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DROP SIZE AND DISPERSED PHASE HOLDUP IN A SPRAY COLUMN

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Drop size and dispersed phase holdup in a spray column for a reactive extraction system (zinc/di (2-ethylhexyl) phosphoric acid) has been obtained under solute transfer from continuous to dispersed and dispersed to continuous phase. A photographic method is used for the estimation of drop size and dispersed phase holdup. The experiments are carried out in a glass spray column operated under semi-batch mode, i.e. the continuous aqueous phase is kept stagnant and the dispersed phase is not recirculated. Dispersed phase velocity through the nozzles is varied from 3.4 x 10^{-2} to 24.5x10^{-2} m/s. Nozzle diameter ranges from 0.8 x 10^{-3} to 3 x 10^{-3}m.

Key words: Drop size, Dispersed phase, Holdup.

Introduction

Knowledge of the interfacial area between droplets and continuous phase is required for the design of continuous counter-current contactors in which droplets of a liquid are passed through another partially or totally immiscible liquid. Interfacial area is important in heat and mass transfer processes and is a function of droplet size and dispersed phase holdup. Interfacial area per unit volume of the column [1] is expressed as:

$$a'_c = \frac{6\,\phi_D}{d_{32}} \quad \text{.............................(1)}$$

whereas the interfacial area per unit volume of the continuous phase can be calculated from the equation [2]:

$$a_c = \frac{a'_c}{1-\phi_D} \quad \text{.............................(2)}$$

The objective of this work is to determine the interfacial area per unit volume of the continuous phase which is required to calculate the metal interfacial flux for extraction of zinc from the acidic aqueous sulphate solutions by di(2-ethylhexyl) phosphoric acid in *n*-heptane.

Theoretical Considerations and Previous Work: *Drop size.* The specific surface diameter or Sauter mean diameter is one of the commonest and most useful quantity for characterizing the drop size of dispersed phase in liquid-liquid systems. It is the diameter of the drop having same volume-surface area ratio as exists in the whole of dispersed phase.

For calculation and design of liquid-liquid extraction columns, it is very important to know the drop size that results from actual operating conditions and different physical properties of the two phases as the drop size influences all relevant parameters related to fluid dynamics as well as mass transfer rates. The rising or falling velocity of the drop swarm depends on the drop size distribution. For that reason mass, transfer efficiency is influenced by drop size as interfacial area and mass transfer coefficients are function of drop size.

Azzopardi [3] reviewed numerous methods for measuring drop size distribution. Photographic methods were recommended for general use when drop sizes in the mm range were involved.

Many investigators took photographs through a window of the column. This technique can be used for high holdups except distances very close to the wall due to interference of drops with the optical path. Chen *et al.* [4] reported the highest holdup possible for photographing in cocurrent flow approximately 7.5%. Beyond this level, drops were overlapping too much and individual drops could not be distinguished. Lin and Ford [5] applied the photographic technique successfully up to a holdup of about 6% in a counter-current flow study in a spray column.

Dispersed phase holdup. Dispersed phase holdup is defined as the volumetric fraction of the dispersed phase which is the total volume of all drops per unit volume of the column. It can be obtained by two methods. At low holdups, ϕ_D<0.03, the holdup can be calculated from experimentally determined drop velocities. At higher holdups, ϕ_D > 0.03, the holdup can be obtained by closing off a section of the column suddenly with quick closing valves, built into the column, and by measuring the height of coalesced dispersed phase after complete phase separation trapped in that section. The ratio of the trapped coalesced phase volume to the volume of enclosed section gives the holdup.

Lapidus and Elgin [6] proposed that for the vertical flow of solid particles in a fluid, for each particle size, there exists a single function correlating the holdup (ϕ_D), the volume concentration of dispersed phase and with slip velocity (U_S), the relative velocity of the dispersed and continuous phases.

$$U_S = f(\phi_D) \quad \ldots\ldots \quad (3)$$

Weaver *et al.* [7] investigated liquid organic drop holdup in a stagnant water phase. Beyaert *et al.* [8] extended the work to a flowing continuous phase. They found good correlations up to flooding point, between holdup and slip velocity for the operation of a spray column.

Thornton [9] correlated the holdup and superfacial velocities of dispersed and continuous phases for counter-current flow in a spray column by the function:

$$\frac{U_d}{\phi_D} + \frac{U_c}{1-\phi_D} = U_O(1-\phi_D) \quad \ldots\ldots \quad (4)$$

where U_0 is a characteristic velocity, obtained by extrapolating the mean relative velocity of the drops to zero flow rates.

The correlation of Zenz [10] for fluidized solids was found to apply for liquid-liquid systems in the non-flooded conditions. Solution with Zenz equation requires a trial and error procedure. Liquid-liquid systems present a rather narrow range on the ordinate of the Zenz plot. At any point on the ordinate, the slip velocity can be defined by the equation:

$$U_S = f(\phi_D)\, U_t \quad \ldots\ldots \quad (5)$$

Hughmark [11] plotted the Weaver's data [7] and gave equations for static holdup, ϕ, as a function of U_d/U_t.

$$\text{For } \frac{U_d}{U_t} < 0.02, \quad \phi = \frac{U_d}{S\,U_t} \quad \ldots\ldots \quad (6)$$

He also recommended that the static holdup, ϕ, can be used to determine holdup for the counter-current system, ϕ_D, by the equation:

$$U_S = \frac{U_d}{\phi} = \frac{U_d}{\phi_D} + \frac{U_c}{1-\phi_D} \quad \ldots\ldots \quad (7)$$

Actually, the Zenz correlation should be used to obtain the holdup for static continuous phase condition, ϕ, and equation (7) should be employed to calculate the counter-current holdup, ϕ_D, rather than the Zenz correlation with slip velocity equation given as:

$$U_S = \frac{U_d}{\phi_D} + \frac{U_c}{1-\phi_D} \quad \ldots\ldots \quad (8)$$

The Hu and Kintner's correlation [12] with the Johnson and Braida's corrections [13] for continuous phase viscosity may be used to calculate terminal velocities. An alternative method for evaluating terminal velocity is obtainable from the correlations of Klee and Treybal [14] and Grace *et al.* [15].

Kumar *et al.* [16] developed a correlation for slip velocity in terms of average drop diameter in a swarm of drops, dispersed phase holdup and physical properties of the system. This relationship can be used in conjunction with equation (8) to calculate a wide range of dispersed phase holdups if continuous and dispersed phase flow rates are known.

More recently, Kumar and Hartland [17] presented empirical equations for slip velocity and dispersed phase holdup independent of each other using a large number of published data and claimed that these equations are more exact and simpler to use than previous empirical correlations which often involve the characteristic velocity which is difficult to accurately predict.

Experimental

Reagents. The zinc sulphate, sodium sulphate and sulphuric acid used were of Analar grade. They were used as supplied by the manufacturers without further purification.

The *n*-heptane supplied by Shell Company was of Knok-testing grade and was used without further purification. The di(2-ethylhexyl) phosphoric acid (DEHPA), was of technical grade obtained from BDH Ltd. This was purified according to the method of Partridge and Jensen [18].

Kodak D-19 Developer, Kodak D-19 Replenisher and Fixer were used for the development of films of drops. Some films were also developed with Kodak D-76 Developer.

Procedure. Experiments were performed in a glass spray column of 1.55m effective height and 0.05m diameter (Fig. 1). The dispersed phase was introduced through 4 nozzles situated at the base of the column. The drops were formed on sharp edged glass capillary nozzles. The four nozzles were located on the corners of a square. Three different nozzle sets were used. The nozzle tips were immersed in the stationary continuous phase, contained in the column. The flow rates through the nozzles were measured and controlled individually. Two flat windows, one at the top and the other at the bottom, allow for the dispersion to be photographed with a cine camera. The column was operated in a semi-batch mode, i.e. the continuous aqueous phase was kept stagnant and the dispersed phase was not recirculated. Full details of the apparatus description and experimental procedure are given elsewhere [2].

Photographic setup for drop size studies. A photographic method was used to measure drop size and drop size distributions during extraction and stripping runs. In order to reduce deformation due to curvature of the wall, flat windows were placed at photographic locations. A scale of 1 mm divisions on transparent plastic sheet was pasted on the front window to provide a reference. A hollow cylinder of black hard paper was fixed around the windows to prevent scattered light from entering the camera. A sheet of tracing paper was placed between the light source and the column window to act as light diffuser, thus providing even, uniform lighting. The diffuser was close to the light source. The best contrast between the continuous phase and drops was obtained by illuminating the glass window from behind with a 200 W lamp at approximately 60 cm from the window. This procedure also prevented unwanted reflections.

Photographs of the drop swarm rising past the plane glass windows in the spray column were taken with a Bollex Paillard cine camera using a 10 mm film. The film speed was 64 frames per second. The area photographed was about 6 cm in diameter. The camera was mounted on a tripod and was held fixed throughout the experiment. During an experimental run, shots were taken in approximately 2 sec bursts at about 15 min intervals. Processed negatives were used directly for the measurement of drop sizes with an image analyzer interfaced with an Apple personal computer. The surface area and volume of the drop were calculated from its major and minor axes.

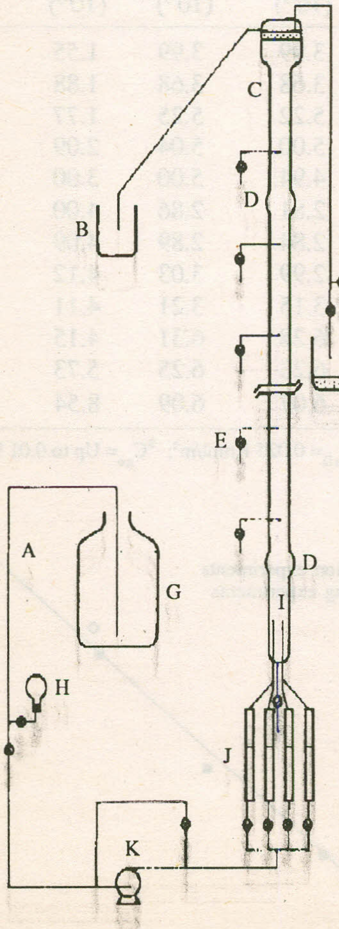

Fig. 1. Schematic flow diagram of the experimental apparatus.
A. Siphon, B. Overflow, C. Column, D. Plane glass windows, E. Sampling tube, F. Continuous phase reservoir G. Dispersed phase reservoir H. Siphon starter (Pipette filler), I. Nozzles, J. Rotameters, K. Magnetically driven centrifugal pumps.

Results and Discussion

Drop size measurement. The dispersion was photographed at two locations along the column height, i.e. 0.14 and 0.9 m from the nozzle. The film clearly showed the presence of shape oscillation, wobbling and zig-zag motion of the drops (Fig. 2). Observed shape oscillations were of the spherical oblate type. An Optomax Image Analyzer (Analytical Measuring Systems, UK) was used employing the standard software and interface for the measurement of major and minor axes of the drops from photographic film for different runs. The scale on the film was used as a reference. In order to simplify the measurement, the drops were assumed to be perfect ellipsoids. The equivalent diameter of single drops, $'d_e$, and the Sauter mean diameter, d_{32}, of the dispersion were calculated by using standard equations [1, 19], namely;

$$'d_e = \sqrt[3]{d_1^2\ d^2} \quad \text{.......................................} \quad (9)$$

$$d_{32} = \frac{\Sigma n\ 'd_e^3}{\Sigma n\ 'd_e^2} \quad \text{.......................................} \quad (10)$$

where d_1 and d_2 are the horizontal and vertical axis lengths of the drops and n is the number of drops of diameter, $'d_e$.

A comparison is shown between drop size determined at two locations in the column in Fig 3. Comparison of the experimentally obtained drop sizes with the predicted ones from different drop size correlations available in the literature is given elsewhere [20].

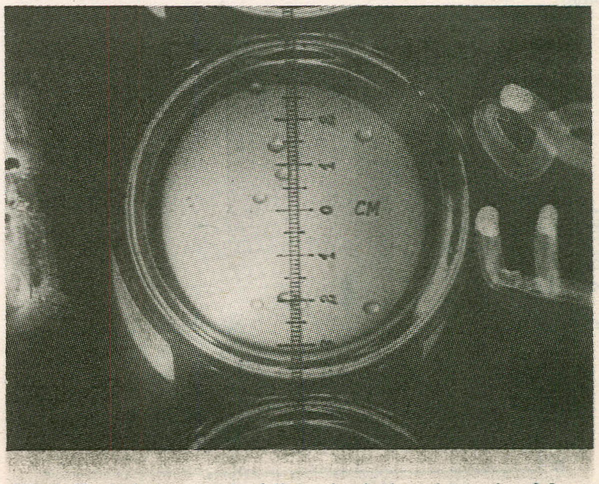

Fig. 2. Typical photograph of dispersion in the column. $d_n = 0.8$ mm, $`L_d = 6.7$ ml/min, $d_e = 3.59$ mm, (extraction experiments). Photographs taken at 90 cm from nozzle tips.

Figure 4 shows the effect of the dispersed phase flow rate on the drop size for different nozzle sizes. It is seen from the figure that the drop size shows a decreasing trend with increasing dispersed phase flow rate in the range studied. The decrease in drop size with increasing nozzle injection velocity is probably due to change in the time of detachment of drop at the nozzle tip as different forces (i.e. forces due to buoyancy, kinetic, drag, interfacial tension and drop acceleration) are acting on a drop during the process of its formation.

Dispersed phase holdup. The dispersed phase holdup is given by:

$$\phi = \frac{n\, \overline{v}_D}{S\, \Delta h} \quad \text{.......................................} \quad (11)$$

where S and v_D are the cross-sectional area of the column and average drop volume respectively.

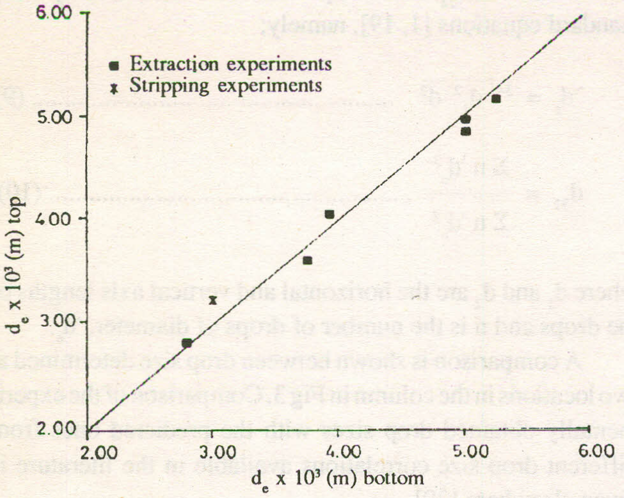

Fig. 3. Comparison of equivalent drop diameter at two locations in the column.

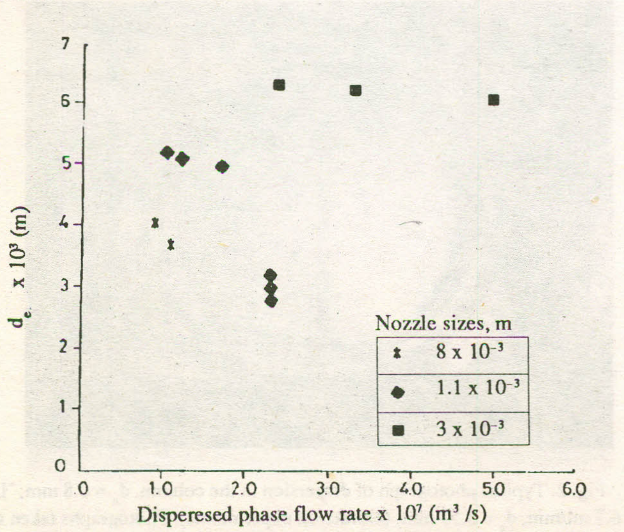

Fig. 4. Effect of dispersed phase flow rate per nozzle on equivalent drop diameter.

The average number of drops, n, contained within a column section of height, Δh, were obtained from the film analysis.

Figure 5 compares the dispersed phase holdup estimated at two heights in the column. The values of hydrodynamic parameters are given in Table 1.

Analysis of the films gave drop size distribution, holdup, specific interfacial area, rising velocity and average residence time. Also the film analysis showed that:

i. dispersed phase holdup does not vary much with height (Fig. 5). The maximum absolute deviation in holdup at 2 locations in the column is about 8%.

ii. specific interfacial area can be taken as independent

TABLE 1. MEASURED AND CALCULATED HYDRODYNAMIC PARAMETERS IN A SPRAY COLUMN FOR ZINC/DEHPA SYSTEM.

d_n, m (10^{-3})	L_d, m^3/s (10^{-7})	d_e, m (10^{-3})	d_{32}, m (10^{-3})	ϕ (10^{-3})	a_c, m^{-1} –
0.8	3.67	3.99	3.99	1.55	2.32
	4.47	3.68	3.68	1.88	3.07
1.1	4.33	5.22	5.25	1.77	2.08
	5.00	5.00	5.04	2.09	2.58
	7.00	4.94	5.00	3.00	3.7
	9.33	2.81	2.86	4.00	8.5
	9.33†	2.84	2.89	4.09	8.61
	9.33*	2.99	3.03	4.12	8.27
	9.33◊	3.15	3.21	4.11	7.8
3	9.67	6.28	6.31	4.15	4.1
	13.3	6.23	6.25	5.73	5.7
	20	6.07	6.09	8.54	8.73

†I = 0.3 Kmol/m^3; *C_{oD}= 0.025 Kmol/m^3; ◊C_{zo}= Up to 0.01 Kmol/m^3 (Stripping).

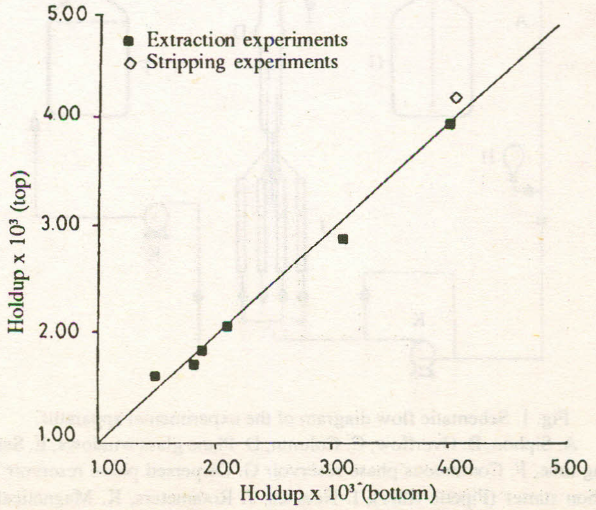

Fig. 5. Comparison between dispersed phase holdup at two locations in the column.

of column height with a maximum absolute deviation of 5% at 2 locations.

Since the terminal velocity of oscillating drops is independent of drop size [12-15], the dispersed phase holdup and specific interfacial area at the 2 locations along the column height does not vary much.

Prediction of dispersed phase holdup. The dispersed phase holdup was predicted from equation (6). The rising velocity in this equation was calculated from different single drop velocity correlations [12-15]. Solution of the single drop velocity correlation led to the calculation of terminal velocity; this was then substituted directly into the equation (6) to predict holdup. The predicted values of holdup are compared with the experimental data as shown in Fig. 6. The experimental values are in good agreement with the predicted ones when calculated terminal velocity from the correlation of Klee and Treybal is used (Fig. 6). Data from all three nozzles are represented in this plot.

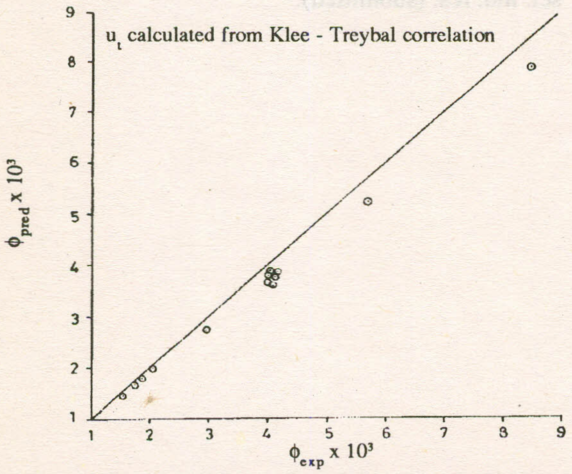

Fig. 6. Comparison of dispersed phase holdups predicted from equation (6) with experimental data.

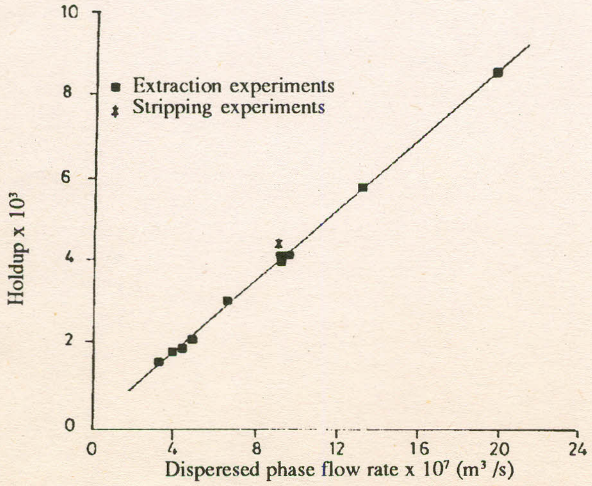

Fig. 7. Effect of dispersed phase flow rate on holdup.

Influence of dispersed phase flow rate in holdup. Figure 7 shows the effect of dispersed phase flow rate on holdup for all nozzle sizes. The holdup increases linearly indicating that terminal velocity of drops is almost independent of the drop size as is the case for oscillating drops [12-15].

Conclusion

Drop size and dispersed phase holdup of the dispersion in a spray column for a reactive extraction system (zinc/di(2-ethylhexyl) phosphoric acid has been obtained.

The drop size shows a decreasing trend with increasing dispersed phase flow rate in the range studied.

The experimental results of dispersed phase holdup are in good agreement with the predicted ones (terminal velocity calculated from Klee-Treybal correlation). The holdup increases linearly with dispersed phase flow rate.

NOMENCLATURE

(The SI system is used throughout this paper)

a_c = Interfacial area per unit volume of continuous phase.
$'a_c$ = Interfacial area per unit volume of column
C = Molar concentration
d_1 = Major axis of drop
d_2 = Minor axis of drop
d_{32} = Sauter mean drop diameter
d_e = Equivalent diameter of the dispersion
$'d_e$ = Equivalent drop diameter of single drops
d_n = Nozzle diameter
I = Ionic strength
L = Volumetric flow rate
n = Average number of drops
S = Cross-sectional area of column
S_D = Average surface area
U = Superfacial velocity through the column
U_o = Characteristic velocity
U_s = Slip velocity of drops relative to continuous phase
U_s = Average rising velocity of drops
U_t = Terminal velocity of drops
V_D = Average drop volume

GREEK LETTERS

ϕ = Dispersed phase holdup for $U_c = o$
ϕ_D = Dispersed phase holdup for countercurrent flow

SUBSCRIPTS

c = Continuous phase
d = Dispersed phase
exp = Experimental values
oD = DEHPA dimer
pred = Predicted values
zo = Organic phase zinc

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References

1. N. V. R. Rao and M. H. I. Baird, Can. J. Chem. Eng., **62**(4), 497 (1984).
2. M. T. Saeed, Ph. D. Thesis, University of London (1989).
3. B. J. Azzopardi, Int. J. Heat and Mass Transfer, **22**, 1245 (1979).
4. G. T. S. Chen, E. H. Young and J. L. York, A. I. Ch. E. Symp. Ser., **70**(178), 144 (1974).
5. F. Lin and J. D. Ford, Can. J. Chem. Eng., **66**, 150 (1988).
6. L. Lapidus and J.C. Elgin, A.I.Ch.E. J., **3**, 63 (1957).
7. R. E. C. Weaver, L. Lapidus and J. C. Elgin, A. I. Ch. E. J., **5** (4), 533 (1959).
8. B. O. Beyaert, L. Lapidus and J. C. Elgin, A. I. Ch. E. J., **7**(1), 46 (1961).
9. J. D. Thorton, Chem. Eng. Sci., **5**(5), 201 (1956).
10. F. A. Zenz; Petrol Refiner, **36**(8), 147 (1957).
11. J. A. Hughmark, Ind. Eng. Chem. Fundam., **6**(3), 408 (1967).
12. S. Hu and R. C. Kintner, A. I. Ch. E. J., **1**(1), 42 (1955).
13. A. I. Johnson and L. Braida; Can. J. Chem. Eng., 165 (1957).
14. A. J. Klee and R.E. Treybal, A. I. Ch. E. J., **2**(4), 444 (1956).
15. J. R. Grace, T. Wairegi and T. H. Nguyen, Trans. I. Chem. E., **54**, 167 (1976).
16. A. Kumar, D. K. Vohra and S. Hartland, Can. J. Chem. Eng., **58**, 154 (1980).
17. A. Kumar, D. K. Vohra and S. Hartland, Can. J. Chem. Eng., **67**, 17 (1989).
18. J. A. Partridge and R. C. Jensen, J. Inorg. Nucl. Chem., **31**, 2587 (1968).
19. F. H. Garner and M. Tayeban, Anal. Real. Soc. Espan. Fis. Quim. (Madrid), **B56**, 479 (1960).
20. M. T. Saeed, M. Jamil and E. S. Perez de Ortiz, Pak. j. sci. ind. res. (submitted).