

OPTIMIZED VALUES FOR A SIEVE PLATE PULSED COLUMN FOR ACETIC ACID, WATER AND KEROSENE SYSTEM

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Abstract

An experimental study was conducted using sieve plate pulsed column. Correlations were developed among different variables with the help of data obtained from operation of the pulsed column in polynomial form. These correlations could be utilized to get an optimum value for acetic acid, water, kerosene system. Flow rate of solvent, concentration of aqueous solution of acetic acid and pulsation frequency were taken as independent parameters; whereas concentration of acid in extract phase (CE_2) and in raffinate phase (CR_1), percentage equilibrium and height of transfer units (HTU) were treated as dependent parameters. A significant effect of pulsation frequency and flow rates of both the streams have been observed on the concentration of extract, raffinate, percentage equilibrium and height of transfer units. Pulsation frequency of 56 (cycles/min) was found critical beyond which a gradual change in all the profiles were observed.

Keywords: *Height of transfer unit, liquid-liquid extraction, pulsation frequency*

Introduction

Liquid-liquid extraction (LLE) is used for separating components in a mixture by virtue of their distribution between two immiscible liquid phases. It involves the transfer of mass from one liquid phase into a second immiscible liquid phase. Transfer of the dissolved component (solute) may be enhanced by the addition of salting out agents to the feed mixture or by adding complexing agents to the extraction solvent. Sometimes a chemical reaction could be used to enhance the transfer. Pulsed Columns are extractors in which a rapid reciprocating motion of relatively short amplitude is applied to the liquid mixture. Produced agitation has been reported to impart improved rates of extraction (Akhtar et al., 2002, Daood et al., 2007). This phenomena help to reduce tower heights and consequently the expense of massive shielding in case of radioactive materials. The pulsing provides a means of agitation thus eliminating the need of any moving parts. This is very useful when highly corrosive and dangerously radioactive liquids are used such as for the extraction and separation of metals from radioactive solutions (Din et al., 2008, 2009). In LLE, it is essential that the feed mixture and the solvent must be immiscible to some extent. In short, LLE involves three stages.

- a) Bringing the feed mixture and the solvent into intimate contact
- b) Separation of the resulting two phases
- c) Removal and recovery of the solvent from each phase

Literature Review

Drop size distribution in a pulsed sieve-plate extraction column for the acetic acid-water-ethyl acetate system has been studied. Different pulsation intensities were maintained for the same flow rates of dispersed and continuous phases. Mean drop size and distribution of drop size were significantly affected by the variation in pulsation intensity. Height of transfer units were observed to be decreased with the decrease in the drop diameter. Based on the results of experimentation a semi empirical correlation for the estimation of mean drop diameter was established. Further it was observed that the drop size was decreased with an increase in pulsation intensity; however the decrease was less pronounced at the higher pulsation intensities. Difference between the maximum and minimum mean drop sizes was decreased with the increase in pulsation intensities whereas concentration of smaller drops was increased. Height of transfer unit was decreased with decrease in drop diameter. These results suggested good mass transfer efficiency at higher pulsation intensities (Hea et al., 2004).

Mass transfer characteristics of a dilute organic acid solution were studied in a pulsed sieve-plate extractor. Mass transfer experiments were conducted on 10% (w/w) acetic-acid solution in water using ethyl acetate as a solvent. Longitudinal concentration profiles were developed for both the continuous and dispersed phases. Effects of pulsation intensity, continuous phase and dispersed phase superficial velocities on apparent heights of transfer units were evaluated. For a range of conditions the column proved to be more efficient at higher pulsation intensities and dispersed phase superficial velocities. It was observed that mass transfer performance can be increased with an increase in pulsation intensity. However after a certain limit of pulsation intensity, the mass transfer performance was not considerably improved. Initial increase in the dispersed phase velocity could significantly enhance the column performance but gradual increase in dispersed phase velocity gradually decreased the advantage expected (Li et al., 2000).

The effect of solvent – solution flow rates, the solvent –solution flow ratio, pulsation frequency and feed concentration on extraction efficiency were studied (Li et al., 2000). Experimental results showed that by increasing the solvent solution flow rates from 0.0974 ml/s to 6.82 ml/s, the extraction efficiency was increased from 6.77 % to 24.73 % respectively. However, increasing solvent – solution flow ratio from 0.583 to 1.0, the extraction efficiency was increased from 18.25 % to 20.53 %. Further increase in flow ratio from 1.0 to 1.55 resulted in the extraction efficiency to decrease from 20.53 to 18.25 % .Increasing the pulsation frequency from 0.367 to 1.8 cycles/s, the extraction efficiency was increased from 18.25% to 31.31%. Comparison between 5% and 10% acid

concentration in feed showed that the extraction efficiency was higher at 10% concentration compared to 5% acetic acid concentration.

The role of pulsation frequency stroke length, continuous phase and dispersed phase, flow rates on mass transfer characteristics i.e. percentage of solute in extract phase was also analyzed using acetic acid, water and ethyl acetate as solvent. Increase in pulsation frequency resulted in substantial decrease in the time required to attain steady state for percentage of solute in extract phase. Steady state values for percentage of solute in extract phase was obtained with 67% saving in time for higher value of pulsation frequency. Increase in the value of percentage of solute in extract phase was rapid initially for values of pulsation frequency from 10 to 35 strokes /min as compared with higher values of pulsation frequency. Also a substantial increase was observed with stroke lengths from 0 to 15 mm and the percentage of acetic acid in extract phase was increased from 6.2% to 7.0% (Usman et al., 2006).

Measurements of flooding in a pulsed sieve plate extraction column were carried out using toluene - water and toluene - acetone - water systems. The influence of plate spacing and initial acetone mass fraction in toluene for three-component system on flooding was studied and empirical correlations for the maximum of these curves are presented. In the ternary system, acetone goes from toluene drops to water: the descending phase. An experimental pulsed column, with an internal diameter of 39.6×10^{-3} m and an operational height of 2,670 m, and with stainless steel perforated plates of thickness 1.5×10^{-3} m, having a hole diameter of 3×10^{-3} m were used. It was verified that the plate spacing and initial acetone mass fraction in the toluene had a great influence on throughput at maximum frequency (Usman et al., 2008).

An attempt has been made to validate the correlations for flooding, drop size and hold-up. The correlations were tested against a set of experimental data gathered from the work of different investigators over the past 50 years. The data covers a wide variation in geometrical and operating parameters of the pulsed sieve plate column and physical properties of the liquid-liquid systems. The exercise helped in identifying correlations that can be used for design of pulsed columns (Usman et al., 2008).

In the present experimental study kerosene is used as solvent for extracting acetic acid from aqueous solution. Experimental data was used to develop correlations between concentration of extract and raffinate phases against solvent and feed solution flow rates, flow ratios and pulsation frequency. The result of present study could be used to reduce experimental work associated with scale up of the pulsed sieve plate column.

Material and Methods

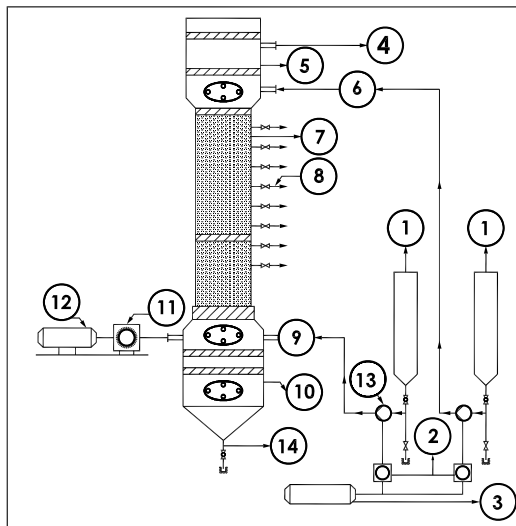
The column has two input streams and two output streams. Aqueous solution (Acetic acid-Water) containing the solute to be extracted was introduced at the top of column. The solvent (kerosene) was introduced at the bottom to extract the solute from the solution. One output stream contains the extracted solute leaving from the top of the column. Second output stream contains only small amount of solute leaving at the bottom of column.

The internal diameter of sieve plate pulsed column was 5 cm with height of 465 cm. It consisted of eighty 1mm thick stainless steel sieve plates. These plates had thirty two, 3 mm diameter circular holes on triangular pitch of 5 mm. 8 sampling points were provided in the column. Settling section was of 10.3 cm diameter with length of 50 cm. Feed cylinders were of 4.5 cm in diameter and 96 cm in length. The column was divided in to three major sections:

- 1) Extraction section
- 2) Extract phase separation section
- 3) Raffinate phase separation section

Separate Feed pumps were available for solvent and aqueous solution from feed cylinders to extraction column. Pulsation can be produced in the system by reciprocating type piston pump. A dial (with safety knob) was available to control the pulsation frequency.

Figure 1: Diagram of sieve plated pulsating Extraction Column



Legends used

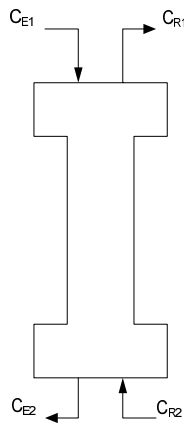
- 1) Feed Cylinders
- 2) Gear box for feed pump
- 3) Motor for feed pump
- 4) Extract outlet
- 5) Kerosene Settling section
- 6) Feed solution inlet
- 7) Extraction Section
- 8) Sampling point
- 9) Solvent inlet
- 10) Settling section for higher liquid
- 11) Gear box for pulsation pump
- 12) Motor for pulsation pump
- 13) Feed pump
- 14) Raffinate outlet

1. Mass Transfer Calculations

Percentage of equilibrium attained and height of transfer unit (HTU) were used as extractor performance criteria. Percentage equilibrium was defined as the percent change in aqueous phase acid composition divided by the change necessary (if the aqueous phase concentration were in equilibrium with the incoming acetic acid- Kerosene feed).

Hence

$$\text{Percentage Equilibrium} = \frac{C_{E2} - C_{E1}}{C_{E2}^* - C_{E1}}$$



C_{E1} = concentration of acid in aqueous feed solution.

C_{R2} = concentration of acid in solvent.

C_{R1} = concentration of acid in raffinate phase.

C_{E2} = concentration of acid in extract phase.

C_{E1}^* = concentration of acid in aqueous feed solution at equilibrium.

C_{E2}^* = concentration of acid in water at equilibrium

The height of the overall transfer unit based on extract phase was also kept as the performance criteria of the system. Procedure used for calculations of height of transfer unit was simplified by the use of log mean concentration driving force which was a reasonable assumption when the solvents were immiscible and concentration changes were small. In the absence of diffusion resistance at the interphase, the number of transfer unit based on water phase could be determined by dividing the change in concentration with the log mean driving force.

$$\text{Number of transfer unit (NTU)} = \frac{C_{E2} - C_{E1}}{\Delta C_{lm}} \quad \text{and}$$

$$\Delta C_{lm} = \frac{(C_{E1}^* - C_{E1}) - (C_{E2}^* - C_{E2})}{\ln\left(\frac{C_{E1}^* - C_{E1}}{C_{E2}^* - C_{E2}}\right)}$$

$$\text{Height of transfer unit (HTU)} = \frac{H}{(NTU)} \quad \text{where, H is height of column.}$$

2. Effect of flow rate variation in solvent and acetic acid-water solution on acid concentration

Flow ratio and pulsation frequency were kept same while flow rates of both solvent and aqueous solutions were gradually changed by controlling the percentage opening of feed pump inlet. Concentration of acid in aqueous solution inlet stream was (C_{E1}) = 0.0 g-mole/l and the concentration of acid in solvent (kerosene) inlet stream was (C_{R2}) = 0.77 g-mol/l. Concentration of acid in extract phase (CE_2) and concentration of acid in raffinate phase (CR_1) were noted and plotted against volumetric flow rate of kerosene and aqueous solution as shown in Figures [2] and [3]. Percentage equilibrium and height of transfer units were calculated and their profiles were plotted against volumetric flow rate of solvent and feed solution as shown in Figures [4] and [5]. The solid line in all the plots is the actual profile made by connecting all points on the graph and dotted line represents the 4th degree polynomial curve. The polynomial equations which follow the curve fitting are given at the bottom of each plot. The “x” values are flow rate of solute and acetic acid-water solution while “y” values are the concentration of acid in extract phase (CE_2), the concentration of acid in raffinate phase (CR_1), percentage equilibrium, and height of transfer units.

Figure 2: Concentration of acid in extract phase vs. flow rate of solvent and feed solution

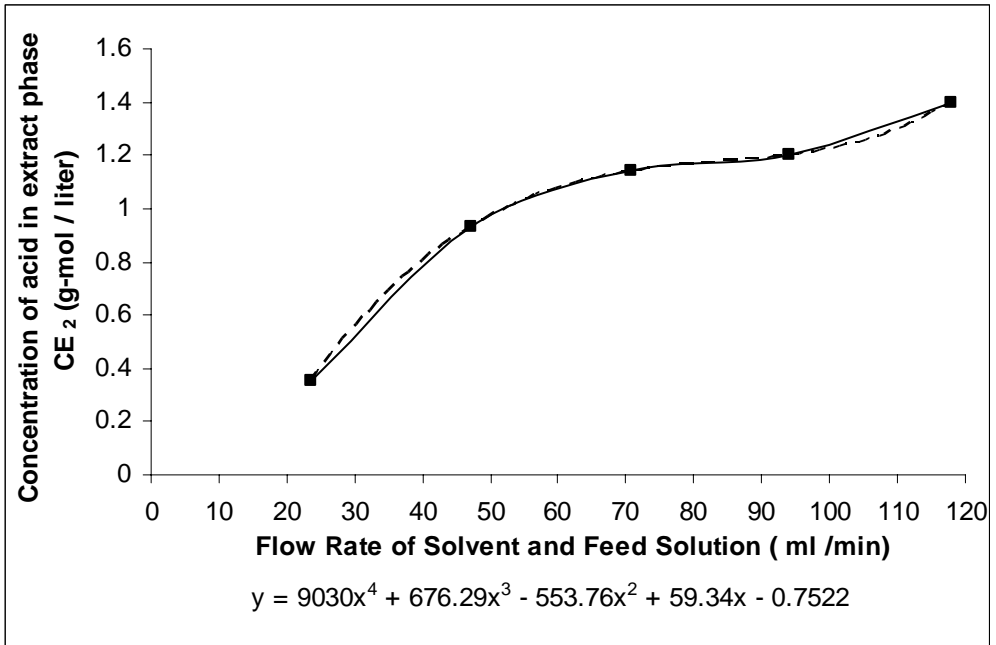


Figure 3: Concentration of acid in raffinate phase vs. flow rate of solvent and feed solution

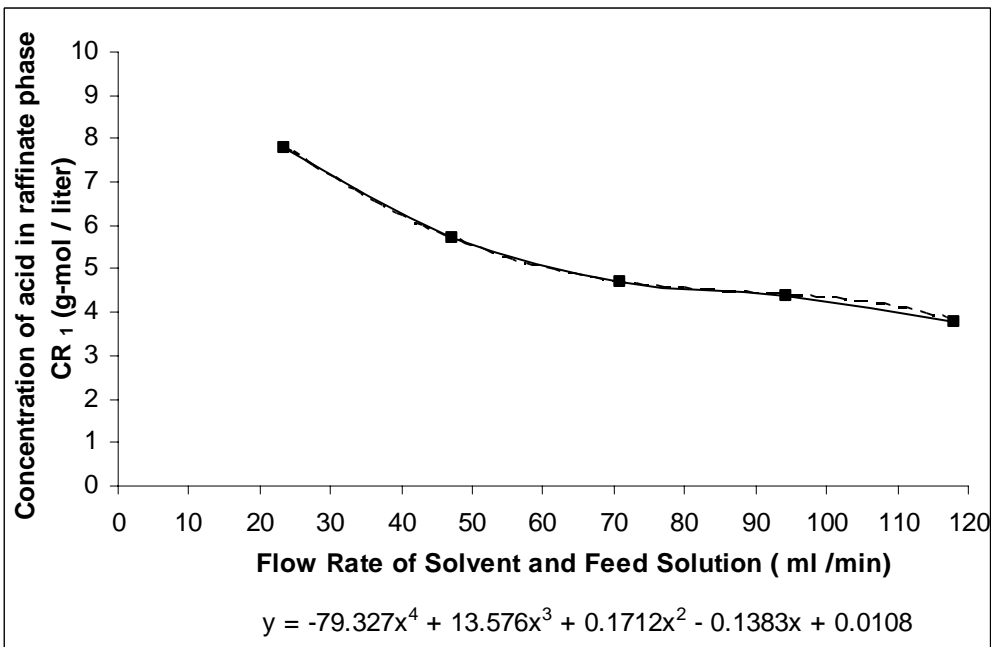


Figure 4: Percentage equilibrium vs flow rate of solvent and feed solution

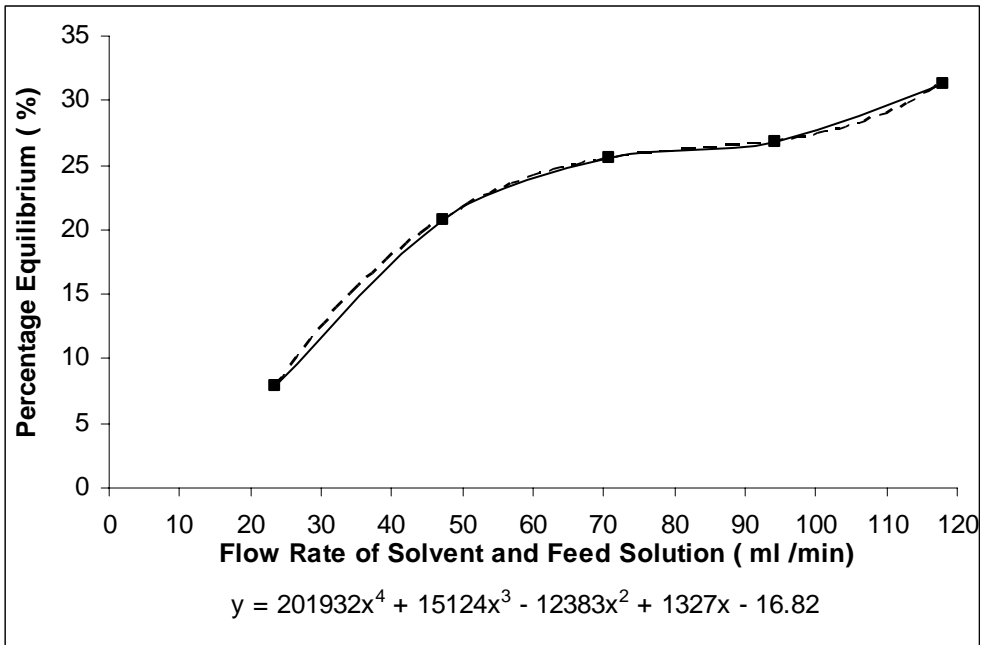


Figure 5: Height of transfer units vs flow rate of solvent and feed solution

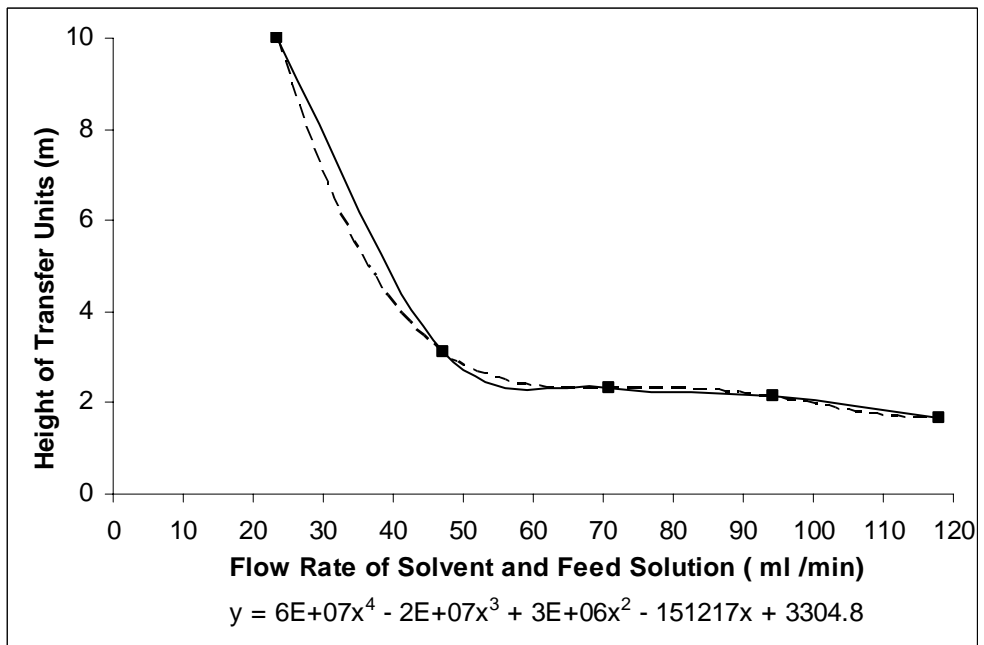


Figure 2 shows a gradual increase in concentration of acid in extract phase (CE_2) with increase in flow rate. Figure 3 shows a gradual decrease in concentration of acid in raffinate phase (CR_1) with increase in flow rate. Figure [4] shows a gradual increase in percentage equilibrium with increase in flow rate. The profile is same as that of concentration of acid in extract phase (CE_2) with respect to flow rate. It could be concluded that percentage equilibrium is directly related with concentration of acid in extract phase (CE_2). Figure 5 shows a sharp decrease in height of transfer units with increase in flow rate initially. However, a constant decrease in profile of height of transfer units could be observed with increase in flow rate of solvent and aqueous solution. A fourth degree polynomial equation is obtained from each of these profiles which could further be used to determine the optimum values.

3. Effect of change in flow ratio of feed solution and solvent

Pulsation frequency and flow rate of solvent at 25% opening of pump were kept constant while flow rate of aqueous solution was gradually changed by controlling the percentage opening of pump to (13.5%, 16%, 25%, 43% and 100%). Concentration of acid in solvent inlet stream was (C_{E1}) = 0.0 g-mole/l and concentration of acid in kerosene was (C_{R2}) = 0.77 g-mol/l. Concentration of acid in extract phase (CE_2) and concentration of acid in raffinate phase (CR_1) were noted and plotted against flow ratio (ratio of solvent and aqueous solution flow rate). Percentage equilibrium and height of transfer units were also calculated and their profiles were plotted against flow ratios of solvent and aqueous solution. Again a fourth degree polynomial equation is obtained for each profile similar to the profiles obtained earlier using curve fitting methods.

Figure 6: Concentration of acid in extract phase vs flow ratio of solvent and feed solution

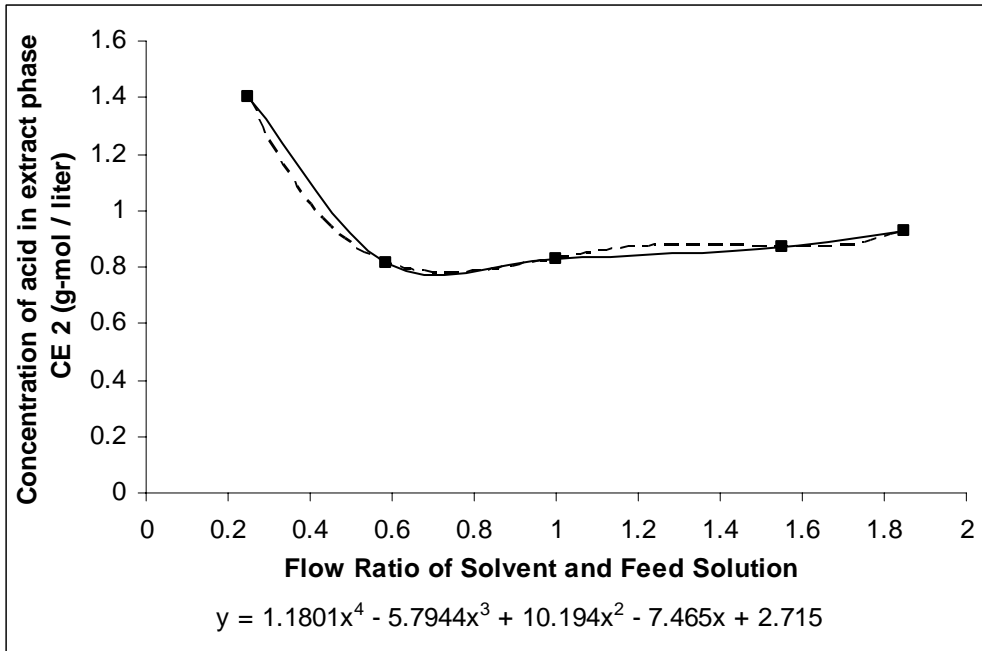


Figure 7: Concentration of acid in raffinate phase vs flow ratio of solvent and feed solution

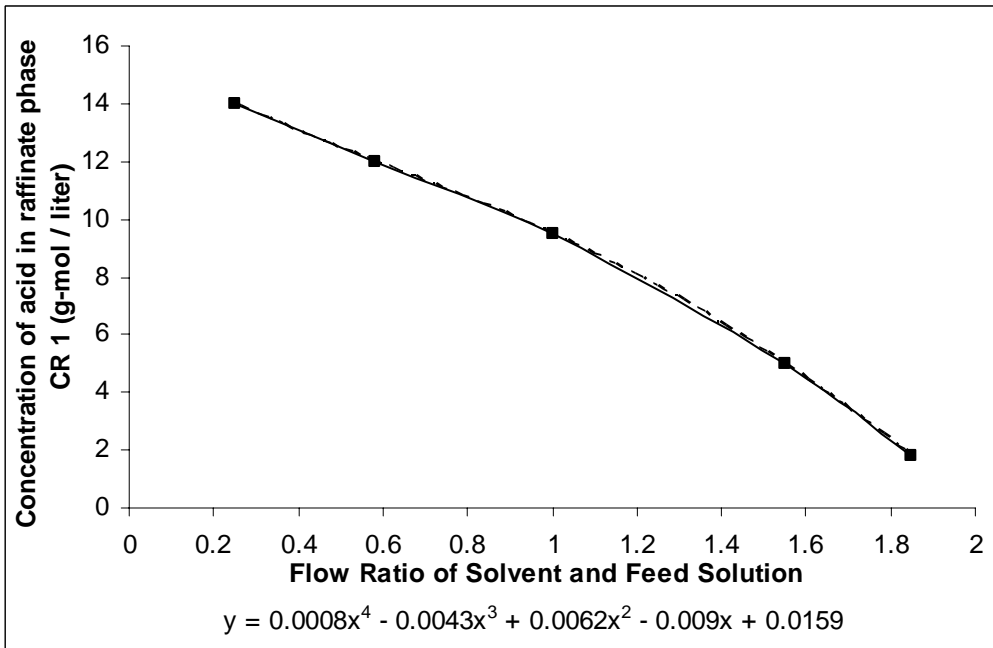


Figure 8: Percentage equilibrium vs flow ratio of solvent and feed solution

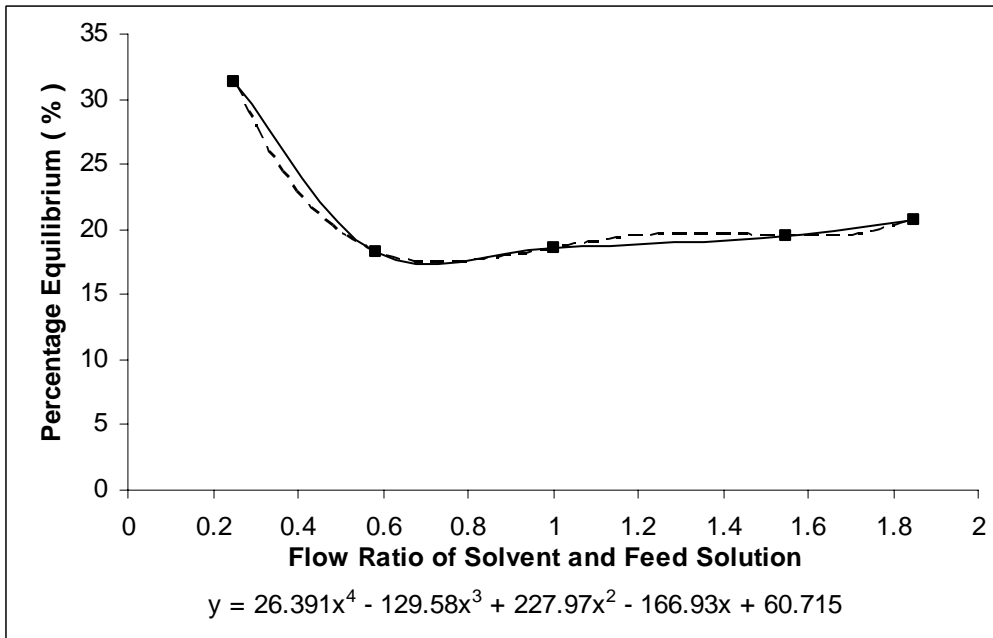


Figure 9: Height of transfer units vs flow ratio of solvent and feed solution

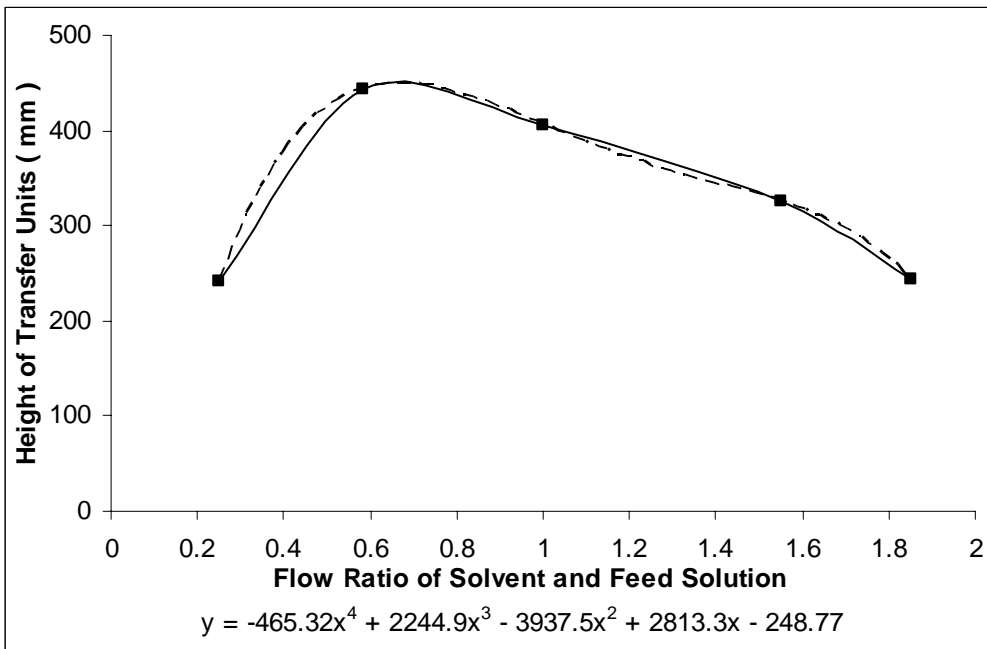


Figure 6 shows initially a gradual decrease in concentration of acid in extract phase (CE₂) with increase in flow rate. However, there is a gradual increase with

increase in flow ratio after the flow ratio of 0.75. Figure 7 shows a gradual decrease in the concentration of acid in raffinate phase (CR_1) with increase in flow rate. Figure 8 shows the same profile as that of Figure [6]. Hence, a conclusion similar to section 3.2 above could be drawn i.e. the percentage equilibrium is directly related with concentration of acid in extract phase (CE_2). Figure 9 shows a sharp increase in height of transfer units with increase in flow rate in the beginning but a gradual decrease with increase in flow ratio after the flow ratio of 0.75. Hence, it could be concluded that height of transfer units is inversely proportional to percentage equilibrium. Similar to the observations made earlier, a fourth degree polynomial equation is obtained for each of the profiles.

4. Effect of pulsation frequency

Flow rates of solvent and aqueous solution were kept constant while pulsation frequency is gradually changed. Concentration of acid in solvent inlet stream was (C_{E1}) = 0.0 g-mole/l and concentration of acid in acetic acid-water was (C_{R2}) = 0.77 g-mol/l. Concentration of acid in extract phase (CE_2) and concentration of acid in raffinate phase (CR_1) were noted and plotted against pulsation frequency. Percentage equilibrium and height of transfer units were calculated and their profiles were plotted against pulsation frequency.

Figure 10: Concentration of acid in extract phase vs pulsation frequency

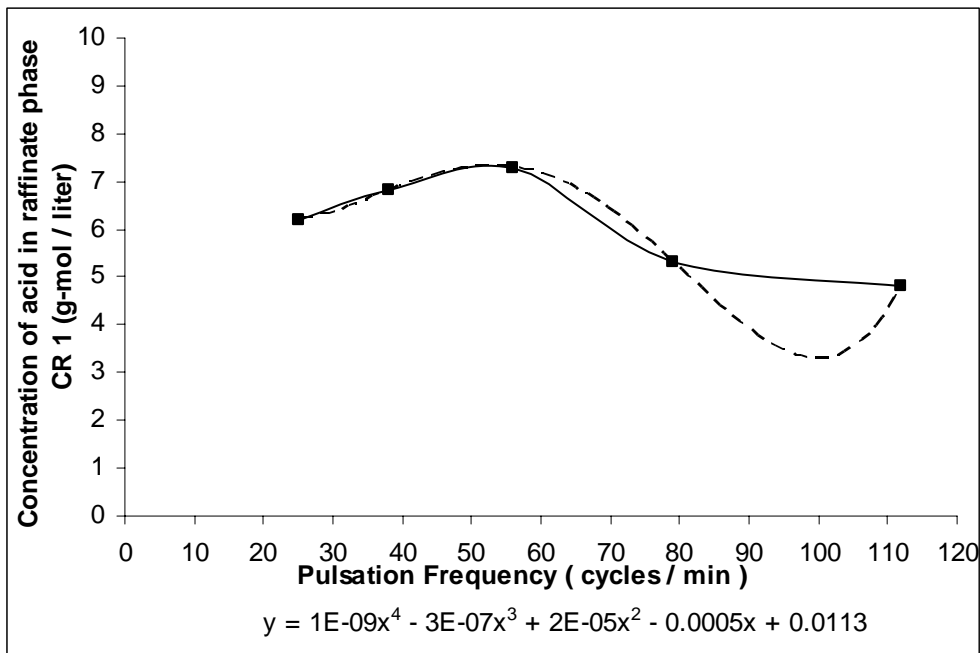


Figure 11: Concentration of acid in raffinate phase vs pulsation frequency

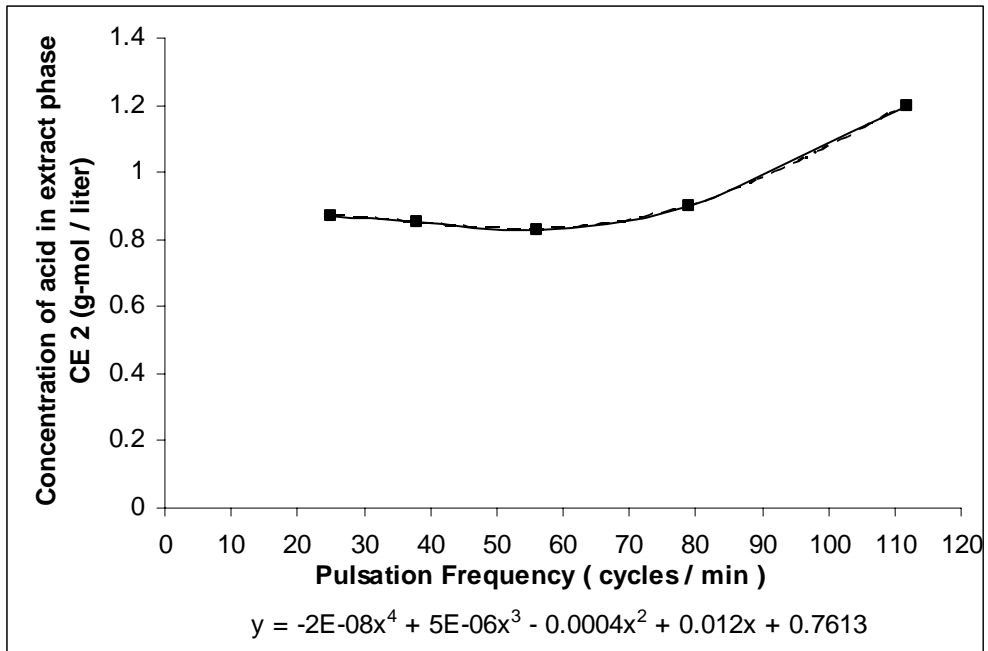


Figure 12: Percentage equilibrium vs pulsation frequency

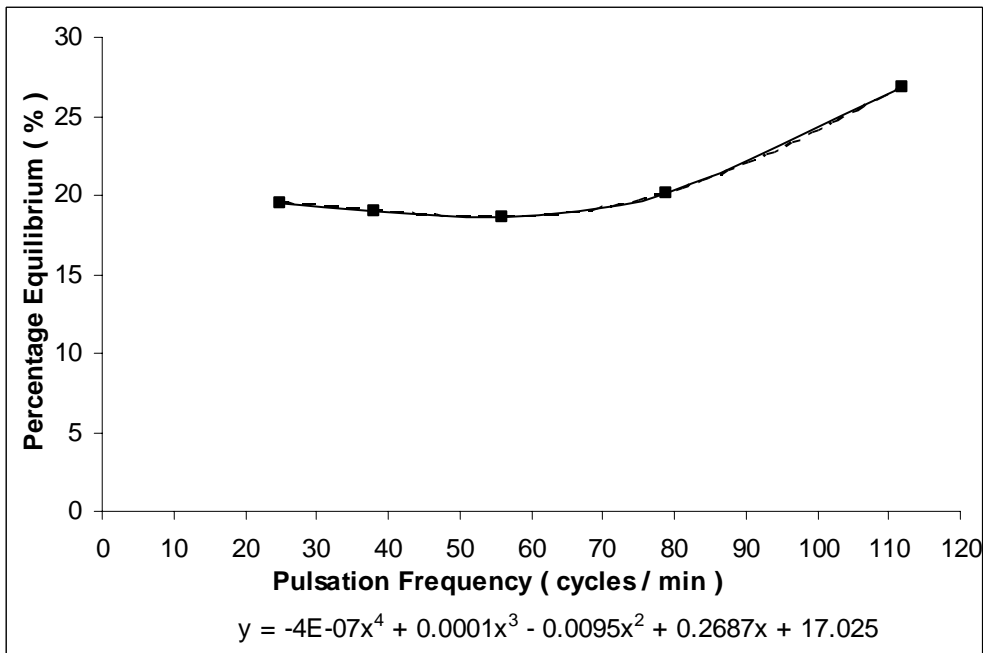
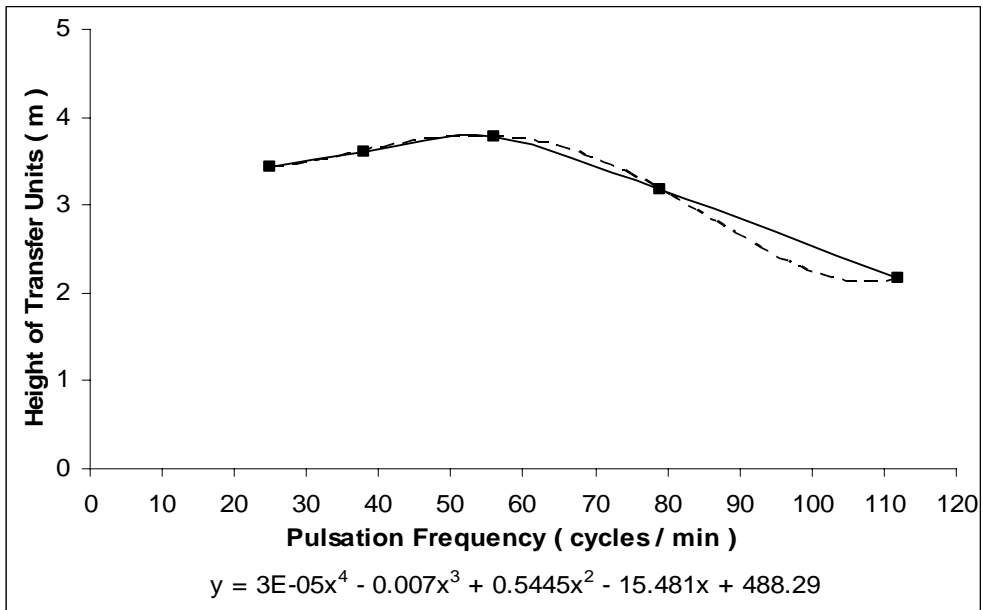


Figure 13: Height of transfer units vs pulsation frequency



It has been observed that pulsation frequency of 56 (cycles/min) is the critical point after which profile of all parameters with respect to pulsation frequency were changed. Figure 10 shows a gradual decrease in concentration of acid in extract phase (CE_2) with increase in flow rate initially but a sharp increase after the critical point. Figure 11 shows a gradual increase in the concentration of acid in the raffinate phase (CR_1) with increase in flow rate in the beginning but a sharp decrease in the profile after the critical point. Figure 12 shows a gradual decrease in the percentage equilibrium with increase in flow rate in the beginning but a sharp increase in the profile after the critical point. Figure 13 shows a gradual increase in height of transfer units with increase in flow rate in the beginning but a sharp decrease in the profile after the critical point.

Results and Discussion

Pulsation frequency and flow rates of both the streams have significant effects on the concentration of extract, raffinate, percentage equilibrium and height of transfer units. Polynomials of specific degree have been plotted to show the behavior of various parameters relative to the operational performance of liquid liquid pulsed extraction column. The parameters were concentration of acid in extract phase (CE_2), concentration of acid in raffinate phase (CR_1), percentage equilibrium and height of transfer unit (HTU). Pulsation frequency of 56 (cycles/min) was observed to be the critical point after which a gradual change was observed in all the profiles plotted with respect to pulsation frequency. Fourth degree polynomial equations were obtained for each of the profiles

obtained for various parameters. These equations could be utilized to determine the optimum values for such systems.

References

- Akhtar, N.A., Butt, M.A., Imran, M., Jameel, A., Aashiq, M. and Sattar, A (2002), "Mass Transfer Characteristics of a Sieve Plate Pulse Column", *Pakistan Institute of Chemical Engineering*. XXXI, 1-4.
- Daood, S. S., Munir, S., Akhtar, N.A. and Butt, M.A.(2007), "Effect Of Operational Parameter on Extraction in Sieve Plated Pulsed Extraction Column", *Proc. Pakistan Acad. Sci.* 44(3):189-195.
- Din, G., Chughtai, I. R., Inayat, M. H. and Khan, I. H. (2008), "Axial dispersion, holdup and slip velocity of dispersed phase in a pulsed sieve plate extraction column by radiotracer residence time distribution analysis", *Applied Radiation and Isotopes*, 66, 1818–1824.
- Din, G., Chughtai, I. R., Inayat, M. H. and Khan, I. H. (2009), "Study of axial mixing, holdup and slip velocity of dispersed phase in a pulsed sieve plate extraction column using radiotracer technique", *Applied Radiation and Isotopes* (Article available online)
- Hea, C.H., Gao, Y. H., Yang, S.H. and Edwards, D. W.(2004), "Optimization of the process for recovering caprolactam from wastewater in a pulsed-sieve-plate column using green design methodologies", *Journal of Loss Prevention in the Process Industries* 17, 195–204.
- Li, H.B., Luo, G.S., Fei, W.Y. and Wang, J.D.(2000), "Mass transfer performance in a coalescence–dispersion pulsed sieve plate extraction column", *Chemical Engineering Journal*, 78, 225–229.
- Usman, M.R., Hussain, S.N., Rehman, L., Bashir, M. and Butt, M.A.(2006), "Mass Transfer Performance in a Pulsed Sieve-Plate Extraction Column", *Proc. Pakistan Acad. Sci.* 43(3):173-179.
- Usman, M. R, Rehman, L. and Bashir, M. (2008), "Drop Size Distribution in a Pulsed Sieve-Plate Extraction Column", *Proc. Pakistan Acad. Sci.* 45(1):41-46.