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Regular Article

Breaking of double emulsions based on electrohydrodynamics principles

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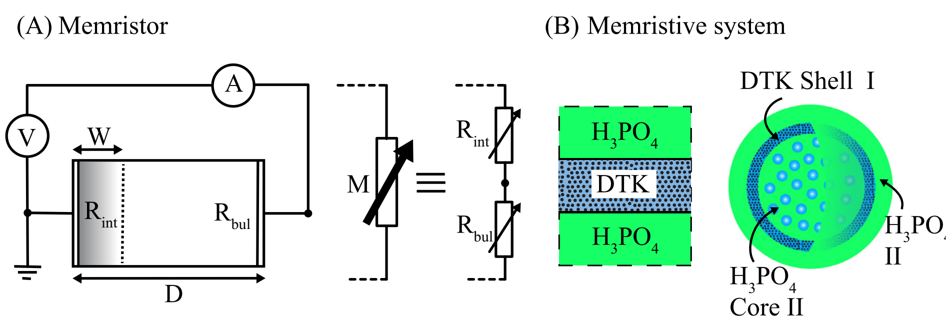
HIGHLIGHTS

- Model of the breaking of double emulsion based on electrohydrodynamics principles.
- Qualitative and quantitative physical picture of the electrocoalescence process.
- Marangoni instabilities of the first and second order possible electrical analogues.
- The electrocoalescence process, in some extent, is elucidated.

GRAPHICAL ABSTRACT



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ABSTRACT

This research focuses on the modeling of the liquid-liquid dispersed system, including particular insight on the electrocoalescence (EC) process that occurs during the breaking of double emulsions. The representative system, used in this work, was taken from the pilot plant for solvent extraction of uranium from wet phosphoric acid. The chosen framework required for elucidation of the EC process is based on the electrohydrodynamic (EHD) principles.

During the model development it was necessary to consider several theoretical concepts for easier understanding and description of the related events. The first is the concept of entities, and corresponding classification of finely dispersed systems. The second concept is an introduction of almost forgotten basic electrodynamics element the memdiode or memristor as a current controlled device, and corresponding memristive systems.

Hence, the conclusions that may be withdrawn from the presented results and findings could enable easier designing of the solutions for a breaking of double emulsions problems, that is, the entrainment problems that may arise in some pilot or industrial plants. Finally, the perspectives and the remaining challenges, considering the here discussed concepts and model based on the EHD principles, are mentioned.

1. Introduction

The evolution of droplet size in liquid-liquid dispersed systems is controlled by break-up, mass transfer, coalescence and electrocoalescence (EC). This research focuses on the modeling of the liquid-liquid dispersed system including the particular insight in the EC process. The EC occurs during the breaking of double emulsions, or the breaking of droplet-film structures submerged into the droplet homophase. One representative experimental example taken for this research is the breaking of double

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emulsions that appear in the solvent extraction of uranium from wet phosphoric acid [1,2].

The double emulsion is formed during the preparation of the liquid-liquid dispersion in the mixer-settlers as an entrained, stable, liquid film around the immiscible liquid droplet. Breaking of these double emulsions or droplet-film structures is important, both from the technical and the economic reasons. Hence, for the solution of the particular entrainment problem the light phase film loop was designed and constructed, and the particular equipment of interest related to this research is the lamellar coalescer [1,2].

The main objective of this research is to provide an approach to understand the EC process, and therefore to enable designing of adequate and relevant methods, and equipment for the solution of the entrainment problems, in particular for the breaking of double emulsions.

The chosen framework required for elucidation of the EC process is based on the electrohydrodynamic (EHD) principles. It was necessary to consider several theoretical concepts for easier understanding and description of the related events. The first is the concept of entities, and the corresponding classification of finely dispersed systems [1–6] and Supporting Information A; this concept permit consideration of the electron transfer phenomenon beside the heat, mass, and momentum transfer phenomena commonly used in chemical engineering. The second concept is an introduction of almost forgotten basic electrodynamics element the memdiode or the memristor (M) as a current controlled device, and the corresponding memristive systems [2,4–9] and Supporting Information B. This concept is needed because the fundamental electrodynamics elements, the resistor (R), the inductor (L), and the capacitor (C) are not sufficient for a satisfactory explanation of the EC process.

Further on, to make a qualitative physical picture of the events comprising the EC process Marangoni instabilities of the first and second order were consulted and their electrical analogues were postulated [1].

For the quantitative physical picture a model of the droplet-film structure submerged into the droplet homophase, considered as the particular memristive system, was developed. This model was used as a fundamental finding in the discussion, and elucidation of the EC process.

This research is supposed to help and to improve previous results obtained during the development, construction and running of the pilot plant for uranium extraction from wet phosphoric acid [1,2,10–23]. The entrainment problems, arise during the running of this pilot plant, in some extent were clarified. Also, the conclusions that may be withdrawn from the presented results and findings could enable easier designing of the solutions for similar entrainment problems that may arise in some pilot or industrial plants.

Finally, the perspectives and the remaining challenges, considering the here discussed concepts and model based on the EHD principles could facilitate or help the understanding of, for example: performances and mechanisms of some chemical and biological sensors, development of some electro-analytical methods, some events and phenomena in biology and/or biomedicine, in particular the hematology (a non-Newtonian fluid flow), the genetics (a DNA models, the electron transfer phenomenon) and the electroneurophysiology (the quantum spin transport).

2. Materials and methods

2.1. Generation of the physical-chemical system

The particular liquid-liquid dispersed system which has been used to corroborate the validity of theoretical predictions was the heavy-phase droplet-light-phase film structure immersed in the heavy-phase continuum or double emulsion. This system was the heavy-phase output from the "pump-mix" mixer-settler battery together with its entrained light phase. The battery was the part of the pilot plant for extraction of uranium from wet phosphoric acid by the D2EHPA-TOPO process [1,2,4–6,10–20]. The heavy liquid was 5.6 M phosphoric acid, and the light liquid was the synergistic mixture of 0.5 M di(2-ethylhexyl) phosphoric acid and 0.125 M tri-*n*-octylphosphine oxide in dearomatized kerosene (DTK).

A polydispersion was generated and the primary separation performed in a laboratory mixer for the batch studies under the conditions applied in the pilot plant [1,2]. The selected hydrodynamic characteristics in the mixer unit were as follows:

- The ratio of the phases in the mixer, light/heavy was equal to 1.1.
- The number of revolutions of the eight-blade double-shrouded impeller in the mixer was equal to 15 s^{-1}.
- The mixing criterion, $\rho n^3 D^2$, was equal to 13.

Thereafter, the sample of the heavy phase, together with the entrained light phase, was isolated and observed *in situ* with an optical microscope. Fig. 1A shows *in situ* photograph of the examined liquid/liquid droplet-film structure immersed in the droplet homophase continuum [1,2]. This isolated system is in hydrodynamic and electrodynamics equilibrium, i.e., the rigid sphere can be observed.

Further on, Fig. 1B shows *in situ* photograph of the system examined at the junction point of the droplet-film structure and

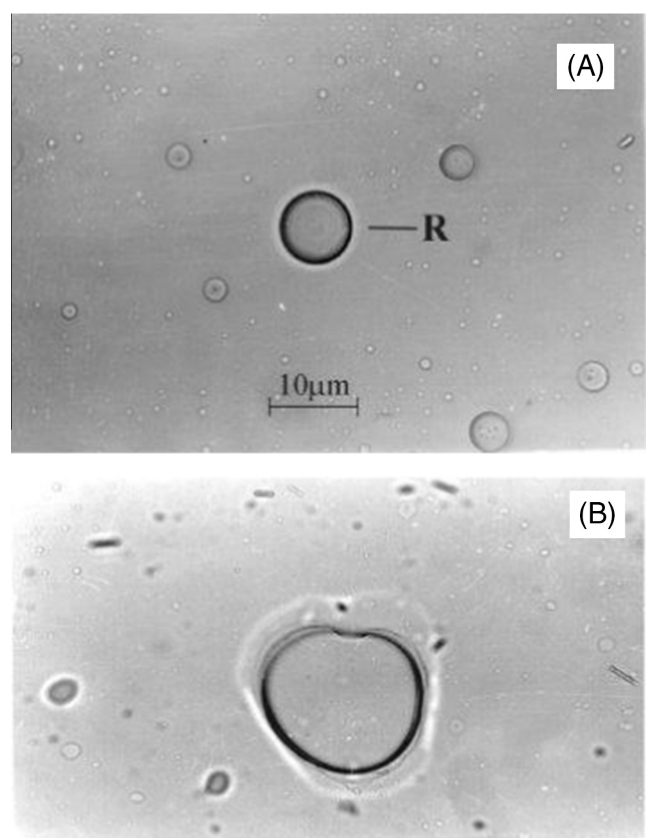

Fig. 1. (A) Photograph *in situ* of the examined liquid/liquid droplet-film structure submerged into the droplet homophase continuum; R – rigid sphere, (B) photograph *in situ* of the examined system at the droplet-film structure-plate junction point; E – elastic sphere.