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INVESTIGATION ON PREPARATION OF PCM EMULSIONS USING A CO-FLOW MICROFLUIDIC METHOD

Lisi Jia [1], Yi'ang Li [1], Ying Chen [1*], Songping Mo [1], Jun Li [1], Gang Liu [1]

[1] School of Materials and Energy, Guangdong University of Technology

ABSTRACT

Phase change material (PCM) emulsions have played increasingly important roles in many industrial fields as thermal energy storage media and heat transfer fluids. Precise size control of PCM emulsions is an important prerequisite for achieving consistent and repeatable performances. The present study introduced a novel co-flow microfluidic method to prepare uniform PCM emulsions in a controllable and reproducible manner. The droplet formation and size distribution of PCM emulsions were recorded in real time by a high-speed camera. The formation mode transition and droplet size variation were investigated by changing the flow rates of dispersed PCM and continuous water phases. The results showed that PCM emulsions with high uniformity and monodispersity can be attained in the squeezing and dripping modes, and the emulsion size increases with increasing flow rate of dispersed PCM and decreasing flow rate of continuous water. This study can provide technology support for future application of microfluidics in size-control of PCM emulsions.

Keywords: PCM emulsion, Co-flow microfluidic, Droplet formation, Size distribution

NONMENCLATURE

Abbreviations	
PCM	phase change material
PDI	polydispersity index
Symbols	
Q	flow rate
D	particle size

1. INTRODUCTION

Phase change materials (PCM) emulsion can be used either as thermal storage materials or heat transfer fluids and offers many advantages, such as high storage capacity during phase change, heat transfer at an approximately constant temperature, high heat transfer rate and low pumping power [1]. Many studies have demonstrated that the size of PCM emulsions is an important parameter determining the thermal storage and heat transfer performance of PCM emulsions. The PCM emulsions in these studies, however, were found to have serious problem in size uniformity, resulting in reduction in consistency and repeatability of experimental results.

The non-uniform size distribution is probably because the conventional emulsion process is usually triggered by mechanical stirring or ultrasonic vibration [2]. In the last two decades, a novel microfluidic technology has been emerged in manipulating well-calibrated droplets and bubbles for material science applications [3]. However, few studies on the utilization of microfluidic technique in fabrication of PCM emulsions are found in the literature. Droplet-based microfluidics has great potential for achieving monodisperse PCM emulsions with controllable sizes.

To date, studies have showed that the size of droplets in a microfluidic system is closely related to the mode of droplet formation [4]. Generally, the microfluidic approaches for droplet generation include five modes: squeezing, dripping, jetting, tip-streaming and tip-multi-breaking. The squeezing mode is dominated by the microchannel confinement. The other four modes arise from the capillary (Payleigh-Plateau) instability as the interfacial tension forces seek to

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minimize the interfacial area according to the thermodynamic principle of minimum interfacial energy. In these cases, viscous and inertial forces that act to deform the liquid interface counteract interfacial tension forces that resist the deformation. It is the competition of these forces that determines the specific mode of droplet formation and the size of droplets formed at a given set of parameters, such as viscosity, interfacial tension, and flow rate [5].

In the present study, a co-flow microfluidic technique was employed to generate PCM emulsions. Given that organic paraffin is the common material used for the PCM emulsions, the droplet formation and size distribution of n-heptadecane emulsions were investigated, emphasizing on the effects of the flow rates of dispersed and continues phases. The results can provide experimental support for controlling the size uniformity of PCM emulsions by microfluidic technology in the future.

2. EXPERIMENTAL

The experimental setup for the preparation of PCM emulsions is shown in Figure 1, which consists of two major components: droplet formation system and visualization observation system. The droplet formation system is composed of two syringe pumps (NE-100, New Era Pump Systems Inc., USA) and a co-flow microfluidic device. The microfluidic device was self-fabricated by coaxially inserting the injection capillary into the collection capillary. The collection capillary with an inner diameter of 900 μm was treated with dipotassium phosphate to make the glass surface hydrophilic in advance to prevent the adhesion of the organic PCM on the capillary wall. The injection capillary was tapered using a micropipette puller, and its orifice was then sandpapered to the final size of 60 μm. The deionized water and n-heptadecane, purchased from Aladdin Chemistry Co., Ltd., China, were used as the continuous and dispersed phases, respectively. Flow rates in the range of 10 to 50 μL/min were set for the PCM phase, and the water phase covers a flow rate ranging from 50 to 2000μL/min. The PCM droplet formation in the coaxial flow was monitored in real time by the observation system, which is made up of an optical microscope (6XB-PC, Shanghai Optical Instrument factory, China), a high-speed video camera (BDS300, Chongqing Optec Instrument Co., Ltd., China), and a computer. The formation mode of PCM emulsion droplets was captured and the corresponding droplet size distribution was recorded.

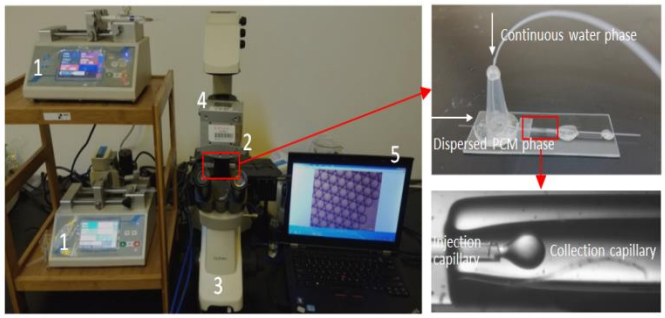

Fig 1 Experimental setup for the preparation of PCM emulsions using a co-flow microfluidic system. (droplet formation system: 1. syringe pump; 2. co-flow microfluidic device; observation system: 3. optical microscope; 4. high-speed camera; 5. computer)

3. RESULTS AND DISCUSSION

Figure 2 illustrates the PCM emulsion droplet formation in the coaxial-flow microfluidic device. Typically, the PCM phase was in contact with the water phase at the orifice of the injection capillary and grew over time, eventually pinching off to form emulsion droplets. Three different modes of droplet formation were observed in the PCM fluid system: squeezing, dripping and jetting. In the squeezing mode (Figure 1a), the PCM phase protrusion obstructs the collection capillary as it grows, thereby restricting the water phase flow around the developing protrusion. The interface of the enlarging droplet is squeezed to deform and necks into a PCM droplet at the orifice. The formed droplet is confined by the capillary wall, adopting a plug-like rather than a spherical shape. In the dripping mode (Figure 1b), the interface shape of PCM droplet remains essentially spherical and the drop attaches to the orifice of the injection capillary. With the increase of droplet size, the PCM droplet is stretched along the flow direction. After reaching an appropriate volume, the PCM droplet is dragged away from the orifice. Differing from the squeezing and dripping modes in which PCM droplet is formed near the injection orifice, the jetting mode shows a long jet before the PCM phase breaks into droplets. As shown in Figure 1c, a liquid jet extends in the same direction as the flow of the water phase, evolving into PCM droplets at a distance from the injection orifice. The resultant PCM droplets then pinch off due to Rayleigh-Plateau instability.

Figure 3 shows the formation mode of PCM emulsion droplets in a co-flow microfluidic device as a function of the volume flow rates of the PCM and water phases. In the PCM fluid system, three distinct types of transition between the squeezing, dripping and jetting

modes are observed: squeezing-dripping transition denoted by the red dividing line, squeezing-jetting transition denoted by the green dividing line, and dripping-jetting transition denoted by the blue dividing line. Previous studies have demonstrated that the droplet formation is governed by three types of forces, namely, inertial force, viscous force, and capillary force. The transition between the dripping and jetting modes is considered as a competition of the surface tension force and the viscous force. With the increase of the flow rate of water phase, the shear stress on the PCM droplet overcome surface tension, leading to the transition from dripping to jetting. The role that the flow rate of PCM phase plays in the dripping-jetting transition is ascribed to the inertial force. The squeezing mode is dominated by the confinement of the microchannel wall. As the flow rate of water phase increases, the viscous force exerted on the PCM droplet gives way to the wall effect of the capillary. The increased flow rate of PCM phase is also beneficial for the occurrence of squeezing mode at low flow rates of water phase.

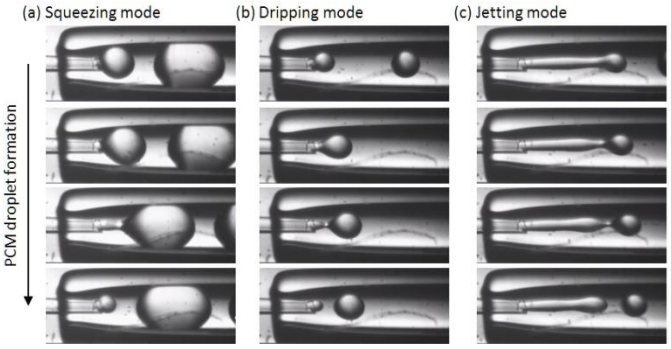

Fig 2 Typical formation process of PCM emulsion droplets in a co-flow microfluidic device. (a. squeezing mode; b. dripping mode; c. jetting mode)

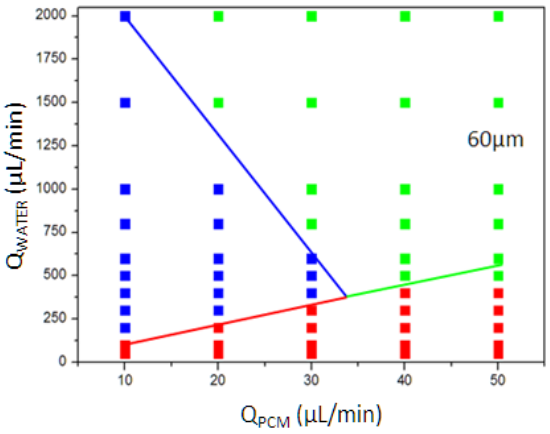

Fig 3 State diagram of the transition in formation mode of PCM emulsion droplets as a function of the volume flow rates of PCM and water phases. The red, blue and green squares denote the modes of squeezing, dripping and jetting, respectively.

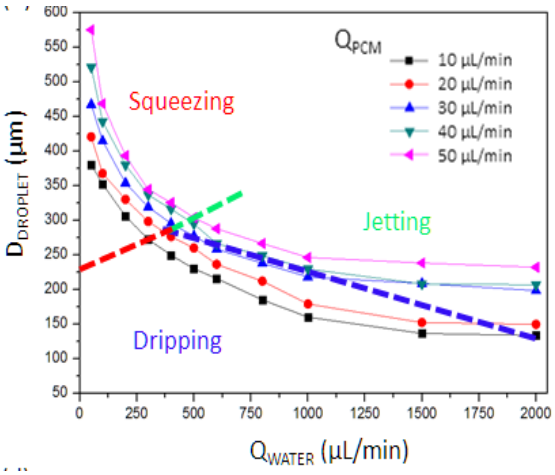

Fig 4 Average diameter of PCM emulsion droplets as a function of the volume flow rates of PCM and water phases. The red, blue and green dashed lines denote the mode transitions of squeezing-dripping, dripping-jetting and squeezing-jetting, respectively.

Figure 4 shows the typical size distributions of the PCM droplets observed in the squeezing, dripping and jetting modes. The droplet size distributions in the squeezing mode were measured when the PCM droplets collected became spontaneously spherical as a result of surface tension. The PCM droplets generated in the squeezing and dripping modes exhibits an extremely uniform size distribution, whereas the droplet size is diverse in the jetting mode. The polydispersity index (PDI) of the PCM droplets was calculated by statistical analysis of the particle size of PCM droplets in the optical micrograph. In the study, the PCM droplets formed in the jetting mode generally have higher PDI values than 0.05, suggesting that they cannot be regarded as a monodisperse system. The high monodispersity of PCM droplets can be achieved in the squeezing and dripping modes. It can be seen from Figure 4 that the PCM droplet size decreases as the flow rate of water phases increases. This is because the higher flow rate of water phase induces a higher velocity gradient on the two-phase interface and a higher viscous drag force that helps the droplet break up from the orifice and thus form smaller droplets. With the increase of PCM phase, the size of PCM droplets increases due to the increased inertia force which pulls the PCM droplet downstream to become larger and larger. The flow rate of water phase has obvious influence in the squeezing- and dripping-dominated

regimes, but little influence in the jetting-dominated regime. The influence of the flow rate of PCM phase is found to be more significant in the squeezing-dominated regime.

4. CONCLUSIONS

In the present study, organic paraffin, n-heptadecane, is used as the dispersed phase, and pure water is used as the continuous phase to prepare PCM emulsion droplets via a co-flow microfluidic method. The formation mode and size distribution of PCM emulsions are investigated, focusing on the effects of the flow rates of the two phases. With regard to the PCM, three modes of droplet formation are observed: squeezing, dripping and jetting, and three types of mode transition are found: squeezing-dripping transition, dripping-jetting transition and dripping-jetting transition. The size of PCM emulsion droplets decreases monotonously with the increase of water phase and the decrease of PCM phase. In the study, monodisperse PCM emulsions with sizes ranging from 90 to 580 μm were attained using the co-flow microfluidic method. Results on the droplet formation modes for the PCM fluid system and the corresponding conditions for size control are beneficial for the application of microfluidic technique in preparation of monodisperse PCM emulsions, which have great potential in electronics, building, textile, food, automotive industries.

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