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PERFORMANCE OF SPRAY, SIEVE TRAY, AND PACKED CONTACTORS FOR HIGH PRESSURE EXTRACTION

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ABSTRACT

The performance of various contactors in high pressure extraction service was studied. The mass transfer efficiency of a spray column and columns filled with sieve trays, 1.27 cm ceramic Raschig rings, and no. 15 metal Intalox saddles were obtained from a "windowed" high pressure extraction column with an internal diameter of 9.88 cm and a column height of 168 cm. The extraction column has an internal diameter of 9.88 cm and a contacting height of 168 cm. Glass windows in the extractor allowed visual observation and photography. Supercritical and liquid carbon dioxide were used to extract isopropanol from water at pressures of 81 to 150 atmospheres and 297 to 318 °K. The sieve tray extractor yielded the highest mass transfer efficiency followed closely by Raschig rings and metal Intalox saddles. The spray column was by far the least efficient. The mass transfer efficiency was observed to be independent of the operating conditions studied in this work. Sauter mean drop diameters and dispersed phase (free) hold-up were also obtained in this work. These data will provide a reliable high pressure mass transfer and hydraulic database for future correlation and design efforts.

Introduction

High pressure extraction is an attractive alternative separation process when other methods such as distillation, adsorption, membranes, and liquid-liquid extraction are impractical or impossible. A great deal of research work has been performed in finding uses for this technology. Some examples are coffee decaffeination (1), extraction of hops resins (2,3), fractionation of heat sensitive compounds (4,5), treatment of hazardous wastes (6), and the deasphalting of petroleum residuals (7). Generally, high pressure extractors operate near or above the critical temperature and pressure of the solvent to take advantage of the enhanced solvating and transport properties associated with this region. Carbon dioxide is a popular choice as a high pressure solvent because of its relatively nonhazardous characteristics along with a reasonably low critical temperature (305°K) and pressure (72.8 atm).

While much of the previous research in high pressure extraction has involved fluid-solid and fluid-liquid phase equilibria, there has been little applied research investigating the hydrodynamics and mass transfer characteristics at these conditions. Thus, the relative performance of different contacting devices is unknown. A variety of countercurrent contacting devices are available for extraction operations. The simplest and least expensive is the spray column which contains only a distributor as an internal. This column is generally inefficient due to backmixing of the continuous phase, resulting in the reduction of the concentration gradients. The sieve tray contactor is an alternative device which may be considered as a series of short spray columns. This arrangement allows reformation of drops as well as prevention of backmixing over the entire column. In high pressure extraction, the geometry of the sieve tray is identical to that used in conventional liquid-liquid extraction service with downcomers (or upcomers), punched holes, and no weir. The packed column is yet another mass transfer contactor. The presence of the packing reduces the backmixing of the continuous phase and also provides a tortuous path for the dispersed phase which contributes to an increase in mass transfer efficiency for the packed extractor.

The purpose of this paper is to provide fundamental hydrodynamic and mass transfer data obtained on a variety of contacting devices under high pressure conditions. These devices include the spray column (distributor only), sieve trays, 1.27-cm ceramic Raschig rings, and no. 15 IMTP (metal Intalox saddles). The experimental data will be presented and the effects of temperature, pressure, and column size on mass transfer efficiency will be discussed.

Previous Work

Only a few studies of both the hydrodynamics and mass transfer at high pressure conditions have been reported. In most cases, overall mass transfer data have been discussed in the literature. Brunner (8) studied the use of supercritical carbon dioxide to separate ethanol from an aqueous stream using a spray column. The effects of pressure, temperature, and solvent to feed ratios were examined. Peters and Tiegs (9,10) investigated the effect of packing in a 2.54 cm diameter column. The separation of a mixture of glycerides of oleic acids with carbon dioxide was studied in this work. Height equivalent to a theoretical stage (HETS) of 30 cm was obtained with wire spirals and gauze packing. Eisenbach (11) investigated the fractionation of a mixture of high boiling ethyl esters from fish oils with supercritical carbon dioxide. A "hot finger" (a heated section of the column) was utilized to improve the separation. Optimum results were obtained at 90°C and 150 atmospheres. In another study (12,13), Critical Fluid Systems (CFS) investigated the performance of a 10.2 cm diameter sieve tray extractor using near critical carbon dioxide. The objective of the study was to evaluate the economic feasibility of separating alcohols (ethanol, isopropanol, and sec-butanol) using pilot-plant-scale equipment. Another pilot-scale study involved the use of liquid carbon dioxide at 60 atmospheres and 25°C (14). A four-centimeter-diameter Scheibel column was used to extract 12 different compounds including ethanol, ethyl acetate, 1-hexanol, and citronella from water.

Recently, Lahiere (15) completed a comprehensive study of the hydrodynamics and mass transfer characteristics of a 2.54 cm diameter spray and sieve tray high pressure extractor. The effects of the two contacting devices on the Sauter mean drop diameter, dispersed phase holdup, flooding, and mass transfer efficiency were analyzed. Three test systems were studied: (1) toluene/acetone/water, operated under conventional ambient conditions, (2) carbon dioxide/isopropanol/water , and (3) carbon dioxide/ethanol/water, the latter two systems operated at high pressures. The high pressure extractor was observed to be 5-6 times more efficient than the extractor operated at conventional ambient conditions. The effects of liquid (82 atm, 24°C) and supercritical (102 atm, 40°C) carbon dioxide were examined. The mass transfer efficiency of dense carbon dioxide was determined to be virtually unaffected by a change in operating conditions. The hydrodynamics at high pressure were observed to behave very similarly to those of the toluene/acetone/water system. Lahiere was also able to utilize correlations developed for liquid-liquid extraction to predict the results obtained from the supercritical fluid extractor. Rathkamp (16), using the same apparatus as Lahiere, studied the performance of 0.64-cm metal Raschig rings. The contrasting effects of the toluene/acetone/water (low pressure) and carbon dioxide/isopropanol/water (high pressure) were also investigated.

Experimental Equipment and Procedure

Figure 1 shows a flow schematic of the pilot plant used in this study. The stainless-steel extraction column was 9.88 cm in diameter (i.d.) with a total height of 215 cm. The column contained 32 high pressure glass windows to allow viewing of the contacting action inside. This windowed extractor was rated to 102 atm. In some cases, data were obtained from an identical column without windows, which was rated to 205 atm. The dispersed phase was distributed with a crossed ring sparger containing nine 0.47-cm diameter holes spaced uniformly to ensure equal distribution. The continuous phase was distributed with an "L"-shaped sparger containing 15 0.32 cm holes. The contacting distance between distributors was 168 cm.

The pilot plant was controlled with a distributed control system. Process flow, pressure, steam, and cooling water valves were controlled pneumatically. The liquid feed was delivered to the extractor with two positive displacement pumps operated in parallel. A dampener was used in the liquid feed line to prevent

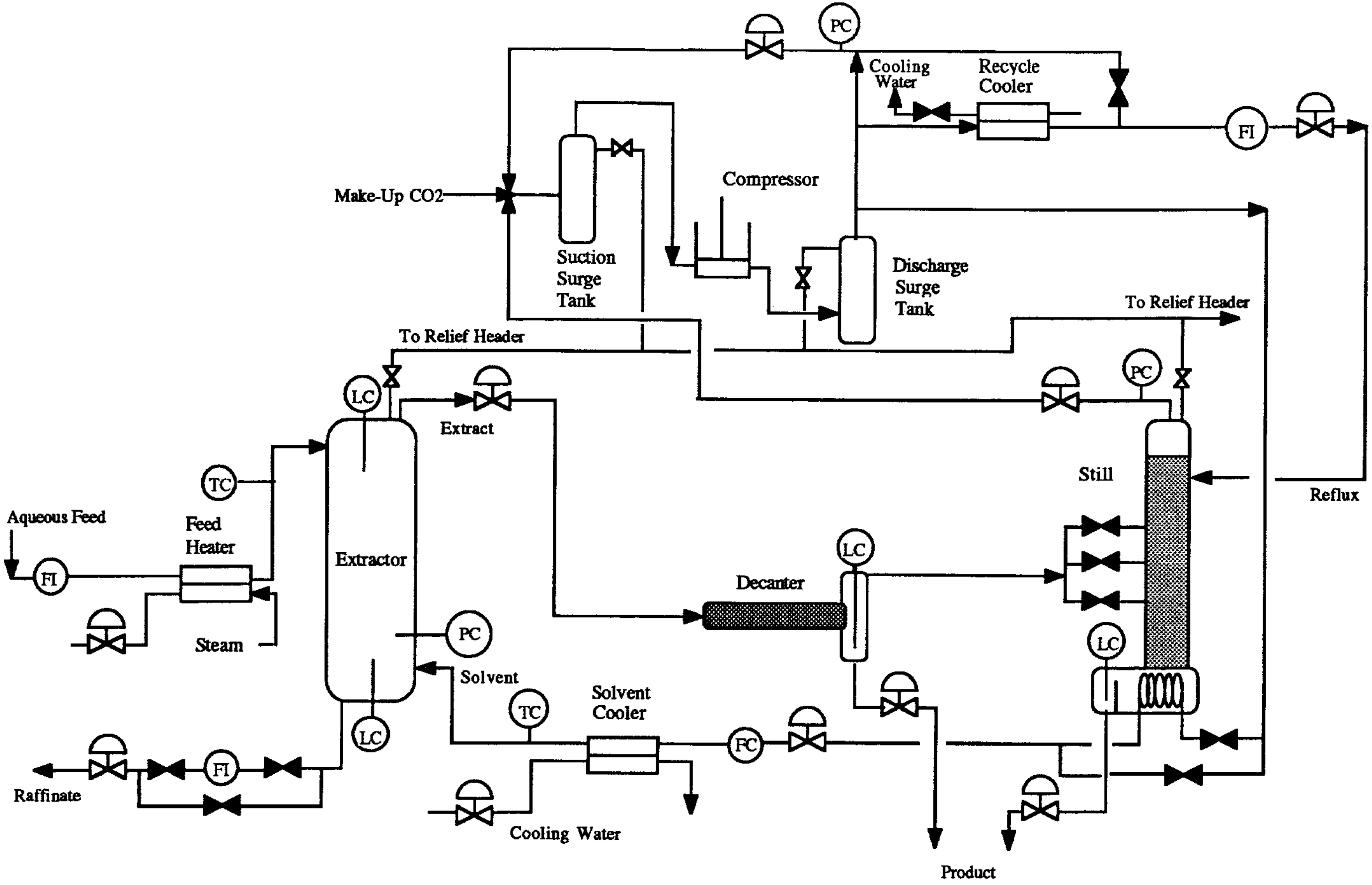

Figure 1. High Pressure Extraction Flow System

flow pulsation. The solvent was delivered to the extractor with a reciprocating compressor. Mass flows of the liquid feed, raffinate, and solvent were measured to a high degree of accuracy using Micro-Motion® mass flowmeters.

Pressures were measured with standard pressure transducers. These transducers were located on the discharge of the compressor, extractor, and solvent fractionator. All vessels contained relief valves which were piped to a common relief header.

The interface level was controlled with a capacitance probe and manipulation of the raffinate flowrate. Likewise, the solvent fractionator and decanter liquid levels were controlled with capacitance probes. The temperatures of the feed and solvent entering the extractor were measured with thermocouples and controlled by heat exchange with cooling water or steam. The compressor recycle stream temperature was controlled in a similar manner.

Analysis of all streams was performed with two online gas chromatographs utilizing thermal conductivity detectors. The samples were collected at process conditions and transferred in 0.318-cm (o.d.) stainless steel tubing to a two microliter sampling loop using a six-port manual sampling valve. This arrangement prevented two-phase sampling and yielded excellent reproducibilities.

The Sauter mean drop diameter was determined using a photographic technique. A material of known (reference) diameter was placed inside the windowed extractor. Photographs of the drops (with reference included) were analyzed with a digitizer. At the end of a run, the solvent flow to the extractor was shut off instantaneously and the mean drop velocity was determined from the drop residence time between the dispersed phase distributor and the top window (150 cm). The dispersed phase holdup was calculated from Equation 1 using the dispersed phase flowrate and the drop velocity.

$$\phi_d = \frac{Q_d}{A_c \, U_{drop}} \qquad (1)$$

Q_d = volumetric flowrate of the dispersed flowrate, cm^3/s

A_c = cross-sectional area of the column, cm^2

U_{drop} = drop velocity, cm/s

A run was performed using the following procedure. At startup, the extractor was initially charged with aqueous feed to three-fourths of the extractor volume. The pilot plant was then pressurized to the desired pressure with carbon dioxide. The continuous aqueous feed flow was then started and analyzed for composition. The overhead from the solvent fractionator and the solvent entering the extractor were analyzed to document their purity. After verifying solvent purity, the extract and raffinate streams were analyzed. Photographs were taken to determine the

Table 1. Geometry of Column Internals

Internal	Geometry
Spray-Distributor	
Ring Sparger	
Hole Diameter	0.5 cm
Number of Holes	9
Sieve Tray	
Tray Spacing	17.0 cm
Fractional Hole Area	0.091
Fractional Downcomer Area	0.015
Hole Diameter	0.32 cm
Holes are Punched Upwards	
Downcomer Length	13.0
1.27 cm Ceramic Raschig Rings	
Packing Surface	3.88 cm^2/cm^3
Void Fraction	0.64
no.15 metal Intalox saddles	
Packing Surface	3.09 cm^2/cm^3
Void Fraction	0.91

Sauter mean drop diameter. Steady-state operation was determined to exist when the composition of the extract and raffinate streams remained constant over a period of 30 - 45 minutes. The feed and solvent flows to the extractor were then discontinued and the dispersed phase holdup was determined. Flows were adjusted and continued for the next run.

One test system was studied in this work: carbon dioxide/isopropanol/water. Three operating conditions were investigated:

(i) 82 atm, 297°K
(ii) 102 atm, 313°K
(iii) 150 atm, 318°K

Isopropanol compositions in the feed and superficial velocities ranged from 4-6 wt.% and 0.034 - 2.0 cm/s. The geometry of the column internals studied in this work.

Data Analysis

Equilibrium data for conditions (i) and (ii) were obtained from Lahiere (15) and Kander (17). These data were verified in this work and additional phase equilibria for 150 atm and 46°C were also obtained. The equilibrium data of this work and Lahiere's study were acquired by pinching the operating line with the equilibrium line, thereby forcing the extract stream into equilibrium with the feed stream. Values of the distribution coefficient (wt/wt basis) ranged from 0.17 to 0.23 for the range of feed concentrations studied. High solvent to feed ratios were required to perform a measurable separation due to the low distribution coefficients, thus carbon dioxide was chosen as the dispersed phase in order to maximize the interfacial area.

At the end of each run, a material balance and a check for equilibrium pinch were performed. If the material balance closure was greater than 95% and the extract solute composition was less than 90% of the equilibrium pinched value, the mass transfer efficiency was determined. The number of transfer units was calculated using Equation 2 (18), which assumes a linear equilibrium relationship, dilute solute concentration, pure solvent, and a straight operating line. This equation is also based on the continuous phase.

$$NTU_{oc} = \frac{\ln\left\{\left(\frac{X_f}{X_r}\right)\left(1-\frac{1}{\lambda}\right)+\frac{1}{\lambda}\right\}}{1-\frac{1}{\lambda}} \tag{2}$$

$$\lambda = \left(\frac{U_D}{U_C}\right) M_{dc} \tag{3}$$

Next, the overall height of a transfer unit was calculated from the contacting height (Z) and the number of transfer units (NTU_{OC}).

$$HTU_{oc} = \frac{Z}{NTU_{oc}} \tag{4}$$

The height equivalent to a theoretical stage (HETS) was determined from Equation 5.

$$HETS = \frac{\lambda \ln \lambda}{\lambda - 1} HTU_{oc} \tag{5}$$

The reproducibility of the data obtained in this work was quite good with an average deviation of 7% as shown in Figure 2. Solute compositions, mass flowrates, and drop residence times were recorded to allow calculation of the superficial velocities, dispersed phase holdup, and mass transfer efficiency.

Experimental Results

The Sauter mean drop diameter was observed to be independent of the type of contactor (spray or sieve tray) and of flowrates, as shown in Figure 3. The dispersed phase holdup was observed to increase linearly at low dispersed phase velocities and then increase nonlinearly with higher velocities as the dispersed phase drops collide with one another. This behavior is also observed in conventional liquid-liquid extractors. The holdup data reported by Lahiere differ from those of this work as shown in Figure 4. This deviation may be attributed to the increased effects of the walls and windows of Lahiere's 2.54 cm diameter column. The effect of the dispersed phase velocity in the spray and sieve tray column is illustrated in Figure 5. The efficiency increases (HETS decreases) as the column is loaded with dispersed phase until a velocity of 1.0 cm/s, is reached, at which point the efficiency is constant. This was likely due to the compensating effects of increased dispersed phase holdup with decreased slip velocity and increased backmixing of the continuous phase. A similar trend was observed with the metal Intalox saddles and Raschig rings as shown in Figures 6. Three operating conditions were explored using metal Intalox saddles. The mass transfer efficiencies of liquid carbon dioxide (82 atm, 24°C) and supercritical carbon dioxide (102 atm, 40°C and 150 atm, 46°C) were observed to be nearly identical. This fact becomes very important when considering the process economics. The performance of the sieve tray column and the column packed with Raschig rings behaved similarly. Greater continuous phase velocities were observed to increase the mass transfer efficiency, as shown in Figure 7. This is likely due to an increase in the slip velocity between phases and better distribution of continuous phase throughout the column.

The relative performance of the various contacting devices is compared in Figure 8. The sieve tray is the most efficient, followed closely by the Raschig rings and the metal Intalox saddles. The spray column is the least efficient. This is due to increased backmixing of the continuous phase and a shorter drop residence time in the column. It is also expected that the Raschig rings would be more efficient than the metal Intalox saddles at the flows investigated, because of the larger available packing surface area. The packing surface increases the tortuous path, thus increasing the residence time of the dispersed phase drops in the column. Although not demonstrated in this work, one would expect that the metal Intalox saddles would have a greater capacity.

The effect of column size on the mass transfer efficiency for the spray and sieve-tray columns is shown in Figure 9. The results of this research are compared with the 2.54 cm diameter column data of Lahiere. The sieve-tray data of each work compare

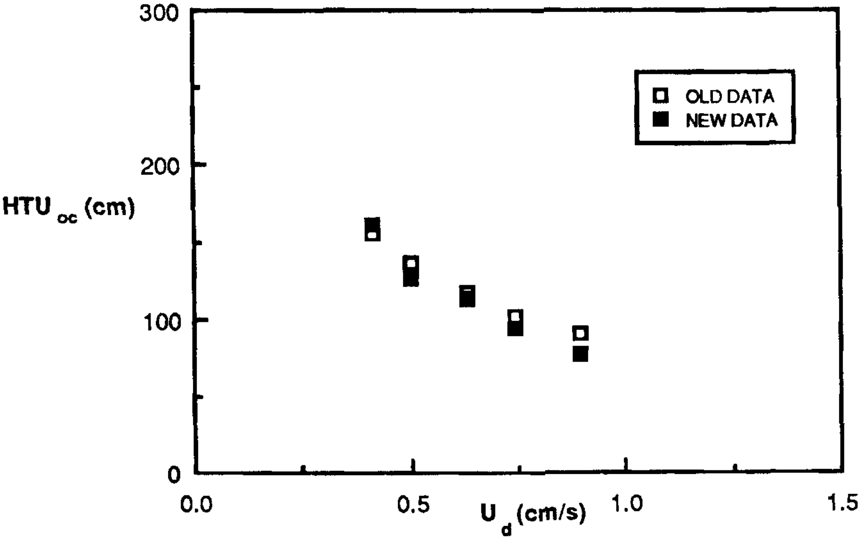

Figure 2. Reproducibilty of Experimental Data. Spray Column. Liquid Carbon Dioxide/Isopropanol/Water(81 atm,297°K). Dispersed: Phase: Carbon Dioxide. U_C: 0.048 cm/s.

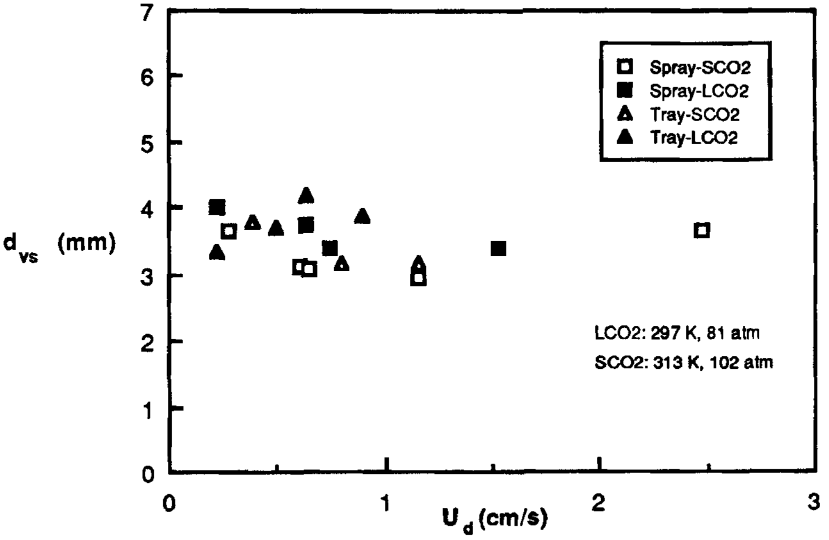

Figure 3. Effect of the Dispersed Phase Velocity on the Sauter Mean Drop Diameter.
System: Carbon Dioxide/Isopropanol/Water.
Dispersed Phase: Carbon Dioxide. U_C: 0.048 cm/s.

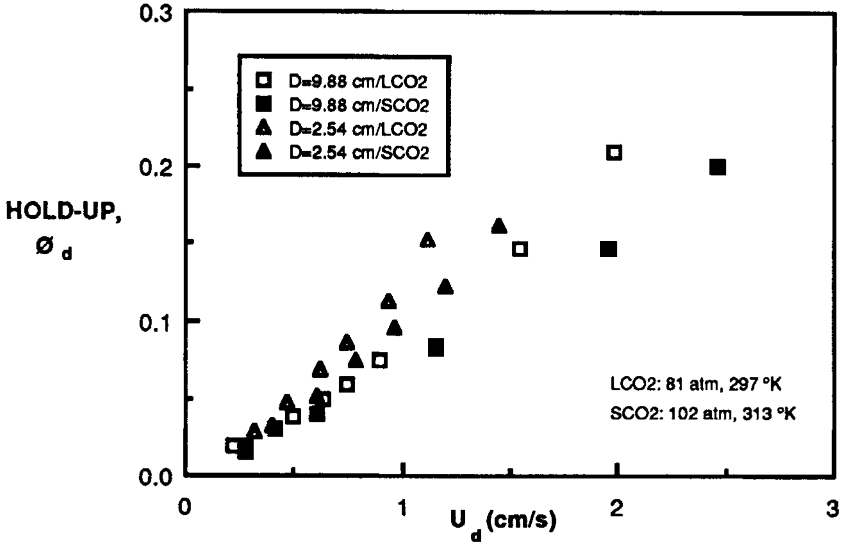

Figure 4. Spray Column Holdup.
System:Carbon Dioxide/Isopropanol/Water.
Dispersed Phase: Carbon Dioxide. U_c: 0.048 cm/s.

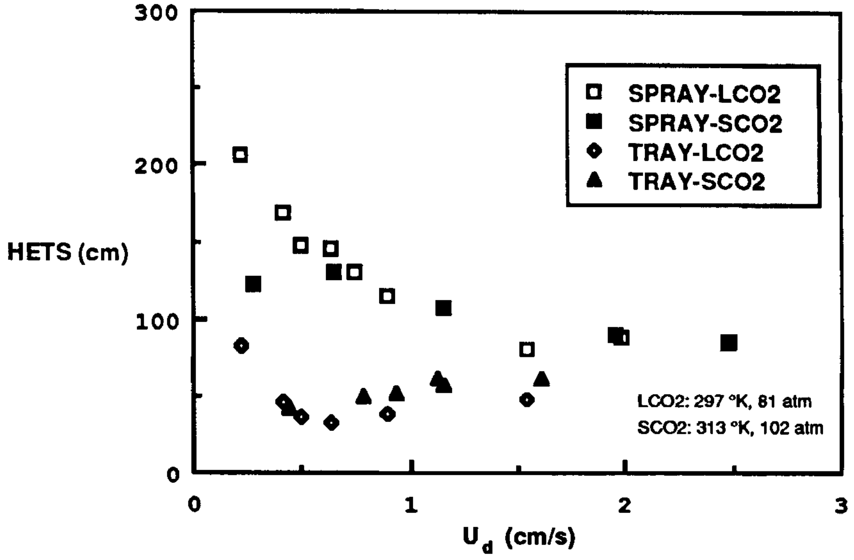

Figure 5. Spray and Sieve Tray Mass Transfer Data.
System: Carbon Dioxide/Isopropanol/Water.
Dispersed Phase: Carbon Dioxide. U_c: 0.048 cm/s.

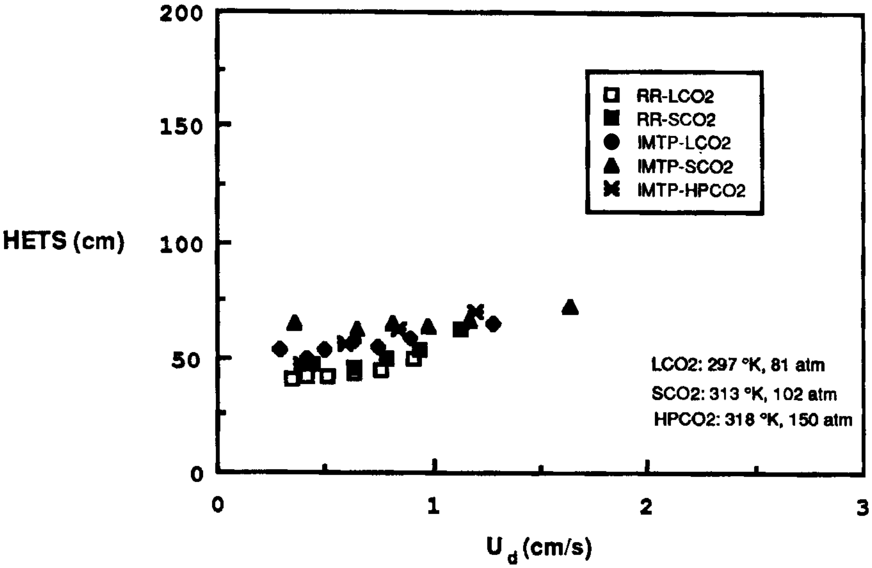

Figure 6. Packed Column Mass Transfer Data.
System: Carbon Dioxide/Isopropanol/Water.
Dispersed Phase: Carbon Dioxide. U_c: 0.048 cm/s.

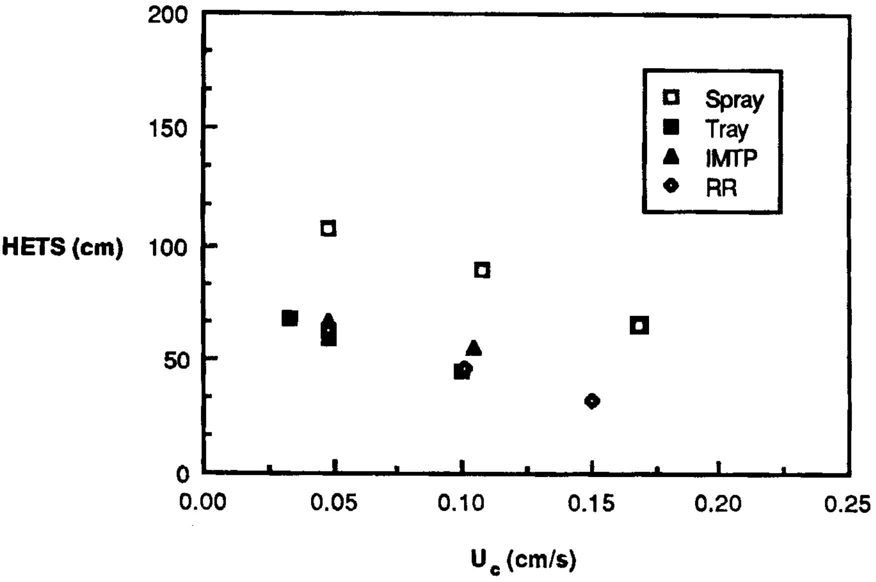

Figure 7. Effect of the Continuous Phase Velocity
System:Carbon Dioxide/Isopropanol/Water(313°K,102 atm)
Dispersed Phase: Carbon Dioxide. U_d: 1.13 cm/s.

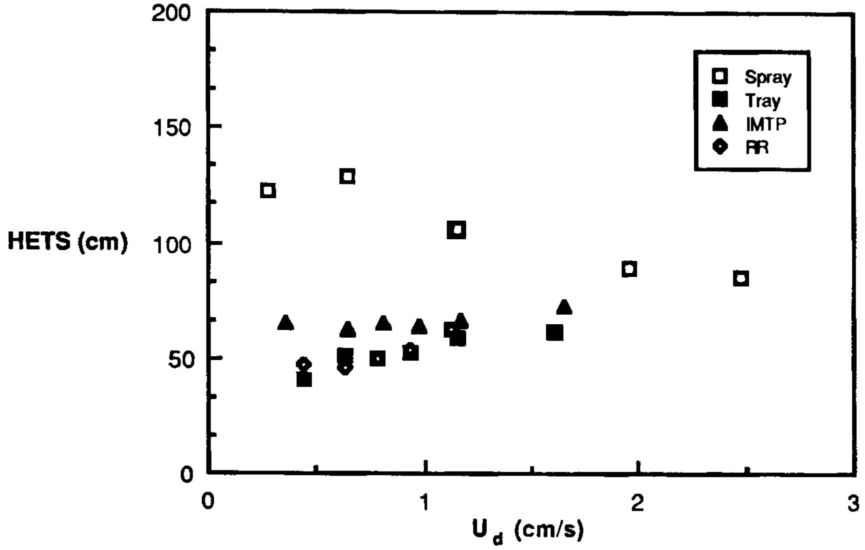

Figure 8. Comparison of Contacting Devices.
System:Carbon Dioxide/Isopropanol/Water(313°K,102 atm)
Dispersed Phase: Carbon Dioxide. U_c: 0.048 cm/s.

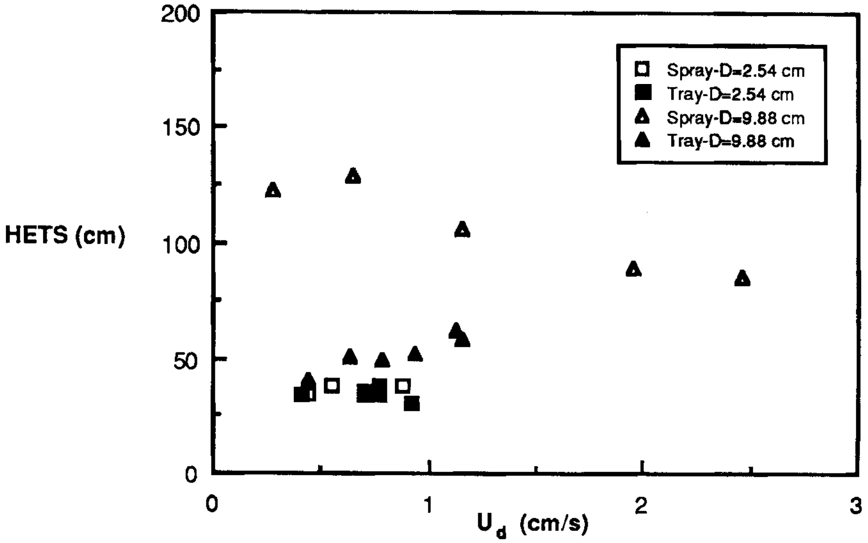

Figure 9. Effect of Column Diameter.
System:Carbon Dioxide/Isopropanol/Water(102 atm,313°K).
Dispersed Phase: Carbon Dioxide. U_c: 0.048 cm/s.

somewhat favorably. However, there is a noticeable difference in the efficiency of the spray column. This is likely due to the decreased holdup and increased backmixing experienced in the 9.88 cm spray column.

Conclusions

The hydrodynamic and mass transfer characteristics were observed to behave similarly to those of conventional liquid-liquid extraction. The mass transfer efficiency of dense carbon dioxide was observed to be essentially independent of operating temperature and pressure. The mass transfer efficiency of each contacting device was observed to improve with increasing dispersed phase flow while greater continuous phase flows yielded slightly greater values of HTU_{OC}. In this work, the spray column was shown to be very inefficient, which differs from an earlier work using a much smaller column diameter.

Nomenclature

A_c	Cross-sectional area of column, cm2·
A_p	Packing surface area, cm2/cm3
C	Concentration, g/cm^3
D	Column diameter, cm
d_{vs}	Sauter mean drop diameter, mm
E	Packing void fraction
HETS	Height equivalent to a theoretical stage, cm
HTU_{OC}	Height of a transfer unit based on the continuous phase, cm
m_{dc}	Equilibrium distribution coefficient (dC_d^*/dC_c)
NTU_{OC}	Number of transfer units based on the continuous phase
Q_d	Volumetric flowrate of the dispersed phase, cm^3/s
U_c	Superficial velocity of the continuous phase based on the column area, cm/s
U_d	Superficial velocity of the dispersed phase based on the column area, cm/s
U_{drop}	Drop velocity, cm/s

X_f	Mass fraction of isopropanol in the aqueous feed
X_r	Mass fraction of isopropanol in the raffinate
Y_f	Mass fraction of isopropanol in the CO_2 feed
Y_e	Mass fraction of isopropanol in the extract
Z	Contacting height, cm
Greek Symbols	
ϕ_d	Dispersed phase holdup, fraction of void volume occupied by the dispersd phase
λ	Extraction factor = $U_d m_{dc}/U_c$

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