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Final Report

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Effect of Electric Fields on Mass Transfer to Droplets

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Project Summary

During the six year funding period, the effects of a direct and alternating field upon single drop hydrodynamics and mass transfer were evaluated both experimentally and theoretically. Direct current field effects upon drop size, velocity and mass transfer rates were also observed for multiple drops formed in a three stage sieve tray column. Drop size, velocity, and mass transfer rates were measured experimentally and compared to simple models for direct current electric fields. Agreement between theory and experiment was found for drop charge, size, and velocity. Drop mass transfer coefficients were substantially larger than theoretical predictions while extraction efficiencies were moderately higher. Drop distortion and oscillation were observed and are thought to result in the experimentally observed higher values.

For alternating current fields, drop flow streamlines and oscillations were measured and found to compare well with predictions from a solved mathematical model. In addition, equipment was constructed to determine mass transfer rates to oscillating drops. Concentration profiles in still and oscillating drops were measured and qualitatively compared to theoretical predictions.

DISCLAIMER

Effect of Electric Fields on Mass Transfer to Droplets

I. INTRODUCTION

Liquid extraction and gas absorption rates are strong functions of the drop surface area and mixing. To increase surface area and mixing, energy is applied as pulsations to the fluid (in the case of liquid extraction) and pressure in spray nozzles. These methods put significant energy into the bulk phases to increase surface area or mixing. One promising and more efficient method to generate high surface areas and mixing is the use of an alternating electric field to charge, form and manipulate drops. Such a field can generate very small droplets (with high surface/volume ratios) and promote mixing (due to alternating current driven oscillations). The research summarized in this report was undertaken to describe the effects of alternating and direct electric fields upon drops in liquid-liquid systems.

Mass transfer rates to drops of one liquid suspended in another are a function of drop size and surface area, drop velocity, mixing in the external and drop phase, interfacial turbulence due to concentration differences along the drop interface and that effect on interfacial tension (Marangoni effect), drop and continuous phase concentrations, transport (viscosity and diffusivity of both phases), and equilibrium (distribution coefficient of transferring solute). A number of these factors can be changed by the imposition of an electric field upon the forming and translating drop.

For the application of a direct current field upon a forming, electrically conductive droplet connected to the field source, the drop will acquire a charge which will produce a force to reduce the drop size relative to no field. The charged drop falling in the direction of gravity will fall at a higher terminal velocity. These effects have been observed and reported in the literature (Vu and Carleson, 1986; Carleson and Berg, 1983; Harker and Amadzadeh, 1974). In some cases the increase in terminal velocity will result in shortened drop residence times and resulting lower absorption efficiencies (for gas absorbing in a falling charged drop; Carleson and Berg, 1983). In some cases (i.e. liquid-liquid systems), the extraction efficiencies will increase perhaps due to drop oscillation or increases in surface area (Bailes, 1981). Electrostatic theory and force balances have been used to determine the effects of the direct current field upon drop size and velocity (Vu and Carleson, 1986; Carleson and Berg, 1983; Takamatsu, et al., 1981).

Incorporating these effects, Vu and Carleson (1986) showed that the drop phase mass transfer coefficient should increase 8% and 35% for the continuous phase. Experimental measurements for dispersed phase control indicate an increase of around 25% (Bailes, 1981), significantly higher than predicted. Corresponding predicted increases in the extraction efficiency are 50% and 90% respectively, for the dispersed and continuous phases. These are comparable to observed effects of between 25% and 120% (Harker and Amadzadeh, 1974; Bailes, 1977, 1981).

To further enhance mass transfer, an alternating electric field can be superimposed on the direct field (Scott and Byers, 1989). The drop charge can interact with the alternating electric field to produce drop oscillation. If the oscillation frequency is near the resonant frequency of the drop, the drop amplitude can be large to result in significant circulation inside and outside the drop. This circulation can enhance the mass transfer rates.

II. RESEARCH RESULTS

The research results summarized in this report are presented in three parts. Part A concerns direct current electric field effects upon single drops. Part B concerns direct current effects on multiple drops. Part C concerns alternating current effects on single drops. For more details concerning the research, the reader is directed to the references cited.

A. Direct Current Effects on Single Drops

Figure 1 depicts the column used to study the effect of a D.C. field on forming and falling single drops. Previous work was performed with this column on the measurement of drop charge and velocity (Vu and Carleson, 1986). In the DOE supported work, drop mass transfer rates were evaluated. Experiments were conducted with water drops in toluene (transferring solute of methanol), furfural drops in heptane (transferring solute furfural) under situations where the dispersed or continuous phase resistance was controlling. Drop mass transfer during free fall was separated from the end effects (transfer during formation or coalescence) by experiments with different free fall lengths and extrapolation of data to zero free fall.

Drop volume was reduced in proportion to the square of the electric field strength, in accordance with simple models (Vu and Carleson, 1986; Yang and Carleson, 1988). Anomalies as much as 20% can occur due to non-linearities in the field strength and drop deformation for low interfacial tension systems (Yang and Carleson, 1988).

The effect of the electric field upon drop velocity appears to follow the predictions from a simple force balance (variation of terminal velocity with field strength). Higher drag coefficients were measured than predicted perhaps due to drop oscillation as the drops fell (Yang and Carleson, 1988).

The single drop extraction efficiencies increased 15% for the toluene water system (at a field strength between 1 and 2 kv/cm) and 51% for the heptane furfural system (at a field strength of around 0.5 kv/cm). The corresponding increase in drop mass transfer coefficient was 80% and 100% respectively for the toluene-water and the heptane-furfural systems. These increases are higher (by 80 - 220%) than predicted ones base upon empirical equations in the literature possibly due to oscillations and drop deformation (observed in the experimental studies).

The effect of drop interfacial turbulence was evaluated by evaluating mass transfer from water drops containing methyl isobutyl ketone falling in air in the column arrangement shown in Figure 1. A VCR camera was used to observe drop interfacial turbulence. The transfer of methyl isobutyl ketone results in a surface tension gradient along the surface of the drop. This gradient results in surface flows and, consequently, interfacial turbulence, which was observed in the photographs of the drop. By imparting a surface charge to the drop, by its formation from a charged needle in the electric field, one can stabilize the surface. The drop charge distribution resists fluid motion. Consequently, it was observed that drop interfacial turbulence and desorption (of methyl isobutyl ketone) decreased. The observations were comparable to those when a surfactant (which also stabilizes the surface) was added to the water. These results agreed with predictions (Carleson and Fuller, 1987).

B. Direct Current Effects on Multiple Drops

Figure 2 depicts the apparatus used for the multiple drops studies. In this apparatus sieve trays were used to coalesce and form the drops. The upper and lower plates were connected to the electric field source while the middle plate was grounded. Drops of water were used to extract methanol from toluene.

Drop volume decreased linearly with the square of the applied field, in accordance with simple models (Yang and Carleson, 1988). However, for multiple drops forming on sieve trays the drop volume reduction with the square of the field variation was higher than for drops forming from single nozzles. This may be due to the effect of the field upon drops forming at orifices rather than at a single needle to result in higher drop charges and consequently smaller drops.

Drop velocities in the sieve tray column varied approximately linearly with field strength. Increasing the field strength resulted in an increase in velocity from 10 cm/sec at no field to 23 cm/sec at 2 kv/cm. These changes are comparable but slightly higher than those observed for single drops.

The extraction efficiency increased about 25% (second stage) at a field of 2 kv/cm). Under the same conditions, the column throughput increased about 26%. The resulting increase in mass transfer coefficient was about 70%.

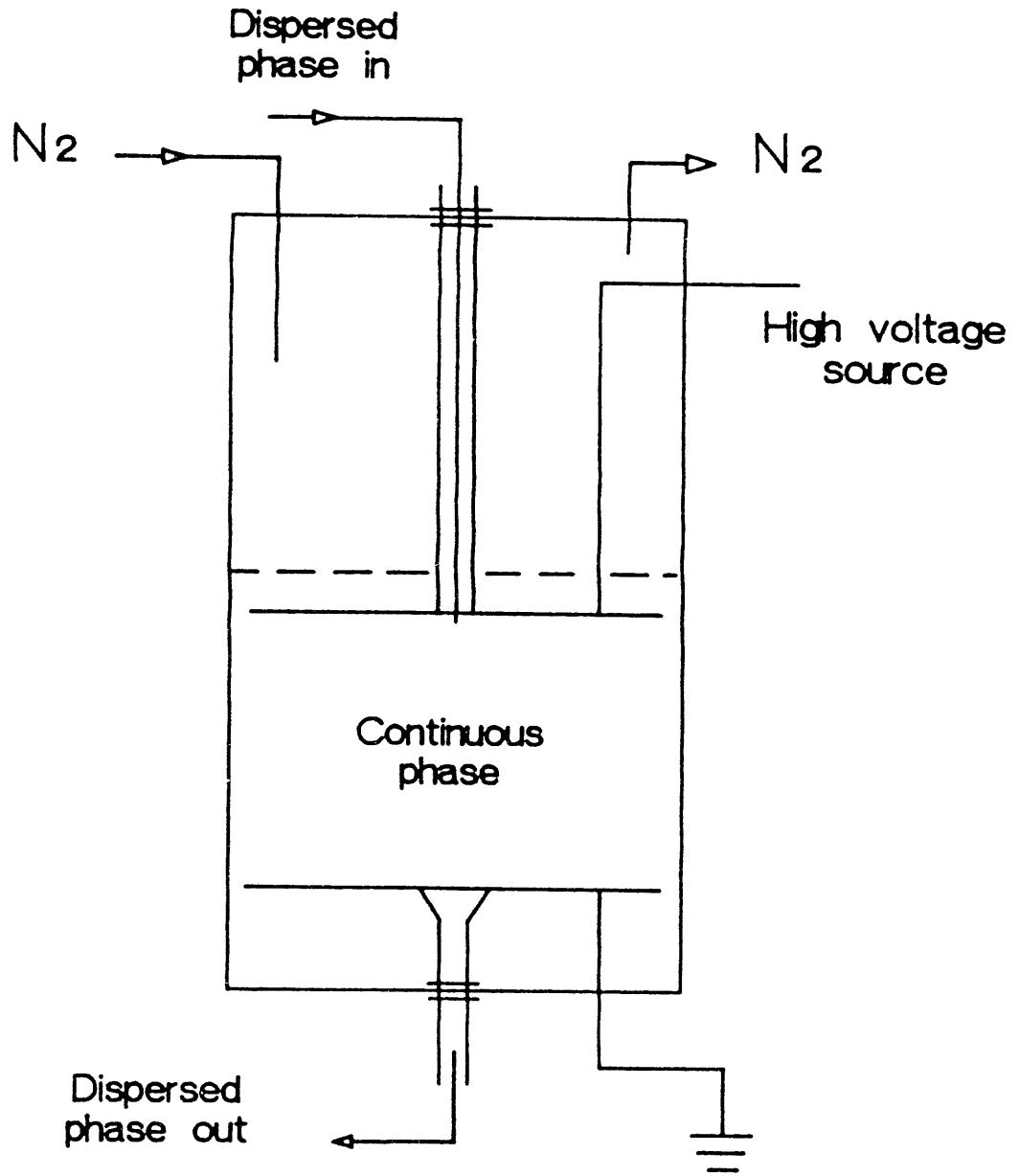

Dispersed
phase in
N2
N2
High voltage
source
Continuous
phase
Dispersed
phase out

Figure 1

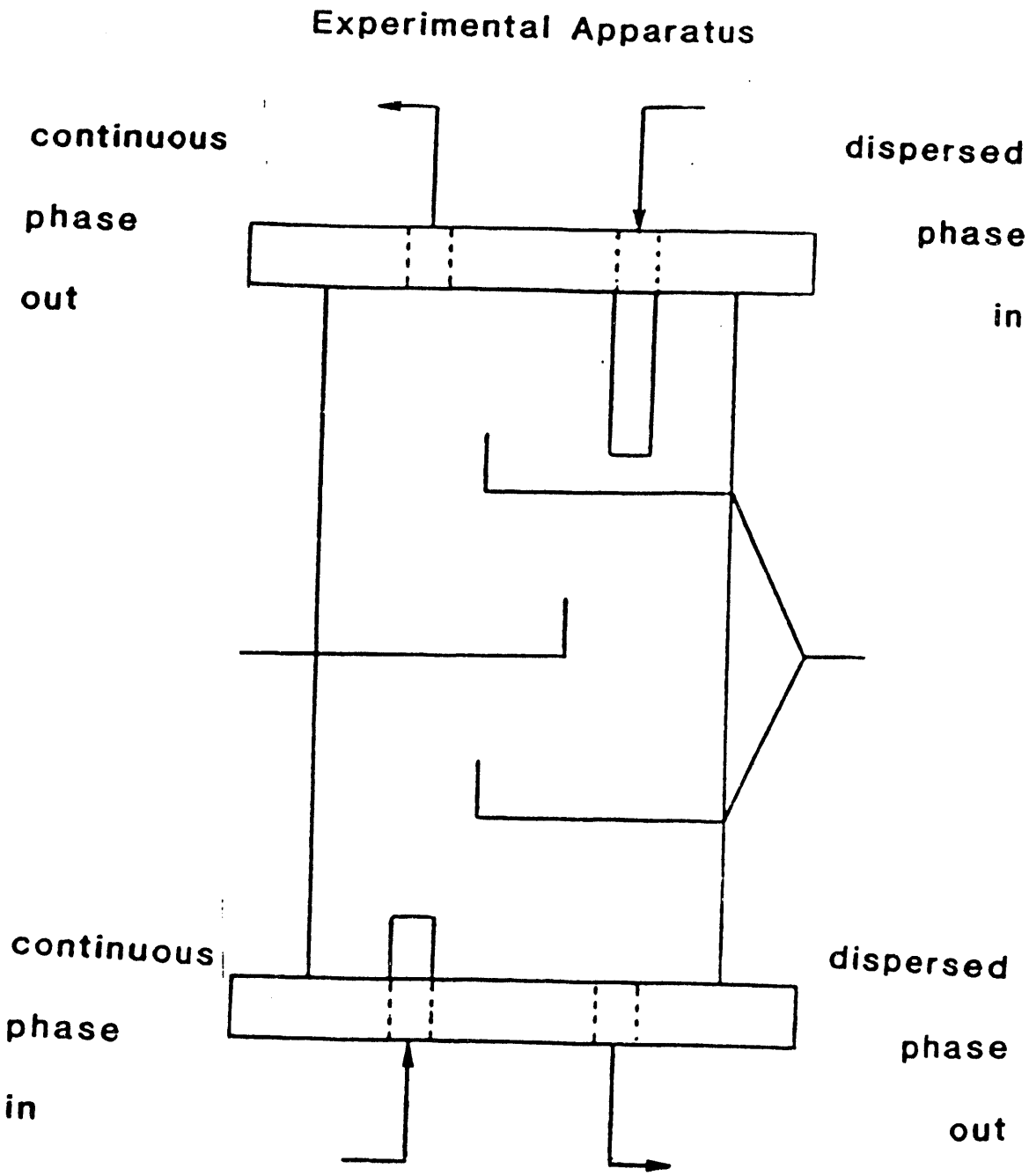

Experimental Apparatus
continuous
phase
out
dispersed
phase
in
continuous
phase
in
dispersed
phase
out

Figure 2

C. Alternating Current Effects upon Single Drops

The equations for drop hydrodynamics were solved for a single charged drop in the presence of an alternating electric field. The simple case of the linear oscillation model (assumed small amplitude deformations to allow neglection of nonlinear terms in Navier Stokes equation) was solved (Yang and Carleson, 1991). In this model drop boundary conditions were assumed for a spherical drop. The linear model predicts the effect of the electric field to produce oscillating linear translations (first mode motion) and prolate-oblate oscillation (second mode motion). The effects of viscosity and density on the drop resonant frequency were investigated. It was found that the resonant frequency of viscous fluids was always smaller than the free oscillation frequency (frequency in absence of an applied field) of the drop. The resonant frequency of second mode oscillations is a function of the densities, viscosities, field strength, interfacial tension, drop size, and permittivity of the continuous phase. Experimental observations of the frequency response curve and drop streamlines were found to agree with the theoretical predictions (Yang and Carleson, 1991) and compared well to other results (Scott and Byers, 1989).

This work was extended to drops undergoing secondary oscillations (oscillations resulting from primary drop distortion and re-distribution of charge) (Yang and Carleson, 1994a). These secondary oscillations correspond to four modes (drop translation, drop prolate-oblate motion, tripole and quadruple motion). Two frequencies are predicted in each mode. The most significant secondary oscillation has the same mode and frequency as the primary second mode oscillation. The resonant frequency of the second mode oscillation decreases with net electric charge density as well as with electric field strength.

Experimental work with an apparatus similar to that depicted in Figure 1 indicates that the first mode oscillation amplitude is proportional to the product of the charge density and field strength. Second mode oscillation amplitude is proportional to the square of the applied field strength (Yang and Carleson, 1994b). As observed and predicted, the amplitude of drop oscillation varies with frequency. A peak occurs at the drop resonant frequency.

An experimental systems (Figure 3) has been fabricated to measure the mass transfer to oscillating drops (Carleson, et al., 1993). In this system a drop is held stationary by an applied acoustic field. The mass transfer of a dye to or from the drop to the continuous phase is followed by using an argon laser to excite the dye. The properties of the phases have to be adjusted to result in a match of the indices of refraction and a moderate density difference. One phase (usually the drop phase) consisted of a mixture of freon and octanol. The other phase (usually the continuous phase) consisted of sugar in water. The intensity of the fluorescence produced is proportional to the dye concentration. A video imaging system was installed to photograph and digitize images of the drop. Images were taken of a stationary drop undergoing dye diffusion into the still continuous phase.

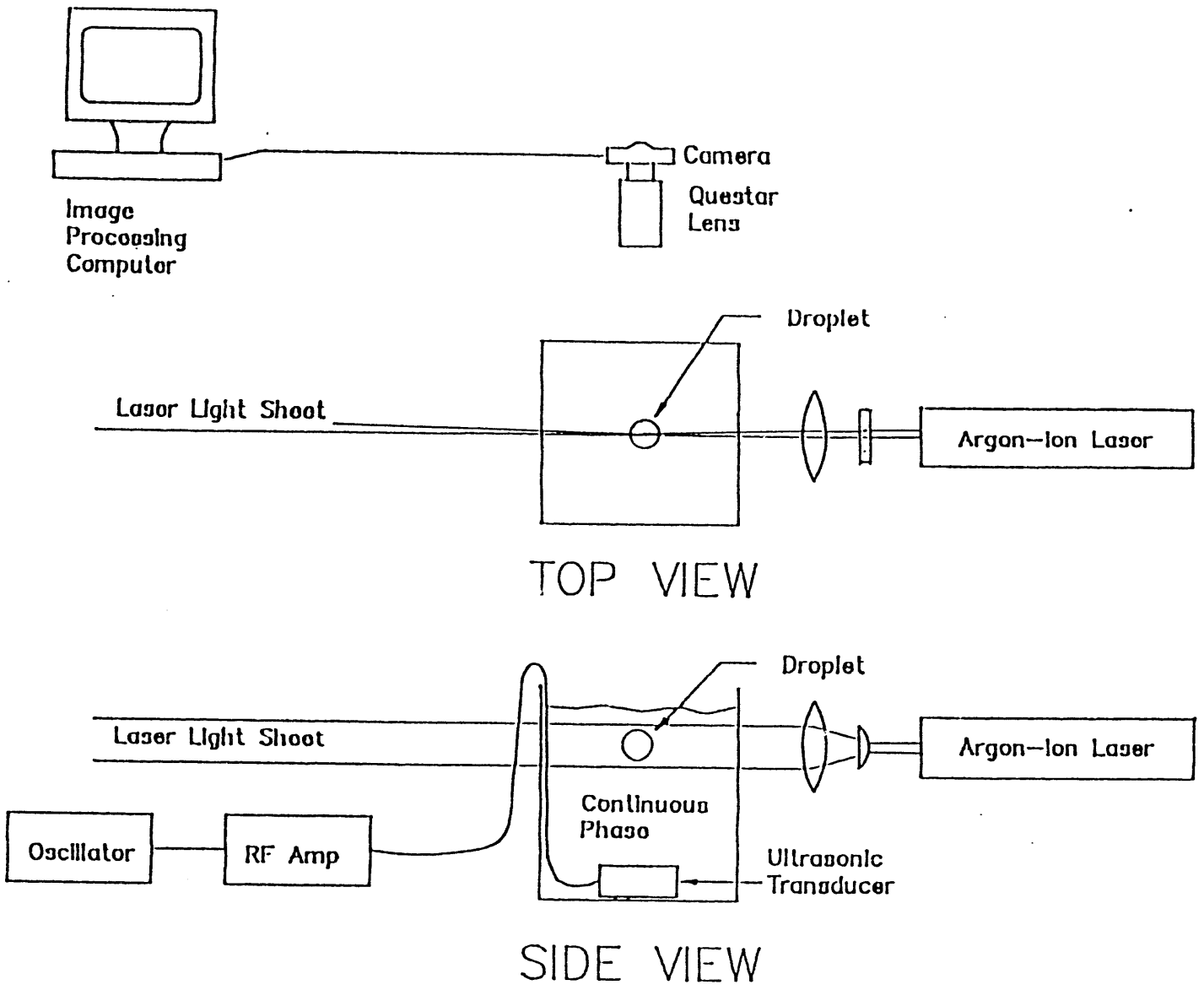
Concentration profiles obtained for a still drop undergoing molecular diffusion agreed qualitatively with theoretical profiles. Digitized images of concentration profiles of an oscillating drop (oscillations acoustically produced) were also obtained. Drop concentration profiles resembled the predicted drop velocity streamlines from the earlier work (Yang and Carleson, 1991).

III. Discussion of Results and Conclusions

A. Single and Multiple Drops in Direct Current Fields

Simple force balances including electrostatics can be used to approximate drop charge, size, and velocity. Deviations from the predictions may be due to non-linear fields in actual equipment configurations and different charging configurations (nozzle versus orifice plates).

Increases in mass transfer rates due to the electric field on single and multiple drops are higher than those predicted based upon empirical models in the literature. The differences may be due to enhanced drop



Image
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Argon-Ion Laser
TOP VIEW
Droplet
Laser Light Shoot
Argon-Ion Laser
Continuous
Phase
Oscillator
RF Amp
Ultrasonic
Transducer
SIDE VIEW

Figure 3

oscillation and deformation under electric field conditions. These effects are not included in the theoretical models used. Increases in mass transfer coefficients are substantial (80 - 100%) whereas increases in extraction efficiency are moderate (30 - 50%) perhaps due to increases in drop velocity to reduce mass transfer enhancements due to higher coefficients.

The application of an electrical charge to a drop surface may reduce the flow of liquid along the surface due to Marangoni effects (interfacial turbulence). The effect of the charge on mass transfer rates is similar to the effect of an adsorbed surfactant.

B. Single Drops in Alternating Current Fields

An applied alternating electric field on a charged drop can cause up and down oscillations as well as shape (prolate-oblate) oscillations. The amplitude of these oscillations are proportional to the square of the applied field strength and vary with the applied frequency. The frequency of the oscillations generally follows that of the applied alternating field. If drops are caused to oscillate at frequencies near their resonant values, the resulting amplitude of the oscillations reaches a maximum. Mass transfer enhancements would be expected to also reach a maximum at this resonant frequency.

Theoretical predictions of the effect of the field strength and drop physical properties on the resonant frequency agree with observations (Yang and Carleson, 1991; Carleson, et al., 1993). Recent work indicates that a laser actuated dye tracer can be used to follow the development of concentration profiles in an oscillating drop (Carleson, et al., 1993).

IV. References (Presented Papers, Publications, and others)

A. Presented papers based upon supported research:

Budwig, R., J. Martinez, T. Carleson, "Hydrodynamic and Mass Transfer Characteristics of Acoustically Levitated and Oscillated Droplets", American Physical Society 1992 Annual Meeting, Washington, D.C.

Carleson, T.E., S. Katamneni, R. Budwig, "Visualization of Mass Transfer to a Charged Drop in an Oscillating Electric Field Such as That Produced by an Alternating Electric Field", AIChE 1993 Annual Meeting in St. Louis.

Yang, W. and T.E. Carleson, "Secondary Oscillations of a Charged Drop in a Alternating Electric Field", 1991 Annual AIChE Meeting in Los Angeles.

Yang, W. and T.E. Carleson, "Linear Oscillations of a Drop in a Uniform Alternating Electric Field", 1990 Annual AIChE Meeting in Chicago.

Yang, W. and T.E. Carleson, "Several Effects of Electric Fields upon Liquid Extraction", 1987 Annual Meeting of the Industrial Applications Society of IEEE in Atlanta.

B. Publications based upon supported research:

Yang, W. and T.E. Carleson, "Linear Oscillations of a Drop in Uniform Alternating Electric Fields", AIChE J., November, 1601(1991).

Yang, W., and T.E. Carleson, "Several Effects of Electric Fields on Liquid Extraction", IEEE Transactions on Industry Applications, Vol. 26, No. 2, 366(1990).

Carleson, T.E. and E. Fuller, "The Effect of Electric Charge upon Mass Transfer from Drops Exhibiting Interfacial Turbulence", Chemical Engineering Communications, Vol. 57, No. 1-6, 277(1987).

Vu, N. and T.E. Carleson, "Electric Field Effects on Drop Size and Terminal Velocity in Liquid-liquid Systems", AIChE J., vol. 32, No. 10, 1739(1986).

C. Publications in preparation for submission

Yang, W. and T.E. Carleson, "Secondary Oscillation of a Charged Drop in an Alternating Electric Field", prepared for submission to AICHE Journal, 1994a.

Yang, W. and T.E. Carleson, "An Experimental Study on Drop Oscillations in Alternating Electric Fields", prepared for submission to AICHE Journal, 1994b.

D. Other references

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Carleson, T.E., and J. Berg, "The Effect of Electric Fields on the Absorption of Pure Sulfur Dioxide by Water Drops", Chem. Eng. Science, Vol. 38, No. 6, 871(1983).

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Scott, T.C., and C.H. Byers, "A Model for Mass Transfer in Oscillating Circulating Liquid Drops", Chem. Eng. Commun., Vol. 77, 67(1989).

Takamatsu, T., M. Yamaguchi, T. Katayama, "Formation of Single Charged Drops in Liquid Media under a Uniform Electric Field", J. Chem. Eng. Japan, Vol. 15, No. 5, 349(1982).

V. Graduate degrees awarded or in progress to supported students:

Jon Martinez, M.S. in Mechanical Engineering 1994, "Visualization of Flow and Concentration Fields in Acoustically Levitated and Oscillated Droplets".

Sudhakar Katamneni, M.S. in Chemical Engineering 1994, "Construction and Evaluation of an Apparatus for Visualization and Measurement of Mass Transfer to a Suspended Droplet in an Oscillating Field".

Wenrui Yang, Ph.D. in Chemical Engineering 1991, "Oscillations of a Charged Drop in an Alternating Electric Field".

Ernest Fuller, M.S. in Chemical Engineering 1986, "Electric Field Enhancement of Liquid Extraction".

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