Experimental Thermal and Fluid Science 81 (2017) 265–276

Contents lists available at ScienceDirect

Experimental Thermal and Fluid Science

journal homepage: www.elsevier.com/locate/etfs

# Influence of DC electric field upon the production of oil-in-water-in-oil double emulsions in upwards mm-scale channels at low electric field strength

Federico Alberini [a,*], Davide Dapelo [b], Romain Enjalbert [a], Yann Van Crombrugge [a], Mark J.H. Simmons [a]

[a] School of Chemical Engineering, University of Birmingham, B15 2TT, UK
[b] School of Civil Engineering, University of Birmingham, B15 2TT, UK

ARTICLE INFO

Article history:
Received 31 March 2016
Received in revised form 18 October 2016
Accepted 20 October 2016
Available online 21 October 2016

Keywords:
Double emulsion
High speed imaging
Image analysis
Drop diameter
Flow regime
Electric field

ABSTRACT

A novel approach to create O/W/O is developed and described to achieve uniform oil drop size coated with thin layers of water. Drops were created using a test cell where the DC field is applied between different internal diameter (ID) needles (from which the O/W emulsion emits upwards into a continuous oil phase) and a grounded metal ring which was located at selected distances from the needle top. The advantages compared to the previous techniques consist of possibility of control on drop size and coating layer of the water using low electric field. A high speed imaging technique has been applied to determine drop size under different flow and electric field conditions. Without the electric field, several flow regimes were observed; stable formation of both the O/W/O emulsion and the O/W emulsion upstream of the cell was possible over a range of Reynolds numbers from 80 to 100. The effect of the electric field was found to be reverse below electric field strength of 60 kV $m^{-1}$, beyond this critical value there was significant impact upon the flow regime, drop size and emulsion structure. The impact of the electric field strength upon flow pattern and emulsion structure and a quantitative analysis of droplet size are presented. The work shows the results for the controlled creation of complex emulsion droplets combining electric field and mm scale channels. The differences with the other physical processes reported in the literature are discussed.



## 1. Introduction

The demand for new complex multiphase liquid products, particularly in the food, pharmaceutical and cosmetic industries, have driven the need for a deep understanding of how emulsions are formed and stabilised [1,2]. A double emulsion is comprised of droplets dispersed in a continuous phase to form a primary emulsion, which is then dispersed within an outer continuous phase forming the secondary emulsion [3]. Industrial applications of double emulsions are diverse, including low fat and low salt food products which maintain the texture and mouth feel of conventional high fat and high salt products [4,5] or controlled drug release mechanisms [6]. Whilst research on single emulsions has been largely developed over the last few decades [7]; recent efforts have focussed on double emulsions so that their potential use in multiphase products can be fully exploited.

Double emulsions can be produced using several different methods. In most cases, multiple emulsions are prepared according to a two-step process [8]; each step comprises the addition of a dispersed phase and its corresponding emulsifying agent (usually a surface active molecule, or particle). Each step may be carried out using processes including, for example, hollow fibre membranes or homogenisers [9]. However, these methods can be difficult to control and possible destabilization pathways include rupturing of the primary emulsion droplets in the second re-emulsification step [10]. This destabilization pathway may be avoided if one-step processes such as micro-fluidics are used [8], which exploit tightly controlled laminar flow conditions [11,12]. Despite their obvious potential, scale-up and fouling (blocking) of the micro channels remain obstacles.

An alternative method is the electro spraying technique which has been investigated extensively over the two centuries [13–19]. Bose and Nollet were the first to investigate the effect of an applied electric field on the formation of liquid droplets from a jet, illustrating a reduction of drop size as a function of the applied field strength. This has been noted in many works since [20–22].

* Corresponding author.
E-mail address: f.alberini@bham.ac.uk (F. Alberini).

http://dx.doi.org/10.1016/j.expthermflusci.2016.10.023
0894-1777/

Normally, electro spraying involves high values of electric field strength usually above $10^6$ V m$^{-1}$ [24,25] and the key phenomenon is the induced charge at the interface between an electrically conducting liquid and the continuous phase. Beyond a certain critical level, the interface becomes unstable and evolves from a generally rounded shape to another including one or several remarkably stable conical features called Taylor cones [17].

Another phenomenon, which can occur when an electric field is applied, is the electro-hydrodynamic flow [23,26,27]. When a neutral leaky dielectric drop is immersed in a continuous conductive phase, flow motion is generated by action of the applied electric field on the continuous phase. Taylor [28] introduced the leaky dielectric model to describe the electro-hydrodynamic deformation of a drop. The theory applies to weak electric fields, where drop deformation is small [29].

In the last few decades, numerical solutions of the governing equations have been made possible using computational methods and these have been used primarily to quantify the dynamics of droplets exposed to electric fields but mostly in single emulsions [23,30,31].

Most recently, analytical work and modelling techniques (finite element) have been used to explore how the dynamics of the creation of double emulsions may be altered by the presence of an electric field [32,33]. Some limited work has been carried out on how double emulsion drops may be made highly concentric by use of an applied AC field [34]. However, in the open literature there still remains a dearth of validated computational and experimental studies on the formation of double emulsions which combine both electric fields and micro or macro channels.

The work presented in this paper is concerned with the experimental investigation of the continuous formation of oil-in-water-in-oil (O/W/O) double emulsions under the effect of a DC electric field. The upwards needle set up allows the formation of stable primary emulsion which was not possible to achieve using the most extensively used downwards set up. The effects of flow rate and low electric field strength ($\sim 10^5$ V m$^{-1}$) upon the flow patterns and types of double emulsion drops are observed and used to catalogue the different flow regimes with reference to the stability and repeatability of double emulsion formation. The observed results have been used to describe the existence of this phenomenon which differentiates from the better known electro-spraying, electro-dripping and electro-hydrodynamic flows. The production of double emulsion using low electric field presents advantages compared with others techniques previously described in the literature. A tighter control of secondary emulsion drop size towards smaller or larger scale can be achieved applying a low electric field. In fact, before a critical value of the electric field is achieved, the water drops tend to increase in size. After the critical value the drops decreases towards a minimum where a thin water coating layer is formed on top of oil drops. If compared with the hollow fibre membrane and homogenisers, this technique has lower potential costs of manufacture and better process control. Finally, compared to micro-fluidic devices, this technique is a valid alternative for the production of double emulsions and it is not much affected by the fouling issues.

## 2. Materials and methods

### 2.1. Fluids and fluid properties

The specification and suppliers of all fluids and chemicals used in the experiments are given in Table 1, together with their bulk properties. For all the experiments, Lytol, a low viscosity transparent mineral oil, was used as the organic phase and distilled water was used as the aqueous phase. The relative permittivity and absolute permittivity of the oil are presented in Table 1. The conductivities of the fluids were measured using a RS CD-4303 conductivity meter to an accuracy of ±0.01 μS cm$^{-1}$. The values obtained for the Lytol oil, pure distilled water and distilled water with SLES at a concentration of 1.4 times the critical micelle concentration (CMC) are reported in Table 1.

**Table 1**
Relevant physical properties of the materials used in the system.

| Material | Lytol | Distilled water | Water + SLES |
|---|---|---|---|
| Density at 20 °C (kg m$^{-3}$) | 800 | 1000 | 1000 |
| Dynamic viscosity (Pa s) | 0.0051 | 0.001 | 0.001 |
| Relative permittivity | 2.1 | ~80 | N/A |
| Absolute permittivity ε (F m$^{-1}$) | $1.89 \cdot 10^{-11}$ | N/A | N/A |
| Conductivity (μS cm$^{-1}$) | 0 ± 0.01 | 0.5 ± 0.01 | 21.64 ± 0.01 |

| Chemicals | Product specification | Supplier |
|---|---|---|
| Lytol oil | Lytol oil | Lytol |
| Distilled water | Distilled water | University of Birmingham |
| SLES | Texapon N701 | BASF |
| Nigrosin | Alcohol soluble | Sigma-Aldrich |
| Span 80 | Span 80 | Sigma-Aldrich |

The organic soluble and aqueous soluble surfactants used were commercial grades of sorbitane monooleate (Span 80) and sodium lauryl ether sulphate (SLES) respectively. Alcohol soluble Nigrosin was used as black dye in the inner oil phase to distinguish clearly between the two different dispersed phases in the images.

The surface and interfacial tensions of the fluids were measured using the Wilhelmy plate method using (Kruss K100 tensiometer) as shown in Table 2. These were first measured as pure fluids and then as fluids with the appropriate concentrations of surfactant used in the experiments. The critical concentration of Span 80 in the oil phase to enable stable formation of the primary O/W emulsion was found to be 0.5 wt%. The CMC of SLES was calculated and a value of 0.2 mol m$^{-3}$ was obtained which corresponds to the literature value [35]. For the experiments, 1.4 times the critical micelle concentration (CMC) of SLES was used to reach a stable primary emulsion. In order to verify the influence of Nigrosin dye on the interfacial tension of the oil phase, measurements with and without the dye were performed. No difference was observed.

### 2.2. Experimental rig

A schematic of the experimental rig is given in Fig. 1a. The specifications of the equipment used are given in Table 3, including the dimensions of the needle electrode. The electric field was generated using a Glassman DC power supply. The applied voltage ranged from 2 kV to 20 kV in steps of 2 kV. The DC cable from the supply was attached to the needle electrode at the bottom of the rig and the ring electrode at the top of the continuous oil chamber was grounded. The entire rig was inserted into a protective Perspex chamber which was interlocked to the power supply to prevent accidental exposure of the operator to the electric field.

With reference to Fig. 1a, the oil phase (dyed with Nigrosin) was pumped into the system using a syringe pump (I) and a gastight glass syringe. The water phase was injected using a syringe pump (L) equipped with a Monoject 140 mL plastic syringe. Due to the buoyancy forces caused by the density differences between the oil and water, the rig was oriented so that the fluids flow vertically upwards. This is a known limitation for wide channels and it can limit the repeatability of the process if the needle is oriented vertically downwards or horizontally.

The single O/W emulsion was produced within a T-piece (A) (ID = $3 \times 10^{-3}$ m) schematised in Fig. 1b. Oil and water were injected separately through 0.37 mm ID needles through each