Dielectrophoretic Trapping of a Floating Liquid Marble

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A liquid marble, a liquid droplet coated with hydrophobic powder, is an emerging digital microfluidic platform. It can potentially serve as a mixer or reactor for chemical and biological assays in the microscale. Automated manipulation of liquid marbles is essential for the implementation of more microfluidic functions in the technology platform of “lab in a marble”. We report the analytical and experimental results of trapping a floating liquid marble using dielectrophoresis in a nonuniform electric field. Liquid marbles with volumes ranging from 5 to 50 μL are effectively trapped by the dielectrophoretic force from a horizontal distance ranging from 10 to 60 mm and under an applied voltage ranging from 1.6 to 5 kV. A one-dimensional analytical model is then proposed for the trapping process and agrees well with experimental data. Finally, based on the relationship between the static friction coefficient and the Bond number of a floating liquid marble, an operation map is derived for successful trapping of the liquid marble, providing a better insight into controlled manipulation of liquid marbles.

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I. INTRODUCTION

Liquid marbles (LMs) are small liquid droplets encapsulated by a shell of hydrophobic micro- or nanoparticles [1–3]. Compared to bare droplets on a superhydrophobic surface, LMs provide a promising alternative to the transport and storage of small liquid volumes. LMs can move across different solid or liquid surfaces with a relatively small initial force, practically zero contamination, good compressive capability, a slower evaporation rate, and consequently less mass loss [4–8]. This feature is attributed to the presence of the protective porous particle shell around the liquid core as well as the air cushions between the LM and the carrier surface [5,9]. Unlike the complicated and costly electrowetting-based digital microfluidics (DMF), LM-based DMF relies on manipulating discrete droplets without being confined in a closed channel or the need for an insulating hydrophobic surface. This unique property of LM-based DMF provides a great flexibility to a wide range of applications.

As an alternative DMF platform, LMs can effectively serve as an individual compartment to provide various microfluidic functions. Recently, LMs have been innovatively used as microreactors for chemical [10–12] and biomedical [13–15] analysis. The three-dimensional geometry, low risk of cross-contamination, and gas permeability make a LM the perfect candidate for culturing

cells [16–18] and microorganisms [19]. Vadivelu *et al.* [16,17] used a floating LM as a bioreactor for growing three-dimensional cell spheroids. The floating LM allows cells to freely associate and interact to produce uniformly sized spheroids. The quality of the spheroids depends on the internal mixing process, which in turn depends on the motion of the LM over the supporting liquid surface. Moving the marble around induces internal flow and promotes mixing. Furthermore, two or more LMs containing different contents can be actuated to collide and coalesce to initiate mixing and reaction [20]. Besides, magnetic LMs [21–24] can be manipulated to implement multiple microfluidic functions such as reaction and detection. This concept is known as “lab in a droplet” [22] or “lab in a marble”. The key challenges of “lab in a marble” are (i) trapping a LM at a specific location and (ii) releasing a LM and transporting it to another location. A reliable and controlled approach for manipulating LMs is essential for the implementation of the above functions.

LMs retain their integrity on a solid surface before dissipating through evaporation. Floating on a free liquid surface prevents evaporation and allows LMs to last much longer for further actuation. There are several actuation schemes for sessile and floating LMs, including utilizing environmental changes such as temperature [25], pH [26], and irradiation [27], and being driven by external fields. These external fields are controllable electric [28–33], magnetic [22–24], and acoustic [34,35] fields and gravitational field [36]. We have successfully demonstrated

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the use of dielectrophoretic (DEP) force to move sessile [29,30] and floating LMs [31] in a nonuniform electric field. These manipulation schemes allow for dispensing, merging, trapping, and mixing droplets and LMs [20]. Among these tasks, trapping of droplets and LMs is one of the most important tasks in DMF. Trapping is a key process for sample preparation, observation, and detection. Passive droplet trapping using specific microchannel designs [37,38] and active droplet trapping using external energy [39,40] have been widely used for various microfluidic applications. However, trapping of LMs has not yet been studied. In many DMF applications, LMs have to be monitored or immobilized for further analysis. Thus, a simple, reliable, and highly efficient trapping method is needed for LM-based microfluidic assays. Because of their significance as a bioreactor, floating LMs are selected for the detailed study of this paper.

Compared to other trapping schemes, electric methods, particularly the DEP method, are practical trapping approaches for nonmagnetic floating LMs. Electric manipulation is attractive for LM-based biochemical applications due to the good biocompatibility without the invasive modification of the liquid core and the coating. The present paper investigates the motion of floating LMs during the trapping process with DEP force. Both experimental data and the analytical model form operation maps with the applied voltage, trapping distance, and LM volume as the parameters. This study aims to fill the gap in knowledge about LM manipulation and facilitate a better understanding of controlled manipulation of LMs.

II. MATERIALS AND METHODS

A. Preparation of liquid marbles

We use water as the working liquid for this study. A deionized water droplet (resistivity of $1.8 \times 10^5 \Omega\text{m}$) is coated by polytetrafluoroethylene (PTFE) powder (Sigma-Aldrich, nominal diameter of $1 \mu\text{m}$) to fabricate a LM. Considering the stability of a floating LM in a high-voltage electric field, marble volumes used in this study are set as 5, 10, 15, 20, 30, and $50 \mu\text{L}$, which are the common liquid volumes used in DMF. As the volume of the coating particles is much smaller than that of the water content, the volume of the coated LM is regarded as the same as of the corresponding bare droplet. The water droplet is first dispensed using a micropipette (Thermo Scientific Finn timer 4500, volume ranging from 0.5 to $10 \mu\text{L}$) on a bed of hydrophobic PTFE powder and then rolled for a few seconds until it is fully coated. The PTFE LM is created and manipulated in the laboratory environment with a temperature of $20^\circ\text{C} \pm 5^\circ\text{C}$ and a relative humidity of $50\% \pm 5\%$ in all experiments. The images of stationary sessile and floating LMs are shown in Appendix A. According to the method proposed by Eshtiaghi *et al.* [41],

the coverage rate of external coating particles is estimated at roughly 90%.

B. Experimental setup

Figure 1 schematically illustrates the customized experimental setup of this study. The setup consists of a container made of 1.5-mm thin polystyrene (PE) plate, a polymethyl methacrylate (PMMA) holder with a cylindrical stainless steel electrode, a set of linear stages, a high-voltage power supply, and a high-speed camera (Photron Fastcam SA3). The PE container is an open-top channel with a dimension of $120 \times 25 \times 10 \text{ mm}$, which is filled with deionized water to support the PTFE LM. The water level to be filled is quantified with two syringes, one with a 10-mL scale for adding deionized water to the container and the other with a 1-mL scale for extracting water to create a receding contact angle between the carrier water and the PE container. The PMMA holder is cut from a 10-mm-thick PMMA plate and has a 0.8-mm-diameter hole with an insulating tape at the bottom to carry the electrode of the same diameter. The positioning platform consists of two programmable linear stages (Zaber Technologies X-LHM150A and T-LS28M, with a travel range of 280 mm and 28 mm, respectively) and two manual linear translation stages (Thorlabs PT1, with a travel range of 25 mm) for adjusting the relative position of the target LM as well as the working distance between the electrode and the water surface. The programmable linear stage with a travel range of 280 mm is designed to carry the electrode holder for trapping the floating LM. DEP trapping of floating LMs relies on a high-voltage dc-dc converter (Eastern Voltage Research HVPS 2) connected to a power supply (Keithley 2200-30-5, maximum voltage/current of 30 V/5 A). The whole trapping process of the floating LM is recorded by the high-speed camera at a frame rate of 60 frames/s. A custom-made automatic triggering device synchronizes the power supply with the high-speed camera, so that video recording starts simultaneously with activating the electric field. The recorded images of 1024×128 pixels are

FIG. 1. The experimental setup for DEP trapping of a floating LM.

analyzed in Matlab (MathWorks) to detect the relative position and to calculate the velocity and acceleration of the floating LM.

C. Liquid marble trapping by dielectrophoresis

To start with, a fabricated PTFE LM with a volume ranging from 5 to 50 μL is gently placed on the water surface using a stainless steel chemical spoon. The LM floats on the carrier water surface. Both the LM and the water surface may deform, causing the LM to sink into the free surface. The high-speed camera is then adjusted to obtain a clear image over the whole range of marble movement. The electrode bottom surface is first moved to a distance of 8 mm away from the free water surface based on preliminary trials with distances ranging from 6 to 10 mm. The electrode height has to be enough to trap larger marbles without touching the electrode. The electrode right over the floating LM is then steadily positioned to specific locations using the 280-mm-range programmable linear stage, corresponding to six horizontal centroid distances between the electrode and the LM, namely, 10, 20, 30, 40, 50, and 60 mm ($\pm 10\%$ measurement and detection errors). The trapping distance setting considers both the image capturing and the effective working range of DEP force. Finally, the high-voltage converter is adjusted to provide five output voltages of 1.6, 2, 3, 4, and 5 kV. These voltage values consider both the effective trigger voltage of the high-voltage convertor and the critical applied voltage before marble rupture. The entire trapping process is recorded by the high-speed camera until the position of the floating LM stabilizes under the electrode. All data are collected and compared systematically using image analysis and simulation in Matlab.

III. ONE-DIMENSIONAL MARBLE TRAPPING MODELING

At a given position relative to an electrode in a nonuniform electric field, a LM experiences a positive DEP force induced by the electric field gradient [Fig. 2(a)]. The horizontal component of the DEP force $F_{\text{DEP}-r}$, as depicted in Fig. 2(c), works as the restoring force to attract the floating LM toward the position directly below the electrode. In addition, the friction force acts along the horizontal direction against the movement of the LM. Furthermore, the floating LM also experiences upward forces, namely, the vertical component of DEP force $F_{\text{DEP}-z}$, the buoyancy force, and the surface tension of the carrier liquid, as well as the weight acting as the downward force. The force balances in r and z directions (considering upward and toward electrode as the positive directions) are respectively

$$F_{\text{DEP}-r} - F_f - ma = 0, \quad (1)$$

FIG. 2. Schematic of the DEP trapping concept and the corresponding electric field distribution from a 2D analytical model. (a) The free body diagram of a floating LM and an electrode under a nonuniform electric field in the cylindrical coordinate system. (b) The 2D axisymmetric electric field generated by the analytical model. (c) The curve of horizontal DEP force $F_{\text{DEP}-r}$ versus LM position r in the electric field.

$$F_{\text{DEP}-z} + F_b + F_s - G = 0, \quad (2)$$

where m and a are the mass and acceleration of the floating LM respectively, $F_{\text{DEP}-r}$ and $F_{\text{DEP}-z}$ are horizontal and vertical components of the DEP force respectively, F_f is the friction force, F_b is the buoyancy force, F_s is the surface tension force, and G is the marble weight. The main focus of the present study is analyzing the dynamic behaviors in the translational motion of a floating LM and does not consider its rotational motion [42].

As the vertical displacement of the floating LM is negligible [43], we argue that the condition of the vertical force balance can always be met and the marble centroid height above the carrier water surface is kept constant during the motion of the floating LM. Thus, the trapping process in the narrow PE container can be regarded as a one-dimensional (1D) problem. We used a two-dimensional (2D) analytical model to describe the nonuniform electric field distribution [Fig. 2(b)]. For a successful trapping case, a stationary floating LM is firstly attracted from the initial equilibrium position from a given initial distance after overcoming the static friction force F_{SF} . When the LM starts moving, the friction then turns into the kinetic type

F_{KF} , which has already been characterized previously by our group [44]. The final equilibrium position of the floating LM is right below the electrode centroid after a few damping oscillations.

Because of the relatively small diameter of the electrode compared to that of the floating LM, we utilize a point-charge model to describe the electric field in the vicinity of the electrode. For a hypothetical point charge suspended above the interface between two different media, namely, air and water, the electric potential field can be described using the method of image charges as follows:

$$\begin{aligned} \Phi(q, d, r, z) \\ = \frac{q}{4\pi\epsilon_m} \left[\frac{1}{[r^2 + (d - z)^2]^{1/2}} - \frac{(\epsilon_w - \epsilon_m)/(\epsilon_w + \epsilon_m)}{[r^2 + (d + z)^2]^{1/2}} \right], \end{aligned} \quad (3)$$

where q is the electric charge, ϵ_m is the air permittivity with a value of approximately 8.85×10^{-12} F/m, ϵ_w is the relative permittivity of water and equals to $80\epsilon_m$, d is the height of the point charge to the ground surface, z is the vertical position to the ground surface, and r is the horizontal position from the point charge. The numerator $(\epsilon_w - \epsilon_m)/(\epsilon_w + \epsilon_m)$ in Eq. (3) has a value (0.975) close to 1. The applied voltage U is changed in experiments instead of determining the magnitude of the point charge. Consequently, Eq. (3) can be rewritten to describe the electric potential experienced by a LM at the position (r, z) , while the units are balanced at both sides of the equation:

$$\begin{aligned} \Phi(q, d, r, z) \\ = \Phi(U, d, r, z) \\ \cong Uh \left[\frac{1}{[r^2 + (d - z)^2]^{1/2}} - \frac{1}{[r^2 + (d + z)^2]^{1/2}} \right] \\ = Uh\sigma, \end{aligned} \quad (4)$$

where U is the applied voltage, h is the distance from the electrode to the water surface ($h = 8$ mm), and σ represents the spatial component of the electric potential field. A correction factor needs to be added as this approach is based on the point-charge model, which is just an approximation for the actual electric potential.

In a specific experiment with a fixed marble volume and applied voltage, U , h , and z remain constant. The potential distribution is 1D and only changes with r . Consequently, the electric field strength can be calculated as

$$\nabla E^2 = \nabla(-\nabla\Phi)^2 = U^2 h^2 \nabla(-\nabla\sigma)^2, \quad (5)$$

where E is the electric field strength. Considering the experimental parameter setting, the gradient part of Eq. (5)

along the r direction can be simplified as

$$\nabla(-\nabla\sigma)^2_r \cong \frac{Cd^2r}{(r^2 + d^2)^4}, \quad (6)$$

where C is the correction factor to the analytical model. As shown by the schematic of the experimental setup in Fig. 1, the electrode is partly inserted in the PMMA holder and its upper part is open for wiring. Based on the experimental data of marble trapping from a relatively long distance, a two-point-charge model is more practical for the electric field. One point of charge represents the bottom part of the electrode embedded in the PMMA holder. The other point of charge represents the upper part of the electrode exposed to the air.

Finally, the horizontal component of DEP force for trapping of the floating LM can be described analytically as

$$\begin{aligned} F_{DEP-r} &= 2\pi\epsilon_m R^3 \nabla E^2_r = 2\pi\epsilon_m R^3 U^2 h^2 \nabla(-\nabla\sigma)^2_r \\ &= 2\pi\epsilon_m R^3 U^2 h^2 \left[\frac{C_1 d_1^2 r}{(r^2 + d_1^2)^4} + \frac{C_2 d_2^2 r}{(r^2 + d_2^2)^4} \right] \\ &= \frac{3}{2} \epsilon_m V U^2 h^2 \left[\frac{C_1 d_1^2 r}{(r^2 + d_1^2)^4} + \frac{C_2 d_2^2 r}{(r^2 + d_2^2)^4} \right] \\ &= \frac{3}{2} \epsilon_m V U^2 h^2 \psi(r), \end{aligned} \quad (7)$$

where R and V are the radius and volume of the undeformed floating LM respectively; C_1/C_2 and d_1/d_2 are, respectively, correction factors and heights to the ground surface of the two-point electrode; and $\psi(r)$ represents the position component containing the horizontal centroid distance r .

IV. RESULTS AND DISCUSSION

The relationships included in the analytical model could explain the dynamic behavior of floating LMs observed in the experiments. A successful trapping process is not an attractive one-way motion with a constant velocity. A floating LM first needs to overcome a barrier of electric potential [Fig. 2(c)] before it can get to the position below the electrode. After escaping the peak of the electric potential, the floating LM accelerates to reach the maximum velocity under the effect of the DEP force and then decelerates slowly until it stabilizes again under the electrode center line. Removing the nonuniform electric field induces the floating LM to move out of the scope of the electrode holder to the next equilibrium position.

A. Marble detection and dynamic data analysis

In a successful trapping case, the floating LM moves from its initial equilibrium position and accelerates toward

FIG. 3. The DEP trapping process of a floating LM (10- μ L volume, 30-mm distance, and 4-kV applied voltage). (a) The position of the LM during the trapping process. (b) The corresponding velocity of the LM. (c) The corresponding acceleration of the LM. (d) The oscillation of the floating LM relative to the electrode and fitting with a DSHM solution (the starting point is set as the first time that the LM is trapped under the electrode center line).

the position below the electrode along the radial direction r . When the floating LM overshoots the electrode center line, it decelerates and oscillates around the electrode. The final movement is similar to the well-known damped simple harmonic motion (DSHM). The magnitude of the initial overshoot is usually less than 5 mm, as depicted in Fig. 3(a). Based on the experimental data of the relative centroid position of a floating LM, we can calculate the corresponding average velocity and average acceleration in a short time period (0.05 s) and regard them as the instantaneous velocity v and instantaneous acceleration a of the floating LM at the start of the time period [Figs. 3(b) and 3(c), respectively]. More data analyses on marble trapping are given in Appendix B.

We discuss here the relevant dimensionless number for the dielectrophoretic trapping process of the LM. First, the

corresponding Reynolds number Re of a floating LM can be defined as

$$Re = \frac{\rho v R}{\mu}, \quad (8)$$

where ρ and μ are the density and dynamic viscosity of the carrier liquid respectively and v is the instantaneous velocity of the floating LM. As the marble velocity varies with the trapping time, the value of Re changes consequently. However, we can determine the order of magnitude of the Reynolds number from experimental data. As $\rho \sim 10^3$ kg/m³, $v \sim 10^{-3}$ to 10^{-2} m/s, $R \sim 10^{-3}$ m, and $\mu \sim 10^{-3}$ kg m⁻¹ s⁻¹, we can see that $Re \sim 1-10^1$. Under the condition of such low Re , the flow of the carrier liquid around the LM during the trapping process can be considered as laminar. Therefore, the friction force term can be estimated by Stokes's law. Secondly, the corresponding capillary number Ca of the floating LM is defined as

$$Ca = \frac{\mu v}{\gamma}, \quad (9)$$

where γ is the effective surface tension of the floating LM with a value of approximately 7.2×10^{-2} N/m. Similarly, we can deduce that the order of magnitude of Ca depends on the value of the velocity. The capillary number approximately ranges from 10^{-5} to 10^{-4} , indicating that the surface tension effect plays an important role in the marble trapping process. The shape of the floating LM therefore remains stable during the trapping process. Thirdly, the corresponding Bond number Bo of the floating LM is defined as

$$Bo = \frac{\Delta \rho g R^2}{\gamma}, \quad (10)$$

where $\Delta \rho$ is the density difference between air and the LM, which is approximately the marble density, and g is the gravitational acceleration. The larger the volume of the floating LM is, the bigger the Bond number. Thus, the value of Bo for this study is estimated up to 0.725 when a 50- μ L floating LM appears. The small Bond number indicates that the surface tension still dominates the marble trapping process in most experiments, especially for LMs with relatively small volumes. The water marbles thus can float with a certain height over the water surface, supporting the assumption of our model.

The kinetic friction between the floating LM and the carrier water can be represented by the Stokes friction with a correction factor f , which takes into account the effect of the deformed meniscus [44]. Combining all the factors that can possibly affect the correction factor f , the kinetic

friction term in Eq. (1) can be expressed as [31]

$$F_{KF} = 6\pi f(U, R, z)\mu Rv, \quad (11)$$

where f is the correction factor as a function of U , R , and z . Thus, Eq. (1) for a successful marble trapping case can be rewritten as

$$F_{DEP-r} - 6\pi f(U, R, z)\mu Rv - ma = 0. \quad (12)$$

To better understand the damping oscillation at the end of the trapping process, we compare the equation of DSHM with Eq. (12). For an unforced DSHM, the first recovery motion of the target object can be described as

$$kr - cv - ma = 0, \quad (13)$$

where c is the viscous damping constant or drag coefficient (kg s^{-1}) and k is the spring constant (kg s^{-2}). Once the floating LM is trapped within the electrode radius, we can consider the LM as the target object, F_{DEP-r} as the effective spring and the carrier water as the damper. Therefore, the viscous damping coefficient term c in Eq. (13) is analogous to the friction term, $6\pi f\mu R$, of Eq. (12) at the end of the trapping process.

We fit the solution of DSHM to the experimental data during the damped oscillation of a floating LM, as shown

FIG. 4. The theoretical electric field distribution of DEP trapping of a floating LM and the corresponding force fitting (15- μL volume and 5-kV applied voltage). (a) The theoretical electric field distribution. (b) The corresponding real-time DEP force curve from experimental data (blue points) and nonlinear parameter fitting outcome (green solid line), as well as the simulation result from the theoretical model (red solid line).

in Fig. 3(d). The fitting curve follows the form of DSHM:

$$r = \lambda_1 e^{-\lambda_2 t} \cos(\lambda_3 t + \lambda_4) + \lambda_5, \quad (14)$$

where λ_1 to λ_5 are the detailed fitting parameters. These fitting parameters determine the values of c and k in Eq. (13), since the fitting curve expression is matched with the solution of the damped harmonic oscillator equation:

$$r = A e^{-(c/2m)t} \cos \left[\sqrt{\frac{k}{m} - \frac{c^2}{4m^2}} t + \varphi \right] + B, \quad (15)$$

where A is the amplitude of the damped oscillation, φ is the phase lag, and B is the displacement adjustment. In each experimental trial of marble trapping, we obtain the value of c and further figure out the magnitude of Stokes friction in the damped oscillation at the end of the trapping process.

As the experiment parameters do not change within a single experiment and the effective centroid height of a

FIG. 5. The calculated DEP force from experimental data in various experiments and the corresponding theoretical data of the model as well as the static friction coefficient fitting. (a) DEP force versus marble volume (5–50 μL) at 5-kV applied voltage and 10-mm distance. (b) DEP force versus the square of applied voltage (1.6–5 kV) at 10- μL marble volume and 10-mm distance. (c) The relationship of the static friction coefficient and the Bond number of floating LMs in DEP trapping (logarithmic coordinates).

floating LM is assumed to be a constant, we deduce that the kinetic friction, or Stokes friction, of the trapping process only depends on the velocity of the floating LM. Based on the analysis mentioned above, we can evaluate the real-time DEP force at various positions in each experiment using Eq. (12). We show the theoretical electric field distribution of DEP trapping of a 15- μL floating LM for an example [Fig. 4(a)]. We plot the corresponding $F_{\text{DEP}-r}$ versus r and then fit it with the form of Eq. (7), as shown by the blue circles and the green solid line in Fig. 4(b).

In experiments with the highest applied voltage (5 kV), floating LMs with various volumes (5–50 μL) can be trapped from a distance ranging from 40 to 60 mm. We characterize the values of C_1 , C_2 , d_1 , and d_2 in these cases and find that the values of d_1 and d_2 , which refer to the peak positions of the DEP force, tend to be constant at $0.0115\% \pm 9\%$ and $0.05\% \pm 15\%$ respectively. Subsequently, we determine the remaining parameters C_1 and C_2 in the analytical model after setting the fixed peak positions of the DEP force curve. Based on the values of correction factors C_1 and C_2 , we set them as integers: 4 and 1000. With all parameters fixed in the analytical model, we then use this model to fit DEP trapping of the 15- μL LM at 5-kV applied voltage, as shown by the red solid line in Fig. 4(b).

B. Validation of the dependence of dielectrophoretic force on marble volume and applied voltage

In the analytical model, we assume that the DEP force is proportional to the marble volume and square of the applied voltage. To validate the relationships of $F_{\text{DEP}-r} - V$ and $F_{\text{DEP}-r} - U^2$, we compare the analytical data with the experimental data of the DEP force.

Figure 5(a) clearly shows that the analytical model agrees well with experimental data, even for large LMs with a large Bond number of over 0.625. For 10- μL floating LMs trapped at different voltages from 1.6 to 5 kV, the relationship between the DEP force and the square of the voltage agrees well with both theory and experiment, as shown in Fig. 5(b). Therefore, it is reasonable to conclude that the analytical model with the form of Eq. (7) is suitable to analyze the data from successful trapping of a floating LM.

C. Determination of static friction coefficient of floating liquid marbles under dielectrophoretic force

As the analytical model for DEP marble trapping is validated, we can determine the minimum initial force to induce the marble motion on the carrier water surface by substituting the experimental parameters of successful

FIG. 6. Operation maps of DEP trapping of floating LMs with various volumes. (a) 5- μL LM trapping. (b) 10- μL LM trapping. (c) 15- μL LM trapping. (d) 20- μL LM trapping. (e) 30- μL LM trapping. (f) 50- μL LM trapping. Blue solid points represent the successful trapping in experiments, while red solid triangles are unsuccessful experimental counterparts. Blue open circles and red open triangles represent the successful and unsuccessful trapping cases predicted by the analytical model, respectively.

trapping cases into the values of V , U , and r in the analytical model. To further figure out the accurate value of the initial DEP force, we take the intermediate value between the successful trapping case with a minimum effort and its failed counterpart with lower applied voltage as the reference. For a successful trapping case, a stationary floating LM can be attracted from the initial equilibrium position after overcoming the static friction. At this boundary point, the magnitude of the trapping DEP force is regarded to be the same as that of the static friction force, namely,

$$F_{\text{DEP}-r} \cong F_{\text{SF}} = \mu_s N_{\text{LM}} = \mu_s \rho_{\text{LM}} V g, \quad (16)$$

where μ_s , N_{LM} , and $\rho_{\text{LM}} (\approx 1000 \text{ kg/m}^3)$ are respectively the static friction coefficient, normal force, and density of the floating LM. Thus, we can deduce that the minimum initial DEP forces of floating LMs with different sizes are similar to their static friction forces. The static friction coefficient can thus be calculated and plotted versus the Bond number as shown in Fig. 5(c). We can clearly see that the dimensionless static friction coefficient is a power-scaling function of the Bond number of the floating LM, which only changes with the undeformed marble radius, or the marble volume, in all experiments.

D. Operation maps of dielectrophoretic marble trapping

According to the fitting function of $\mu_s(\text{Bo})$ in Fig. 5(c), we can combine Eqs. (7) and (16) and simplify to obtain the required applied voltage to induce the motion of a resting floating LM as a function of trapping distance r and marble volume V , as follows:

$$U = f(r, V) = \sqrt{\frac{2\mu_s \rho_{\text{LM}} V g}{3\epsilon_m V h^2 \psi(r)}} = \sqrt{\frac{2E - 3\text{Bo}^{1.5} \rho_{\text{LM}} g}{3\epsilon_m h^2 \psi(r)}} \\ = \sqrt{\frac{5E - 4(\rho_{\text{LM}} g)^{2.5} V}{\pi \epsilon_m h^2 \gamma^{1.5} \psi(r)}}. \quad (17)$$

For each LM with a given marble volume, the applied voltage only depends on the trapping distance under the DEP force. Figure 6 shows the curve of minimum applied voltage for DEP trapping and compares it with experimental data in the $U-r$ space for different LM volumes.

The green solid lines in Figs. 6(a)–6(f) indicate the boundary for successful marble trapping for different LM volumes. The proposed analytical model can precisely predict whether a floating LM can be moved under a given applied voltage at different loading conditions with an accuracy of roughly 92% (the ratio of successful prediction outcomes to all experimental trials). The critical values of

FIG. 7. (a),(b) Images of sessile and floating 10- μL LMs and (c),(d) sessile and floating 30- μL LMs.

the applied voltage could be used as a reference in further study of DEP manipulation of floating LMs.

V. CONCLUSIONS AND PERSPECTIVES

Floating LMs with 5 to 50- μL volumes, serving as a three-dimensional cell culture DMF platform, can be effectively trapped by the DEP force from a distance ranging from 10 to 60 mm and under an applied voltage ranging from 1.6 to 5 kV. The precise trapping approach demonstrated in this paper shows a huge potential for automated manipulation of LMs. The general boundary condition of successful DEP trapping of floating LMs is proposed by utilizing a modified one-dimensional analytical model. The on-demand DEP trapping of floating LMs can be accurately controlled via changing the applied voltage. This method provides an alternative to the transport of small liquid volumes at a relatively long working distance. Future works could consider the use of the DEP force for trapping of two or more LMs and mixing through marble oscillation.

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FIG. 8. Dynamic analysis on DEP trapping of floating LMs with various experimental parameters. (a) The trends of displacement, velocity, and acceleration with time of 10- μ L LMs trapped from 10, 30, and 50-mm distances under 4-kV applied voltage. (b) The trends of displacement, velocity, and acceleration with time of 30- μ L LMs trapped from 10, 30, and 50-mm distances under 4-kV applied voltage. (c) The trends of displacement, velocity, and acceleration with time of 50- μ L LMs trapped from 10, 30, and 50-mm distances under 4-kV applied voltage. (d) The trends of displacement, velocity, and acceleration with time of 10- μ L LMs trapped at 1.6- to 5-kV applied voltages from 50-mm distance.

APPENDIX A: IMAGES OF SESSILE AND FLOATING LIQUID MARBLES

The images of stationary sessile and floating LMs are shown in Fig. 7. The static images of both smaller (10- μ L) and larger (30- μ L) PTFE water marbles on a clean glass slide [panels (a) and (c)] and the free water surface [panels (b) and (d)] are captured for comprehensive comparison.

APPENDIX B: DYNAMIC DATA ANALYSES ON DIELECTROPHORETIC LIQUID MARBLE TRAPPING

More dynamic data analyses on DEP trapping of floating LMs with various experimental parameters are accomplished for better understanding of marble controlled manipulation. Figure 8 consists of four panels that show the trends of displacement, velocity, and acceleration

with times of 10, 30, and 50- μ L LMs trapped from 10, 30, and 50-mm distances under 4- or 5-kV applied voltages.

APPENDIX C: VIDEOS OF SUCCESSFUL DIELECTROPHORETIC LIQUID MARBLE TRAPPING

Videos 1 to 6 show the DEP trapping process of typical cases (experimental parameters are listed in each caption). All videos are recorded at a rate of 60 frames/s and played at the same frame rate.

VIDEO 1. A 5- μ L LM trapped at 2-kV voltage from a 40-mm distance.

VIDEO 2. A 5- μ L LM trapped at 5-kV voltage from a 40-mm distance.

VIDEO 3. A 5- μ L LM trapped at 5-kV voltage from a 60-mm distance.

VIDEO 4. A 10- μ L LM trapped at 5-kV voltage from a 40-mm distance.

VIDEO 5. A 30- μ L LM trapped at 5-kV voltage from a 40-mm distance.

VIDEO 6. A 50- μ L LM trapped at 5-kV voltage from a 40-mm distance.

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