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Mass Transport from Single Droplets in Imposed Electric Fields

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ABSTRACT

The influence of electric charge phenomena on mass transfer in systems involving droplets in a continuous medium is of increasing importance. The enhanced transport behavior of droplets in imposed fields on mass transfer and the influence of naturally occurring charges on droplets, particularly in the atmosphere, are both subjects of current interest. The objective of this study is to elucidate the hydrodynamic and mass transfer effect of imposed fields on charged single droplets. Investigations of both static and pulsed electric fields are reported.

The experimental approach is based on the suspension of single droplets in a precisely machined expanding channel. A stationary, saturated water droplet is fluidized by the upflow of 2-ethylhexanol. This arrangement allows precise photographic monitoring of droplet size and shape as well as the hydrodynamics of both phases in the region of the interface.

Baseline continuous-phase mass transfer studies with internally circulating droplets corroborate earlier studies with the same system. The effect of the accumulation of unavoidable surfactant on droplet circulation and mass transport rate at long exposure times is reported. The imposition of a static electric field in the axial direction caused distortion of droplets into prolate spheroidal shape.

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An electric field pulsed at a tuned frequency is observed to cause oscillation of droplets between prolate- and oblate-spheroidal shapes. In addition to frequency and field amplitude, the interfacial properties of the fluid system as well as viscosities and densities of the two phases influence the oscillation. The effect of the pulsation of the field on mass transfer to the continuous phase is reported. These data are compared with results for both non-oscillating mass transfer and theoretical models.

INTRODUCTION

Mass transfer between immiscible liquid phases, generally referred to as solvent extraction, is a technique used for separation and purification of chemicals into final form. Solvent extraction is used in many industries such as petroleum refining, chemicals manufacture, metals processing, and nuclear fuel reprocessing. Therefore, any improvement in the efficiency of solvent extraction will have broad implications.

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One serious problem encountered in solvent extraction is that the processes are rate limited by interfacial mass transfer. In addition to diffusional properties, rate depends on the hydrodynamics of the contacting phases and the extent of contamination of the interface with surfactants. Since transport between droplets and the continuous phase accounts for most of the mass transfer between phases in extractors, it is important to the various regimes of convective diffusion which occur. These include: (1) stagnant drop, where the drop is in a quiescent solution experiencing only diffusion and natural convection; (2) rigid drop, where fluid flows past the drop but the interface does not move; (3) circulating drop, where fluid flows past the interface and the interface moves with the fluid, establishing flow cells in the drop; and (4) oscillating drop, where drops are stretched periodically into oblate and prolate ellipsoids. Obviously dispersion and coalescence associated with high shear fields are important contributors to mass transfer but are more properly discussed separately.

Most processes rely on mass transfer enhancement through an increase in surface area by using small drops or by an increase in convection from mixing the streams. Both of these can have serious drawbacks and complications. When very small droplets are used, the drops can become entrained in the continuous phase hindering coalescence and make phase separation difficult and, in the limit, impossible. Because of the scale of turbulence imposed on the continuous phase, small drops can act like rigid spheres with very little interfacial motion and, hence, low local transport rates. Mixing involves mechanical agitation and internal moving parts.

Mechanical agitation requires energy input to the bulk fluid, most of which is inefficiently used. Generally, fluid close to the impeller

is well mixed but fluid far away is poorly mixed. When the mechanical device breaks down the unit must be shut down and opened up for service — sometimes a dangerous and costly operation (particularly where hazardous materials are used).

Thus, one potential method of enhancing mass transfer is to promote convection which is focused on the interfacial region. The use of drop oscillation is one such means. Droplet oscillation occurs naturally as a consequence of wake shedding. When a large drop falls under the influence of gravity in a continuous medium, the drop develops a wake and forms an oblate spheroid. At some point the drop will shed its wake causing it to alter its shape toward a prolate form after it releases the wake. The process repeats itself as the drop falls through the medium. The change in shape is minimal in most cases. However, if conditions are right, that is, if the drop is the correct size for its natural frequency to match the shedding frequency, it will oscillate as it falls. Lamb (1) performed a stability analysis on droplet oscillation which predicted oscillation frequency as a function of viscosity, interfacial tension, and density difference between the two phases. Schroeder and Kintner (2) developed an empirical modification to Lamb's equation which accounted for the magnitude of the stretch.

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Thus it can be seen that droplet oscillations can enhance mass transfer but continued oscillations require an energy input to the droplet. Energy input can be mechanical, electrical, or magnetic. Loshak and Byers (3) found that mechanical energy in the form of sound waves is not an effective method of sustaining oscillations. Loshak and Byers studied droplet oscillation frequency and decay of oscillation using sonic energy to force droplet oscillation. They found that influence of the sonic energy damped out rapidly as the drop fell away from the oscillating surface where it was formed, thus droplet oscillation decayed. The nature of the oscillation decay indicated the presence of surfactant at the interface.

The current required to produce a controlled magnetic field raises the possibility that high energy costs might detract from this means of driving oscillation. Electric fields cause droplets to deform in the direction of the field and require high voltage but very little current assuming that the continuous phase is relatively nonconductive. The effects of electric fields on the deformation and stability droplets was studied by Lord Rayleigh (4). Lord Rayleigh reported that an aqueous drop in an electric field will deform along the field lines. The drop stability studies allow us to predict a maximum charge to mass ratio which is referred to as the Rayleigh limit or Q_{Ra}. When the critical charge to mass ratio is exceeded, the drop will fission off small daughter drops to remove a portion of the charge.

At ORNL mass transfer from aqueous drops to an organic continuous phase with continuous phase control was studied by Clinton (5) for the case of rigid and fully circulating drops. Clinton used a

tapered column fabricated from Lucite which had a converging section at the bottom and diverging section at the top where the drops were fluidized as shown in Fig. 1. He found that the converging section at the fluid entrance was critical in maintaining droplet hydrodynamic stability. Clinton fluidized a denser aqueous drop by an upflow of continuous phase organic varying flow rate so that the drop could remain stationary with respect to the laboratory reference frame. A box camera with a 32-mm macro lens was used to observe drop size during an experiment. First Clinton studied water droplets on whose surface surfactant had been sorbed, halting any internal circulation. The Sherwood number, Sh, for those rigid drops could be related to the drop Reynolds number, Re, and the continuous phase Schmidt number, Sc, as

$$Sh = 0.99\ Re^{0.42}\ Sc^{0.33}$$

$$0.2 < Re < 20\ . \qquad (1)$$

For fully circulating water drops, Clinton studied only a system which with a constant Sc (35,700) in the Re range 1.7 to 13 yielded the following relationship

$$Sh = 114\ Re^{0.57} \qquad (2)$$

The mass transfer coefficient, K, at a given Reynolds is a factor of approximately four to five higher in the circulating case.

Given the work of Clinton on nonoscillating droplets and reorganizing that, it would be preferable to observe droplets in a stationary frame of reference; we build our experiment around this experimental idea. The imposition of a field (electric in this study) to cause oscillation of droplets which could be predicted to enhance mass transport is the subject of the remainder of this paper.

THEORY

The phenomenon of droplet oscillation has been observed by researchers for many years. Lamb (1) developed a linearized oscillatory solution of the continuity equation caused by gravitational oscillations of a liquid sphere with the objective of predicting the natural frequency. Lamb assumed that the liquids were incompressible and inviscid. The oscillation frequency depends on drop size and density difference between the two phases and is given below:

$$\omega_L = \frac{1}{2\pi} \sqrt{\frac{\gamma}{r^3} \frac{n(n+1)(n-1)(n+2)}{(n+1)\rho_D + n\rho_C}}\ . \qquad (3)$$

The solution predicts several modes of oscillation with the n = 2 mode being the dominant mode. For n = 2 the drop shape oscillates between prolate and oblate forms, while for n = 3 a 3-lobe oscilla-

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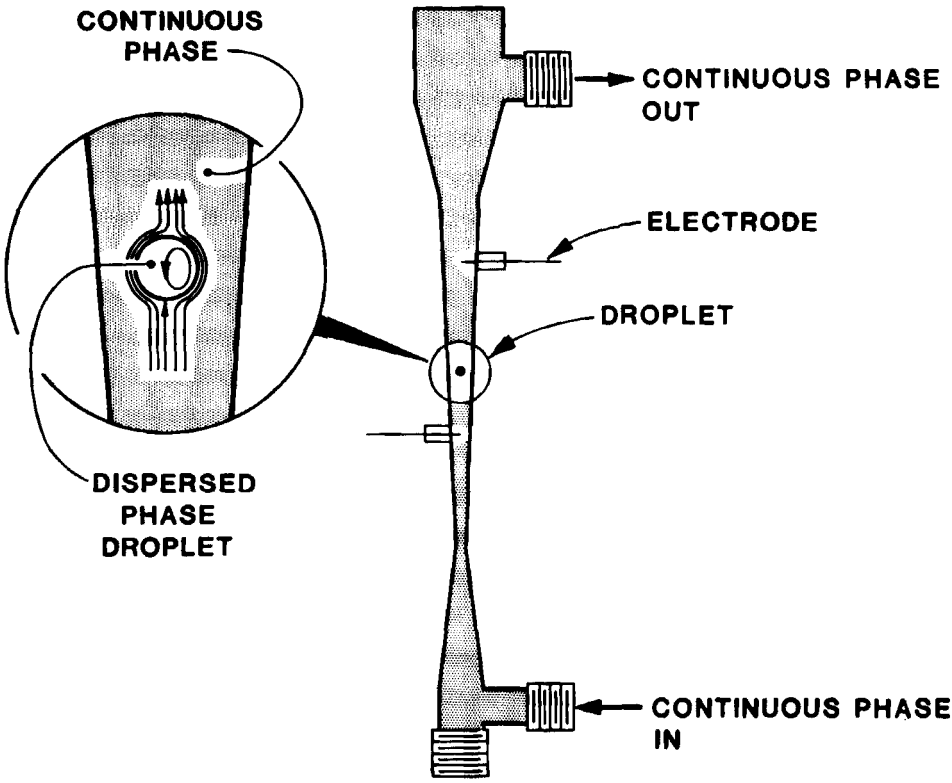

Fig. 1. The experiment centers around a fluidized water drop in a continuous phase of 2-ethylhexanol.

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tion occurs, n = 4 has 4 lobes and so on. A value of n = 0 describes compression, while a value of n = 1 is translation, and thus are not applicable to oscillation.

Schroeder and Kintner (2) extended the analytical solution of Lamb to account for amplitude of oscillation. Their work, which is empirical, was based on high speed films of oscillating drops in several systems. The variation in amplitude of oscillations over a cycle was assumed to be sinusoidal from oblate to spherical and then prolate form. The modification proposed to the Lamb equation was an empirical factor b in the frequency correlation.

$$\omega_{SK} = \frac{1}{2\pi} \sqrt{\frac{\gamma b}{r^3} \frac{n(n+1)(n-1)(n+2)}{(n+1)\rho_D + n\rho_C}} \quad . \tag{4}$$

where

$$b = 0.805\ r^{0.225} \ . \tag{5}$$

Based on first principles, Miller and Scriven (6) extended the work of Lamb for the general case of viscous fluids under the influence of gravity. The results account for the viscosity of the two phases.

$$\omega_{MS} = \omega_L - \frac{(2n+1)^2}{2\sqrt{2r}\ \Gamma} \frac{(\omega_L\ \mu_D\mu_C\rho_D\rho_C)^{1/2}}{(\mu_D\rho_D)^{1/2} + (\mu_C\rho_C)^{1/2}} \ . \tag{6}$$

$$\Gamma = \rho_D(n+1) + n\rho_C \ . \tag{7}$$

The Miller and Scriven analysis also takes into account damping of natural oscillations due to viscous dissipation of energy in a boundary layer near the surface of the drop. The damping coefficient, τ, is given by

$$\tau = \frac{(2n+1)[(n-1)\mu_D + n(n+2)\mu_C]}{r^2\ \Gamma} \ . \tag{8}$$

Loshak and Byers studied droplet oscillation and decay of oscillation using an oscillating syringe needle to force droplet oscillation (3, 7). The oscillation frequencies agreed well with Miller and Scriven but decay rates were higher than predicted, possibly due to the accumulation of surfactant impurities.

Ramabhadran (8) expanded the work of Loshak and Byers to examine the effects of shear elasticity on the decay of oscillations. The primary thrust of the study was droplet stability during oscillation and not drop hydrodynamics. Ramabhadran found that surfactants at the interface dominated the oscillation decay rate.

Turning to mass transfer from droplets enhanced by oscillations, Rose and Kintner developed a model based on penetration theory and sinusoidal oscillations (9). The mass transfer coefficient, K, is

$$K = \left[-\frac{2\pi D_E}{V} \int_{t_o}^{t_f} \frac{1}{f_I(t)} \left\{ \left(\frac{3V}{4\pi(a_o + a_p \left| \sin \omega t \right|)^2} \right)^2 \right. \right.$$

$$\left. \left. \frac{1}{2\alpha} \ln \frac{1+\alpha}{1-\alpha} + (a_o + a_p \left| \sin \omega t \right|)^2 \right\} \ dt \right] \tag{9}$$

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in which

$$\alpha^2 = \frac{(a_o + a_p \mid \sin \omega t \mid)^2 - \left(\frac{3V}{4\pi(a_o + a_p \mid \sin \omega t \mid)^2}\right)^2}{(a_o + a_p \mid \sin \omega t \mid)^2} \tag{10}$$

$$f_1(t) = \frac{[a_o{}^2 b - (a - x_o)^2(b_o - x_o)] - 2abx_o + bx_o{}^2}{a^2 - 2ax_o - x_o{}^2} . \tag{11}$$

Angelo and Lightfoot extended penetration theory to account for stretch at the interface (10). They developed a model based on the assumption that velocity at the interface was related to the change in surface area and the velocity component, v_y, in the continuity equation caused the mass transfer enhancement. Symbolically, the continuity equation becomes:

$$\frac{\partial C}{\partial t} + v_y \frac{\partial C}{\partial y} = \mathcal{D} \frac{\partial^2 C}{\partial y^2} \tag{12}$$

and using the assumption that velocity in the y direction is proportional to the rate of change in surface area, S,

$$v_y \doteq -y \frac{\partial}{\partial t} (\ln S) . \tag{13}$$

After substituting Eq. (13) into Eq. (12), separating variables, and integrating, the average mass transfer coefficient K is given as

$$\overline{K} = \frac{2}{\tau} \sqrt{\frac{\mathcal{D}}{\pi t_o}} \sqrt{\int_o^\tau \left(\frac{S(t)}{S_o}\right)^2 dt} . \tag{14}$$

The authors reported that their model and the Rose and Kintner model agreed within about 15%.

Other approaches to modeling mass transfer from oscillating drops have been highly empirical. One such approach to convective mass transfer is presented by Boyadzhiev et al. (11). Boyadzhiev studied mass transfer from oscillating drops and suggested a factor $\overline{R}$ be used as a premultiplier of diffusivity. The product is then an effective diffusivity which depends on convection. Boyadzhiev plotted the factor $\overline{R}$ vs Reynolds number for $10 < Re < 200$. Hence, the effective diffusivity is an empirical approach to attempting to account for drop hydrodynamics.

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Recent efforts by Chalabreysse (12) and Martin and Vignet (13) indicate that droplet oscillations yield significant increases in mass transfer. The experiment is based on application of high voltage AC current to the inlet nozzle of an extraction column. Droplet size varied from ~.08 cm to ~.45 cm depending on the applied field. The applied field has two effects, the droplet size is now controlled by the application of an electric field and the column can operate in the droplet size range where the droplets are likely to oscillate. The electric field will tend to increase droplet velocity through the column thus increasing convection. Mass transfer coefficients increased dramatically (200%) but there appears to be no delineation by Martin and Vignet between the regimes of mass transfer (i.e., rigid drops, circulating, or oscillating) or the extent of convective mass transfer which are used as a baseline case. Hence, it is difficult to distinguish between the effects of oscillation, internal circulation, and convective mass transfer.

The effect of electric fields on droplets suspended in fluids has been examined by Stratton (14) and O'Konski and Harris (15). When a conducting liquid droplet is placed in a quiescent nonconducting liquid and exposed to an electric field, the drop deforms in order to balance electric and surface forces. The deformation of the drop shape can be characterized by the drop eccentricity e where

$$e = \left[1 - \left(\frac{b}{a}\right)^2\right]^{1/2} \tag{15}$$

(a is the major axis of the drop and b is the minor axis). The sum of electric forces and surface forces can characterize the drop shape in terms of e.

For small values of e, $0 \leq e \leq .7$

$$e = \frac{3}{4} E_0 \left(\frac{r\varepsilon_1}{\pi\gamma}\right)^{1/2} \left[\frac{(\kappa_2-\kappa_1)(\kappa_2^2 + 7\kappa_1\kappa_2 - 2\kappa_1^2(1 + 3\,\varepsilon_2/\varepsilon_1)}{(\kappa_2 + 2\kappa_1)^3}\right]. \tag{16}$$

Upon inspection of the previous equation one finds, for a given system and drop size

$$e \propto \frac{E_0}{\left(\frac{r}{\gamma}\right)^{1/2}} . \tag{17}$$

The form of the equation for e given above indicates that interfacial tension is a factor which determines the amount of stretching (i.e., amplitude of oscillation) at a given field strength.

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EXPERIMENTAL APPARATUS

The experimental apparatus (Fig. 2) consists of equipment to fluidize a water drop in the continuous medium, static and pulsed DC power supplies for electric field sources, and equipment for viewing and recording droplet oscillations. A detailed description is given elsewhere [Wham and Byers (16)].

Droplets are fluidized in a tapered column fabricated from glass tubing and polished to assure smoothness. The 2-ethylhexanol (2EH) enters at the bottom and exits at the top. The converging section at the bottom serves to help induce laminar flow throughout the top diverging section. Droplets are introduced through a hole in the top of the column. Other items of importance not shown in the figure are a rotameter for controlling 2EH flow rate, a small laboratory pump, and a purged reservoir of 2EH.

Droplets are subjected to electric fields, both static and pulsed, through electrodes inserted radially into the tube. The electrodes are positioned ~2.5 cm apart and are made of 0.32 cm stainless steel rods. This was found to yield the most stable fluidized droplets in both stretching and oscillating modes. Configurations using axially opposed electrodes caused flow instability problems because of turbulence introduced by the bottom electrode. A configuration where the electrodes were radially opposed to each other also yielded flow instability problems because the charged droplet tended to be attracted to the high potential electrode which caused droplet instability and breakup.

A static DC field is provided using a Hipotronics DC power supply with a range of 0-20 kV. A pulsed DC field is produced by an automotive distributor and coil. Power is supplied by a variable power supply (0-12 V) and the distributor is turned by a variable speed motor. The arrangement allows pulses to be generated at a frequency in the range of 0-150 pulses/sec with a fixed duration of each pulse of ~4 msec. Generally, the potential between electrodes is ~10 kV but can be controlled between 5-20 kV. A Digistrobe strobe light/tachometer is used to calibrate the frequency of the DC pulse. A Tektronix type 504 oscilloscope is used to check input to the column. Data from the oscilloscope provide frequencies and pulse amplitudes.

In order to view oscillating drops, a camera with a shutter speed which is fast compared to drop oscillation frequency (150 pulses) and pulse duration (4 msec) is required.

A Tritronics model PC5600 high speed shuttered video camera was used to record the experiments. Shutter speed can vary from 4 msec to 0.1 msec with framing rates of 60-300 frames per second. A Sony VO-5800 video recorder is used for recording the video image and playback of the results for analysis. A FOR.A video timer VTG-33 is used to record the date and time (to within 0.01 sec) on the video

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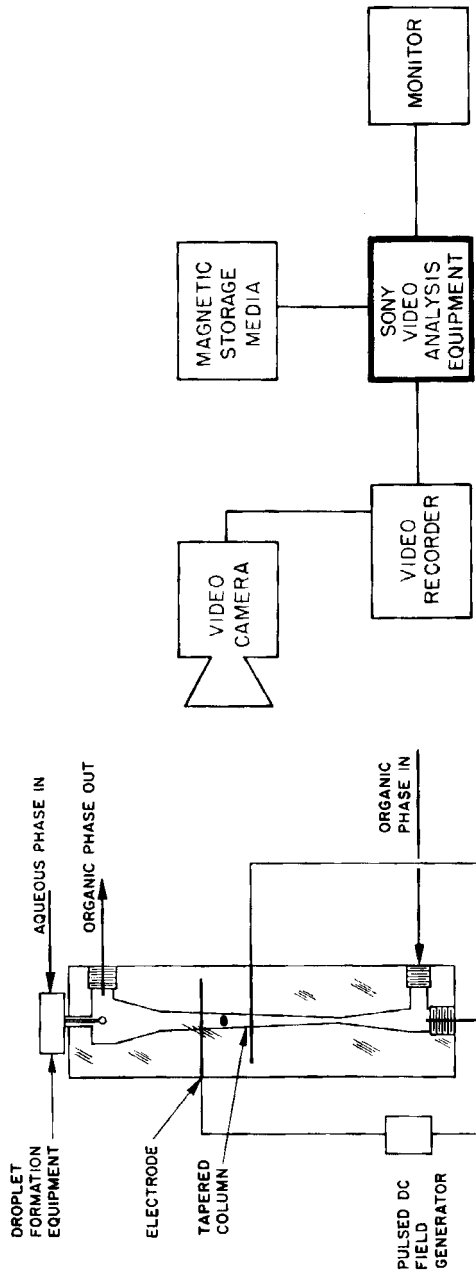

Fig. 2. The experimental equipment incorporates microprocessor electronics for video analysis. It allows for interactive data acquisition and analysis, as well as for recording the data on magnetic storage media.

tape during each experiment. A VPA-100 position analyzer is used for determination of droplet size for the video image. An Apple IIe microcomputer is used for data analysis, as well as a Sony microcomputer with Genlocking capability. The Genlocker allows an overlay of microcomputer graphics on the video image. This combination provides selective digitizing of the video image.

An experiment consists of charging a cleaned reservoir with ~400 ml of 2EH. The 2EH is pumped through the system. At time zero a droplet of water is released into the column. The video camera is started just prior to droplet introduction. When the drop is fluidized in the center of the column, the DC field starts and the drop is stretched if the field is static and oscillates in a pulsed field. Observation of the size and oscillation behavior of the drops continues for up to 10 min.

Data on drop size are analyzed by measuring the change in the projected area of a backlighted drop per unit time. Assuming the drop is axisymmetric, the drop volume and drop surface area is calculated yielding mass transfer and hydrodynamic data. The flux of water from the drop to the continuous phase (2EH) is calculated using the following equation:

$$N = -\frac{1}{A}\frac{d(\rho V)}{dt} = K(C^* - C_b) \quad . \tag{18}$$

In those experiments which do not subject the drop to an electric field, the drop remains approximately spherical and the above equation can be modified as shown:

$$-\frac{1}{A}\frac{d(\rho V)}{dt} = -\frac{\rho}{4\pi r^2}\frac{\partial}{\partial t}\left(\frac{4\pi}{3} r^3\right) = -\rho\frac{dr}{dt} \quad . \tag{19}$$

From an experimental viewpoint, the observation of a single parameter, such as diameter, is only valid in near-spherical situations. Where significant distortion or oscillation occurs, a volume measurement is required. Since the shapes of the droplets are not regular, it is necessary to perform a computer analysis to determine drop volumes as a function of time.

RESULTS AND DISCUSSION

Our analysis of electrically driven oscillations is divided into three areas. These include:

1. The effect of static fields on drop shape and system physical properties.

2. The changes in drop shape in response to a pulsed electric field.

3. Mass transfer effects in electric fields.

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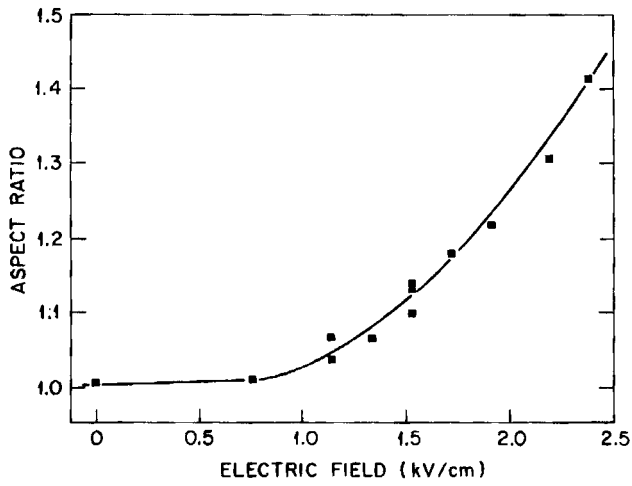

Fig. 3. Droplet distortion vs the electric field strength.

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Turning first to the effect of the electric field on droplet shape and physical properties, the limits of electric field strengths of interest are from minimal observable changes in drop shape, the point where it is disrupted by the imposition of a field which exceeds the stability limit. Figure 3 shows droplet aspect ratio as a function of electric field strength expressed in terms of applied voltage per cm electrode separation. When a static axisymmetric electric field is applied, a drop distorts into a prolate spheroid and remains stable to a range of 4 kV/cm. Up to approximately 1 kV/cm, the drops are not stretched to any significant extent. The nonunity aspect ratio at zero field reflects the distortion caused by this fluidizing continuous phase. It causes an oblate distortion, thus the field must balance this force on the drop before it can impose a prolate shape on the drop. This type of stretch is manifested at higher field strengths. This indicates that the electric field generator which will promote oscillation of droplets must be capable of imposing a field strength of approximately 3 kV/cm and optimally be reduced to less than 1 kV/cm in order to achieve an oscillation cycle. The maximum time frame during which this fluctuation must occur is on the order of the reciprocal of the desired oscillation frequency.

The most critical physical property we were concerned with is the interfacial tension of the water/2-ethylhexanol system. In order to develop a force balance on the drop, we must account for the electric field effect. There is a possibility that the concentration of electric charges at the interface affects interfacial tension

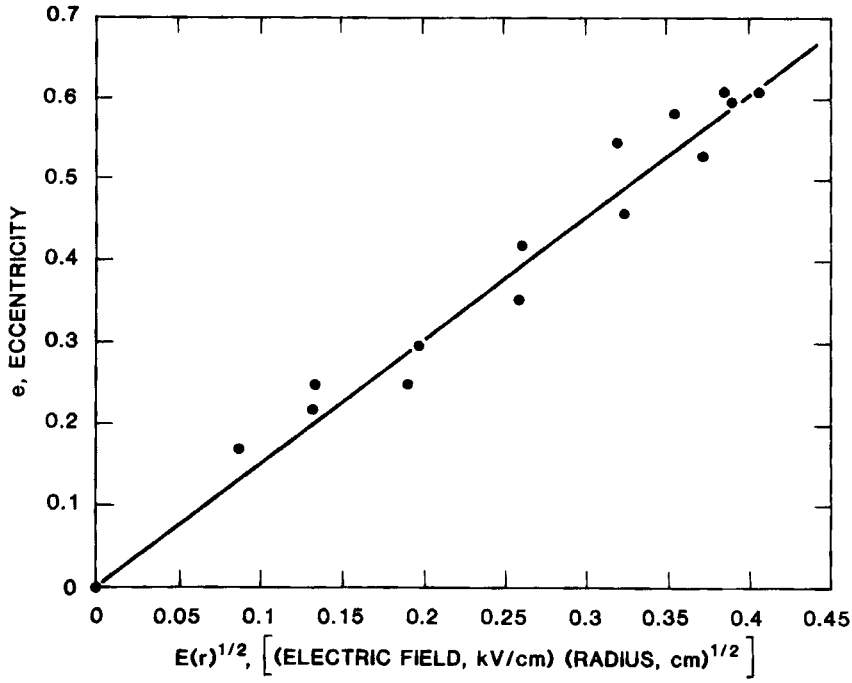

Fig. 4. Electric field strength vs droplet eccentricity.

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either directly or indirectly by modifications of the distribution of chemical species at the interface. If interfacial tension is constant, then the electric field distorts the drop into a prolate spheroid at field strengths below the Rayleigh limit. Otherwise we must account for the variation interfacial tension as a function of field strength. The analysis of O'Konski and Harris [Eq. (7)] indicated that the droplet eccentricity is a linear function of the applied electric field provided droplet radius and interfacial tension remain constant. The next figure, Fig. 4, shows the same data as Fig. 3 but this time plotted as eccentricity vs applied field strength times the square root of the radius of the drop. As can be seen from the figure, the curve is linear over the range studied. At low fields one would expect hydrodynamic effects to render this relationship inaccurate. It should also be noted that the data appear to be skewed at higher field strengths. The skew results from the fact that at the higher field strengths the drop has been fluidized in the column for longer periods of time and tends to accumulate surfactant at the interface, thus lowering the interfacial tension. The change in the slope is in the correct direction for a lowering of interfacial tension.

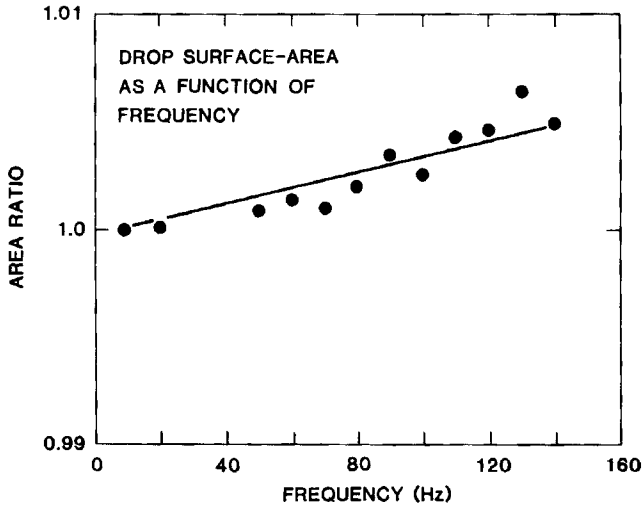

Fig. 5. The ratio of the surface area of an oscillating drop to a sphere of equivalent volume vs frequency oscillation.

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The next area of interest is the change in droplet surface area as a function of pulsed field. Data were taken using fluidized water drops in a saturated solution of 2 EH. The droplets were subjected to oscillation frequencies of 10-120 Hz and a field strength of ~2.5 kV/cm. Drop aspect ratios and surface area were calculated from videotapes of the experiments. The time averaged surface area was then calculated. The ratio of droplet surface area to the surface area of a sphere of equivalent volume is plotted vs oscillation frequency in Fig. 5. The droplet surface area is seen to change less than 1% from that of an equivalent sphere over the range of frequencies shown, and a linear fitting of the data is within experimental error. Therefore, the calculated relation between droplet surface area and drop distortion (aspect ratio) was plotted in Fig. 6. The range of aspect ratios greater than 0.7 and less than 1.3 is highlighted because that is the general range of aspect ratios observed during droplet oscillation. Again, the surface area does not vary significantly from the surface area of a sphere of equivalent volume.

Finally, mass transfer for oscillating droplets was measured by transferring water from a water droplet into the 2 EH. The initial concentration of water in 2 EH was 0.3 wt % as compared to equilibrium concentration of 2.6 wt %. Figure 7 is a comparison of drop mass (computed from volume) vs time for an oscillating drop to the baseline case of a nonoscillating drop. The droplets examined here had an equivalent initial diameter of 0.3 cm and the Reynolds number, based

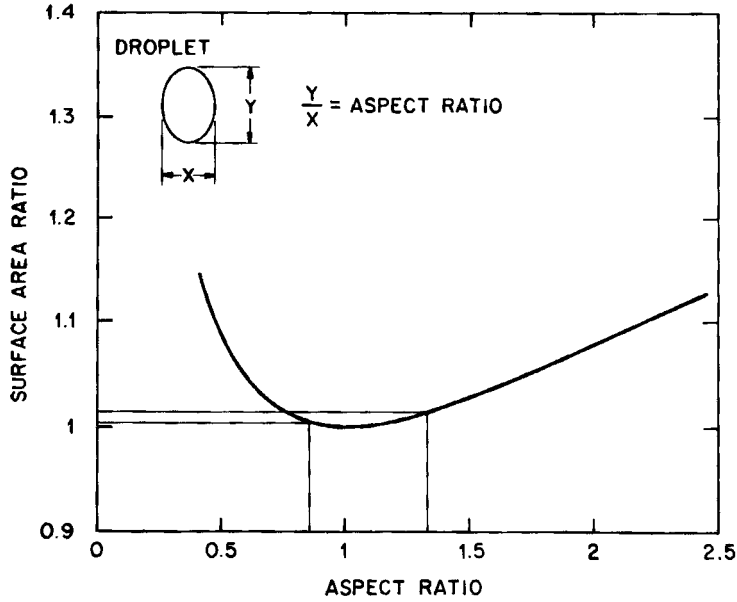

Fig. 6. The ratio of surface area of an elliptical drop to a sphere of equivalent volume vs the aspect ratio of the elliptical drop.

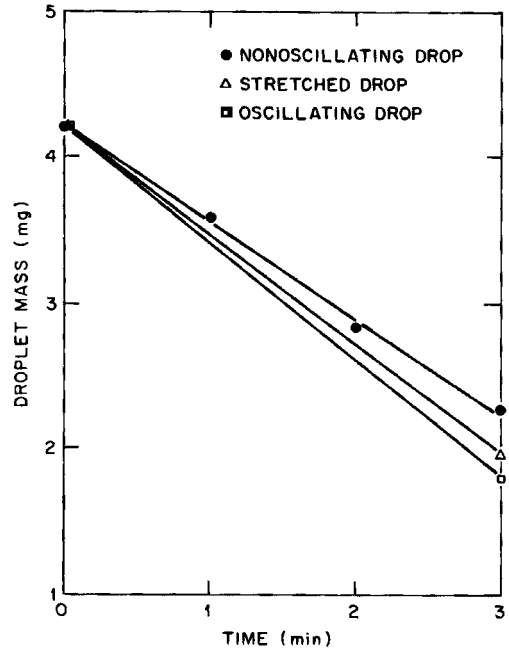

Fig. 7. Decrease in droplet mass vs time.

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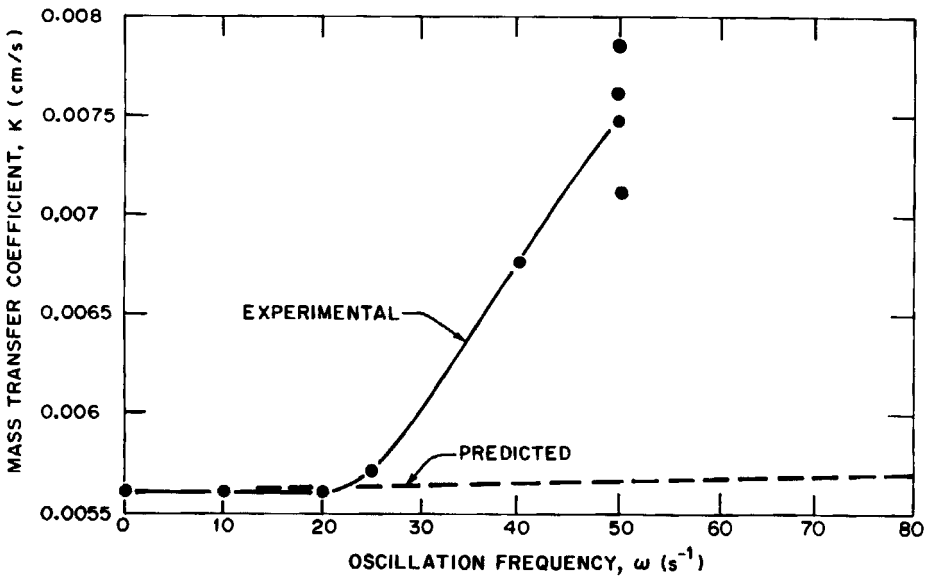

Fig. 8. Mass transfer coefficient vs oscillation frequency for 0.3-cm water drop fluidized in 2-ethylhexanol.

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on continuous phase properties for the baseline case was .0055 cm/s. The data shown are for initial rate. Previous experiments [Clinton (1972)] indicate that surfactant accumulation does not affect the slope for the first 6 to 10 minutes. To remain conservative we selected the first three minutes of the process as our free-circulation period. This was confirmed by our own duplication of selected experiments done by Clinton. Forced droplet oscillations yield a 35% increase in mass transfer coefficient. This is a significant increase in mass transfer for a continuous phase controlled system with no contamination of the interface. Additionally, it should be noted that the droplet is forced to oscillate well below its natural frequency of 100 Hz. Based on the observed improvement in the continuous phase, the dispersed phase controlled case should show significant enhancement. Finally, it is worth mentioning that oscillation of the droplets disrupts the accumulation of surfactant and promotes internal circulation of the droplet where it would normally be noncirculating.

The early results, shown in Fig. 7, were followed by an investigation of the effect of frequency on mass transfer coefficient. Shown in Fig. 8 is a plot of mass transfer coefficient (averaged over area and time) vs frequency. There is very little enhancement over the base case at low frequencies, 10-30 Hz, but a 35% increase

occurs at 50 Hz which is the limit of our equipment. At the bottom of the figure is a plot of the theory of Angelo and Lightfoot based on a time averaged surface area. The theory does not match the data because the theory assumes that surface stretch is the basic cause for enhancement and as is shown in Fig. 3, the time averaged surface area changes only slightly and cannot account for the observed increase in mass transfer. Based on experimental observation, it appears that the mass transfer model must take into account the hydrodynamic changes due to droplet oscillation. This is the subject of forthcoming work.

CONCLUSIONS

Based on the experimental work presented here, we can develop the following conclusions:

1. Interfacial tension is constant over the range of electric field strengths considered for this paper.

2. Oscillations can be forced by a pulsed electric field.

3. Mass transfer can be enhanced using pulsed electric fields. As much as 35% was seen in the continuous phase controlled studies undertaken in this program.

4. Average surface area effects on mass transfer are minimal for the system presented in this paper. Droplet surface area changes are much smaller than the observed change in mass transfer.

5. Extensions to mass transfer theory are needed to account for the hydrodynamics of the oscillating droplet. In particular, the equation of continuity should be reworked using knowledge of droplet oscillations.

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NOMENCLATURE

n = mode of oscillation, integer

R = drop radius, cm

C = concentration, moles/cm^3

v = velocity, cm/s

$\mathcal{D}$ = diffusivity, cm^2/s

K = mass transfer coefficient

t = time, s

t_o = time period for one oscillation, s

S(t) = time dependent surface area, cm^2

S_o = surface area for sphere, cm^2

$\overline{E}$, E_n = electric field, kV/cm

Q = charge

a, b, c, d = constants, value depends on equation used

Sh = Sherwood number
Gr = Grashoff number
Sc = Schmidt number
Re = Reynolds number
} Dimensionless quantities

t^* = dimensionless time, t/t_o

Greek

ω = oscillation frequency, s^{-1}

γ = interfacial tension, dynes

ρ = density, gm/cm^3

μ = viscosity

τ = characteristic damping time [Eq. (8)], s

ε_o = dielectric constant of a vacuum

κ = electric permittivity

ε = dielectric constant

Subscripts

c = continuous phase

D = dispersed phase

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L = Lamb

SM = Scriven and Miller

RK = Rose and Kintner

x = x direction, Cartesian coordinate

y = y direction, Cartesian coordinate

z = z direction, Cartesian coordinate

Ra = Rayleigh limit

B = Boyadzhiev

REFERENCES

1. Lamb, H., Hydrodynamics, 6th Ed., Cambridge University Press, Reprinted by Dover, New York (1945).

2. Schroeder, R. R. and R. C. Kintner, "Oscillations of Drops Falling in a Liquid Field," AIChE J. 11, 5-8 (January 1965).

3. Loshak, J. and C. H. Byers, "Forced Oscillations of Drops in a Viscous Medium," Chem. Engr. Sci. 28, 149-56 (1973).

4. Rayleigh, Lord, "On the Equilibrium of Liquid Conducting Masses Charged with Electricity," Phil Mag 14, 184 (1882).

5. Clinton, S. D., "Mass Transfer of Water from Single Aqueous Sol Droplets Fluidized in a Partially Miscible Alcohol," ORNL/TM-3771 (July 1972).

6. Scriven, C. A. and L. E. Miller, "Oscillations of a Droplet Immersed in Another Fluid," J. Fluid Mech. 32, 417 (1968).

7. Loshak, J., "Forced Oscillation of Drops in a Viscous Medium," M.S. Thesis, University of Rochester, Rochester, N.Y., 1969.

8. Ramabhadran, T. E., "Inference of Dynamic Interfacial Properties from a Study of an Oscillating Drop and Bubble," Ph.D. Dissertation, University of Rochester, Rochester, N.Y., 1972.

9. Rose, P. M. and R. C. Kintner, "Mass Transfer from Large Oscillating Drops," AIChE J. 12, 530-534 (May 1966).

10. Angelo, J. B., E. N. Lightfoot, and D. W. Howard, "Generalization of the Penetration Theory for Surface Stretch: Application to Forming and Oscillating Drops," AIChE J. 12, 751-760 (July 1966).

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11. Boyadzhiev, L., D. Elenkov, and G. Kyuchukov, "On Liquid-Liquid Mass Transfer Inside Drops in a Turbulent Flow Field," Can. J. Chem. Engr. 47, 42-44 (1969).

12. Chalabreysse, M. C., R. Neige, J. Aubert, and G. Folcher, Kinetics Measurements in Liquid-Liquid Exchange Applied to Isotopic Separation," Sep. Sci. Technol. 15(3), 557-566 (1980).

13. Martin, L., P. Vignet, C. Fombarlet, and F. Lancelot, "Electrical Field Contactor for Solvent Extraction," 1983.

14. Stratton, J. A., Electromagnetic Theory, McGraw-Hill, New York, 1941.

15. O'Konski, C. T. and F. E. Harris, "Electric Free Energy and the Deformation of Droplets in Electrically Conducting Systems," J. Phys. Chem. 61, 1172 (1957).

16. Wham, R. M. and C. H. Byers, "Mass Transfer from Single Drops in Pulsed Electric Fields," presented at the AIChE Meeting, San Francisco, California, Nov. 1984.

17. Scott, T. C., "Visualization of Flow Fields and Interfacial Phenomena in Liquid-Liquid Solvent Extraction," Sep. Sci. Technol. (this issue).

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