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Breakup modes and criterion of droplet with surfactant under direct current electric field

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ABSTRACT

The breakup behaviors of aqueous droplets with surfactant in oil under direct current (DC) electric field are investigated in this paper. Four different droplet breakup modes were observed, namely conical shape-conical jetting, ellipsoidal shape-conical jetting, ellipsoidal shape-filamentous breakup and cylindrical shape-filamentous breakup. The conversion of breakup modes was discussed with the electric capillary number (*Ca*)-surfactant concentration (C_{SDBS}) phase diagram and the droplet elongation process. The mechanism validated here is that the lobes appear in all breakup modes, and the liquid volume at the both ends of droplet differentiate the breakup modes under different conditions. Different from previous studies, the criterion of droplet breakup modes was proposed based on the interfacial dynamics instead of the bulk phase properties. By analogy with stress–strain relationship, it is found that the electrostatic pressure shows a linear relationship with droplet specific surface area during the droplet elongation process, and the slope *K* could be employed to identify different breakup modes. These findings are of great significance to the promotion and application of electric-induced droplet breakup.

1. Introduction

The droplet breakup induced by electric field has been widely applied in medicine (Fedosov et al., 2011), mass spectrometry (Monge et al., 2013; Seifert et al., 2015), electro spraying (Zhang and Wang, 2013; Bhushani and Anandharamakrishnan, 2014), and droplet manipulation (de Ruiter et al., 2014; Chokkalingam et al., 2014). However, the droplet breakup in electric field, which is defined as the electrodispersion, has a negative effect on oil production because it significantly reduces the water-in-crude oil emulsion separation efficiency. The crude oil products contain several natural interfacial agents, notably wax crystallites, asphaltenes and resins, or artificial ones, such as sodium dodecyl benzene sulfonate (SDBS). When these interfacially active compounds adsorb on the oil–water interface, they considerably change rheological properties of the oil–water interface, as a consequence the breakup behaviors of droplet are obviously changed under electric field (Eow et al., 2001a,b). Therefore, it is important to figure out the breakup criterion of droplet with surfactant under electric field.

Previous researches on the droplet breakup under electric field mainly focus on the critical conditions and the breakup modes from the aspect of bulk phase properties. There are three research hotpots in terms of the critical conditions: the cone angle of droplet, the critical deformation degree and the critical electric field intensity. According to Taylor (1966), the critical static semi-angle for a conductive droplet under direct current (DC) electric field is about 49.3°, which is known as Taylor cone. It is found that when the ratio of permittivity is above 17.6, the corresponding static semi-angle decreases to about 30° (Stone et al., 1999). However, the semi-angle of dynamic cones for droplet breakup under alternative current (AC) electric field is lower and about $19^\circ \pm 1^\circ$ according to Ye (2010) and Luo et al. (2015). The viscous stress should be responsible for the difference between the dynamic cones and the static cones (Karyappa et al., 2014).

The critical electric field intensity above which the droplet breakup occurs is another competing research area. Based on the experiments (Nishiwaki et al., 1988), numerical simulations (Wright et al., 1995; Basaran and Scriven, 1989) and theoretical analysis (Taylor, 1988), the

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Table 1 – The physical properties of two phases used in the experiments.

Phase	Density/kg m^{-3}	Viscosity/mPa s	Relative permittivity	Conductivity/S m^{-1}
Silicone oil	963	980	2.26	10^{-12}
Water	996	0.89	82	4.3×10^{-4}

critical electric field intensity was obtained for different oil–water emulsions. Besides the electric field parameters, the droplet breakup also depends on the interfacial tension and the original size of droplet (Eow et al., 2001a,b; Eow et al., 2002). The electric capillary number Ca, which represents the ratio of electric stress ($\varepsilon_0\varepsilon_c E_0{}^2$) and capillary stress ($\gamma/r$), is thus a crucial criterion for predicting the probability of droplet breakup under electric field (Allan and Mason, 1962):

$$Ca = \frac{\varepsilon_0\varepsilon_c r E_0{}^2}{\gamma} \tag{1}$$

where ε_0, ε_c, r, E_0 and γ are the vacuum permittivity, the relative permittivity of continuous phase, the droplet radius, the uniform electric field intensity and the interfacial tension, respectively. The critical electric capillary number Ca_c above which the droplet breakup occurs is clarified to be 0.2 ± 0.02 (Taylor, 1964; Basaran and Scriven, 1989; Varshney et al., 2012; Ha and Yang, 2000; Moriya et al., 1986; Sherwood, 1988; Dubash and Mestel, 2007a,b; Ptasinski and Kerkhof, 1922), and the corresponding critical aspect ratio is in the range of 1.5–2.3 (Ye, 2010; Basaran and Scriven, 1989; Eow et al., 2001a,b; Eow et al., 2002; Dubash and Mestel, 2007a,b).

The breakup modes have been qualitatively described and classified by a few researchers. According to the experimental observations, Eow et al. (2002) found two breakup modes of droplet in DC electric field: (1) filaments ejected from both ends and (2) filament ejected from one end and Taylor cone formed at the other one. Vizika and Saville (1992) and Moriya et al. (1986) observed another two breakup modes: (1) droplet breakup with two lobe ends and (2) droplet breakup with two Taylor cones. Ha and Yang (2000) demonstrated that the breakup modes of non-Newtonian droplet are associated with the viscosity ratio of the dispersed phase to the continuous phase λ and the viscoelastic properties of fluid. They observed that the pinch-off mode gradually turned to tip streaming mode with increasing λ. Three axisymmetric shapes prior to breakup were experimentally and numerically analyzed by Karyappa et al. (2014), and the corresponding range of Ca and λ for each breakup mode was presented. Droplet breakup mode also depends on the ratios of conductivity and permittivity. Sherwood (1988) indicated that when the conductivity of droplet is greater than that of continuous phase, droplets would breakup with lobe ends, but when the permittivity of droplet is greater than that of continuous phase, droplets would breakup with pointed ends. For the second mode, when the electric field intensity exceeds the critical value, the cones become unstable and the liquid filaments eject from both ends (Petrin, 2007). Moreover, the length of filament is proved to be associated with the electric field intensity (Garzon et al., 2014). In AC electric field, Torza et al. (1971) reported three breakup modes relating to the electric field frequency, the increase rate of electric field intensity, the electrical properties of the system and the drop radius. Nishiwaki et al. (1988) observed four types of droplet breakup behaviors and they clarified the importance of viscosity ratio to the droplet breakup under AC electric field.

In summary, the previous studies mainly focused on the relationship between the droplet breakup and the bulk phase properties, such as the ratios of viscosity, conductivity and permittivity, while the effects of the interfacial properties on the droplet breakup have not received sufficient attention. Although the breakup modes of droplet in electric field have been described and classified in detail, the conversion for different breakup modes of droplet is still undetermined.

Motivated by the above descriptions, the breakup behaviors of droplets with surfactant subjected to DC electric field are investigated with the aid of high-speed photography. Firstly, the breakup modes observed in our experiments are summarized, then the conversion of different modes is discussed. At last, based on the stress-strain analysis, the criterion of different droplet breakup modes is explored.

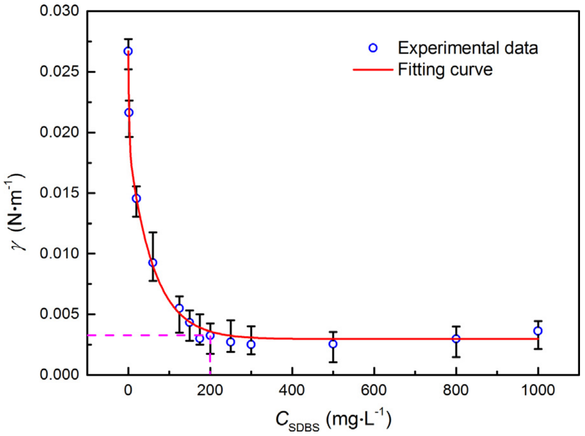

Fig. 1 – The relationship between oil–water interfacial tension γ and concentrations of SDBS C_{SDBS}.

2. Experiment

2.1. Materials

In this paper, the dispersed phase was prepared by adding sodium dodecylbenzene sulfonate (SDBS, Aladdin) into the distilled water, and the silicone oil (Aladdin) was the continuous phase. The physical properties of two phases used in the experiments are listed in Table 1. The liquid viscosity was measured by a stress-controlled rheometer (Anton Paar MCR 301). The conductivity was determined by conductivity meters, INESA DDS-11A for dispersed phase and CEARI YX1154B for continuous phase.

Since the viscosity of the continuous phase is higher, it is inaccurate to measure the interfacial tension using conventional techniques, such as du Noüy ring method or Wilhelmy plate method. According to Karyappa et al. (2014), the interfacial tension can be precisely and conveniently determined by the linear relationship between the droplet deformation degree and Ca when the droplet deformation is small (1966). Thus this strategy was applied to measure the oil–water interfacial tension for different surfactant concentration C_{SDBS}. For each C_{SDBS}, three repeated experiments were conducted. The oil–water interfacial tension is plotted in Fig. 1.

2.2. Experimental setup

The experimental setup was composed of a high-speed video camera, a waveform generator, a high-voltage power amplifier, a transparent test cell and a LED light source, as displayed in Fig. 2. The droplet breakup was recorded by the high-speed video camera (NAC Hotshot 1280) equipped with a 100× lens (Mitutoyo 5× objective with a 20× tube made by Pomeas). The camera was utilized with a frame rate of 1000 frames per second and the resolution was 1280 × 512 pixels. The pixel length was 2.8 μm.

The test cell was made of Perspex and had two stainless steel electrodes with the dimensions of 40 mm × 30 mm (about 1 mm in thickness), as shown in Fig. 3. The high voltage electrode was connected to the high-voltage power ampli-