



Self-division of a mineral oil–fatty acid droplet



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ARTICLE INFO

Article history:

Received 18 August 2015

In final form 30 September 2015

Available online 9 October 2015

ABSTRACT

Self-division of a mineral oil–fatty acid droplet placed in an alkaline solution was investigated. The initially homogeneous mineral oil droplet containing various amounts of 2-hexyldecanoic fatty acid underwent a division process resulting in the formation of two droplets. One formed (‘daughter’) droplet contains middle-phase microemulsion (surfactant-rich phase), while the other contains mineral oil with 2-hexyldecanoic acid (surfactant-low organic phase). We found that the pH of the water phase has negligible effect on the ratio of the sizes of the ‘daughter’ droplets. However, the contact angle between two droplets highly depends on the pH of the alkaline solution.

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1. Introduction

Investigation of division of micro- and macroscopic chemical objects such as micelles, vesicles and fatty acid droplets has gained much interest in the last few years due to the importance of creating a minimal artificial cell [1–12]. These chemical objects can mimic some aspects of a living cell and display ‘biological-like’ responses, i.e. they can react to the environmental stimuli. For instance they can move either randomly or directionally in a gradient of chemical substances (acid, salt, etc.), which can be considered as an artificial chemotaxis [13–24]. Moreover, some of these droplets can self-divide driven by environmental factors such as pH and chemical composition [9,10]. Probably the most ingenious idea illustrating the self-replication of microscopic compartment is DNA replication in giant vesicles followed by vesicle growth and division triggered by addition of membrane precursor to DNA-amplified giant vesicles [9].

Significance of such investigations can be highlighted by the fact that simple models of protocells can be considered as simple bilayer vesicle structures or oil microdroplets placed in a water phase. These latter mentioned structures could represent even more primitive and simple examples of life on Earth than layer protected vesicles [25].

In this letter we show a new, simple example for self-division of a mixed mineral oil–fatty acid droplet in an aqueous environment. The droplet placed in an alkaline water phase can undergo self-division due to development from a two-phase system into a thermodynamically stable three-phase system.

2. Experimental

A drop of mineral oil (MO, Sigma–Aldrich) (typically $\sim 10 \mu\text{L}$) that contains 5–90 v% of 2-hexyldecanoic acid (HDA, Sigma–Aldrich) and a small amount (ca. 5 mg mL^{-1}) of oil red dye (Sigma–Aldrich) for better visualization was placed in a Petri dish (diameter of 12 cm) filled with an alkaline solution of potassium hydroxide (KOH, Sigma–Aldrich). The depth of the alkaline solution in the Petri dish was 1 cm. The drop was placed at the liquid–air interface and the division process was monitored by a digital camera (Canon EOS 20D). All experiments were carried out at room temperature ($22 \pm 0.5^\circ\text{C}$). MO and HDA are not soluble in water, however, they are miscible in all proportions. Deprotonated form of HDA acts as a surfactant in water and has critical micelle concentration (CMC) of $3.6 \times 10^{-6} \text{ M}$ at pH 10.5 [26].

3. Results and discussion

The process of droplet self-division is shown in Figure 1 (see also Movie 1 in Supplementary Content). After placing a homogeneous droplet containing the mixture of MO and HDA (yellow color) in the alkaline solution, at the perimeter of the droplet becomes blurry due to deprotonation of the HDA at the water–oil (w/o) interface (Figure 1). Several seconds later the appearance of a new phase (red color) can be sparsely observed in the oil phase due to microemulsion formation at the w/o interface of the droplet (Figure 1a). In parallel, there is deprotonation of HDA at the w/o interface of the droplet, and due to this process HDA becomes water soluble and acts as a surfactant, enriching the liquid–air interface and producing micelles in the water phase. However, the amount of HDA in the droplet is much higher than can be dissolved in the water

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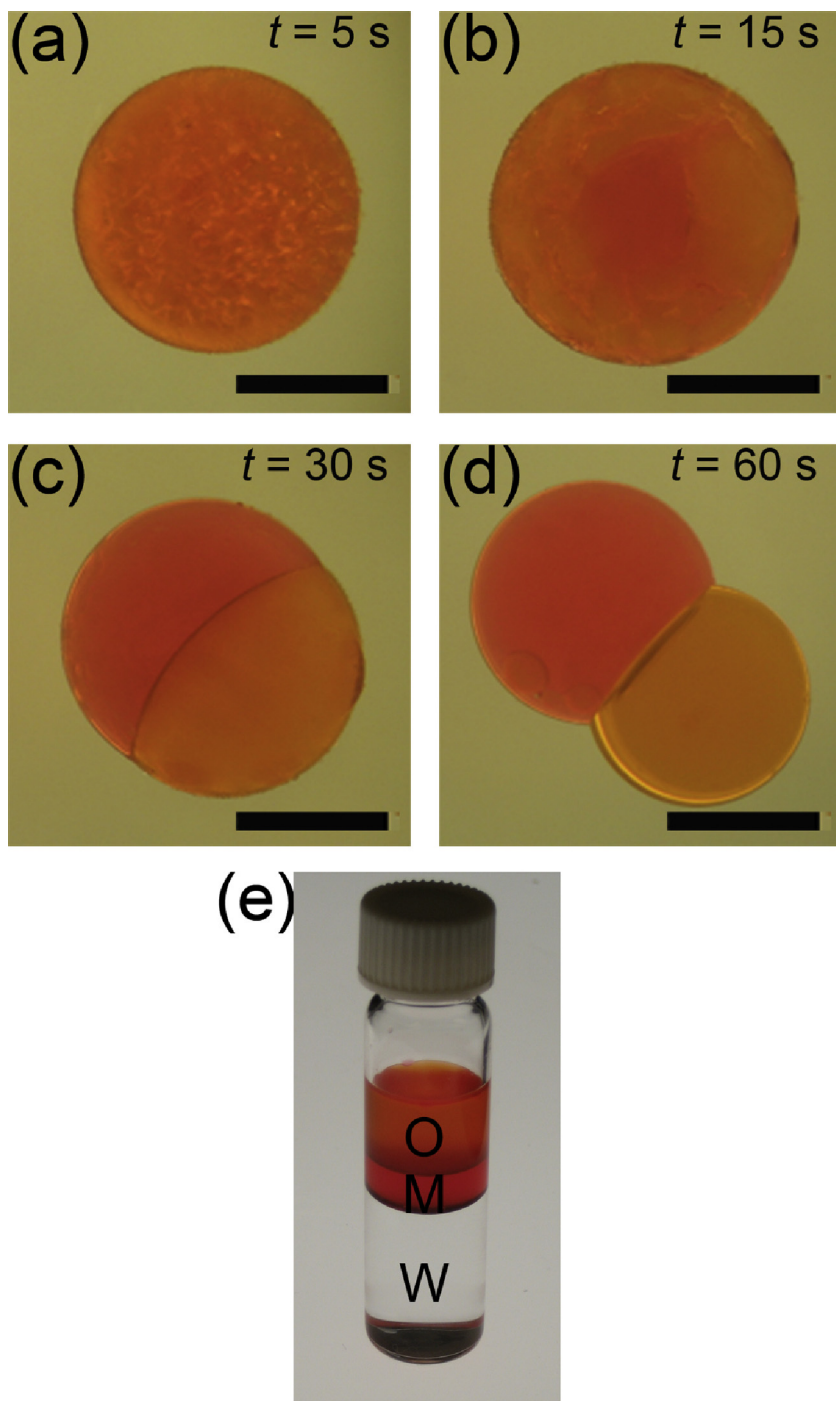


Figure 1. Self-division of a mineral oil–fatty acid droplet in an alkaline solution of KOH (2M, pH 14.3). The mineral oil droplet contains initially 30 v% 2-hexyldecanoic acid. The scale bar represents 1 mm (a–d). Thermodynamically stable three-phase system (e); W: water phase with deprotonated 2-hexyldecanoic acid, M: microemulsion (surfactant-rich middle-phase), O: mineral oil with 2-hexyldecanoic acid (surfactant-low organic phase).

(bulk) phase, and thus it produces a thermodynamically stable new phase (Winsor type III microemulsion), the so-called surfactant-rich middle-phase (M), which is insoluble both in organic (O) and water (W) phases (Figure 1e) [27–29]. In other words, the system transforms from a two-phase state into a three-phase state, in which the three phases, O, M and W, are thermodynamically stable. Middle-phase microemulsion contains both oil and water that form a bicontinuous structure. This middle-phase microemulsion has an intermediate density between the oil and water phases.

Oil red dye is yellow in the oil phase and red in the microemulsion phase, and thus the self-division process can be perfectly monitored.

As the interfacial reaction between HDA and base progresses, more small droplets (size of $\sim 5\text{--}10\ \mu\text{m}$) containing microemulsion form at the interface of MO droplet ('mother' droplet) (Figure 2). Later these small droplets can coalesce resulting in the formation of two distinct phases (O and M, Figure 1b), and finally the droplet divides into two 'daughter' droplets (Figure 1c and d).

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