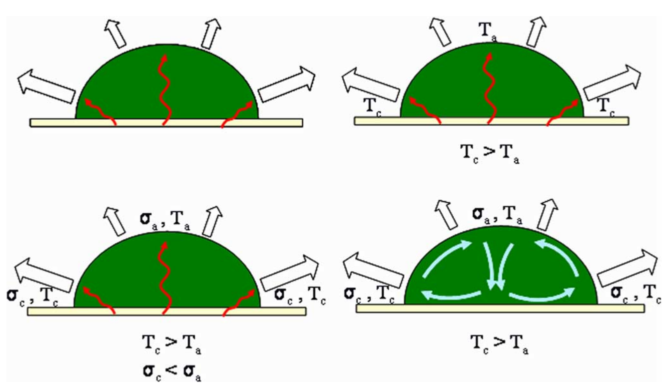

Fig. 2. Schematic depicting the Marangoni flow induced by temperature gradient in an evaporating drop.

ring formation is still a topic of much debate. For example, experimental observations of ring structures have been reported in deposition of dilute colloidal drops on atomically flat hydrophobic substrates [9], [10] in the absence of initial pinning or particle jamming of the contact line.

In addition to the evaporation-driven flow, Deegan *et al.* [4] also reported observing a weak Marangoni flow in their experiments. A Marangoni force manifests itself when there is a gradient in surface tension σ along a liquid-gas interface—regions of higher surface tension "pull" liquid along the interface. Since surface tension is a function of temperature, a nonuniform temperature profile along the drop surface during evaporation would set up corresponding surface tension gradients to drive the flow. As depicted in Fig. 2, thermal Marangoni flow is induced inside an evaporating drop as a result of a higher contact line temperature T_c than that of the drop apex T_a due to the longer thermal conduction path from the substrate to replenish the latent heat at the surface.

Hu and Larson [11], [12] included the thermal Marangoni effect in their numerical simulations and the results show that Marangoni flow depends strongly on the evolving contact angle of the drop, with the flow being driven along the interface towards the drop apex. Their theory also predicts a flow reversal if the contact angle is below a critical value. At that point, the temperature gradient along the interface is reversed—the small height of the apex at low contact angles allows heat conduction to more sufficiently replenish the thermal energy lost to evaporation. This, in turn, allows for the higher evaporation rate at the contact line to cause the temperature there to be lower than at the apex. In an evaporating inkjet-deposited colloidal drop, the interplay of evaporation-driven and Marangoni flows plays an important role in determining the final particle deposition morphology [13]. Park and Moon [14] summarized droplet evaporation and SiO_2 particle deposition profiles in water, water/diethylene glycol, and water/formamide solvent systems respectively as a result of complex flow patterns during drop evaporation.

In order to better control the drop dynamics and subsequent particle deposition, the substrate is often deliberately patterned into different strips such that either the energy level in each strip can lead to different wetting properties [15] or the substrates are physically patterned into a series of grooves for guiding the ink to flow into certain directions [16]. For example, a predetermined pattern of hydrophilic and hydrophobic strips can be created to cause the impacting drop to selectively wet part of the substrate, thereby depositing the particles in the hydrophilic regions.

The important physical processes after the drop impact phase are summarized in Fig. 3. A fundamental understanding of these mechanisms will help optimize process conditions of printable electronics for more reliable deposition patterns with better edge definition, higher resolution, and improved electrical-thermal-mechanical properties. A mesoscale study,

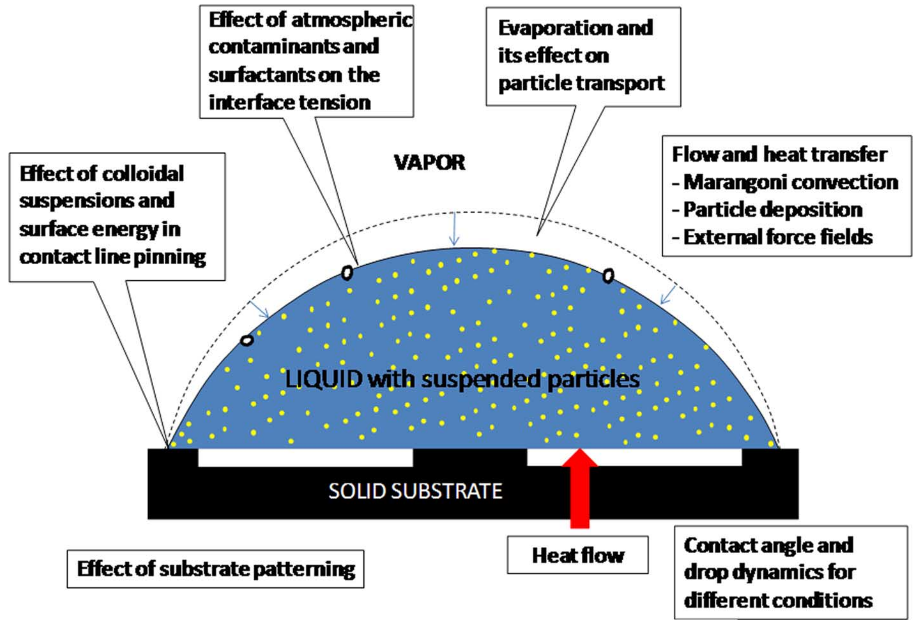

Fig. 3. Physical processes occurring as a liquid drop containing suspended particles impacts on and spreads on a patterned substrate. This work examines the effect of suspended particles on contact line motion and is a first step towards a fundamental understanding of this challenging problem.

such as the lattice Boltzmann simulation, that directly models the microflows, evaporation, particle self-assembly, and other complex transport phenomena present during an inkjet-printed evaporating drop serves such a need.

II. Numerical Model

Lattice Boltzmann models of particle suspensions were first introduced by Ladd [17], [18] and over the years, they have been proven to be computationally efficient tools for various applications ranging from colloidal suspensions to biofluids [19]–[22]. In this paper, we study colloidal particles suspended inside a liquid drop that interacts with solid substrates. We demonstrate the contact angle hysteresis and the stick-slip behavior of the contact line motion as a liquid drop wets and evaporates on a patterned substrate. In addition, the deposition dynamics of a colloidal drop containing many suspended particles are examined with and without evaporation. The liquid drop is assumed to be in equilibrium with its vapor phase (Please note an on-going debate about whether a sessile drop is in *strict* equilibrium with its saturated vapor and the possible change in contact angle upon evaporation [8], [23], [24].) and the multiphase lattice Boltzmann model of Shan and Chen [25], [26] is used to model the multiphase dynamics. Interaction of the liquid drop with solid substrates is modeled via the use of adhesive forces between the fluid (i.e., liquid and vapor phases) and solid. A similar adhesive force is introduced between the solid particles and this force, in addition to the hydrodynamic interactions, determines the motion of each suspended particle within the drop. This combined multiphase particle suspension model allows for a fundamental study of the physical processes taking place within the evaporating colloidal drop as the drop spreads on various substrates.

In this paper, we: 1) extend the two-dimensional (2D) multiphase particle suspension LBM model of Joshi and Sun [27] to three-dimensional (3D); 2) extend the treatment discussed in [27] to include particle rotational dynamics; and 3) improve the treatment of inter-particle forces when the gap between neighboring particle surfaces falls below the grid resolution. For computational efficiency, the 3D LBM model was implemented on a parallel computer using the message passing interface (MPI). A typical 3D simulation on a $200\times200\times100$ lattice using 20 processors takes about 10 hours.

III. Results and Discussion

Following Joshi and Sun [28], the initial momentum of the drop and the resultant impact dynamics on the substrate (see Yarin [29]) have been neglected in the present work as well. The effect of gravitational forces is assumed to be negligibly small. For a recent simulation of drop impact dynamics (without particle suspensions) using the LBM, the reader is referred to Mukherjee and Abraham [30]. Dimensionless groups that are important in drop dynamics include the Reynolds number, $\mathrm{Re} = \rho \mathrm{UD}/\mu$, Weber number, $\mathrm{We} = \rho \mathrm{DU}^2/\sigma$, Ohnesorge number, $\mathrm{Oh} = \mu/(\rho\sigma D)^{1/2}$, and Capillary number, $\mathrm{Ca} = U\,\mu/\sigma$, where U is the velocity scale (e.g., averaged contact line velocity during drop spreading), D is the drop diameter, μ is the dynamic viscosity of the liquid drop respectively, σ is the interface tension between liquid and vapor, and ρ is the liquid density. Because gravitational effects are negligible, the Bond number (Bo) is assumed to be zero. Additional parameters include the density ratio between liquid and vapor, volume fraction of the suspended particles, and particle Peclet number ($\mathrm{Pe} = U_\mathrm{p} D_\mathrm{p}/\alpha_\mathrm{p}$, where $U_\mathrm{p}, D_\mathrm{p}, \alpha_\mathrm{p}$ are the velocity, diameter, and thermal diffusivity of the particle, respectively). Here, the Pe number is assumed to be large and the particle Brownian motion is neglected. In theory, the above mentioned dimensionless numbers can be used to correlate LBM parameters with real physical parameters encountered in experiments [31]. For example, in our LBM simulation, we use $g_f = -0.65$ (corresponds to $\sigma = 0.13$), $D = 40, \nu = 1/6, \rho = 2.53, \mu = 0.4216$, which lead to an Oh number of 0.11. However, because of limitations in the parameter space where the LBM simulations

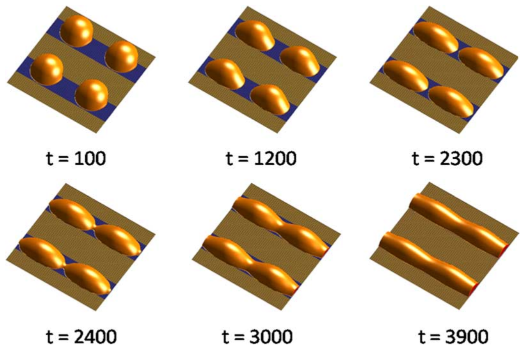

Fig. 4. LBM simulation of drop dynamics on a patterned substrate. The initial shape of the drop is hemispherical and the initial position of the drop is on the hydrophilic (blue) band. Subsequent evolution causes the drop to spread along the hydrophilic band and coalesce with adjacent drops forming a fiber shape. Time is given in lattice units [27].

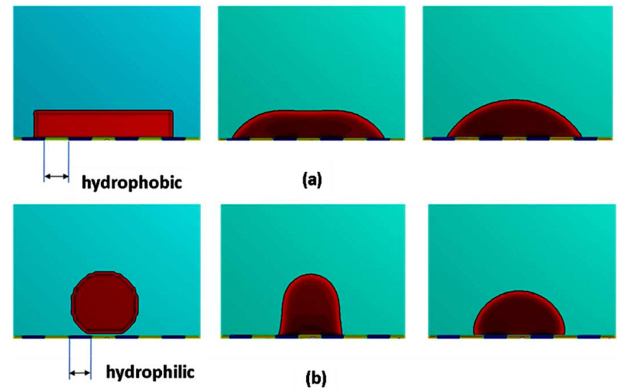

Fig. 5. LBM simulation of contact angle hysteresis (a) An initially rectangular liquid phase relaxing to equilibrium (receding contact angle) (b) An initially circular liquid phase of the same volume relaxing to equilibrium. (advancing contact angle). The final equilibrium shape shows markedly different contact angles.

work without numerical instabilities, not all parameters in the simulations can be matched perfectly with experiments. Also, additional dimensionless numbers related to drop evaporation are not considered here because the current model is isothermal.

The multiphase LBM model without suspended particles was first used to simulate drop wetting dynamics on a patterned substrate. The results are shown in Fig. 4. The initial condition in this simulation is a hemispherical liquid drop located on a patterned substrate made up of a series of hydrophilic and hydrophobic bands. Periodic boundary conditions are used with a grid size of $81 \times 81 \times 81$. The center of the drop is slightly offset from the centerline of the hydrophilic band. As expected, the liquid drop is repelled by the hydrophobic part of the substrate and attracted by the hydrophilic part. The drop thus gradually moves into the hydrophilic part and is subjected to further stretching due to attractive surface forces. If adjacent droplets are spaced close enough, they merge to form a liquid fiber in the hydrophilic band, as shown.

We next examined the advancing and receding contact angles of a liquid drop wetting and de-wetting on a patterned substrate. In Fig. 5(a), the initial shape of the liquid phase is rectangular and in Fig. 5(b), the initial shape is circular. Note that the volume of the liquid is the same in both cases. The substrate was made up of alternating hydrophobic and hydrophilic bands. The final equilibrium shape of the liquid phase for the case of receding contact angle in Fig. 5(a) is clearly different and smaller than that observed for the advancing contact angle case of Fig. 5(b).

Using a thermal LBM to simulate drop evaporation remains a challenging problem. The main difficulty is in accurately incorporating the phase change behavior accounting for the latent heat of vaporization. Following the recent work in Kusumaatmaja and Yeomans [32], we have carried out LBM simulations that model the receding contact line as a liquid drop shrinks on a substrate (Fig. 6). Mass is removed at a rate of 0.1% from the liquid in a quasi-static manner, allowing the liquid and vapor phases to attain the equilibrium density ratio after every such mass removal. It is found that the liquid drop shrinks in a self-similar fashion, maintaining the same contact angle throughout.

If a rough substrate is used in place of the smooth substrate of Fig. 6, a stick-slip motion of the contact line is observed, as illustrated in Fig. 7. Thus, these types of LBM simulations can provide a first approximation to the drop drying process and provide some insight into the contact line pinning during drop evaporation [33]–[35], especially when simulations also include a discrete particle phase within the liquid drop. Because actual evaporation is not modeled, we refer to this mass removal as *pseudo* evaporation.

Our LBM results so far are consistent with some recent LBM simulations that examine drop dynamics on patterned substrates [36], [37]. However none of the studies till date include suspended particles in the liquid drop. We now illustrate the dynamics of a liquid drop containing many suspended particles.

If the drop is brought into contact with a flat substrate (Fig. 8), the interface attaches to and spreads onto the substrate until the equilibrium contact angle is reached. During this process, the suspended particles move because of the bulk motion of the liquid inside the drop. Particles that are close to the moving liquid–vapor interface get attached to the interface and remain there throughout the subsequent drop dynamics. The particles in the interior remain relatively steady although they adjust their positions slightly as the liquid spreads on the substrate. The relative position of the suspended particles changes only if there is a substantial movement of the liquid phase. In many applications of colloidal drops settling onto a substrate, the suspended particle dynamics is influenced by the flow fields within the liquid due to thermal or surface tension gradients and because of additional long-distance forces between the particles themselves or particles and the substrate. Adding in these effects is a part of ongoing work in our group and the results will be reported in a future publication.

Next, we illustrate a case where a free standing liquid drop containing suspended particles is shrinking via a mechanism similar to that discussed earlier with reference to Fig. 6. We artificially reduce the density at each vapor node by 1% after every 100 time steps. This causes mass to be transferred from the liquid phase to the vapor phase, maintaining the same density of the liquid phase and thus causing the liquid phase volume to shrink. Fig. 9 shows the different stages as the liquid drop shrinks. As the interface shrinks, it traps particles within it and eventually a stage is reached where the entire liquid-vapor interface is jammed with particles. A similar particle jamming might be responsible for contact line pinning during evaporation of a 3D colloidal drop on a substrate, leading to the coffee-ring patterns observed in our experiments [31]. Note that this type of

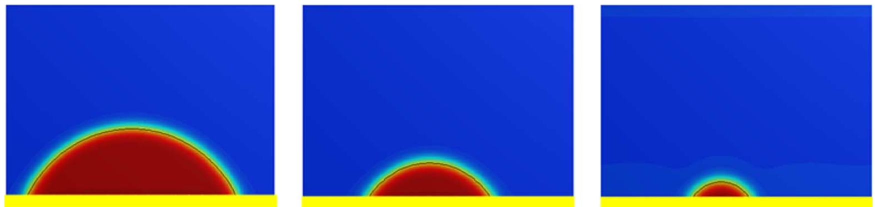

Fig. 6. Simulation of shrinkage (pseudo-evaporation) of a liquid drop using the lattice Boltzmann method. Mass is removed slowly (0.1% at a time) and the system (liquid + vapor) is allowed to return to equilibrium. The contact angle does not change as the drop shrinks.

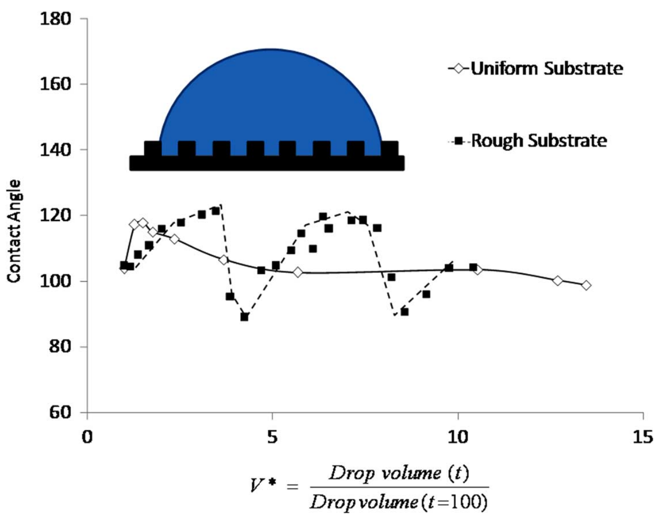

Fig. 7. LBM simulation of a liquid drop shrinking on a rough substrate. The inset illustrates the square-wave profile of the rough substrate and the initial shape of the liquid drop. Te graph depicts the stick and slip behavior of the contact angle as the drop shrinks. Because the volume is reducing with time, the initial contact angle corresponds to the maximum $V*$ and contact angle evolution with time proceeds from high $V*$ to low $V*$ (or from right to left on the graph).

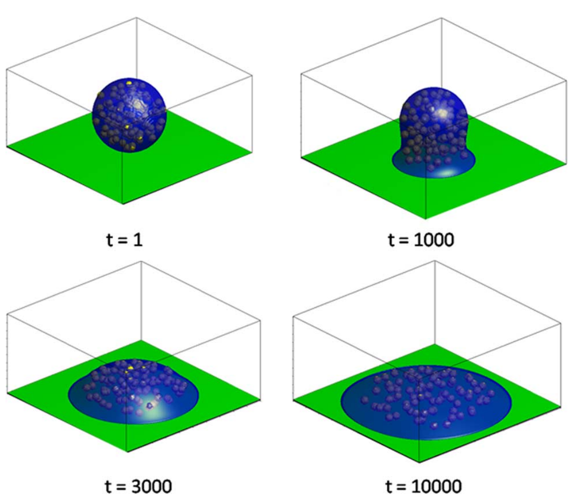

Fig. 8. LBM simulation of a liquid drop containing 90 suspended particles wetting a flat substrate. The particles close to the liquid-vapor interface attach to the interface and the drop eventually attains its equilibrium contact angle of 30°.

pinning effect cannot be captured using a 2D particle suspension model.

To examine the effect of particle jamming and consequently pinning of the contact line, in Fig. 10, we present an example where a 3D hemispherical drop containing 36 suspended particles, equally spaced along at the contact line, shrinks due to intermittent mass removal (pseudo-evaporation). In experiments, evaporatively-driven migration of particles towards the liquid-vapor interface has been observed by several researchers [4], [31]. While the LBM cannot currently simulate this thermally induced particle migration, it can help model subsequent particle deposition on the substrate as the drop shrinks. It was observed that the contact angle remains constant during this shrinkage (i.e., the contact line is not pinned as observed in Deegan *et al.* [4]). But the particles located at the liquid-vapor interface eventually jam the interface. It can be seen from Fig. 10 that the interface jamming is broken by ejecting particles from the shrinking contact line. The deposited particles can be seen to form a hexagonal packed structure.

Finally, we consider a case similar to that shown in Fig. 4, but with suspended particles present in the liquid phase. We use a $201 \times 201 \times 101$ lattice with 87 suspended particles of diameter 13 each (30% particle volume fraction). The density ratio between liquid and vapor is 11 and that between the suspended particle and liquid is 2. The wetting and deposition dynamics of the colloidal drop are shown in Fig. 11. Like the earlier case without particles, the liquid drop is repelled from the hydrophobic band and is attracted towards the hydrophilic band in the center of the substrate as shown in Fig. 11. As the drop wets the substrate, some of the suspended particles attach themselves to the liquid–vapor interface and subsequently remain attached. The drop simultaneously spreads and evaporates. After the carrier liquid completely evaporates, the suspended particles will deposit on the central (hydrophilic) band. Thus, the LBM model can be used to optimize the substrate design, e.g., the width of the hydrophilic/hydrophobic bands and the surface energy step, for an improved particle deposition for a variety of colloidal inks and applications.

IV. Conclusion

Numerical modeling can play a key role in clarifying the underlying physical phenomena and in optimizing process parameters during inkjet-printed electronics fabrication on flexible substrates. In the present work, the multiphase LBM) was combined with the particle suspension LBM to simulate deposition dynamics of liquid drops containing suspended particles on homogeneous and patterned substrates. The model was used to simulate motion of drops on patterned substrates due to hydrophilic and hydrophobic surface forces. The suspended particles in a liquid drop were found to move according to the hydrodynamic velocity fields set up during drop spreading and particles close to the liquid–vapor interface attached to and remained in the interface. Simulation of evaporation was carried out in an indirect fashion and a self-similar shrinkage of the liquid–vapor

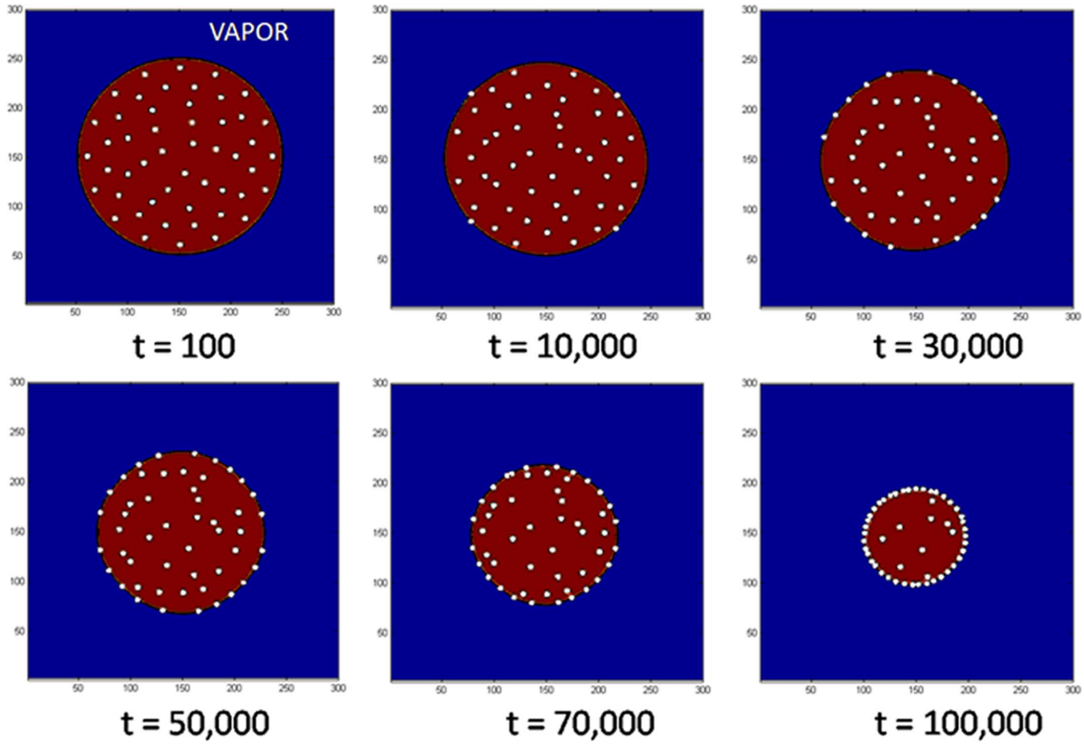

Fig. 9. Shrinking of a free standing, 2D liquid drop containing particles illustrates the tendency of particles to adhere to the shrinking liquid-vapor interface and cause jamming of the interface. A similar jamming effect might help explain contact line pinning with reference to a 3D drop evaporating on a substrate.

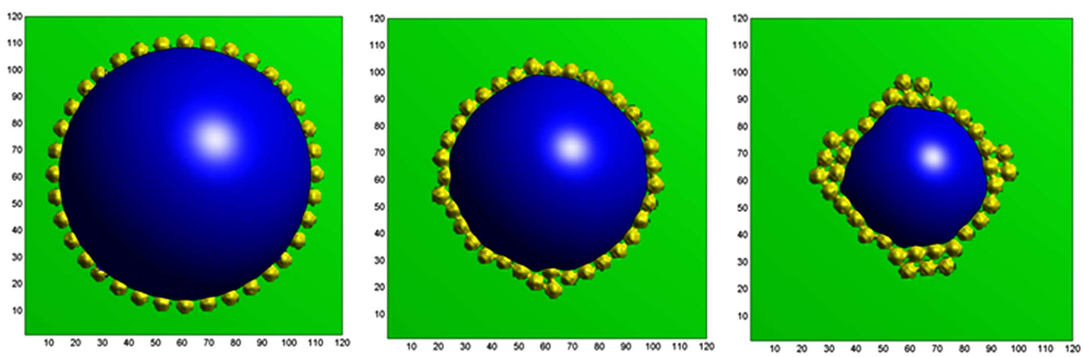

Fig. 10. Shrinking of a 3D liquid drop with particles at the contact line illustrates the tendency of particles to cause jamming of the interface. Particles are eventually ejected from the jammed structure and the drop continues shrinking. The particles eventually form a hexagonal close-packed structure.

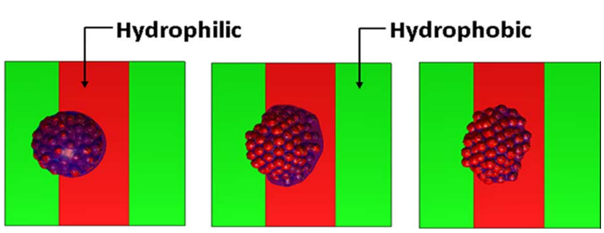

Fig. 11. Colloidal drop dynamics on a patterned substrate comprising of alternating hydrophilic and hydrophobic bands. The drop is initially at rest and contains 87 suspended particles throughout the liquid phase. The snapshots from left to right show the preferential wetting induced by surface energy steps on the substrate and the evaporating process.

interface was observed for a smooth substrate. If the surface is rough, a stick and slip behavior of the contact line is observed, consistent with previous studies. Evaporation of a freely suspended liquid drop containing suspended particles led to interesting results of colloidal jamming at the liquid–vapor interface that may help explain the mechanisms of coffee-ring deposits observed in experiments. Examples of this jamming phenomenon were presented via 3D simulations with suspended particles on the contact line.

Future work in this area involves a more detailed analysis of the evaporation process including non-isothermal LBM with the latent heat release and the extension of the suspended particle model to include long range inter-particle forces present in real colloidal suspensions. Simulations will also focus on the interaction of drops in forming a line pattern [38], [39]. A detailed parametric study and comparison with experiments will also be carried out. The LBM model described in this study thus has the potential to vastly improve our understanding and control of inkjet-printed colloidal drop dynamics for printable electronics fabrication.

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