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# On the successful encapsulation of water droplets into oil droplets

Xiaodong Chen<sup>a</sup>, Yingnan Sun<sup>b</sup>, Chundong Xue<sup>a</sup>, Yude Yu<sup>b</sup>, Guoqing Hu<sup>a,\*</sup>

<sup>a</sup>State Key Laboratory of Nonlinear Mechanics, Institute of Mechanics, Chinese Academy of Sciences,  
No.15 Beisihuanxi Road, Beijing 100190, China.

<sup>b</sup>State Key Laboratory on Integrated Optoelectronics, Institute of Semiconductors, Chinese Academy of Sciences,  
P.O. Box 912, Beijing 100083, China.

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## Abstract

Compound water-in-oil microdroplets can serve as microreactors in chemical and biological analyses. The inkjet printing is a useful technique to generate compound microdroplets by droplet impact. To understand the underlying physics during the droplet impact, a combined experimental and numerical study is carried out. The effect of spreading condition, impact velocity, and oil viscosity are investigated. The balance of the triple-line among the three interfaces dominates primarily the stable morphology of the compound droplet. Reducing oil viscosity can reduce the required impact velocity. High impact velocity is necessary to reduce the side-slipping of the water droplet.

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## 1. Introduction

Compound water-in-oil microdroplets can be used to isolate the active ingredient from the ambient. As the water droplets are encapsulated and protected in an oil shell, the compound droplet can be used as microreactors to perform chemical and biological analyses in picoliter or nanoliter volume [1]. In our previous study [2,3], a novel approach based on the double-inkjet printing technique are used to generate picoliter compound water-in-oil

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\* Corresponding author. Tel.: +86-10-82544298; fax: +86-10-82543977.

E-mail address: [guoqing.hu@imech.ac.cn](mailto:guoqing.hu@imech.ac.cn)

microdroplet arrays. The method is contact-free during sample loading. Ingredients can be easily added to the droplet on-demand. To understand the key parameters that influence the successful encapsulation of water droplets, a combined experimental and numerical study is performed. We consider the effect of the relationship of surface tensions of the three kinds of interfaces, impact velocity and oil viscosity.

## 2. Problem statement

Figure 1(a) shows the schematic of the problem considered here. A water droplet with an initial velocity,  $U$ , impacts onto an oil droplet resting on a plate in an air ambient. The impact dynamics is controlled by the geometrical and physical parameters of the two droplets, as well as the surface tensions between each two of the three phases. Two spreading conditions are considered in this paper,  $\sigma_{wa} < \sigma_{wo} + \sigma_{oa}$  and  $\sigma_{wa} > \sigma_{wo} + \sigma_{oa}$ , where  $\sigma_{wa}$  and  $\sigma_{wo}$ ,  $\sigma_{oa}$  are the surface tensions on water/air, water/oil and oil/air interfaces, respectively. As shown in Figure 1(b), the balancing of the three surface tensions at the triple-line determines the morphology of the compound sessile droplet when the droplets merge. For the Condition I, the water droplet floats on the top of the oil droplet. For Condition II, the water droplet will be completely engulfed into the oil droplet.

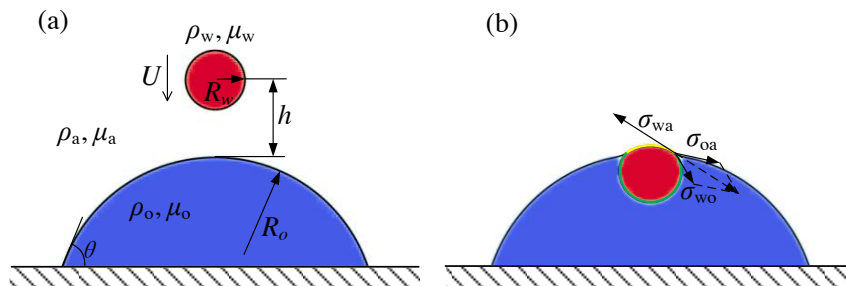


Fig. 1. (a) Problem setup; (b) Balancing of three surface tensions at triple-line.

## 3. Experimental setup

The inkjet printing methods described in our previous studies [1, 4] are used in the present paper to generate the compound sessile droplet. The detail information about the experimental platform can be found in Ref. [1]. Using a  $50 \mu\text{m}$  nozzle, Mineral oil (M8410, Sigma-Aldrich, USA) droplets are deposited on a silanized silicon dioxide surface to form an sessile droplet with diameter of about  $340 \mu\text{m}$ . Water droplet with diameter of about  $45 \mu\text{m}$  is printed to the top of the oil droplet using a  $30 \mu\text{m}$  nozzle. The impact dynamics is imaged using a microscope with a long-working-distance Nikon objective (S Plan Fluor ELWD 20 $\times$ /0.45) and tube lens with a cemented achromatic doublet. The evolution of the interfacial flow is recorded with a high-speed camera (Phantom v7.3, Vision Research Inc., USA) at frame rates of about 28 k fps. For Condition II, Span® 80 nonionic surfactant (S6760, Sigma-Aldrich, USA) is added to the oil phase to reduce the surface tension of the water/oil interface when the droplets merge. As shown in Table 1, the surface tensions of different interfaces are measured with pendant drop method by an optical tensiometer (Attension Theta, KSV Instruments, Finland). The viscosities of the water and oil are respectively 1 and 26 mPa·s measured by a viscometer.

Table 1. Surface tension for different spreading conditions.

Surface tension (mN/s)	$\sigma_{wa}$	$\sigma_{wo}$	$\sigma_{oa}$
Condition I	56.03	56.57	22.08
Condition II	56.03	6.68	22.07

#### 4. Numerical methods

To understand the impact dynamics, direct numerical simulations are also carried out to reveal the droplet deformation that is difficult to observe in experiments. The problem considered here involves three phases and three kinds of interfaces. A three-phase volume-of-fluid (VOF) method is developed based on of the Gerris open-source flow solver [2]. The overall numerical methods are validated by cases of the fluid lens spreading between two other fluids following Refs. [3, 5]. Contact angles at the three-phase contact line are in good agreements with the Young's relation. Since the change of topology of the interfaces can handle automatically through VOF method, the forming and vanishing of the triple-line among the three phases can be obtained without any additional model.

#### 5. Results and discussions

##### 5.1. Effects of spreading condition

Figure 2 shows the dynamics for three cases captured by the high-speed camera. Controlled by the spreading condition of the triple line, two types of impact outcomes are observed experimental. For Condition I (Figure 2(a) and (b)), the water droplet floats on the top of the oil droplet. For Condition II (Figure 2(c)), the water droplet is engulfed completely into the oil droplet.

When the impact velocity is low (as shown in Figure 2(a)), the water droplet contacts with the oil droplet with small deformation. The gas film between the droplets ruptures to form the water/oil interface and the triple-line among the three phases. Since the moving speed of the triple-line is faster than the impact velocity of the water droplet, a clear dark horizontal line in the third image indicates the quick rising of the triple-line. After that, a stable morphology is reached in the last image. The water droplet floats on the top of the oil droplet.

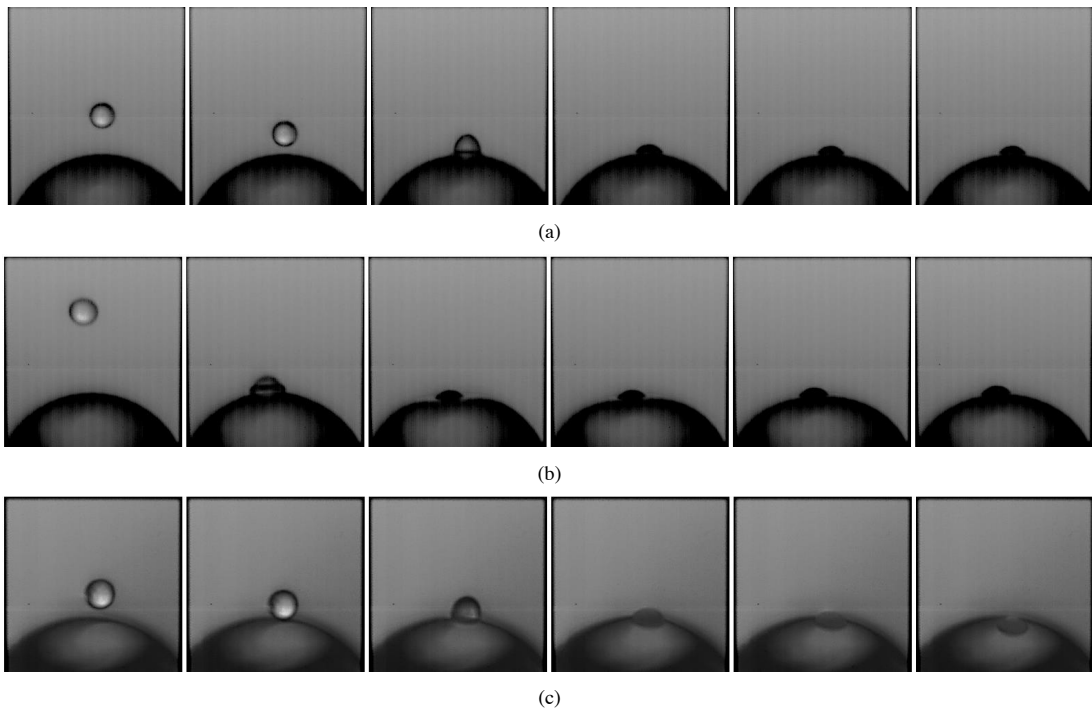


Fig. 2. Experimental images of a water droplet colliding with an oil droplet resting on a plate.

(a)  $\sigma_{wa} < \sigma_{wo} + \sigma_{oa}$ ,  $U = 0.8$  m/s; (b)  $\sigma_{wa} < \sigma_{wo} + \sigma_{oa}$ ,  $U = 3.8$  m/s; (c)  $\sigma_{wa} > \sigma_{wo} + \sigma_{oa}$ ,  $U = 0.5$  m/s.

When the impact velocity is high (as shown in Figure 2(b)), large deformation is observed in the second image. After the gas film ruptures, the water droplet penetrates into the oil droplet. A clear deformation of the oil droplet is observed in the third image. The deformation of the oil droplet recovers in the sequent images. Meanwhile, the water droplet moves up from the top of the oil droplet to relax to the stable morphology. A further increasing of impact velocity cannot increase the penetration distance of the water droplet. This is because the viscosity of the oil phase is much larger than that of the water phase. Major deformation consumes the most of kinetic energy of the water droplet. On the other hand, the increasing of acting area caused by the large deformation of the water droplet reduces the pressure acting from the water droplet to the oil droplet.

For Condition II (Figure 2(c)), even the impact velocity is low, the triple-line moves slowly to cause engulfment of the water droplet. In the application point of view, adding surfactant to the oil droplet for the successful encapsulation may have undesirable effect to the reagent in the water droplet.

### 5.2. Effect of oil viscosity

For the abovementioned reason, adding surfactant may not be an option to promote in some condition. In order to promote encapsulation, one may need to use an oil phase with low viscosity. To test this concept, a few numerical simulations are carried out. To validate the numerical simulations, the case in Figure 2(b) is reproduced numerically. The interfacial dynamics showed in Figure 3(a) is in good agreement with the experimental images.

The viscosity of the oil phase in Figure 2(b) is reduced to 2 mPa·s. The impact dynamics is shown in Figure 3(b). With a low viscosity, the deformation of the oil droplet is large to allow the water droplet to penetrate completely to change the morphology of the compound droplet. The opening of the oil phase closes under the recovery motion of the oil/air interface. The water droplet is then engulfed completely into the oil droplet. Another advantage of using less viscous oil is that the water droplet can settle directly onto the plate in the bottom. The possibility of the floating water droplet slip along the interface of oil droplet (as shown in Figure 2 (c)) is avoided. The position of the water droplet can then be more controllable.

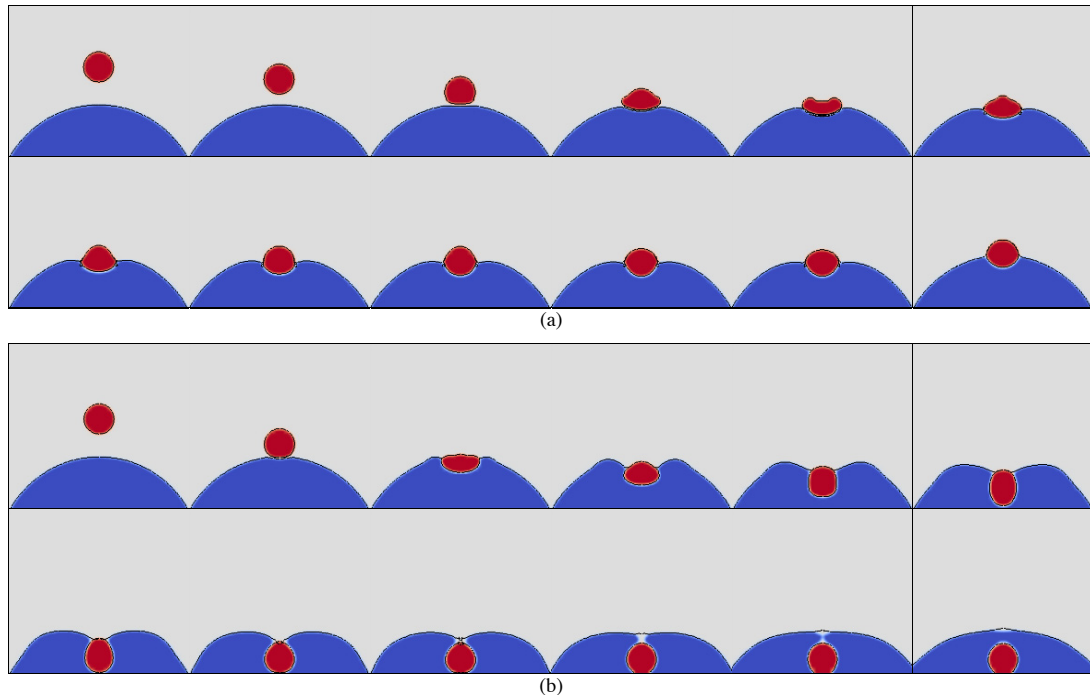


Fig. 3. Simulation snapshots of a water droplet colliding with an oil droplet resting on a plate for different oil viscosity.

(a)  $\mu_o = 26 \text{ mPa}\cdot\text{s}$ ; (b)  $\mu_o = 2 \text{ mPa}\cdot\text{s}$ .

## 6. Concluding remarks

The impact dynamics of a small water droplet into a large oil droplet are investigated experimentally and numerically. The effects of various physical and geometrical parameters are addressed to seek for the proper setup for the successful encapsulation of the water droplet. For the case considered in this paper, several mechanisms are coupled to effect the encapsulation. The gained understandings can be used to guide the design and optimization of the related applications.

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