DROPLET SIZE CHARACTERISTICS AND ENERGY INPUT REQUIREMENTS
OF EMULSIONS FORMED USING HIGH-INTENSITY-PULSED ELECTRIC FIELDS*

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DISCLAIMER

Droplet Size Characteristics and Energy Input Requirements
of Emulsions Formed Using High-Intensity-Pulsed Electric Fields

ABSTRACT

Experimental methods have been developed to measure droplet size

characteristics and energy inputs associated with the rupture of aqueous

droplets by high-intensity-pulsed electric fields. The combination of

in situ microscope optics and high-speed video cameras allows reliable

observation of liquid droplets down to 0.5 μm in size. Videotapes of

electric-field-created emulsions reveal that average droplet sizes of less

than 5 μm are easily obtained in such systems. Analysis of the energy

inputs into the fluids indicates that the electric field method requires

less than 1% of the energy required from mechanical agitation to create

comparable droplet sizes.

INTRODUCTION

In general, separation processes are based upon exploiting differences

in the physicochemical properties of the species to be separated. Although

the driving force for separation is based upon these differences, in most

cases one must still provide a significant energy input into the system

to force mass transfer to occur at an acceptable rate. This energy input

can take the form of pumping, heating, cooling, mixing, or other similar

operations. When attempting to improve the performance of a given

separation technique, one could either change or intensify the physical

properties driving force or refine the method of energy input to manipulate

the system.

Solvent extraction separation systems are one of the basic unit operations employed in the chemical process industries. The driving force for separation is the preference of certain chemical species to interact with one liquid phase over another; hence, separation is accomplished through selective partitioning into immiscible liquid phases. To obtain satisfactory mass transfer rates, some form of energy input is required to create interfacial surface area and promote interfacial and intrafacial convection.

Two major problems which limit the use of solvent extraction in industrial applications are the efficient creation and control of interfacial mass transfer surface area. In practice, interfacial area is usually created by a form of mechanical agitation. This general approach may take on such forms as extrusion through sieve plates, use of impellers in baffled tanks, or forced countercurrent flow through packing. As varied as these operations may appear, they all share a requirement for an energy input into the bulk of each of the liquids to create a dispersed phase with a reasonable amount of surface area. This represents a possibly inefficient use of energy because the continuous phase must be manipulated to create the desired effect on the dispersed phase.

When attempting to maximize surface area production in an apparatus, many mechanical agitation techniques tend to form polydisperse emulsions which are difficult to characterize and control in mass transfer operations. A method needs to be found that will not only efficiently create large amounts of interfacial surface area for mass transport, but will also allow for adequate control of phase disengagement once the mass transfer operations have been completed.

Vast amounts of interfacial surface area can be created via electric field-induced droplet rupture. In order to develop concepts for solvent extraction systems driven by electric fields, one must first address some important issues concerning the properties of emulsions formed by the droplet rupture method. In this paper the average droplet size, droplet size distribution, and energy requirements are characterized for these types of emulsions.

BACKGROUND

Placing a spherical, conducting droplet surrounded by a nonconducting continuum in an electric field will cause stresses to develop on the droplet. As the strength of the field is increased, the droplet will deform into an ellipsoid whose major axis lies parallel to the electric field lines (Fig. 1). If sufficient field strength is provided, the droplet will become unstable and disintegrate into a large number of smaller daughter droplets. The same effect occurs when a pulsing electric field is used. The droplet deforms when the field is on and relaxes back toward the spherical shape when the field is off; hence, the droplet is forced to oscillate about the spherical form. Augmentation of direct-contact heat transfer has been carried out using pulsing electric fields in the 2 to 8 Hz frequency range by Kaji et al. (1978, 1980). Results from these studies indicate that low-frequency, high-displacement pulsing of liquid droplets enhances the heat transfer process. This type of technique has also been used in our laboratory under higher frequency conditions which are near droplet natural oscillation frequencies (20 to 60 Hz) to study the effects of droplet oscillation on the rate of

DROP OSCILLATION IN PULSING ELECTRIC FIELD
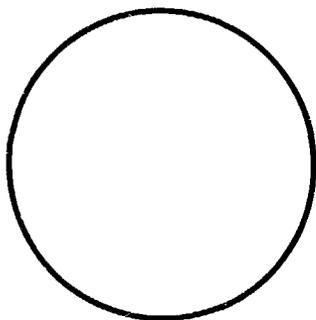
SPHERE

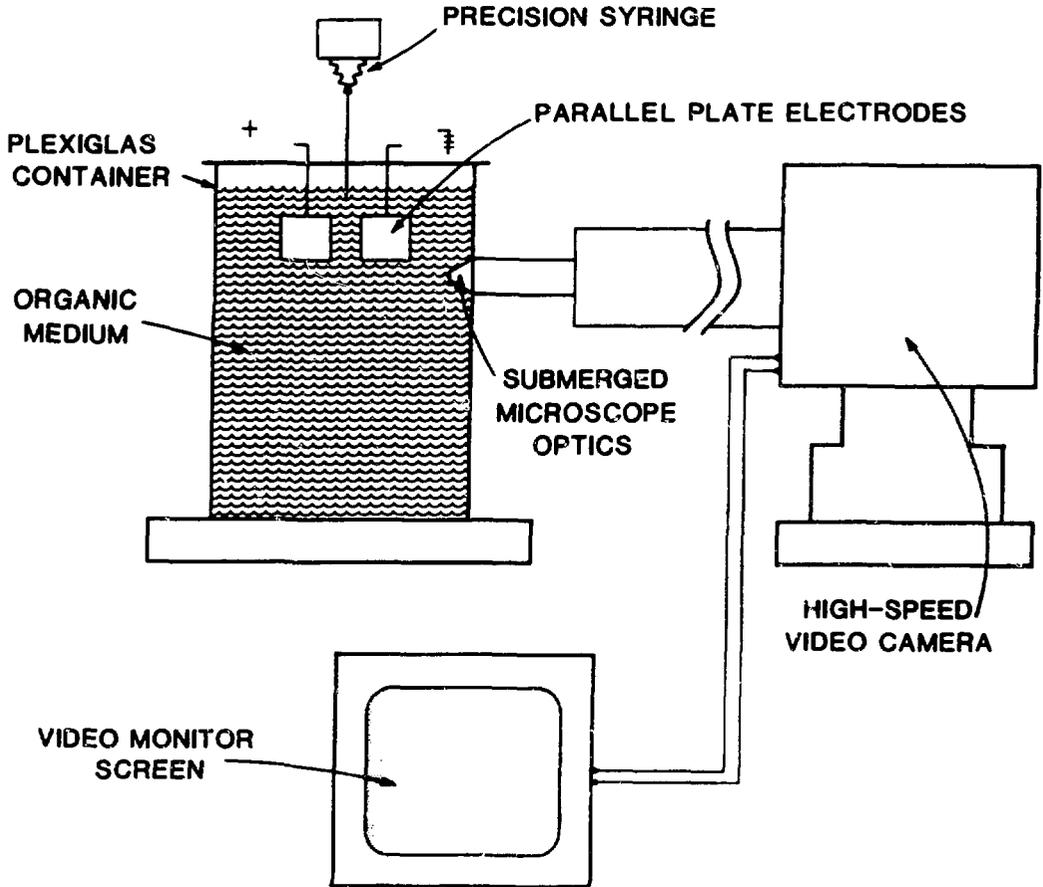(NO FIELD)
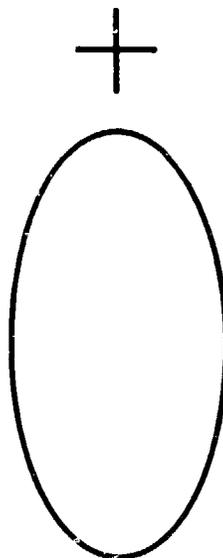
PROLATE SPHEROID

(FIELD ON)

mass transfer for the case of continuous phase control (Wham and Byers 1987, Scott 1986, Scott 1987a, Scott and Byers 1987). If the proper combination of pulse rate and field strength is utilized, the droplet will shatter, thereby creating a vast amount of interfacial surface area. Initial data obtained in our laboratory indicate that one should be able to choose a drop size or a range of drop sizes which will be allowed to exist in a given region of a mass transfer apparatus while creating large amounts of interfacial surface area (Scott, 1987b).

In order to assess possible process applications, experimental methods have been developed to measure droplet size characteristics and energy inputs associated with the rupture of aqueous droplets by high-intensity, pulsed electric fields. The combination of in situ microscope optics and a high-speed video camera allows reliable observation of liquid droplets down to 0.5 μm in diam. Figure 2 contains a schematic diagram of the experimental system. Aqueous droplets can be emulsified between precision-machined parallel plate electrodes. Knowledge of electric field conditions and observations of the resulting microscopic droplets lend insight into the properties of emulsions which would be formed in solvent extraction vessels.

EXPERIMENTAL EQUIPMENT AND PROCEDURES

Two optical units were constructed for use with the TRITRONICS model DCSM-5600 high-speed video camera (Fig. 2). One was a 45X microscope objective, placed at the end of a 1-in. I.D. barrel, with a 10X microscope eyepiece placed at the other end. The second assembly was the same except that a 100X microscope objective was used. The units were 33.7 cm in

APPARATUS FOR DROPLET-SIZE AND SIZE-DISTRIBUTION MEASUREMENTS

PRECISION SYRINGE

PARALLEL PLATE ELECTRODES

PLEXIGLAS CONTAINER

+

ORGANIC MEDIUM

SUBMERGED MICROSCOPE OPTICS

HIGH-SPEED VIDEO CAMERA

VIDEO MONITOR SCREEN

length. These optics were attached to the camera, and the objective was inserted into a Plexiglas container through a neoprene O-ring for sealing. This allowed the lens to be immersed in the organic solution for better resolution. The camera signal was then fed to a FOR.A model UTG-33 video timer, a FOR.A model VPA-1000 video position analyzer, and finally to a video monitor screen for visual observation.

The system was calibrated with a 50-μm stainless steel wire. This gave a calibration of 5.7 divisions/μm on the video screen using the 100X objective and the 10X eyepiece. Resolution of the video position analyzer on the screen was two divisions; therefore, it is possible to detect a minimum droplet size of approximately 0.5 μm. The video camera has a magnification feature that, when energized, can raise the calibration to 8.7 divisions/μm. Visible light cannot be used to reliably observe objects less than 0.5 μm; hence, this optical system was able to operate at the limit of detection.

The electrodes were 1.27-cm squares which were 0.32-cm thick. Spacing between the electrodes was maintained at 1.0 cm. The metallic microscope objective lenses were placed 2.0 cm away from the electrodes to avoid sparking. A triangular trough was fabricated to direct the generated droplets into the focal area of the lens. Lighting for this application proved to be critical. An intense fiberoptics lamp was directed through the rear wall of the container and through the organic media to provide back lighting for the lens.

The electric field was created by a custom-built pulsed voltage generator. This generator utilizes a variable speed drive motor which rotates a typical automotive spark distributor that delivers 6 pulses/ revolution. Rotation is monitored by a digital tachometer. A 12-V dc

automotive high-voltage coil is used with the distributor. A variable primary dc voltage supply of up to 15 V is used to energize the coil. This system is capable of pulses up to 25 kV at 0-200 Hz.

Emulsions comprised of microscopic water droplets in the continuous organic phase were created by electric field disruption of a stream of water flowing from the precision syringe. Dispersions of this type were generated at pulse frequencies of 20, 40, and 60 Hz, using field strengths of 5.1, 7.7, and 9.5 kV at each frequency; therefore, nine different operating conditions were created for each aqueous/organic fluid pair. Videotapes were taken of the emulsions at a shutter speed of 500 frames/s. Two minutes of videotape were recorded for each operating condition. For each run, fifty droplets which were in focus were measured using the video position analyzer.

RESULTS AND DISCUSSION

Droplet size measurements were taken for dispersions of water formed in four different organic solvents: 2-ethyl-1-hexanol, hexanol, octanol, and 30% tributyl phosphate (TBP) / 70% dodecane. Table 1 contains a summary of the results obtained for the water - 30% TBP/70% dodecane system. The tabular entries display the largest and smallest droplets observed as well as the average droplet size for each of the nine experimental conditions. It is evident that extremely small average droplet sizes (<5 µm) and tight size distributions (1 to 10 µm) are possible when using the electric field method. Similar droplet sizes and size distributions were obtained for all of the fluid systems.

TABLE 1

Examples of Droplet Sizes and Size Distribution
from Electric Field Disruption

Organic Phase: 30% Tributyl Phosphate/ 70% Dodecane
Aqueous Phase: Distilled Water (Mutual Saturation)

Droplet sizes in μm.

5.1 kV/cm		7.7 kV/cm	9.5 kV/cm
20 Hz			
High	36.0	15.4	9.8
Average	9.6	5.4	3.4
Low	2.6	2.1	1.4
40 Hz			
High	6.8	11.3	5.6
Average	3.8	4.5	3.1
Low	1.9	1.8	1.6
60 Hz			
High	33.3	11.6	8.8
Average	5.9	4.7	2.9
Low	1.6	1.9	0.9

Figure 3 contains the droplet size distribution obtained for the highest energy run shown in Table 1 (60 Hz, 9.5 kV). The distribution is slightly skewed toward the larger droplet regime, which appears to indicate that some coalescence is taking place to form larger droplets in the system. In order to properly address this hypothesis, more extensive droplet size data sets will have to be obtained.

Assuming that the emulsion and the electrodes make up a parallel plate capacitor, energy usage in the system can be approximated by measuring the voltage drop across the electrodes and mathematically treating the transient behavior as the charging and discharging of the "capacitor."

The total energy expended in charging the capacitor is given by (Sears, 1958):

$$E_c = \frac{1}{2}CV^2, \tag{1}$$

where E_c is the energy per pulse (joules), V is the measured voltage, and C is the capacitance. This is calculated using the following relationship:

$$C = K\varepsilon_0 \frac{A}{L}, \tag{2}$$

where K is the dielectric constant of the emulsion, ε_0 is the permittivity of a vaccum, A is the electrode area, and L is the distance between electrodes.

Table 2 contains the values used for the constants in Eqs. 1 and 2. Taking the measured voltage to be 9.5 kV and inserting the appropriate values into eq. 1 yields a total energy per pulse (E_c) of 6.41 x 10^{-5} W·s. Assuming a 60-Hz pulse rate, one expends 2.38 x 10^{-3} W/cc of fluid to create the emulsion shown in Fig. 3.

NUMBER OF DROPLETS IN SIZE RANGE

DROPLET DIAMETER (MICRONS)
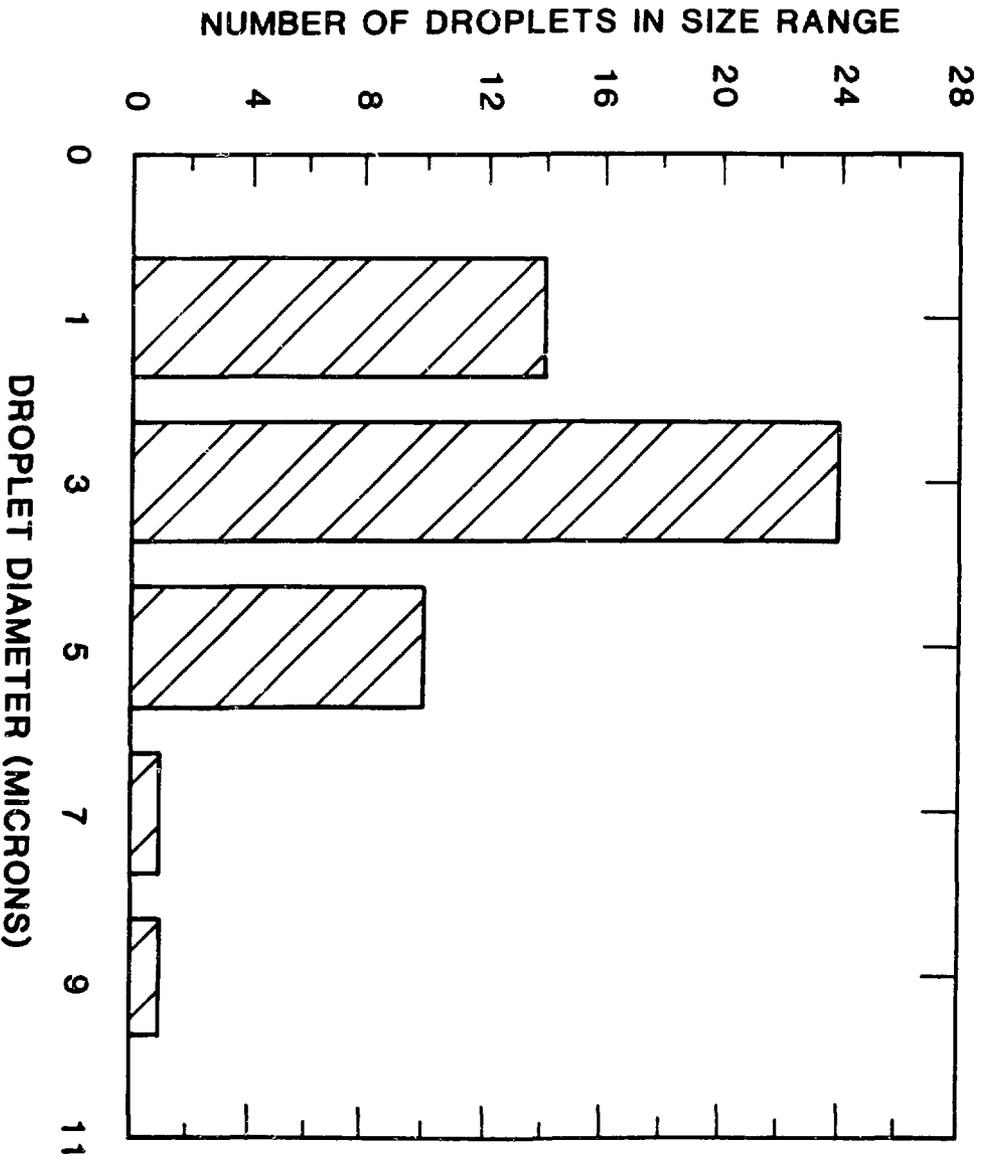
DROPLET SIZE DISTRIBUTION
(WATER – TBP/DODECANE; 60 Hz@9.5 kV)

ORNL DWG 87-1332

TABLE 2

Values of Parameters Used to Caculate Electric
Energy Input to the Emulsions

Parameter	Value
K	10
ε_o	$8.85 \times 10^{-12} \ C^2 N^{-1} m^{-1}$
A	$1.61 \times 10^{-4} \ m^2$
L	$1.0 \ \times 10^{-2} \ m^2$

For comparison, the power input required for a mechanically agitated baffled tank was calculated. The correlations of Chen and Middleman (1967) were used to obtain the power number required in the vessel, and the actual input per unit of fluid was then calculated using the correlations of Rushton et al (1950 a, b). The calculated power input was 25 W/cc of fluid for an average droplet size of 5 µm. This illustrates that the electric field method is substantially more energy efficient in this particular instance. The agitator data had to be extrapolated in order to obtain a value for the power input. The resultant impeller speed was over 3000 rpm; hence, the 5 µm size represents conditions which are probably beyond the capability of an agitated tank.

CONCLUSION

The use of pulsing electric fields to create mass transfer surface area appears to be an attractive possibility for improved solvent extraction operations. The emulsions are comprised of micron-sized droplets which have a narrow size distribution and therefore may be amenable to hydrodynamic control in a mass transfer apparatus. The initial data presented seem to indicate that some coalescence does occur after dispersion. This point needs to be addressed in future studies in order to assess possible uses of electric fields to induce phase separation. Energy requirements for creation of the vast amount of surface area are substantially lower than mechanical agitation while operating in regimes which are probably beyond the capability of typical agitation systems.

The adaptation of such schemes to industrial applications could provide systems which are an order of magnitude more efficient than present-day extractors. In order for this type of technique to be useful in industrial applications, one must demonstrate hydrodynamic control of the emulsion and subsequent coalescence of the droplets to enable phase separation.

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FIGURE CAPTIONS

Fig. 1. Drop oscillation in pulsing electric field.

Fig. 2. Apparatus for droplet-size and size-distribution measurements.

Fig. 3. Droplet size distribution (water - TBP/dodecane; 60 Hz at 9.5 kv).