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## Short communication

# Hydrodynamic control of droplets coalescence in microfluidic devices to fabricate two-dimensional anisotropic particles through boundary element method

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## A B S T R A C T

Multicompartment and patchy particles attract much attention recently due to their great potentials in many fields such as drug delivery systems and photonic crystal materials. The controlled coalescence of droplets might be a promising approach to fabricate such particles since different droplets might be composed of distinct components or might contain diverse functional solutes. Through a two-dimensional boundary element method, we investigate the controlled adhesion of small droplets to specific locations of a main drop via careful flow control at inlets and outlets of a skillfully designed microfluidic device at low Reynolds numbers. This paper presents an original procedure to construct anisotropic particles assisted by microfluidics. By alternating extensional and rotational flow at the central cavity of the micro-device, it is possible to make small droplets adhere to the main drop one by one at expected location and thus fabricate patchy particles with multiple patches at expected angles in two dimensions.

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**Keywords:** Anisotropic particles; Microfluidics; Droplet coalescence; Boundary element method

## 1. Introduction

During the past ten years, anisotropic particles attracted much attention due to their great potentials in many fields, including drug delivery systems, photonic crystal materials and multiplexed bioassays (Du and O'Reilly, 2011; Lahann, 2011). Especially for multicompartment and patchy particles (see Fig. 1), their site-specific surfaces provide a great possibility to engineer the next generation of materials with expected internal structures. Simulation (Zhang and Glotzer, 2004; Doppelbauer et al., 2010) and experiment studies (Chen et al., 2011) have shown that assembly of patchy particles might generate diverse novel structures such as the kagome lattice. Some skillful preparation techniques have been

developed to fabricate such fantastic particles, such as templating, colloidal assembly, particle lithography, glancing-angle deposition, nanosphere lithography and capillary fluid flow (Pawar and Kretzschmar, 2010).

Recently, the rapid development of microfluidics provides a great platform to generate complex particles with diverse morphology and internal structures (Teh et al., 2008; Wang et al., 2011). Highlight news in Nature Physics about particle assemblies assisted by microfluidics to generate complicated aggregates in 2011 presented more about the great power of microfluidics in the construction of complex structures (Schneider et al., 2011; Gerstner, 2011). However, no serious effort has been put on the droplets assembly manipulated via controllable flows in skillfully designed microfluidic devices.

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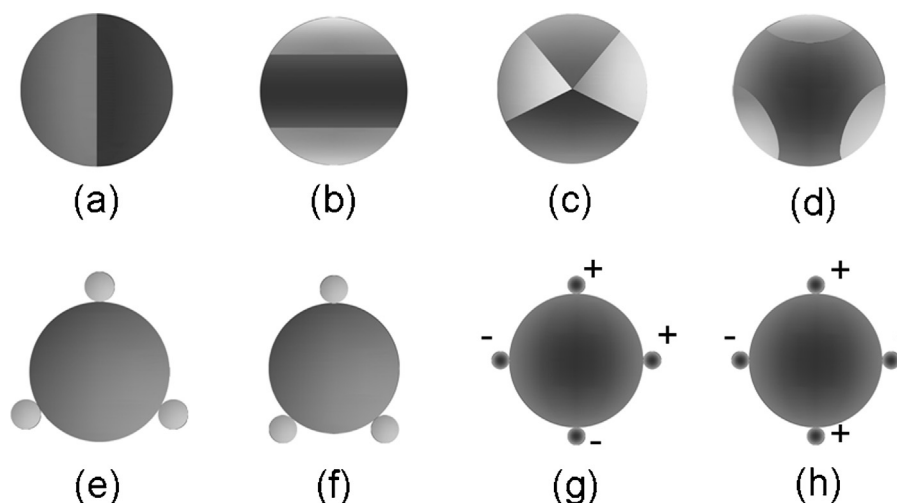


Fig. 1 – Illustration of the multicompartment and patchy particles.

Fine microfluidic manipulation of droplets might create a flexible approach to fabricate compartmentalized and patchy particles since those droplets can be composed of diverse materials or contain distinct functional solutes of various concentrations.

Fialkowski et al. (2005) have made use of the coalescence of droplets dyed by distinct colors to construct polymeric microspheres of complex internal structures. However, although the concept is presented, how to control the droplets in micron-scale and fabricate anisotropic particles in an efficient procedure is never given. The fine capability of microfluidics to manipulate particles and droplets makes it possible to fabricate anisotropic particles through the droplets coalescence. In droplet-based microfluidics, the fusion of droplets containing different reactants has been employed by many groups to carry on the chemical reaction in microchannels (Hung et al., 2006; Liu et al., 2007), in which the quick mixing of various components is expected. On the contrary, in order to retain the sharp interface of distinct materials, mixing should be avoided as best as possible in the fabrication of anisotropic particles. Besides, how to control the number and location of patches, especially the precise control of the angles between different patches, is another key issue in this process. Thus, a promising microfluidic device must be capable to generate flow fields which can adjust the coalescence location of various droplets, especially for the adhesion angle of the small droplet (SD) to a big main drop (MD). Hence, the geometry of the device and the control of flow rates at its inlets and outlets will be critical to achieve the goal.

A hydrodynamic trap of the cross-slot is the common device to trap particles, drops or cells in order to execute operations on them (Tanyeri et al., 2010). However, the simple flow fields generated in a cross-slot cannot fulfill the controlled coalescence of multiple droplets to fabricate patchy particles. Thus, we designed several hydrodynamic traps with arms more than four to execute more complex operations. Nevertheless, they are still inadequate to achieve the final goal. Fortunately, microfluidic four-roll mill (MFRM) was designed by Lee et al. (2007), which can generate flow fields from extensional, simple shear to rotational flows (Lee et al., 2007; Wang et al., 2012) at its central cavity. Based on MFRM, we designed a micro-device as shown in Fig. 2. Four pairs of inlet and outlet with the same width  $2w_0$  are arranged symmetrically around a central circular cavity with a radius  $r_c$ .  $w^*$  is the half width of four uniform bafflers and equals to  $0.4w_0$ . Unlike MFRM, there

are two more horizontal channels with width  $2w_0$  which could be used as the entrance of small droplet.

## 2. Mathematical formulations

Lately, Wang et al. (2013a,b) developed a more generalized boundary integral method (GBID) which can treat the rheology of multiple emulsions with diverse internal structures in microchannels with various geometries. In the current paper, since we only investigate the interaction among simple droplets (without any internal structure) in microchannels, a simplified form of GBID is employed. Validation of the numerical method GBID had been done in the paper (Wang et al., 2013b). In that paper, we successfully repeated the results of Stone and Leal (1990) for the deformation of a concentric double-emulsion globule under an axisymmetric extensional flow. It is well known that the boundary integral method is only valid for Stokes flows for which the inertia effects can be neglected. Thus, the calculation in this paper is only useful to the droplets coalescence for which the entire merging process is in Stokes regime (Yao et al., 2005; Sprittles and Shikhmurzaev, 2012). This means that all involved droplets must have very high viscosities.

As shown in Fig. 2,  $S_0$  is the boundary including the wall, inlet and outlet of the micro-device with unit normal vectors  $\mathbf{n}$  pointing inside. The two-dimensional (2D) droplet is driven by the continuous phase (CP) with viscosity  $\mu$  and density  $\rho$ .

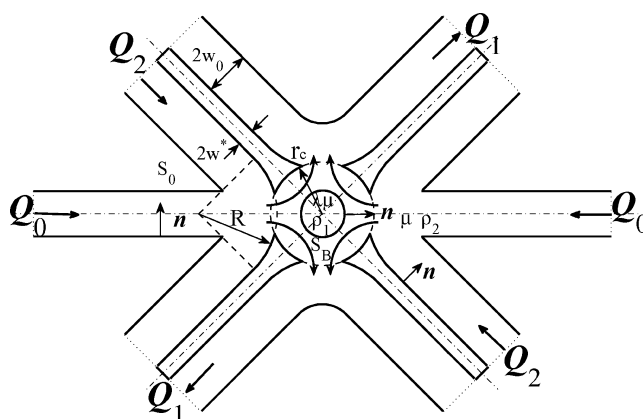


Fig. 2 – Illustration of a microfluidic four-roll mill.

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