Colloids and Surfaces A 553 (2018) 652–659

Contents lists available at ScienceDirect

Colloids and Surfaces A

journal homepage: www.elsevier.com/locate/colsurfa

Coating of solid particles with liquid layer by microfluidics

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GRAPHICAL ABSTRACT

ARTICLE INFO

Keywords:
Microfluidic T-junction device
S/W/O compound droplets
Flow regimes
Surface treatment
PS-PVA double-layer shells

ABSTRACT

A simple, low-cost and reliable method for coating of solid particles with liquid layer, namely formation of S/W/O compound droplets, is proposed and demonstrated experimentally via a vertical microfluidic T-junction device. At low flow rate ratio of oil (O) to water (W) phase, S/W/O compound droplets containing two solid cores are observed. With the increase of the flow rate of oil phase, S/W/O compound droplets with one solid core inside, also called single encapsulation, can be obtained. In single encapsulation, three typical flow regimes, which are squeezing regime, dripping-like regime, and jetting-like regime, can be quantitatively distinguished by the flow rates of both phases. S/W/O compound droplets, fabricated in the dripping-like regime, show a high monodispersity. Finally, polystyrene coated with polyvinyl alcohol layer (PS-PVA double-layer shells), used as a fuel container in inertial confinement fusion (ICF) experiments, can be successfully prepared via solidification of such compound droplets.

1. Introduction

Over recent decades, microencapsulation technologies have attracted a great deal of interest and now it possesses many potential applications for various fields, such as drug encapsulation, materials synthesis, crystallization and chemical reactions [1–5]. In the applications mentioned above, the liquid phase is usually encapsulated into a continuous phase, by which single droplet (W/O or O/W) and compound droplets (W/O/W or O/W/O) were successfully prepared [6–9]. However, with the increasing demand and development in the pharmaceutical industry, bioreactors and inertial confinement fusion (ICF) experiment, there is a growing interest in the coating of solid particles with liquid layer, namely fabricating of solid in water in oil (S/W/O) compound droplets [10–19].

S/W/O compound droplets are special droplets where the inner water phase acts as a barrier that separates the inner solid particle from the outer oil phase. These compound droplets have been applied in drug delivery for coating cells in which the coating layer acts as a drug carrier and a protective barrier. Besides, PS-PVA double-layer shells, prepared by solidifying the S/W/O compound droplets, also can be widely used as a fuel container in ICF experiments because of the low coefficient of gas permeability. However, one technically challenging application of such S/W/O compound droplets is the preparation of the droplets which meet the specifications for the precise control over the

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https://doi.org/10.1016/j.colsurfa.2018.05.096
Received 26 April 2018; Received in revised form 29 May 2018; Accepted 30 May 2018
Available online 30 May 2018
0927-7757/

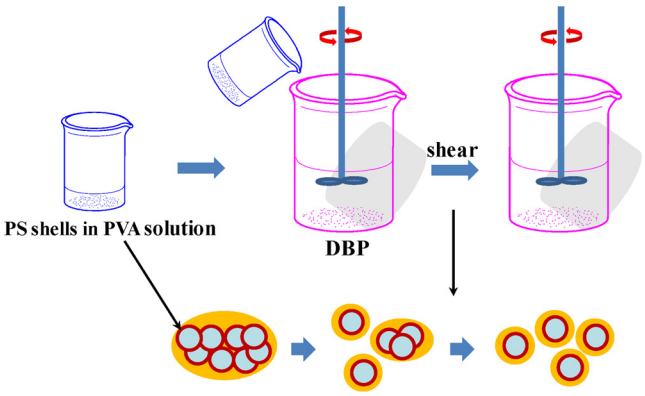

Fig. 1. Fabrication of S/W/O compound droplets by stirring method.

liquid layer thickness and monodispersity.

A two-stage emulsification method is now mostly used in the practical formulation of S/W/O compound droplets (Fig. 1). This technique exploits the turbulent shearing force generated by vigorous mixing so as to break off the droplets. However, the resulting S/W/O compound droplets, fabricated by a conventional stirring method, show a broad size distribution, poor controllability of liquid layer and reproducibility [20–22]. To overcome the shortcomings of such stirring method, Tsai et al, proposed a new method for the conformal coating of particles by driving the paramagnetic particles with magnetic force to pass through the W/O interface in a microchannel [23]. However, this method is only useful for coating of small paramagnetic particles and the coating thickness of the liquid layer is always thin enough (about 1 μm). As an alternative, various microfluidic method have been developed for manufacturing the droplets due to the controllability and repeatability, such as cross-flowing shearing, perpendicular flowing shearing, hydrodynamic flow focusing and geometry-dominated breakup [24–28]. Owing to the simple structure, easy facture, and low cost, a T-junction microfluidic device is therefore proposed to prepare S/W/O compound droplets in this work [29–35]. Pan et al., investigated the formation mechanisms of S/W/O compound droplets in a horizontal T-junction device [36]. However, when they are transported through a horizontal channel, the water phase can be easily peeled off from the surface of the solid particles, resulted by the density difference between three phases, causing a damaged compound droplets. Note that the essential applications involving such S/W/O compound droplets require the generation of many individual monodisperse droplets that do not disperse and lose their integrity. Accordingly, a vertical channel is used to effectively inhabit the contact of compound droplets with the channel wall so that integrated compound droplets can be obtained.

A new method for coating of solid particles via a T-junction microfluidic system was proposed in this work. Self-assembly T-junction device was fabricated through a pouring method, in which the PDMS precursor and cross-linker at ratio of 10:1 were initially poured into a mold, and then the PDMS was solidified at 80℃ for further reinforcement. After that, high fidelity intact chips can be prepared by disassembling the mold and cutting needless PDMS. PS shells with inner water drop and PVA solutions were regarded as solid (S) and water (W) phase, respectively. PS shells were transported by the W phase in the side channel. To meet the density-match demand, the mixture of dibutyl phthalate (DBP) and dioctyl sebacate (DOS) was employed as oil (O) phase, which was served as continuous phase and flowed in the main channel. When two phases meet at the T-junction inlet, the dispersed phase was sheared to form S/W/O compound droplets. The flow rates in the side and main channel were controlled by two syringe pumps. The interface evolution during S/W/O compound droplets formation process at T-junction were recorded by means of a high-speed camera.

2. Experimental

2.1. Materials

Polydimethylsiloxane (PDMS) (Suzhou Wenhao Lab-on-a-chip Technology Co.Ltd.), PS ($\overline{M}_W$ = 250 kg mol^{-1}, ρ_s = 1.05 g mol^{-1}, Acros Organics Inc), PVA ($\overline{M}_W$ = 13 kg mol^{-1} ~ 23 kg mol^{-1}, 87%–89% mole hydrolyzed, Aldrich Company), anhydrous calcium chloride ($CaCl_2$) (Chengdu Kelong Chemical Reagent Factory), DBP (Chengdu Kelong chemical Reagent Factory) and DOS (Chengdu Kelong Chemical Reagent Factory) were all used as received without further purification. Fluorobenze (FB) (Shanghai Jingchun Reagent Ltd.) was purified by distillation. Deionized water was used for all aqueous phases.

2.2. Fabrication of T-junction device

The single T-junction microfluidic device was fabricated by pouring PDMS precursor and cross-linker at ratio of 10:1 into a mold as depicted

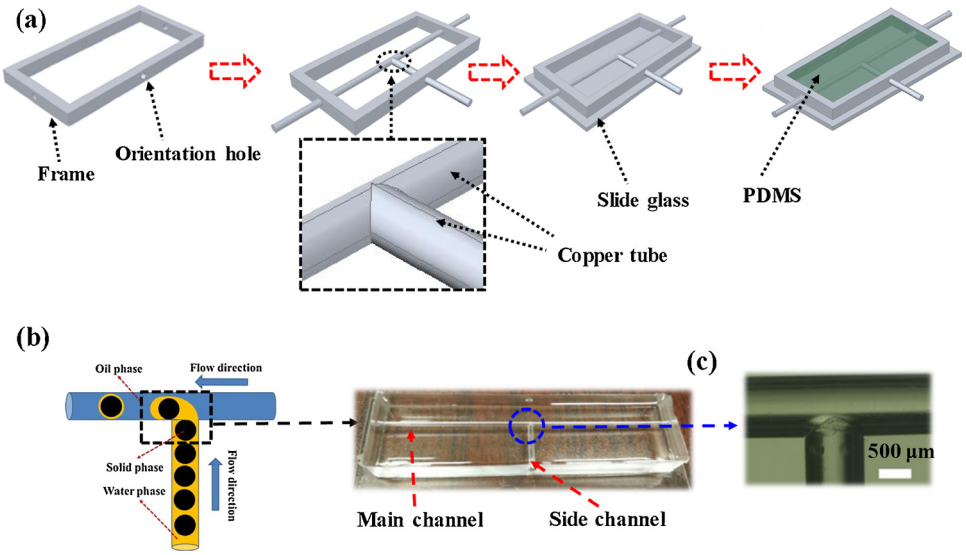

Fig. 2. Fabrication of T-junction device: (a) flow chart of the T-junction device fabrication, (b) 3D sketch map of S/W/O compound droplets formation and physical map of the T-junction device and (c) optical image of T-junction, the scale bar is 500 μm.