

# **Environmental Management Science Program**

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## **Phase Equilibria Modification by Electric Fields**

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## Phase Equilibria Modification by Electric Fields

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### Research Objective

The objective of this project is to use electric fields to favorably manipulate the thermodynamic and transport properties of mixtures so that higher separation efficiencies can be achieved. The main focus is to understand and quantify the influence of electric fields on vapor-liquid, liquid-liquid, and solid-liquid systems. It is expected that this program will lead to greater separation efficiency in a wide range of environmental treatment processes, including solvent extraction, sorption, distillation, and stripping. Such processes are widely used by DOE for treatment of wastes and sites contaminated with heavy metals, radionuclides, and organic solvents. Particular examples of applications of vapor-liquid-equilibria modification can be found in the separation of volatile organic compounds by either stripping or distillation. Improvements can also be made in liquid-liquid-extraction processes of TRU, Sr, Tc, and Cs by both thermodynamic and transport enhancements. Separations of metal ions by electro-sorption can be used to remove such metal ions as Cs, Sr, Co, Cr, Cd, Hg, and TRU.

### Research Progress and Implications

Following is a summary of the work after 1 and 2/3 years of a 3-year project:

**Vapor-Liquid Equilibria:** Studies during the first year of this program showed that the separation factor for polar-polar and polar-nonpolar liquid mixtures was significantly increased by applying an electric field across the vapor-liquid interface (Blankenship et al., 1997). This work has been continued in the second year with focus on the geometry and distance of the electrodes, direction and strength of the electric field, and electrical properties of the components. Results consistently showed that, at low applied voltage (ca. 4-6 kV) there was no significant effect on the chemical composition of the two phases. At higher applied voltage, the separation factor of the mixture increased. It was found that the applied voltage is the key to vapor-liquid-equilibria modification, while the distance between the electrodes had little effect. These experimental results, coupled with numerical solutions of Maxwell's equations suggested that the mechanism of equilibria modification by electric fields is related to the charge density at the interface (Blankenship et al., 1998).

**Liquid-Liquid Studies:** Various binary and ternary liquid systems, including cyclohexane-water, cyclohexane-isopropanol-water, tributyl phosphate-water, and water-MIBK-acetic acid, have been employed in liquid-liquid-equilibria experiments. Unlike vapor-liquid systems, it is difficult to isolate an equilibrium effect of an electric field, since the application of a high voltage difference across a liquid-liquid interface causes significant electrohydrodynamic (EHD) flows. Such EHD flows have been extensively used in the past to enhance heat transfer in various processes. Experiments were conducted to further investigate the use of EHD flows in transport enhancement. It was found that electric fields cause simultaneous pumping, spraying, and mixing of fluids (Shin et al., 1997; Tsouris et al., 1998a). These phenomena were observed using various polar solvents including water, ethanol, propanol, 2-propanol, and butanol. Such phenomena may be employed to significantly enhance transport rates in solvent-extraction treatment processes.

**Solid-Liquid Equilibria:** It has been shown by other investigators that electro-sorption may lead to an efficient process for the removal of ions, including heavy metals and radionuclides, in which the sorbent is readily regenerated by removing the external electrical potential. The mechanism of electro-sorption is due to transport of ions from the aqueous solution to the solid-liquid interface and capture of the ions in the electric double layer formed at the surface of the electrodes. Electrochemical reactions may also occur at the electrodes. Equilibrium and kinetic experiments are conducted in order to better understand the mechanisms of electro-sorption and develop predictive models for

metal ion uptake by this phenomenon. In these experiments, the electrodes are made of carbon aerogel, a material composed of interconnected particles with microscopic interstitial pores (<50 nm), which is ideal for electro-sorption because of its high specific surface area (400 to 1100 m<sup>2</sup>g<sup>-1</sup>) and low electrical resistivity (< 40 mWcm). Preliminary experiments conducted with NaCl solutions showed that the conductivity of the solution decreased during applied electric field and increased after removing the field. These results indicate that the ions are captured in the electric double layer when the field is applied and are released back to solution when the field is removed.

## Planned Activities

The following activities are planned for the remaining of the second and third year:

**Gas-Liquid Systems:** Studies will be focused on understanding the mechanisms of electric-field effects on vapor-liquid-equilibria. Raman spectroscopy will be used to study the interface with and without electric fields applied. Additional polar-polar and polar-nonpolar systems will be used in the experiments to determine the characteristics of suitable chemical systems, and testing will be conducted to determine approaches to effectively employ electric fields.

**Liquid-Liquid Systems:** Experiments will be conducted to quantify the effect of electric fields on phase equilibria of binary and ternary systems. Exploratory experiments will be undertaken in collaboration with B. A. Moyer (EMSP 55087) to determine the effect of applied electric fields on transport and equilibrium in solvent-extraction systems with advanced solvents developed in that program. In parallel, the work on EHD flows will be continued with the objective to better understand the underlying phenomena and develop devices that could be used for efficient separations.

**Solid-Liquid Systems:** A surface complexation model will be developed to predict equilibrium and kinetics of metal-ion uptake. This model will consider: (i) surface ionization, (ii) complexation of surface sites with ionic species, and (iii) the formation of an electrical double layer adjacent to the surface. The capabilities of this model are expected to include prediction of the effect of the various solution variables, such as pH, ionic strength, and metal ion concentration, as well as the effect of the electric field on the extent of electro-sorption. Experimental data will be obtained for model verification.

## Other Access To Information

- Shin, W.-T, S. Yiacoumi, and C. Tsouris, "Experiments on Electrostatic Dispersion of Air in Water," *Ind. Eng. Chem. Res.* 36, 3647-55 (1997).
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- Tsouris, C., W.-T. Shin, and S. Yiacoumi "Spraying, Pumping, and Mixing of Fluids by Electric Fields," in press, *Can. J. Chem. Eng.*, June, 1998a.
- Blankenship, K.D., D.W. DePaoli, and C. Tsouris "Effect of Electrode Configurations on Phase Equilibria with Applied Electric Fields," submitted for publication in *Sep. Purif. Technol.*, April, 1998.