

# Optimization of isothermal low-energy nanoemulsion formation: Hydrocarbon oil, non-ionic surfactant, and water systems



Jennifer Komaiko, David Julian McClements\*

*Biopolymers and Colloids Research Laboratory, Department of Food Science, University of Massachusetts, Amherst, MA 01003, USA*

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## ABSTRACT

Nanoemulsions can be fabricated using either high-energy or low-energy methods, with the latter being advantageous because of ease of implementation, lower equipment and operation costs, and higher energy efficiency. In this study, isothermal low-energy methods were used to spontaneously produce nanoemulsions using a model system consisting of oil (hexadecane), non-ionic surfactant (Brij 30) and water. Rate and order of addition of surfactant, oil and water into the final mixture were investigated to identify optimal conditions for producing small droplets. The emulsion phase inversion (EPI) and spontaneous emulsion (SE) methods were found to be the most successful, which both require the surfactant to be mixed with the oil phase prior to production. Order of addition and surfactant-to-oil ratio (SOR) influenced the particle size distribution, while addition rate and stirring speed had a minimal effect. Emulsion stability was strongly influenced by storage temperature, with droplet size increasing rapidly at higher temperatures, which was attributed to coalescence near the phase inversion temperature. Nanoemulsions with a mean particle diameter of approximately 60 nm could be produced using both EPI and SE methods at a final composition of 5% hexadecane and 1.9% Brij 30, and were relatively stable to droplet growth at temperatures <25 °C.

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## 1. Introduction

Oil-in-water (O/W) emulsions are utilized in a wide range of industries to encapsulate, protect, and/or deliver lipophilic components, e.g., pharmaceuticals, cosmetics, foods, agrochemicals, and petrochemicals. Emulsions are formed when one of two immiscible liquids is dispersed in the other liquid as small spherical droplets [1,2]. The resulting systems are thermodynamically unstable and may breakdown through a variety of instability mechanisms, including gravitational separation, coalescence, flocculation, and Ostwald ripening. Nanoemulsions are emulsions whose droplet diameter typically falls in the range of 20–200 nm [3]. Unlike microemulsions, which may have similar particle sizes, nanoemulsions are also thermodynamically unstable systems that have a tendency to breakdown over time. There has been growing interest in the formation, stabilization and utilization of nanoemulsions due to their novel physicochemical properties, high optical clarity, good stability to gravitational separation and aggregation, and ability to increase the bioavailability of encapsulated active ingredients [4–6].

Nanoemulsions can be fabricated using both high energy and low energy approaches. High energy approaches utilize specialized equipment (“homogenizers”) capable of generating intense mechanical forces that disrupt and intermingle the oil and water phases. The main variables that impact nanoemulsion characteristics using high energy methods are the energy intensity and duration, the surfactant type and concentration, and the physicochemical properties of the oil and water phases [7]. In contrast, low energy approaches rely on the spontaneous formation of emulsions based on the phase behavior of certain surfactant, oil, and water systems [8]. There is interest in using lower energy techniques in the emulsion formation process due to economic benefits [2] and increasing amounts of research have been conducted to investigate the utility of different low-energy approaches [8–11]. However, the goal of using low energy in a high product throughput industry setting has yet to be fully realized [12] with few studies investigating the effect of scaling-up from a laboratory setting [13].

Low energy approaches can be broadly categorized as either thermal or isothermal methods. Thermal methods rely on emulsion formation due to changes in surfactant properties with temperature, whereas isothermal methods rely on emulsion formation due to changes in local system composition at a fixed temperature. Spontaneous emulsification (SE) and emulsion phase

\* Corresponding author. Fax: +1 (413) 545 1262.

E-mail address: [mccllements@foodsci.umass.edu](mailto:mccllements@foodsci.umass.edu) (D.J. McClements).

inversion (EPI) methods fall into the category of isothermal methods [9,10], while the phase inversion temperature (PIT) method is an example of a thermal method [2]. In the SE method, an emulsion is formed when an oil–surfactant mixture is added to water, whereas in the EPI method, an emulsion is formed when water is added to an oil–surfactant mixture [13]. In the PIT method, an emulsion is formed when a surfactant–oil–water mixture is rapidly cooled below the phase inversion temperature (PIT) with continuous mixing [14].

One of the main objectives of the current study was to investigate the formation of nanoemulsions by low energy isothermal methods using a well-defined model system: hydrocarbon oil, non-ionic surfactant, and water. A substantial amount of research has already been carried out on optimizing emulsion formation by emulsion phase inversion [15] and spontaneous emulsification [10,16] methods, but few studies have directly compared these two approaches with each other and with other possible isothermal methods [3,8,13,17]. In principle, there are six different methods of forming nanoemulsions from surfactant (S), oil (O) and water (W) by injecting one liquid into another liquid at fixed temperature: (SO) → W; (W) → SO; (SW) → O; (O) → SW; (OW) → S; and, (S) → OW. Here, the material in parentheses is initially placed in an injector, while the other material is initially placed in a reaction vessel. In this study, we investigated all six possible methods of forming nanoemulsions using this approach. Previous studies have compared two or three of these methods. For example, Forgiarini reported that nanoemulsions could be formed using the (W) → SO method but not with the (O) → SW method [18]. Studies using similar surfactants, oils, and water phases have reported differences in the size of the droplets produced by the (W) → SO method (EPI) and the (SO) → W method (SE) [9,10]. In addition to examining order of addition effects, we also investigated the influence of surfactant-to-oil ratio, addition rate, stirring speed, and storage temperature on the formation and stability of emulsions formed by isothermal low energy methods.

## 2. Materials and methods

### 2.1. Materials

Hexadecane (Sigma–Aldrich, St. Louis, MO) was used as the hydrocarbon oil phase. Polyoxyethylene (4) lauryl ether (Brij 30)

(Acros, Thermo Fisher Scientific, NJ) was used as the non-ionic surfactant. Distilled and deionized water was used as the aqueous phase to prepare all solutions and emulsions (Milli-Q®). For convenience, we use the symbols S to refer to surfactant, O to refer to oil, and W to refer to water in the remainder of the manuscript.

### 2.2. Emulsion preparation

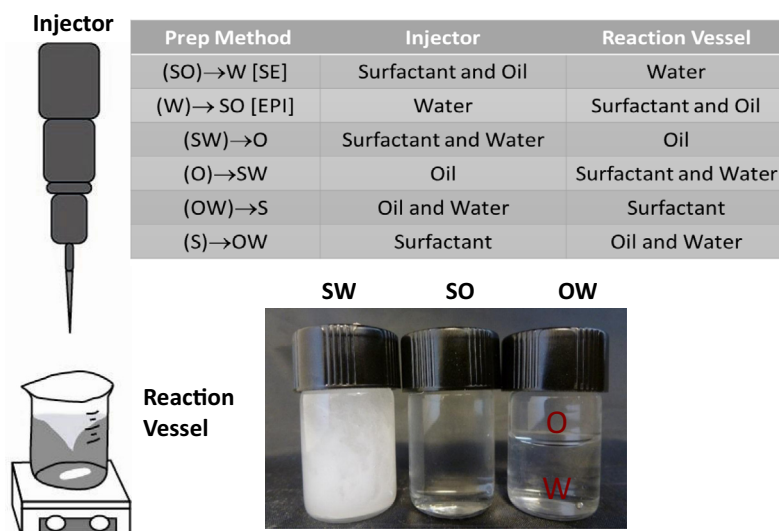
#### 2.2.1. Influence of order of addition

Emulsions were prepared by simple addition of 1 or 2 components (S, O and/or W) from an automated electronic pipette (Rainin SE4, Mettler Toledo, Oakland, CA) into a beaker containing 1 or 2 components (S, O, and/or W) and stirring at 700 rotations per minute (RPM) using a magnetic stir bar at room temperature (~20 °C). All combinations of water, oil and surfactant were tested for a total of 6 methods: (SO) → W; (W) → SO; (SW) → O; (O) → SW; (OW) → S; and, (S) → OW (Fig. 1). The material in parentheses was initially in the pipette (injector), while the other material was initially in the beaker (reaction vessel). The titration was done over 20 min and the sample was allowed to stir for an additional 5 min for a total mixing time of 25 min. Method (SO) → W is also known as Spontaneous Emulsification (SE), while Method (W) → SO is also known as Emulsion Phase Inversion (EPI). Prior to emulsion production, initial phases containing two components were mixed for a minimum of 30 min at 500 RPM. These experiments were carried out at a fixed surfactant-to-oil ratio (SOR) of 0.375. These preliminary experiments indicated that only the SE and EPI methods were able to produce very fine droplets, and so only these two methods were used in later studies.

The aliquot volume, interval time, and dispense speed of the electronic pipette used to titrate the systems were controlled. All pipetted aliquots were divided into 100 increments and the interval time was varied to attain the desired addition time. To obtain the same final SOR, the aliquot and total volumes had to be adjusted for each system.

#### 2.2.2. Influence of surfactant-to-oil ratio

The influence of surfactant concentration was investigated by varying the surfactant-to-oil ratio (SOR). The total oil content in the final systems was held constant at 5%, while the SOR was varied by altering the amounts of surfactant and water content in the final system:



**Fig. 1.** Visual representation of the order of addition screening study. Table shows the six different preparation methods tested. The photographs show vials containing various two component mixtures: surfactant and water formed a gel; surfactant and oil formed a homogenous solution; and, oil and water were immiscible (oil phase on top of water phase).

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