

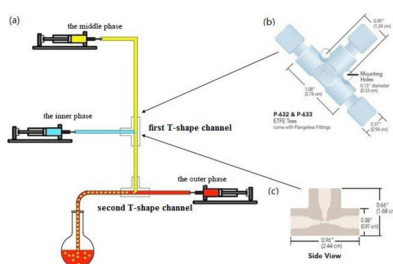


^b Laser Fusion Research Center, China Academy of Engineering Physics, Mianyang 621900, PR China

- Three various double emulsions made by one double T-junction microfluidic device.
- This device states remarkable advantages comparing to PDMS microchip.
- The size of these three double emulsions is associated with the flow rates.
- The prepared hollow microspheres shows perfect compact structure.

GRAPHICAL ABSTRACT

The preparation of different kinds of hollow microcapsules based on same assembled double T-shape device. The size of hollow microcapsules can be controlled by adjusting the flow rates of the three phases.



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ABSTRACT

A simple way to prepare water-in-oil-in-water ($W_1/O/W_2$) and oil-in-water-in-oil ($O_1/W/O_2$) double emulsions which is based on commercial double T-junction tubes was set up, which shows the remarkable advantages of simplicity, operability and cost-efficiency comparing with the traditional poly(dimethylsiloxane) (PDMS) microchip. By this device, the stable polyacrylonitrile (PAN) microbubbles have also been produced based during the experiments. The success in producing PAN microbubbles provides a new possibility for the preparation of hollow microspheres with polymer materials, which have the further potential applications in numerous particular fields.

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1. Introduction

Empirical researches have established the importance of microbubbles as they provide wide potential for applications in

various specific areas, including lightweight materials [1], aerated food products [2], wastewater treatment [3], ultrasound contrast agents [4], and pharmaceuticals [5,6]. Additionally, the methodology adopted in the manufacturing process of microbubbles is also applicable to other fields which have already attracted universal awareness (i.e. energy-storage materials [7], catalyst supports [8], and drug-delivery carriers [6,9]). Normally, microbubbles can be obtained through some specific techniques, which mainly include the approaches such as sonication, high shear emulsification, membrane emulsification, inkjet printing and coaxial

* Corresponding authors at: State Key Laboratory of Bioelectronics, Biological Science and Medical Engineering Department, Southeast University, Nanjing 210096, PR China. Tel.: +86 025 83619983; fax: +86 025 83795635.

E-mail addresses: lb6711@126.com (B. Li), lqge@seu.edu.cn (L. Ge).

electrohydrodynamic atomization (CEHDA). However, the major weakness of these traditional methods is the low monodispersity when generating the microbubbles [10–12]. More recently, microfluidic devices have been broadly accepted to apply in the formation of stable monodispersed microdroplets and microbubbles [13–18]. After the comparison of advantages and limitations of all these available methods, microfluidic devices have been selected to be the generation equipment of the microbubbles in this research.

Conventionally, the microfluidic devices mainly include three different types: flow-focusing, T-junction and co-flowing geometry. The separate types of devices, which have multiple intersections and flows with the majority following the microfluidic principle, are ideally designed for manufacturing uniform-size microbubbles and similar products [19–24]. Furthermore, the major reason for us to use a T-junction microfluidic device to generate microbubbles is that it increased the controllability of the flow rate, which enabled the repetition of the microbubble formation process [25]. More importantly, the T-junction microfluidic device provides additional remarkable advantages of simplicity, well operability, reusability, and cost-efficiency. It also increases the efficiency and controllability when producing the droplets. With the T-junction microfluidic device, both the quantity and the size are easily controllable.

In this paper, the major objective is to describe how to use a simple double T-junction microfluidic device which was constituted by oleophobic and hydrophobic materials to produce water-in-oil-in-water ($W_1/O/W_2$), oil-in-water-in-oil ($O_1/W/O_2$) and gas-in-water-in-oil (G/W/O) double emulsions. The device not only ensures monodisperse droplets, but can also be at a high pressure inlet flow conditions, the liquid droplets produced with a high frequency. As in the system of G/W/O double emulsions withstand

air pressure [26]. This will be followed by a discussion on how to control the size of the microbubbles. It is also notable that in this experiment, the formation process of G/W/O double emulsions was largely depended on the specific techniques developed for the first two. In the system of $O_1/W/O_2$ and G/W/O, the middle phase liquid was PAN solution. However, it is very difficult to find out the best way to dissolve PAN, the most commonly adopted solvents are DMF, dimethyl sulfoxide (DMSO), and concentrated salt solutions (zinc chloride, sodium thiocyanate, etc.) [14]. Among these solvents, DMF is a good choice due to its relatively large solvability and lower boiling point for evaporation [27–29]. Finally, without expensive materials and complicated processes such as microfluidic chips fabricated from flexible poly(dimethylsiloxane) (PDMS) through soft-lithograph, the experiment can be easily repeated due to the low marginal cost, which provides a more persuasive reason for this choice of technique.

2. Experimental

2.1. Materials

Benzene, 1,2-dichloroethane, N,N-dimethylformamide (DMF) and calcium chloride (CaCl_2) were obtained from Sinopharm Chemical Reagent Shanghai Co. Ltd, P.R. China. Polystyrene (PS) particles were acquired from Nacalai Tesque, Japan. Polyvinyl Alcohol (PVA, Mw = 40,000) solid and Aliquat 336 was bought from Alfa Aesar, England. Polyacrylonitrile (PAN, Mw = 60,000) was purchased from Tri-high Membrane Technology Co. Ltd, China. The silicon oil was procured from Shinetsu Chemical Co. Ltd., Japan. Deionized water was laboratory homemade. All the materials were used without further purification.

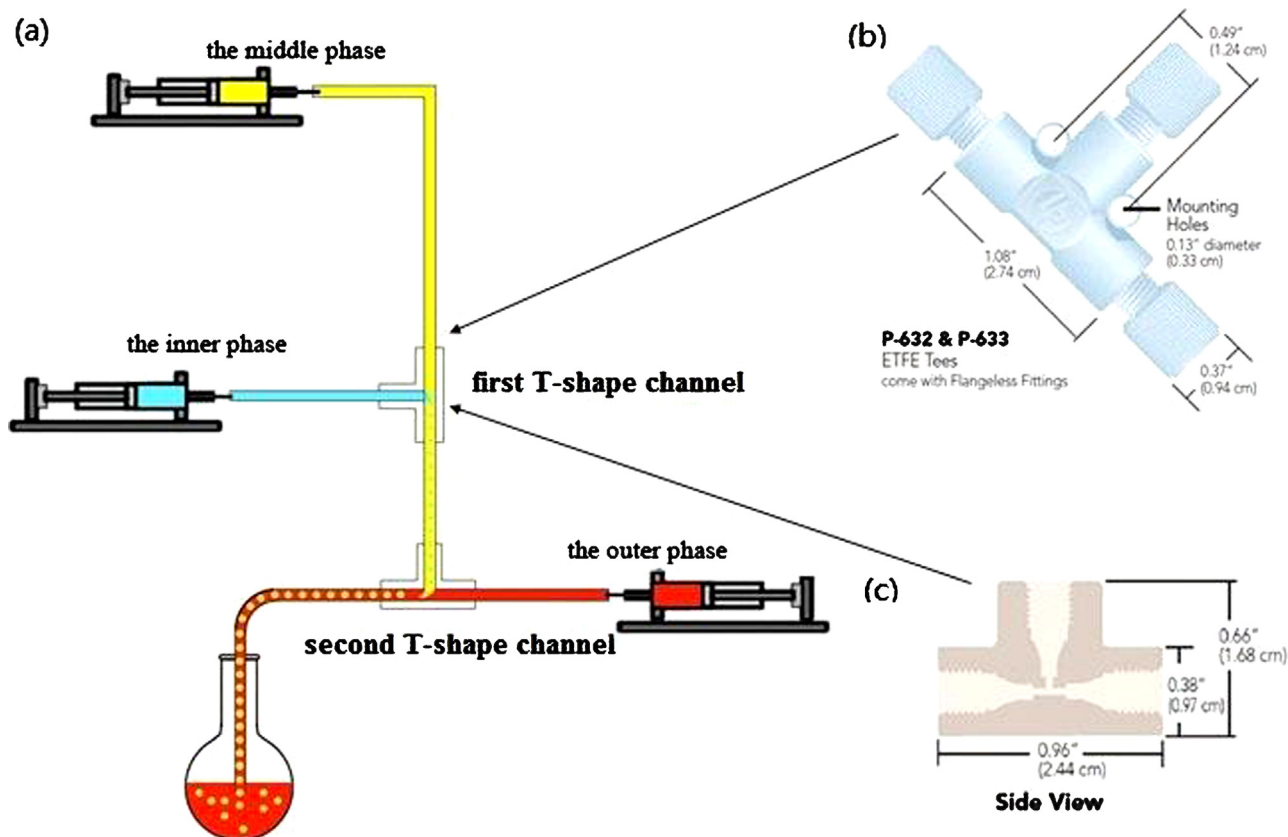


Fig. 1. Sketch of the double T-junction microfluidic device and droplet formation in the microfluidic device. (a) Sketch of the double T-junction microfluidic device. (b) The T-shape channel used. (c) The side view of T-shape channel.

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