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THE INDIVIDUAL MASS TRANSFER IN THE ETEROGEN LIQUID-LIQUID SYSTEMS

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ABSTRACT. The improved Lewis cell was used as an efficient device to determine the mass transfer coefficient for any ternary multi-component systems. In this paper the individual mass transfer coefficients were determined for the ternary systems: water-acetone-carbon tetrachloride, using a Lewis cell, operating continuous and batch regime, with a constant stirring rate.

1. INTRODUCTION

The mass transfer intensity can be quantified using the individual mass transfer coefficients for each phase. For the liquid-liquid extraction, the determination of these coefficients is a difficult task that was approached by lots of researchers, using two different ways (Colburn and Welch, 1942; Pratt, 1983).

In one of these ways, two partially miscible liquids were used. One of them is saturated with the other, whose saturation degree is monitored when they are contacted. This method is not very precise and can be applied in a limited number of situations.

In another way, ternary systems are used, having different values of the partition coefficients that are also different from 1. In this case, it may be considered that the mass transfer resistance is located in the phase with the lower equilibrium concentration. This hypothesis involves a great degree of uncertainty (Apreotesei and Tudose, 1998) and the method cannot be applied to any system.

In the both methods shortly described above, one of the contacted phases is dispersed as drops into the other, so that the interfacial area is difficult to determine, because the drops are deformable and the mixing intensity between the two phases is variable (Atagunduz et al., 1968; McManamey, 1961; Harikrishnan et al. 1994).

In this paper, these disadvantages are overcome by using an improved Lewis cell, which operates continuously. The experimental set-up and the operating procedure allow to determine the interfacial areas (contact surface between phases) precisely, while the mixing can be well controlled for every phase.

The Lewis cell (Lewis, 1954) with several improvements remains the most efficient method to determine the mass transfer coefficients for any ternary or for systems with more components, as the G.L. Standard stated. Numerous researchers conceived such modified cells, especially for reactive extraction study (Pratt, 1983).

The Lewis cell presented in this work was used to determine the individual mass transfer coefficients for the ternary system water acetone-carbon tetrachloride (Tudose and Apreotesei, 2000).

To asses the behaviour of the mean residence times in the cell compartments, the extraction of the acetone from carbon tetrachloride, during continuous and batch operation modes, respectively, for the same stirring intensity (850 rot/min) and solute initial concentration (7% weight).

2. EXPERIMENTAL TECHNIQUE

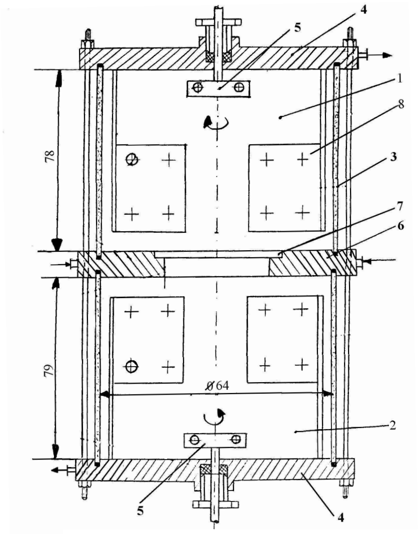

Fig. 1 Extraction cell

The cell used for the study of the mass transfer in continuous and batch operation mode was made out of Cr-Ni stainless steel and glass (Fig. 1). The compartments (1) and (2), where the aqueous and organic phases are introduced have the side walls made out of glass (3). The two edges are equipped with metallic seals (4), which support the bearings of the blade mixer (5), driven by separate engines, with changeable speed, between 80÷1200 rpm. The compartments are separated by a metallic plate (6), which has a central hole with the diameter of 30 mm, where the interface between the two liquids is located (7). The hole diameter can be modified using several rings.

Several orifices were also made in the central plate to assure the feed of the compartments with liquids.

To avoid the interface deformation by the turbulence, three baffles were placed inside each compartment. Each baffle has four holes of a 4 mm diameter. Similar holes were made on the agitator blade surfaces.

The experiments were performed using the following ternary system water-acetone-carbon tetrachloride in continuous and batch operation mode. Both liquids, water and carbon tetrachloride were purified previously by distillation, considering that carbon tetrachloride forms an azeotrope with acetone.

The feed flow rates in continuous operation mode ranged between 1,5÷6,6 kg/h organic phase and 0,87÷3,5 kg/h aqueous phase. In the batch operating mode, we studied the behaviour of the concentrations at mean residence time values ranged between 11÷90 min.

The acetone concentrations at the entrance and exit for both phases were determined using the refractometric method. Acetone balance was used to verify the measuring accuracy.

To establish the dominant resistance, the experiments occurred in the following conditions:

- mixing of the aqueous phase only;
- mixing of the organic phase only;
- mixing of both phases.

Experiments were performed using stirrer speeds of 850 rpm, when a uniform distribution of solute concentration in the liquid bulk of each compartment results and the specific transferred flux becomes independent on stirrer speed.

3. RESULTS AND DISCUSSIONS

The cell used for experimentation has a very small specific surface (namely 1.4 m^2/m^3) calculated as a ratio of the interface area between phases to the volume of both compartments of the cell. This type of specific surface calculation applies for industrial extraction columns, as well. In these conditions it is expected that the specific flux of the transferred solute is small and dependent of the cell compartments. (Tudose and Apreotesei 2000).

The dependence between the phase concentrations and the mean residence time was assessed in both discontinuous and continuous operation modes. The experiences were performed using a solution of acetone in carbon tetrachloride (7% weight) and distilled water. The concentrations were determined in mixing and non-mixing conditions of a single or both phases, at the same mixing intensity (850 rpm), when the transferred flux do not depend on the stirrer speed.

Figures 2 and 3 evidence the changes in the both phases exit concentrations as functions on the mean residence time in the cell compartments.

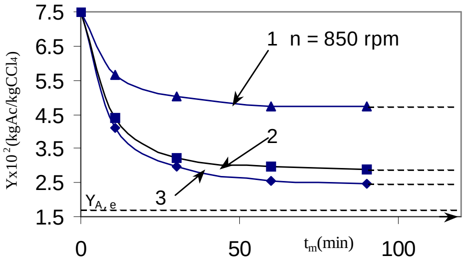

Fig. 2 The variation of the acetone concentration in the organic phase with the mean residence time in the cell in batch operating mode: 1- organic phase agitated, aqueus phase non-agitated, 2- aqueous phase agitated, organic phase neagitated, 3- both phases agitated

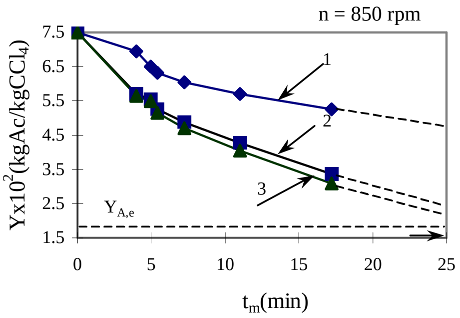

Fig. 3 Variation of acetone concentration in organic phase with the mean residence time in the cell, in the continuous operation mode

First of all, it was observed that the concentration s tends slowly to the equilibrium state owing to the small specific areas.

Second, when only one of the phases -the organic one- was mixed, the diminishing of the exit concentration is very slowly, this fact sugesting that it exists a dominant resistance in the non-agitated phase. When the aqueous phase was stirred, the concentration diminishes more rapidly ouring to a higher transfer resistance and a lower value of the molecular diffusion coefficient of this phase, comparative with the organic phase.

As was expected, the mixing of the both phases leads to an increased mass transfer intensity and the exit concentration tends to the equilibrium value.

From figs. 2 and 3 it results that the equilibrium state was reached wery slowlyouring to a very small specific area, so that it can be concluded that the mean distribution times could be chosen at very different values in the continuous operation mode.

The equilibrium data for the system water -acetone-carbon tetrachloride are presented in fig.4 (curve LF). Any point P on the ordinate represents the initial concentration of the solute in the organic phase, considering the hypothesis that the solvent (water) is pure ($Y_{A,i}$). The equilibrium solute concentration ($Y_{A,e}$) is represented by the point Q. The line PQ is the operation curve for the solute transfer from organic phase to aqueous one. The concentration of the phases at the cell exit will be done by the point M, on the line PQ.

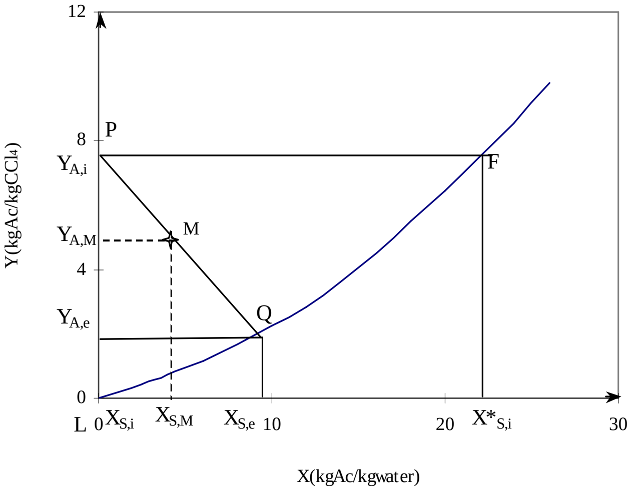

Fig.4 The operating and equilibrium curves

For this situation, the equations for the specific flux of the transferred solute will be the following:

$$n_{B,YX} = k_{X,YX} \cdot \left(X_{S,e} - X_{S,i}\right) = k_{Y,YX} \cdot \left(Y_{A,i} - Y_{A,e}\right) \qquad (1)$$

They were calculated based on the rafinat and extract concentration at the exit of the cell, knowing the contact surface between phases. The individual coefficients were obtained from eqs.1.

The variation of the individual mass transfer coefficients with the residence time in the cell, in the continuous separation mode is represented in fig. 5.

The values of the coefficient decrease slowly with the cell residence time, ouring to a more rapidly decrease of the specific flux of the transferred solute, compared to the driving force. Higher individual coefficient values in the organic phase were obtained when a single phase was stirred (fig. 5a, curve 1), while these values were smaller when only the aqueous phase was stirred (curve 2).

The process intensity is diminished when the phase were the transfer resistance is predominant was not stirred, and this fact explains the small values of the transfer coefficients, when the organic phase was not agitated.

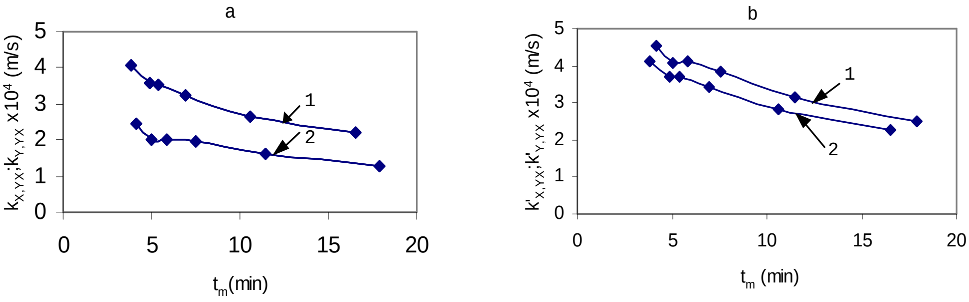

Fig. 5 The variation of the individual mass transfer coefficients with the residence time in the cell
a - a single phase stirred   b - both phase stirred
1- $k_{Y,YX}$, only the organic phase stirred   1- $k'_{Y,YX}$
2- $k_{X,YX}$, only the aqueous phase stirred   2- $k'_{X,YX}$

When both phases are agitated, the specific phases are agitated the specific flux of the transferred solute becomes independent on the stirrer speed and the individual coefficient values phases are different in the two phases.

For the system water-acetone carbon tetrachloride (fig. 5b) presented in this paper, the mass transfer coefficients in the phase were the equilibrium concentration of the solute is smaller (organic phase) are smaller than those in the adjacent phase.

This conclusion apparently contradicts the considerations of other authors (Harishnan et al., 1994). The individual coefficients were calculated from relations 1, were the driving forces were used, expressed through the difference between the feed concentration and the equilibrium value. For higher mixing intensity in both cell compartments, the solute is uniform distributed and the concentration. In this situation, the true driving forces are represented by the differences in the concentration values in cell compartments (point M, fig. 4) and an equilibrium values: $(X_{S,e}-X_{S,M})$ şi $(Y_{A,M}-Y_{A,e})$.

Table 1 presents the individual coefficient values in the two phases calculated using the true driving forces, when both phase are agitated intensely.

Table 1.

The individual mass transfer coefficients calculated using the true driving forces

t_m(min)	3.99	4.96	5.62	7.25	11.01	17.21
$k_{X,YX}$ (m/s)	10.37x10^{-4}	7.94x10^{-4}	7.39x10^{-4}	7.00x10^{-4}	6.8x10^{-4}	6.41x10^{-4}
$k_{Y,YX}$ (m/s)	9.70 x10^{-4}	7.13x10^{-4}	6.72x10^{-4}	6.30x10^{-4}	6.0x10^{-4}	5.57x10^{-4}

The individual coefficients in the organic phase, were the solute concentration is lower, have smaller values than those in the aqueous phase. The results are in concordance with those obtained by other authors.

The experiments performed in botch operation mode, for mean residence time values in the cell ranged between 11÷90 min allowed the calculation of the individual coefficients using the relationship (2), proposed by Lewis (Lewis 1954).

$$k_{X,Y} = \frac{V}{a \cdot t_m} \ln\left(c^*_{X,Y} - c_{X,Y}\right) \tag{2}$$

When the residence time is increased up to 30 min, the individual coefficients in the aqueous phase as well as those in the organic phase decrease significantly (figs. 6, 7). For longer residence times, the decreasing is insignificant as a result of the diminishing of the transferred solute flux.

As in the continuous operation mode, the experiments in the batch operation mode were performed with a single or both phases stirring.

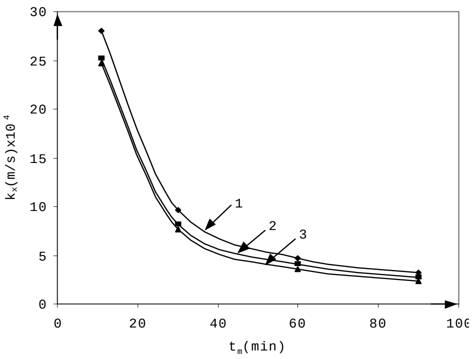

Fig. 6 The variation of the individual mass transfer coefficients in the aqueous phase with the residence time in the cell: 1- organic phase agitated, aqueus phase non-agitated, 2- aqueous phase agitated, organic phase neagitated, 3- both phases agitated

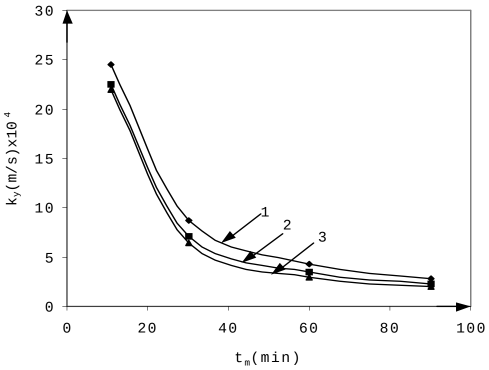

Fig. 7 The variation of the individual mass transfer coefficients in the organic phase with the residence time in the cell

The highest values of the individual mass transfer coefficient in the batch operation mode were obtained when the organic phase was mixed, because in the determinatt resistance to mass transfer is located in that phase.

The individual mass transfer coefficients obtained in this situation were a little higher than those presented in literature, in both operation modes (continuous and batch). Other authors have chosen such systems were the dominant resistance to the mass transfer was located in a single phase. In fact they determined the overall mass transfer coefficient, which was considered equal with the individual coefficient. In this way, the resulted values are a litle lower than the true value of the individual coefficients obtained in this paper. McManamey obtained in 1961 a value of the individual mass transfer coefficient of k_x=0.233x10^{-4}m/s using the method Colburn and Welsh in a batch Lewis cell, with the system i-butanol-water. This value has the same order as that determined in this paper, were the value of the coefficients are dependent on the mean resistance time in the cell.

4. CONCLUSIONS

- The improved Lewis cell represents one of the most efficient method for determination of the mass transfer coefficients, in any ternary system;
- The mass transfer area is strictly measurable and can be modified under certain limits.
- The cell allowed the operation is bach and continuous mode.
- The individual mass transfer coefficient for acetone extracted from CCl_4 and water were calculated both in continuous and batch operation mode.
- The results were compared with those from literature. It was evidenced that the individual mass transfer coefficients presented in this paper are sensibly higher than those reported in literature, for binary systems, obtained using a aproximate method, that can be applied for a limited number of situations.

NOTATIONS

a- mass transfer surface, m^2;
Ac- acetone;
B- notation for solute;
c- acetone concentration (kg/m^2);
c'- acetone equilibrium concentration (kg/m^2);
k- individual mass transfers coefficient when only one phase is stirred, m/s;
k'- individual mass transfer coefficient when both phases are stirred, m/s;
n- stirrer speed, rpm;
n_B- the specific flux of the transferred solute, kgAc/m^2s;
S- notation for solvent;
t_m- mean residence times in the cell compartments (s);
X- acetone concentration in the aqueous phase, kgAc/kgwater;
Y- acetone concentration in the organic phase, kgAc/kgS;

SUBSCRIPT

ag- agitation;
A- initial solution;
B- solute;
e- equilibrium;
s- solution;
S- solvent;
X- aqueous phase;
Y- organic phase;
YX- the transfer direction is from organic to aqueous phase

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