高度な製造のための多成分液体の熱流体物理の解明

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Revealing Thermofluid Physics of Multicomponent Liquids for Advanced Fabrication

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The Toyota Riken Scholar program proposed by the author aims to reveal the thermofluid physics, including wetting, dewetting, and phase change, in the evaporation-type fabrication process based on multicomponent liquids, such as OLED or perovskite thin films. During the program, I have focused on the wetting dynamics for various liquid-solid combinations, flow structure near three phase contact line of evaporating liquids, as well as, controllable crystallization from saline solution droplets by tuning the mass flux and internal flow. The findings have lead to fruitful results with scientific and application impacts.

1. Background

Fabrication based on solvent evaporation, represented by inkjet printing, is a common technique used in various fields such as material science, chemistry, and pharmaceuticals. This method involves the use of a solvent to dissolve a polymer or other substance and then allowing the solvent to evaporate, leaving behind a solid structure or film. The process of solvent evaporation and the induced self-assembly of functional materials are therefore of great importance to the properties of the fabricated thin films or structures.

While the fabrication process is often optimized from the engineering point of view, the thermofluid physics during the evaporation of multicomponent liquids (artificial inks for OLED, QLED, etc.) is far from a good understanding, which is a bottleneck issue in controllable fabrication of electronic devices (evaporation-type fabrication)[1].

For this reason, the Toyota Riken Scholar program, proposed by the author, aims to reveal the thermofluid physics, including wetting, dewetting, and phase change, in the evaporation-type fabrication process of OLED or perovskite thin films.

2. Research Progress

Several research topics have been conducted during the program, including exploration of wetting dynamics for various liquid-solid combinations, flow structure near three phase contact line, as well as, controllable crystallization from saline solution droplets by tuning the mass flux and internal flow.

In the aspect of liquid wetting, with both mathematical modeling and experiments, we reveal that, the wetting dynamics of volatile droplets can be scaled by the spatial-temporal interplay between capillary, evaporation, and thermal Marangoni effects (図1). We elucidate and quantify these complex interactions using phase diagrams based on systematic theoretical and experimental investigations.

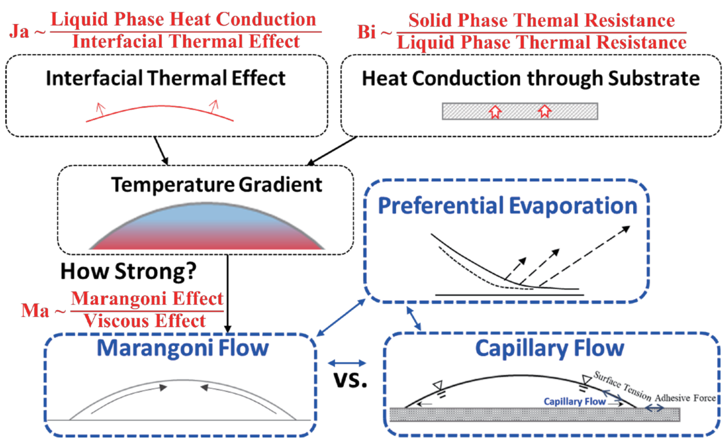

図 1　Decomposition of the dominating mechanisms in wetting dynamics of evaporative droplets.

A spreading law of evaporative droplets is derived by extending Tanner's law (valid for non-volatile liquids) to a full range of liquids with saturation vapor pressure spanning from 10^1 to 10^4 Pa and on substrates with thermal conductivity from 10^{-1} to 10^3 W/m/K. Besides

2024年2月29日　受理
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its importance in fluid-based industries, the conclusions also enable a unifying explanation to a series of individual works including the criterion of flow reversal and the state of dynamic wetting, making it possible to control liquid transport in diverse application scenarios.

In the case of flow structure near three phase contact line (TPCL), we indicate that for less volatile liquids such as butanol, the flow pattern is dominated by outwarding capillary flow (図2(a)). With increasing liquid volatility, e.g., alcohol, the effect of evaporation cooling, under conditions, induces interfacial temperature gradient with cold droplet apex and warm edge. The temperature gradient leads to Marangoni flow that competes with capillary flow, resulting in the reversal of interfacial flow and the formation of a stagnation point near TPCL (図2(b)). The spatiotemporal variations of capillary velocity and Marangoni velocity are further quantified by mathematically decomposing the tangential velocity of interfacial flow. The conclusions can serve as a theoretical base for explaining deposition patterns from colloidal suspensions, and can be utilized as a benchmark in analyzing more complex liquid systems, such as quantum dot inks or perovskite solutions for practical application.

In the aspect of controllable crystallization from saline solution, we have combined transparent observation and micro-particle image velocimetry (μPIV) for relating the crystallization process with the transition of flow field. Approaches with vapor field control and substrate heating have been applied for controllable nucleation formation and crystal growth, which can provide direct theoretical supports for high quality crystallization in the fabrication of perovskite thin films.

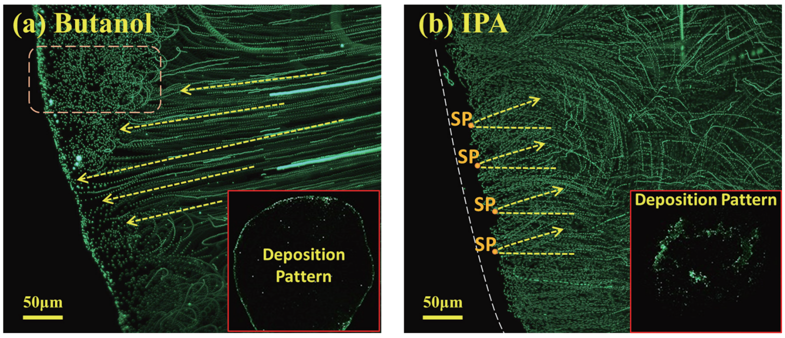

図２ Trajectory of tracing particles reveals the flow field near three phase contact line of evaporating drops.

3. Achievements, Conclusions and Perspectives

The above research findings have lead to one Editor's Pick paper in Applied Physics Letter[2], one paper with minor revision in Journal of Fluid Mechanics[3], and one paper under preparation for Physical Review Fluids.

To conclude, analyzing and understanding the behavior of multicomponent liquids is crucial in fields such as chemistry, chemical engineering, pharmaceuticals, and material science. With extensive knowledge of the fundamental thermal-fluid science, we will be able to take full control of the liquid behaviors and therefore high-quality fabrication of solvent-evaporation-based surface structures and thin films.

With more to explore, I would like to thank the Toyota Riken Scholar program for providing an excellent interdisciplinary platform for researchers from different fields and with different experts to communicate, discuss, and work together on important and mutually interested scientific or technical problems. I am also looking forward to solid collaborations with researchers with fabrication and biochemical backgrounds through Toyota Riken Scholar Joint Research in the future.

REFERENCES

1) Z. Wang, D. Orejon, K. Sefiane and Y. Takata, *Physics Reports,* **960** (2022) 1-37.
2) Z. Wang, G. Karapetsas, P. Valluri and C. Inoue, *Applied Physics Letters,* 124.10 (2024). (Editor's Pick)
3) Z. Wang, G. Karapetsas, P. Valluri and C. Inoue, arXiv:2304.04030v3 (2024).