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ACCEPTED MANUSCRIPT

Dielectric behavior of oil-water emulsions during phase separation probed by electrical impedance spectroscopy

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Abstract

Electrical impedance spectroscopy (EIS) is applied to studying the stability and phase behavior of oil-water emulsions. The method involves EIS on 500-micron-thick samples of emulsions in a parallel-plate capacitor test cell. The frequency response data is fitted to an equivalent circuit model to estimate the electrical impedance of the samples. The technique is used for investigating the capacitance and resistance of emulsions during phase separation. A theoretical explanation based on the capacitance of the emulsion layer is provided to understand the reason behind the variation in the dielectric constant of the samples during phase separation. It is observed that creaming and sedimentation can be captured as a permittivity decay, so permittivity measurements can be utilized to sense phase separation. The technique is shown to be more effective for oil-in-water emulsions rather than water-in-oil ones.

Keywords: phase separation monitoring, liquid system sensing, emulsion, electrical impedance spectroscopy, equivalent circuit, milli-fluidic

1. Introduction

Characterization of emulsions and sensing emulsion stability are crucial for a variety of industries ranging from cosmetics[1] and pharmaceutical[2], to the petroleum industry[3, 4]. The structure of the dispersed phase affects electrical properties of emulsions[5, 6, 7, 8, 9]. Therefore, variations in electrical properties of emulsions in different frequency ranges can be used for detecting changes in the structure and arrangement of the dispersed phase. This makes electrical (electrochemical) impedance spectroscopy (EIS) a sensing technique and a powerful experimental tool for investigating emulsion physics as well as characterization and stability sensing of emulsions[10, 11, 9].

Dielectric spectroscopy has been applied to investigation of the structure of emulsions. Sjoblom et. al. [5] investigated the effect of flocculation on dielectric properties in water-in-oil emulsions by means of shear-driven droplet floc disintegration. They also studied the effect of water fraction and salt concentration on static and high-frequency permittivity of water-in-oil (W/O) emulsions. The effect of temperature and high DC electric fields on percolation of W/O microemulsions has also been studied [6]. More recently, measurement of dielectric properties has been applied to waterin-crude-oil emulsions[12, 4] and multiple emulsion production [3]. Theoretical explanation of dielectric properties of heterogenous media has been attempted by researchers in different areas such as colloidal science, solid electronics

¹⁵ and material science[13, 11, 14, 15]. The initial models are based on Maxwell-Garnett's[16] effective medium theory for homogenous distribution of the dispersed phase and are valid for low concentrations (below 2%). Bruggeman[17], Hanai[18], and Boyle[8] proposed models for higher concentrations of the dispersed phase in homogenous emulsions. Their studies show that coagulation effects may cause drastic deviations from the homogeneity assumption. Skodvin

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