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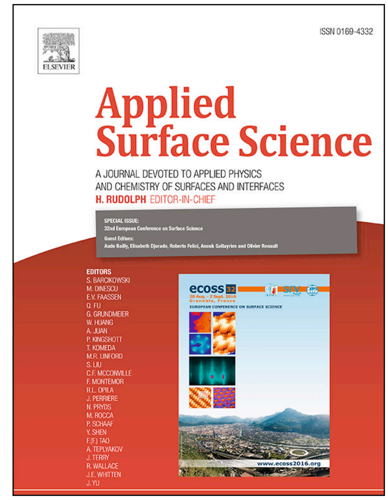

Full Length Article

High-throughput controllable generation of droplet arrays with low consumption

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High-throughput controllable generation of droplet arrays with low consumption

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We describe a controllable sliding method for fabricating millions of isolated femto- to nanoliter-sized droplets with defined volume, geometry and position and a speed of up to 375 kHz. In this work, without using a superhydrophobic or superoleophobic surface, arrays of droplets are instantly formed on the patterned substrate by sliding a strip of liquid, including water, low-surface-tension organic solvents and solution, along the substrate. To precisely control the volume of the droplets, we systemically investigate the effects of the size of the wettable pattern, the viscosity of the liquid and sliding speed, which were found to vary independently to tune the height and volume of the droplets. Through this method, we successfully fabricated an oriented single metal-organic framework crystal array with control over their XY positioning on the surface, as characterized by microscopy and X-ray diffraction (XRD) techniques.

1. Introduction

The generation of droplet arrays possesses great significance, both for the fundamental study of wetting[1, 2], dewetting[3, 4] and thermodynamics[5, 6] as well as various applications to compartmentalize chemical reactions[7, 8], biological analyses and materials syntheses[9, 10]. Many methods have been developed to fabricate droplets, such as laser microfabrication[11-14], inkjet printing[10, 15], microfluidic pen lithography[16, 17], electrowetting-on-dielectric (EWOD) actuation[18,19], trapping droplets by microcavities[3], solvent exchange[20-22], and discontinuous dewetting on a patterned surface. Among these methods, discontinuous dewetting is a straightforward method to produce high-density microdroplet arrays at low cost by dewetting the liquid along a hybrid surface with regions of different wettability[2, 16]. Huizeng Li and co-workers fabricated femtoliter droplet arrays by splitting an aqueous mother drop on a patterned superhydrophilic-superhydrophobic substrate, thereby isolating single cells[23]. Shaozhou Li et al. and Kerui Xu et al. deposited aqueous droplet arrays on a (super) hydrophilic/hydrophobic surface using the dip-coating method, respectively[24, 25]. Jin-Liang Zhuang et al. deposited droplet arrays of dimethylsulfoxide (DMSO) solution on Au substrates with patterned self-assembled monolayers by spin-coating[26]. Wenqian Feng et al. produced low-surface-tension liquid droplets arrays by moving a bulk drop on highly wettable and nonwettable slippery areas on smooth glass or flexible polymer films[27]. Using this method, which did not use a superoleophobic surface, a wide range of low-surface-tension liquids, including ethanol, methanol and even hexane, could be used to generate separated droplets arrays. These single-step formations of thousands of droplets in precise locations in an array format with desired volumes provide a unique solution for high-throughput applications. However, large-scale droplet arrays require uniform, precise and controllable dewetting speed and additional reagent consumption because the hybrid surface must first be wetted with bulk liquid.

In this work, we developed a facile, ultra-high-throughput droplet array generation method by sliding a liquid strip along the substrate in a controllable manner with low reagent consumption. Millions of femtoliter to nanoliter droplets with defined positions, geometry and volume could be produced in seconds (up to 375kHz). This method enables the fabrication of droplet arrays compatible with polymer, oil, water, mixture and organic liquids with surface tension as low as γ_{lv}=18.8 dyn/cm. In addition, the key factors in controlling the droplet volume, including the wettable pattern size, the viscosity of the liquid, and the relative sliding speed of the liquid strip, were also discussed systematically. These factors can also be regulated to control the volume of the droplet, which is significant for materials synthesis, especially for the oriented growth of single crystals. In addition to providing insights into the fundamental mechanism underlying the control of the droplet volume, we also demonstrate, as a proof-of-concept application, a simple technique to fabricate arrays of single-crystal metal-organic frameworks (MOFs). This method is based on evaporation-induced self-assembly crystallization in the femtoliter droplet arrays.

2. Material and methods

2.1. Chemicals and materials

AZ9260 photoresist, AZ400K developer and SU8 2002 photoresist were purchased from Suzhou Wenhao Microfluidic Technology Co., Ltd. (Suzhou, China). 1H,1H,2H,2H-Perfluorooctyltriethoxysilane (POTS) was

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