Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

ELECTRODISPERSION OF FINE AQUEOUS DROPLETS IN SUPERCRITICAL CARBON DIOXIDE

K. D. Heath[a]; H. D. Cochran[b]
[a] Chemical Technology Division, Oak Ridge National Laboratory*, Oak Ridge, Tennessee, U.S.A. [b] Department of Chemical Engineering University of Tennessee, Knoxville, Tennessee, U.S.A.

To cite this Article Heath, K. D. and Cochran, H. D.(2001) 'ELECTRODISPERSION OF FINE AQUEOUS DROPLETS IN SUPERCRITICAL CARBON DIOXIDE', Separation Science and Technology, 36: 5, 1211 — 1224
To link to this Article: DOI: 10.1081/SS-100103646
URL: http://dx.doi.org/10.1081/SS-100103646

SEPARATION SCIENCE AND TECHNOLOGY, 36(5&6), 1211–1224 (2001)

ELECTRODISPERSION OF FINE AQUEOUS DROPLETS IN SUPERCRITICAL CARBON DIOXIDE

K. D. Heath[1] and H. D. Cochran[2]

[1]Chemical Technology Division, Oak Ridge National
Laboratory,* Oak Ridge, Tennessee 37831-6224
[2]Department of Chemical Engineering University of
Tennessee, Knoxville, Tennessee 37996-2200

ABSTRACT

This first study of electrospraying of a liquid into a dense, supercritical fluid has demonstrated a new method for dispersing aqueous liquid into supercritical carbon dioxide (SC-CO_2) using pulsed high-voltage electric fields. The electrodispersion cell (EDC) was equipped with a laser light scattering system to determine the dependence of mean droplet size on operating variables such as field strength, pulse frequency, and liquid flow rate. Over a range of operating conditions, electrospraying produced micron-size aqueous dispersions with high interfacial area, and the micron-size dispersions were very efficiently coalesced by the wire-mesh electrode so that entrainment of liquid in the CO_2 flowing from the cell was negligible. In related work elsewhere, results have been presented on the effects of temperature and pressure of the SC-CO_2 on the droplet size and on the extraction of ethanol from an aqueous so-

---

* Managed by Lockhead Martin Energy Research Corp. for the U.S. Department of Energy under contract DE-AC05-96OR22464.

lution into SC-CO_2 using the EDC. This new method of contacting an aqueous phase with SC-CO_2 may find initial applications in cases where small quantities of a valuable component must be separated from water.

INTRODUCTION

For some time governments and chemical process industries have been pursuing alternate substances to replace hazardous organic solvents which are currently used in most industrial synthesis and separation operations. SC-CO_2 has been identified as a leading choice for solvent substitution (1). Technologies have been developed for many separation and reaction processes using SC-CO_2 as a solvent, and several large commercial applications are in operation throughout the U.S., Europe, and Japan. However, because of the low solubility of water in SC-CO_2 and poor mass transfer performance of conventional contactors between water and SC-CO_2 phases, many potential commercial processes involving aqueous solutions have so far been beyond the range of commercial application of SC-CO_2.

Techniques for producing aqueous dispersions in SC-CO_2 have been sought for over a decade, and only recently (2-7) have a few successful surfactants been found that result in reverse micelles in SC-CO_2 with small quantities of water in their cores. These successes with surfactants have attracted a great deal of attention because of the widespread importance of their potential applications. To date, the highest water-to-surfactant molar ratio attained has been 32, which allows a maximum of 2 wt. % water to be soluble in CO_2 (2). This value is 10 times the amount of water that is soluble in pure CO_2. Although much progress has been achieved, the known surfactants (perfluoro polymers) are very costly, both to manufacture and to separate from the final product when finished.

An alternate approach may exist to produce dispersions of submicron-sized aqueous droplets in SC-CO_2 without the use of surfactants. Over the past fifteen years, researchers have explored drop and bubble formation behavior in electric fields from fundamental and applied perspectives. Techniques have been developed for producing aqueous dispersions in non-conducting organic solvents using intense electric fields; these developments have resulted in patents for an emulsion phase contactor to be used in separations (8, 9) and for an electric dispersion reactor (10-12). A number of applications have been explored (13-21), and successful technology transfer has resulted in commercialization. To date the solvents in which the submicron-sized dispersions have been produced have included many organic liquids but never dense supercritical fluids. The goal of this work was to test feasibility of this technique, which would broaden the range of separation and reaction processes in which SC-CO_2 is applicable.

In the following section, prior work on electrospraying is briefly reviewed. Then, the experimental approach of the present study is described. This is followed by results and discussion of the electrohydrodynamic performance of the electrodispersion cell (EDC) for fine dispersion of aqueous liquids in SC-CO_2. Finally, some brief concluding remarks are offered. In related work elsewhere (22, 23), results are presented on the effects of temperature and pressure of the SC-CO_2 on the droplet size and on the extraction of ethanol from an aqueous solution into SC-CO_2 using the EDC.

BACKGROUND

Electrospraying is a process that relies on electrical forces to break a liquid into fine, charged droplets. One of the simplest ways of implementing such a process is to feed a liquid with sufficient electric conductivity through a grounded metal capillary a few centimeters away from a conducting mesh screen, which is charged to a sufficiently high electric potential. Under the influence of the electric field, the liquid may emerge in different flow modes: dripping, microdripping, cone-jet, spindle, simple jet, and ramified jet. In the cone-jet mode, the liquid forms a cone-shaped interface at the outlet of the capillary, through the apex of which a fine thread of liquid is ejected. The liquid thread breaks up farther downstream to disperse into a spray of droplets, which often shows a remarkably narrow size distribution (24-27). The reason for the monodispersity of the cone-jet electrospray is that axisymmetric surface wave instabilities dominate the liquid breakup, which results in a constant ratio between the primary droplet diameter and the thread diameter (28). For liquids with low viscosity, the ratio has been measured at about 1.89 (24, 26-29), consistent with the prediction of Rayleigh's capillary instability theory (29). Electrospraying is capable of producing monodisperse droplets in the size range from 5 nm up to 100 μm when operating in the cone-jet mode (24-27, 30-34).

When the mode of electrospraying extends past the cone-jet mode into the spindle, simple jet, or ramified jet mode, monodispersity is no longer observed as instabilities at the liquid apex cause a large range of different droplet sizes to be emitted. The applied voltage is a key parameter in establishing the cone-jet mode; various modes of spraying can be observed depending on the voltage applied (35-37). However, droplet size has been reported to be virtually independent of the applied voltage for liquids of relatively high electrical conductivity (38, 39). Tang and Gomez (28) observed that at low flow rates, the applied voltage seems to have a modest effect, and at larger flow rates, the droplet size decreases almost linearly with the increase of the applied voltage. This effect appears to be more pronounced for lower conductivity solutions. Within the range of applied voltage that ensures a stable cone-jet, as much as 50 % reduction in size was observed for a fixed flow rate of the lower conductivity solution. If the mode of electrospray is beyond the cone-jet mode, polydispersity is observed, and an increase in droplet size can be seen upon increasing the field strength (35).

The frequency of a pulsed electric field affects the size of the droplets (40-42) as well. The most interesting behavior is found to occur when the electric field pulse frequency is in the vicinity of the natural oscillation frequency of the droplet (43). Near this natural frequency, a significant increase in field strength is required for droplet rupture (40); hence, this implies that this is the condition of maximum stability for the droplet. In addition, stability of the droplet drastically decreases on either side of the natural frequency.

The flow rate is the dominant factor controlling the droplet size of the electrospray in the cone-jet mode; droplet sizes ranging more than two orders of magnitude can be easily generated from the cone-jet electrospray solely by varying the liquid flow rate (28). Various researchers (26, 28, 31, 32, 37, 38, 44) have studied the droplet size dependence on liquid flow rate. When the electrospray is operated at the onset voltage conditions, defined as the minimum voltage required for a cone-jet spray, the droplet size increases monotonically with the liquid flow rate; and the dependence can be fitted by a power law (25, 34, 45). If the mode of electrospray extends beyond the cone-jet mode, polydispersity dominates the dispersion; however, similar trends of increasing mean droplet size with flow rate are still observed (35).

Stable electrosprays in the cone-jet mode have been investigated for several decades now. The effects of electrospraying in liquids has been well documented (20, 22, 40-42) as well as electrospraying into gases (24, 28, 39, 46, 47). No literature has been found on electrospraying into supercritical fluids, which is the focus of this work; in supercritical fluids the density can be controlled to achieved properties ranging from those of liquids to those of gases.

EXPERIMENTAL APPROACH

The central component in the experimental apparatus is a high-pressure electrodispersion cell (EDC). The pressure cell obtained from Pressure Products Industries, Inc. is capable of operating at pressures up to 34.47 MPa and temperatures from -30 to 175 °C. An EDC was fabricated from this pressure cell by installing fluid supply and withdrawal lines, electrical feedthroughs, and internal electrodes (see Fig. 1). The inside chamber is in the shape of a cylinder with a diameter of 5.08 cm and a height of 17.78 cm.

The electric field was created by a custom-built pulsed dc voltage generator (see Fig. 2). This generator utilizes a Global Specialties 4001 pulse generator and an Electronics Measurements, Inc., SCR dc power source that are supplied to a custom built pulser. The pulser then energizes a 12 V dc automotive high-voltage

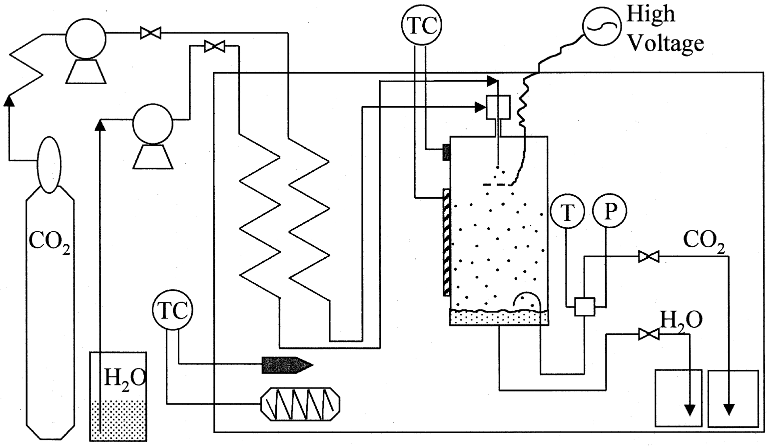

Figure 1. Full schematic of electrodispersion system.

coil (Mallory Promaster). The coil produces the high voltage, which is sent to the system through high-voltage diodes (Collmer Semiconductor, Inc., CS4107X30). The voltage is measured using a high-voltage step-down probe (Farnell HV40B; 1000:1) coupled with a Tektronix Inc., Type 504 oscilloscope to accurately determine the optimum voltages and frequencies. The oscilloscope was calibrated to

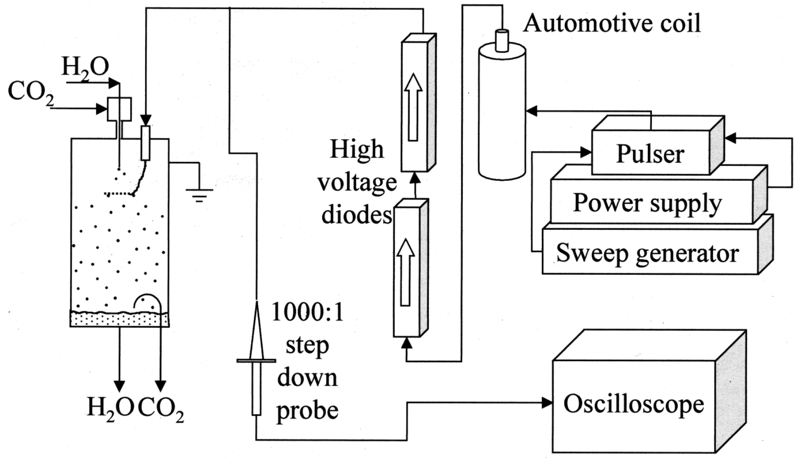

Figure 2. Electrical schematic for providing pulsed, high-voltage electric fields.

within 2%, less than the width of the trace line. This system is capable of pulses up to 30 kV at 0-10 kHz.

Water flow is established with a SSI 222C high-pressure liquid chromatograph (HPLC) pump, and the level of accumulated water in the cell is maintained by adjusting the water let down valve (Autoclave Eng., micrometering valve 30VRMM4812). The CO_2 flow is established with a Teledyne Sprague Engineering Model S-86-JN-60 booster compressor, and the pressure of the system is maintained by adjusting the CO_2 let down valve (Autoclave Eng., micrometering valve 30VRMM4812). For the recovery of extract, the CO_2 outlet stream is sent to a series of U-tube condensers (1.27 cm OD and 0.635 cm ID), which contain glass wool for increased effectiveness. A Fisher Porter gas flow meter is used for online flow rate indication, and a wet test meter (Singer American Meter Division Model 802, serial no. P-843) is used for precise volumetric measurement of the CO_2 outlet stream. A bypass valve is located before the condensers so that the system can reach steady state before the condensers are operated. System operation proved to be simple and stable so that controlled electrodispersion experiments could be performed successfully.

The EDC was operated with an aqueous medium and CO_2 flowing in a cocurrent mode. The carbon dioxide and water entered the cell through ports in the top with no pre-mixing. There are two outlet ports, one for draining the accumulated aqueous solution, the second, an extended crooked sheltering tube, for CO_2 removal. The pressure of the system was measured with an analog gauge supplied by Sprague Engineering Corp. with an accuracy of ± 0.34 MPa. An air bath with a high-intensity circulating fan was used to control temperature and minimize electrical hazards. Since the cell consists of an almost solid block of stainless steel with an approximate weight of 34 kg, a separate electrical resistance heater (Omegalux, 505/5-P) and digital temperature controller (Omega; custom built) have been adapted to the cell itself to assist in temperature control. Temperature control was maintained to ± 0.1 °C.

To characterize mean droplet size, a laser light scattering system was used. A 10-mW He-Ne laser was positioned to enter the cell through a contractible iris to control the beam diameter and to block extraneous light. On exiting the cell, the high intensity of the beam was blocked with a beam stop, and the remaining scattered light was displayed on a screen designed especially for uniformity. The image of the scattered light on the screen was then acquired through a video camera (Pulnix TM-200) and sent to a DigitalVision RT Mono frame grabber for computer analysis. A two dimensional profile was created based on the observed pattern on the screen. A profile of the image is displayed as intensity versus distance from the center of the beam. For analysis, these data were converted to light intensity versus the magnitude of the scattering vector. Monodispersed samples of 10 and 2 μm sizes were used for calibration. The distribution of aqueous droplets in the EDC has been assumed to be Lorentzian, and the droplets have been assumed to be spherical.

RESULTS AND DISCUSSION

The series of frames in Fig. 3 displays the effect of increasing the aqueous flow rate to the EDC while maintaining a constant voltage of -8.8 kV (3.5 kV/cm), pressure (7.58 MPa), temperature (40°C), and frequency (400 Hz). Figure 3a uses a flow rate of 4.0 ml/min; Fig. 3b, 5.0 ml/min; while the last, Fig. 3c, 6.0 ml/min. The dispersion effect remains in all three cases. The only noticeable difference between the three is the extension of the initial stream length. This and later results show that the process is feasible over a range of flow rates at least 1-9 ml/min. The limit of the dispersion is balanced by the flow of the aqueous solution and the electric field, which shatters it. The mode of electrospray for this figure appears to be that of the ramified jet (35), characterized by the liquid thread widening at the apex and its edges emitting many fine jets.

Figure 4 shows the relationship of field strength (2-5 kV/cm) and mean droplet diameter $<d_p>$ for three pulse frequencies (100, 1,000, and 10,000 Hz) while holding the aqueous flow rate constant at 3 ml/min. Here $<d_p>$ increases with increasing field strength. Specifically an increase from 0.9 μm at 2 kV/cm to 1.75 μm at 5 kV/cm was observed. This result appears contrary to prior work, which was preformed in the cone-jet mode (32). However, the result in Fig. 4 and those in the following paragraphs were likely not obtained in the cone-jet mode. Prior work has established that increasing the field strength while in the cone-jet mode causes a decrease in droplet size. However, an electrospray which operates beyond the onset of cone-jet mode (at higher field strength, flow rate, or both) becomes unstable and polydisperse. Polydispersity is believed to have caused the observed increase in $<d_p>$ with increasing field strength seen in Fig. 4. The results in Fig. 4 also show the effect of increasing the pulse frequency. All three pulse frequencies give similar $<d_p>$ at lower field strength with higher frequency giving a larger $<d_p>$ at higher field strength.

Figure 5 displays the effect of increasing pulse frequency (1-10,000 Hz) on $<d_p>$ for two aqueous flow rates (4 and 7 ml/min) while holding the field strength constant at 4 kV/cm. Here it is shown, that at the lower pulse frequency

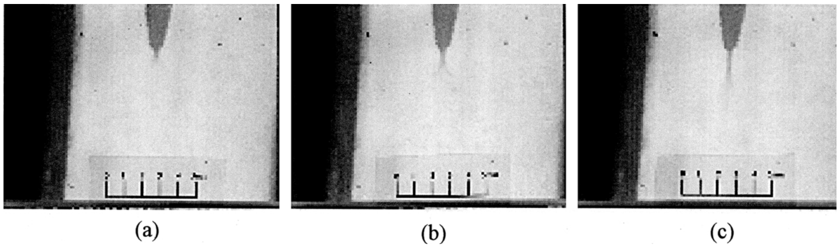
(a) (b) (c)

Figure 3. Series of frames depicting the effect of increasing the aqueous flow rate on a dispersion into supercritical CO_2 at 40°C, 3.5 kV/cm, and 400 Hz. (a) 4.0 ml/min, (b) 5.0 ml/min, (c) 6.0 ml/min. Inserted scale is 5 mm with each division being 1 mm.

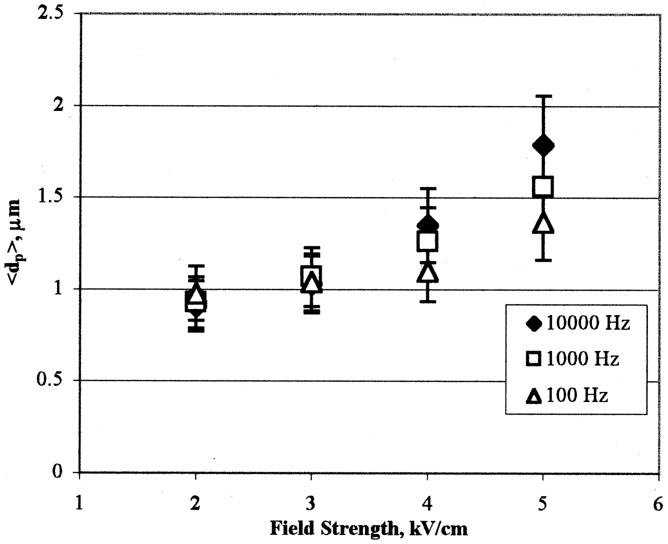

Figure 4. Effect of field strength on $<d_p>$ with distilled H_2O at flow rate of 3 ml/min, 10.34 MPa, and 40°C.

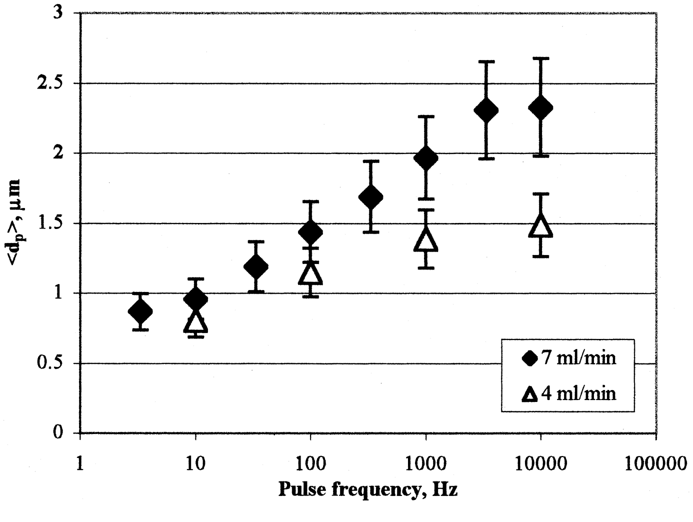

Figure 5. Effect of pulse frequency on $<d_p>$ at 4kV/cm, 10.34 MPa, and 40°C for distilled H_2O.

of 10 Hz, $<d_p>$ is smaller than 1.0 μm for both flow rates. However, as the pulse frequency is increased to 10,000 Hz, the $<d_p>$ for both flow rates increase. The 7 ml/min data set shows a larger dependence with an increase to 2.3 μm for increased pulse frequency, while the 4 ml/min data set has a more modest dependence with a $<d_p>$ of 1.5 μm at the highest pulse frequency.

Figure 6 gives the effect of increasing flow rate (1-9 ml/min) on $<d_p>$ for two pulse frequencies (10 and 1,000 Hz) while holding the field strength constant at 4 kV/cm. At lower aqueous flow rates (1-6 ml/min), $<d_p>$ appears fairly constant for these conditions, but increases once the flow increases. For all flow rates the 10 Hz data set gives smaller $<d_p>$ than the 1,000 Hz data set. The 10 Hz data set shows $<d_p>$ less than 1.0 μm until a flow rate of 8 ml/min where it climbs to 1.5 μm with a maximum of 2 μm at 9 ml/min. For the 1,000 Hz data set the $<d_p>$ stays below 1.5 μm until the flow rate is raised to 8 ml/min where it increases to almost 3 μm with a maximum of 3.5 μm at 9 ml/min. Findings of increased $<d_p>$ with increasing flow rate similar to these have been observed by many researchers (32-38) for electrospraying liquid into gas.

In prior paragraphs the focus of attention has been the characterization of the electrohydrodynamic performance of the EDC by studying the effects of operating variables on $<d_p>$. In subsequent work the focus shifted to using the EDC to perform an extraction of ethanol from a 10 vol. % aqueous ethanol solution using SC-CO_2. The results at a condition of 35°C and 10.34 MPa are presented and

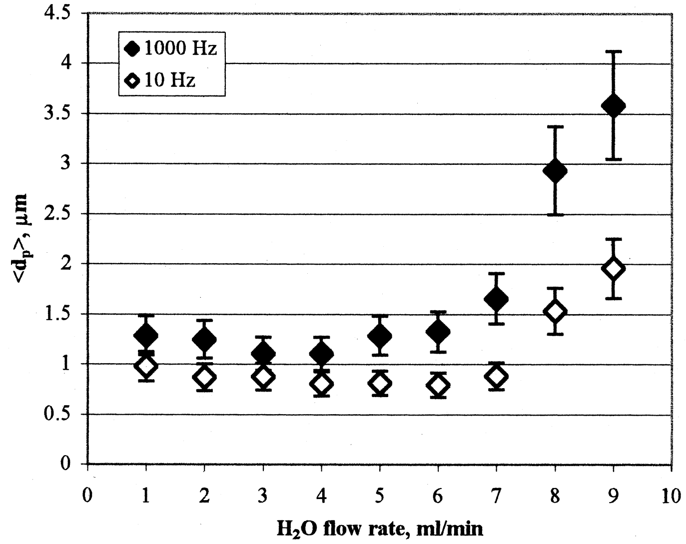

Figure 6. Effect of flow rate on $<d_p>$ at 4kV/cm, 10.34 MPa, and 40°C.

compared to theoretical equilibrium predictions based on flash calculations using the Peng-Robinson (PR) equation of state model. Figure 7 displays the effect of increasing CO_2 flow rate on the separation factor, α, defined as $(y_2/x_2)/(y_1/x_1)$ where x and y denote mole fractions in the liquid and supercritical phases, respectively. Points represent the experimental data and the theoretical predictions are represented as a line. There is an important conclusion that can be drawn from the observation that the separation achieved in the EDC is determined only by equilibrium for all conditions studied up to about 5 mol/hr CO_2 flow rate. Entrainment of micron-size liquid droplets from the EDC is apparently negligible; otherwise the measured extract ethanol concentration would have been lower than the equilibrium concentration. The liquid feed concentration and the liquid effluent concentration were always much lower than the extract concentration. Any entrainment of the liquid would have decreased the measured extract concentration below the equilibrium value. It is generally believed that the micron-size droplets produced by electrospraying are electrically charged. Charged droplets would be strongly attracted to the wire mesh electrode. Apparently, a high degree of droplet coalescence at the wire mesh electrode was achieved in the EDC, resulting in negligible liquid entrainment. Figure 8 gives the rate of extraction of ethanol and water at 10.34 MPa and 35°C. Because of the limited capacity of the condensers used

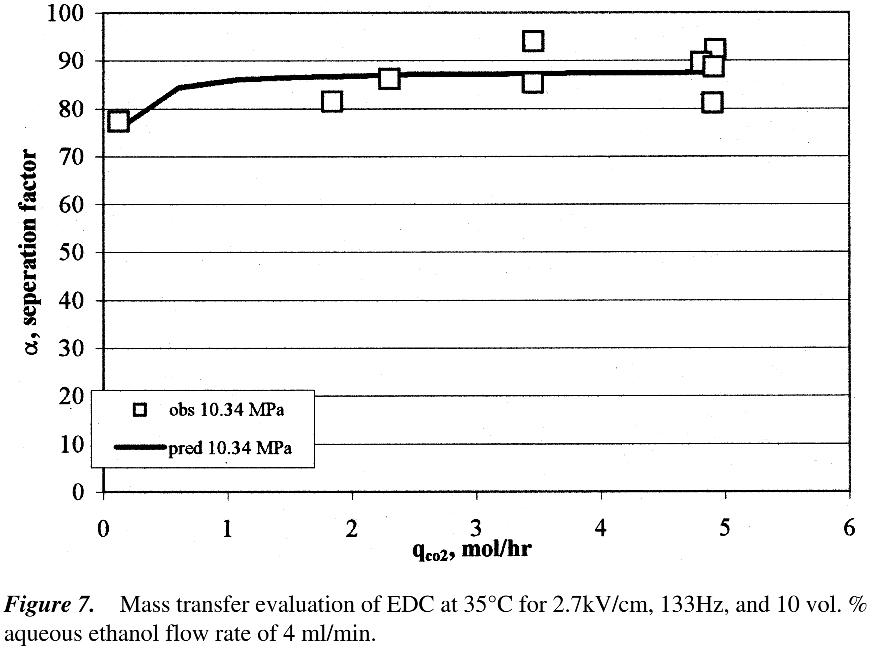

Figure 7. Mass transfer evaluation of EDC at 35°C for 2.7kV/cm, 133Hz, and 10 vol. % aqueous ethanol flow rate of 4 ml/min.

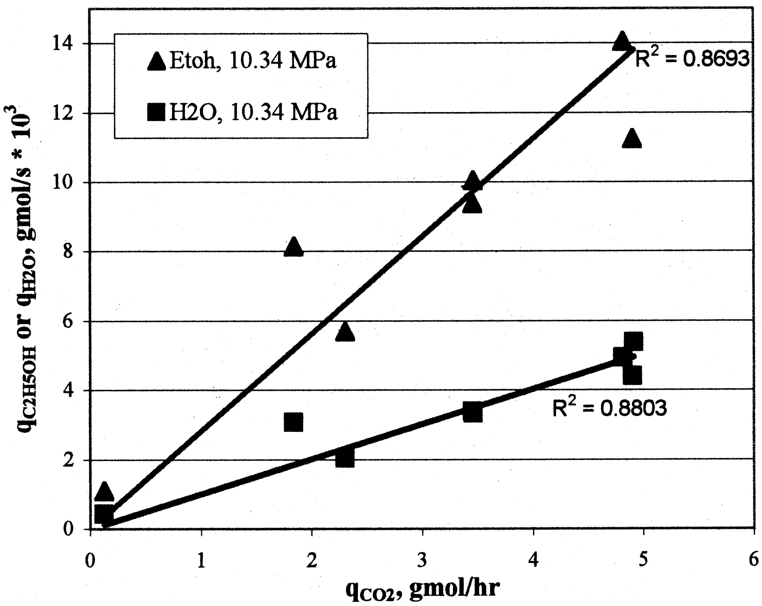

Figure 8. Rate of extraction of C_2H_5OH and H_2O from a 10 vol. % aqueous ethanol flow rate of 4 ml/min, 2.7 kV/cm, and 133 Hz at 35°C with null experiment included.

to recover the extracted ethanol and water, extraction rates that would characterize the intrinsic mass transfer performance of the EDC could not be achieved in the present study.

CONCLUDING REMARKS

This first study of electrospraying of a liquid into a dense, supercritical fluid has demonstrated a new method for dispersing aqueous liquid into SC-CO_2 using pulsed high-voltage electric fields at 35°C and 10.34 MPa. With a laser light scattering system it was possible to determine the dependence of mean droplet size on operating variables such as field strength, pulse frequency, and flow rate. Contrary to prior studies of electrospraying liquid into gas and operating in the cone-jet mode, increased field strength caused $<d_p>$ to increase. Likely this was the result of polydispersity in the electrospray at conditions beyond the cone-jet mode. In all cases, whether increasing flow rate or field strength, lowering the pulse frequency appeared to give smaller $<d_p>$. The effect of increasing $<d_p>$ with increasing flow rate is consistent with results by other researchers studying electrospraying of liquid into gas. Extraction of ethanol from a 10 vol. % aqueous ethanol solution using SC-CO_2 in the EDC was limited by equilibrium for all conditions studied up to about 8 mol/hr CO_2 flow rate. Entrainment of micron-size liquid droplets from the EDC is apparently negligible. It is generally believed that the micron-size droplets produced by electrospraying are electrically charged. Charged droplets would be strongly attracted to the wire mesh electrode. Apparently, a high degree of droplet coalescence at the wire mesh electrode was achieved in the EDC, resulting in negligible liquid entrainment.

In future work it is planned to measure the mass transfer coefficient. One approach would be to design and install condensers capable of operating at substantially higher CO_2 flow rates so that the intrinsic mass transfer performance of the EDC can be studied. Since the current operation measures a number average droplet size, it is difficult to accurately determine the total surface area in contact with the gas phase. It would be beneficial to disperse a water-soluble dye so that water holdup could be measured photometrically. If it were possible to measure the suspended liquid volume, then a calculation could be made of total surface area. This would then allow the determination of mass transfer coefficients.

The use of pulsed electric fields to produce a fine dispersion of aqueous droplets into SC-COl_2 appears to be an attractive possibility for improved operations where an environmentally friendly system is desired. The present work established a basis for (a) broadening the range of applicability of solvent substitution by SC-CO_2, (b) contributing to the fundamental knowledge of droplet formation in a unique experimental way (SC-CO_2 affords a range of variables not available in most experiments), and (c) exploring the behavior of micro-droplets in supercritical fluids. These dispersions may have a variety of applications including extractions, dyeing operations, reactions, and the formation of fine particles. The adaptation of such schemes to industrial applications could provide systems which are both more efficient and environmentally beneficial. Likely initial commercial applications of this technology might be found for systems where a small volume of high-value product must be extracted from an aqueous medium. Possible applications of this type might be found in the pharmaceutical industry.

ACKNOWLEDGMENT

We are pleased to acknowledge support for this work by the Division of Chemical Sciences of the U.S. Department of Energy.

REFERENCES

1. C.L. Widrig, J. Tao, and D. Allen, *Basic Research Needs for Environmentally Responsive Technologies of the Future,* results of a workshop sponsored by NSF and DOE, Jan 4-6, 1996, page 13.

2. K.L. Harrison, J. Goveas, and K.P. Johnston, *Langmuir,* 10**,** 3536 (1994).
3. M.J. Clark, K.L. Harrison, K.P. Johnston, and S.M. Howdle, *J. Am. Chem. Soc.,* 119**,** 6399 (1997).
4. M.P. Heitz, C. Carlier, J. deGrazia, K.L. Harrison, K.P. Johnston, T.W. Randolph, and F.V. Bright, *J. Phys. Chem. B, 101,* 6707 (1997).
5. J.L. Fulton, D.M. Pfund, J.B. McClain, T.J. Romack, E.E. Maury, J.R. Combes, E.T. Samulski, J.M. DeSimone, and M Capel, *Langmuir*, *11,* 4241 (1995).
6. E.J. Beckman, *Science*, *271,* 613 (1996).
7. K.P. Johnston, *et al*., *Science*, *271,* 624 (1996).
8. T.C. Scott and R.M. Wham, Surface Area Generation and Droplet Size Control in Solvent Extraction Systems Utilizing High Intensity Electric Fields, U. S. Patent 4,767,929 (1988).
9. T.C. Scott, Magnetic/Electric Field Solvent Extraction, U. S. Patent 4,941,959 (1989).
10. M.T. Harris, W.G. Sisson, and O.A. Basaran, Improved Nozzle for Electrical Dispersion Reactor, U. S. Patent 5,464,195 (1995).
11. M.T. Harris, T.C. Scott, and C.H. Byers, Method and Apparatus for Preparing Metal Oxide Powders, U.S. Patent 5,122,360 (1992).
12. M.T. Harris, T.C. Scott, and C.H. Byers, Method and Apparatus for the Production of Metal Oxide Powder, U. S. Patent 5,207,973 (1993).
13. T.C. Scott and R.M. Wham, *Ind. Eng. Chem. Res*., *28,* 94 (1989).
14. M.T. Harris, T.C. Scott, O.A. Basaran, and C.H. Byers, *Mat. Res. Soc. Symp. Proc*., *180,* 153 (1990).
15. M.T. Harris, T.C. Scott, O.A. Basaran, and C.H. Byers, *Superconducting Engineering, AIChE Symp. Ser., 88,* 44 (1992).
16. M.T. Harris, W.G. Sisson, T.C. Scott, O.A. Basaran, C.H. Byers, W. Renand, and T. Meek, *Mat. Res. Soc. Symp. Proc*., *271,* 945 (1992).
17. M.T. Harris, T.C. Scott, and C.H. Byers, *Mat. Sci. Eng*., *A168,* 125 (1993).
18. T.C. Scott, D.W. DePaoli, and W.G. Sisson, *Ind. Eng. Chem. Res*., *33,* 1237 (1994).
19. M.T. Harris, W.G. Sisson, S.M. Hayes, S.J. Bobrowski, and O.A. Basaran, *Mat. Res. Soc. Symp. Proc*., (1995).
20. T.C. Scott, J.M. Cosgrove, D.W. DePaoli, and W.G. Sisson, *Appl. Biochem. Biotech*., *51/52,* 283 (1995).
21. E. Borzabadi and A.G. Bailey, *J. Electrostatics, 5,* 369, (1978).
22. K.D. Heath and H.D. Cochran, *Ind. Eng. Chem. Res*., submitted (1999).
23. K.D. Heath, "Carbon Dioxide Solvent Substitute: Fundamental Study of Aqueous Dispersions in Supercritical Carbon Dioxide." Ph.D. Dissertation, University of Tennessee, (1999).
24. K. Tang and A. Gomez, *Phys. Fluids A*, *6,* 2317 (1994a).
25. J. Fernandez de la Mora, J. Navascues, *et al*., *J. Aerosol Sci*., *21,* S673 (1990).

26. D. Chen, D.Y.H. Pui, and S.L. Kaufman, Electrospraying of Conducting Liquids for Monodisperse Aerosol Generation in the 4 nm to 1.8 μm Diameter Range, *J. Aerosol Sci. 26,* 963 (1995).
27. M. Choupeau and B. Prunet-Foch, *J. Electrostatics*, *22,* 135 (1989).
28. K. Tang and A. Gomez, *J. Colloid Interface Sci., 184,* 500 (1996).
29. A.R. Jones and K.C. Thong, *J.Phys. D. Appl. Phys. 4,* 1159 (1971).
30. B. Vonnegut and R.L. Neubauer, , *J. Colloid Sci., 7,* 616 (1952).
31. J. Rossell-Llompart and J. Fernandez de la Mora, *J. Aerosol Sci., ??,* 1093 (1994).
32. D. Chen and D.Y.H. Pui, *Aerosol Sci. and Tech.*, *27,* 367 (1997).
33. I.G. Loscertales and Fernandez de la Mora, *J. Aerosol Sci. 27,* 695 (1993).
34. A.M. Ganan-Calvo, *J. Aerosol Sci. 25S,* S309 (1994).
35. M.B. Cloupeau and B. Prunet-Foch, *J. Electrostatics 25,* 165 (1990).
36. M.B. Cloupeau and B. Prunet-Foch, *J. Aerosol Sci. 25,* 1021 (1994).
37. I. Hayati, A.I. Bailey, and T.H.F. Tadros, *J. Colloid Interface Sci. 117,* 205 (1987).
38. J. Fernandez de la Mora and I.G. Loscertales, *J. Fluid Mech. 260,* 155 (1994).
39. K. Tang and A. Gomez, *J. Aerosol Sci. 25,* 1237 (1994).
40. T.C. Scott, *AIChE Journal, 33,* 1557 (1987).
41. R.M. Wham and C.H. Byers, *Sep. Sci. Tech., 22,* 447 (1987).
42. T.C. Scott, in *Chemical Separations, I: Principles,* C.J. King, J.D. Navratil, Eds., Litarvan, Denver (1986).
43. H. Lamb, *Hydrodynamics,* 6th ed., Dover, N.Y., 475 (1945).
44. K. Tang and A. Gomez, *J. Colloid Interface Sci. 175,* 326 (1995).
45. K. Tang, " The electrospray: Fundamentals and Feasibility of Its Application to Targeted Drug Delivery by Inhalation." Ph.D. Dissertation, Yale University, 1994.
46. J. Zeleny, *Proc. Cambridge Phil. Soc. 18,* 71 (1915).
47. X. Zhang and O.A. Basaran, *J. Fluid Mech. 326,* 239 (1996).

MARCEL DEKKER, INC.
270 Madison Avenue, New York, New York 10016

Downloaded At: 10:49 25 January 2011

Request Permission or Order Reprints Instantly!

Interested in copying and sharing this article? In most cases, U.S. Copyright Law requires that you get permission from the article's rightsholder before using copyrighted content.

All information and materials found in this article, including but not limited to text, trademarks, patents, logos, graphics and images (the "Materials"), are the copyrighted works and other forms of intellectual property of Marcel Dekker, Inc., or its licensors. All rights not expressly granted are reserved.

Get permission to lawfully reproduce and distribute the Materials or order reprints quickly and painlessly. Simply click on the "Request Permission/Reprints Here" link below and follow the instructions. Visit the U.S. Copyright Office for information on Fair Use limitations of U.S. copyright law. Please refer to The Association of American Publishers' (AAP) website for guidelines on Fair Use in the Classroom.

The Materials are for your personal use only and cannot be reformatted, reposted, resold or distributed by electronic means or otherwise without permission from Marcel Dekker, Inc. Marcel Dekker, Inc. grants you the limited right to display the Materials only on your personal computer or personal wireless device, and to copy and download single copies of such Materials provided that any copyright, trademark or other notice appearing on such Materials is also retained by, displayed, copied or downloaded as part of the Materials and is not removed or obscured, and provided you do not edit, modify, alter or enhance the Materials. Please refer to our Website User Agreement for more details.

Order now!

Reprints of this article can also be ordered at

http://www.dekker.com/servlet/product/DOI/101081SS100103646