論文の内容の要旨

Abstract of Dissertation

Title of Dissertation: Droplet Manipulation Using Liquid Dielectrophoresis on Electret with Superlyophobic Surfaces
（エレクトレット上の液体誘電泳動を用いた超撥液表面における液滴操作に関する研究）

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Micro total analysis system (μTAS) or laboratory on a chip (LOC) is an active MEMS field integrating many subjects like mechanics, electronics, chemistry, biology and medicine, and aims at exploring micro/nano scale world and developing novel microfluidic devices for biophysics, biomedical and many other applications. Droplet-based microfluidics attracts much attention in recent years featured with high efficiency and high flexibility. Several electric actuation methods such as electrowetting on dielectric (EWOD) and liquid dielectrophoresis (L-DEP) are popular for droplet manipulation since surface tension and electric force are more significant in micro scale than in macro scale. However, high voltage requirement (typically higher than 30 DCV for EWOD or ~100 ACV for L-DEP) is disadvantageous for them to be commercialized. High actuation voltages root from dominance of surface tension in micro scale, leading to relatively large contact angle hysteresis (CAH).

Low wetting surfaces or super surfaces including superhydrophobic surfaces and superlyophobic surfaces (SLS) are preferred to improve microfluidic performance with extreme high contact angle (CA) and low CAH simultaneously. Whereas superhydrophobic surfaces have been well studied for decades, most of them are repellent only to water or high-surface-tension liquid. On the other hand SLS are superhydrophobic as well as superoleophobic (repellent to oil), and thus repellent to any liquid. SLS is a new field to be explored and only a few reports are available, lack of understanding for designing, fabricating and modeling. Conventional CA models fail to predict liquid behavior on SLS, and there is no theory available to predict droplet behavior on SLS.

Based on the above consideration, the objectives of the present study are

- To develop a new droplet manipulation method with a low-voltage actuation
- To develop robust, high performance SLS for microfluidics applications
- To establish a wetting theory to model the droplet behavior on SLS

- To integrate SLS with the proposed droplet manipulation method

A novel CMOS-compatible low-voltage droplet manipulation method named "Liquid Dielectrophoresis on Electret (L-DEPOE)" has been developed. By using a quasi-permanently charged material called electret as the voltage source, a dielectric droplet can be moved between two electrodes by switching electric relays/switches with a low voltage. Its actuation mechanism is clarified by employing Maxwell-stress-tensor method, and L-DEP is recognized as a kind of positive molecular-based DEP originated from Maxwell pressure in non-uniform electric field. To model the droplet motion in L-DEPOE, circuit and hydrodynamic models are developed, and then the governing equation of droplet motion is numerically solved.

The L-DEPOE droplet manipulation is demonstrated by microfluidic prototypes. Electret made by parylene-C and CYTOP with new polymer coatings was developed and tested, showing much better charge stability in hexadecane than pure CYTOP electret. A ~5 nL dielectric droplet (silicone oil or hexadecane) can be observed to switch between two electrodes, when an external capacitor switches its connection to the bottom electrodes by a 5 DCV relay. For the first time, the actuation voltage for droplet transport has been successfully reduced to 5 DCV. Hence, L-DEPOE achieves the potential to greatly broaden microfluidics applications with various digital circuits. However, high CAH of oil is responsible for the limited droplet speed and deformation, leading to complicated droplet motion traces.

To reduce CAH for low-surface-tension liquid for broader microfluidics applications, SLS are successfully fabricated by MEMS approach on silicon substrates with highly uniform 3D overhang structure. CA as high$^\circ$ as 158° and low CAH $\Delta\theta$~8° are achieved simultaneously for both water and hexadecane as well as good robustness and low adhesion. A systematic method including design and fabrication criteria for SLS is put forward for the first time, and open the way of high performance SLS for various applications in liquid operations.

A universal wetting theory has been established to model droplet behavior on SLS. Equilibrium, maximum/ minimum pinning CAs as well as advancing and receding CAs for smooth and rough surfaces are deduced from surface free energy analysis. Energy minimum principle and energy conservation principle is applied to analyze solid/liquid/vapor apparent contact line (ACL) thermodynamics, and is extended by adopting effective surface tension to investigate droplet behaviors on rough surfaces especially on the Cassie-Baxter (C-B) state, in which droplets are sitting on partially wetted solids with air cushion entrapped underneath. It is predicted from the present theory that a droplet will advance or recede mainly in the rolling mode on superhydrophbic or lyophobic surfaces, and low-surface-tension liquid will tend to leave residue film on SLS when receding. Such rolling behavior on superhydrophobic surfaces have been supported by previous reported experimental observation, and oil residue was also experimentally observed in present study.

From the present static CA model, four basic thermodynamic models including Young, C-B and Wenzel models as well as Gibbs inequalities have been deduced. Hence this wetting theory enables comprehensive understanding for droplet behavior on C-B rough surfaces. The formulas for static and advancing/receding CAs on the C-B state are verified by present experimental data of water and hexadecane CAs on SLS as well as previous CA data of superhydrophobic surfaces, and found they are in a nice agreement .

Finally, SLS with embedded electrodes are fabricated for the integration with L-DEPOE for faster droplet manipulation. And hexadecane CAs on the SLS are used to verify the universal wetting theory. Again the data are in good agreement with the theoretical prediction. Dynamic CAs tested with a wide range of droplet motion speed do not significantly change and thus the universal wetting theory can be applied with reasonable accuracy for faster droplet motion. By applying the threshold voltage through water to the SLS electrodes, quantitative evaluation of pressure stability is realized.

The fabricated SLS greatly reduce CAH in L-DEPOE configuration, but the pressure robustness is not enough. Simulation of L-DEPOE on isotropic SLS with f_s=0.01 shows that both static and dynamic CAH forces on SLS with embedded electrodes are much reduced, and the velocity of droplet motion is ~0.8 mm/s, increased by ~1.6 times for the droplet size on 500 µm order compared with L-DPEOE on flat electrodes. To further increase the velocity, larger droplet and electrode area are preferred, and ~2 mm/s velocity can be obtained when droplet is on 1000 µm order. SLS with embedded electrodes are expected to effectively improve the microfluidics performance and open a new way for low-consumption, high-efficiency µTAS to extend its applications with various liquids.