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Correspondence Letter to the Editor

Dear Editor,

I enjoyed reading Dr. Lee's paper "The effects of a surfactant on the mass transfer in spray-tower extraction column" [1]. He indicated that their results for Ka are not identical to those of Hong and Maa [2] and Chu et al. [3]. The discrepancy, he mentioned, may be due to the effect of different apparatus, in addition to the solvent–solute systems and surfactants used.

I think, being a complex system, it has certain problems with respect to the interpretation of experimental data and to the comparisons with others. The main source of the discrepancy is the lack of good characterization of the system. Obtaining reproducible results for a complex system like spray tower extraction is only possible when various parameters are clearly specified. Unfortunately, this point has not been given enough care.

I would like to review some important aspects of the subject and remind key parameters for those who are interested to collect more experimental data for developing accurate correlation.

End effects

In this system, at least, for two reasons the end effects are very important.

Firstly, mass transfer during formation of drops at an orifice can be a very significant fraction of the total mass transfer in extraction [2]. Although the times of drops formations are very short, they have a crucial role in mass transfer irrespective of the height of tower.

Secondly, the surface contamination theory implies that all drops, no matter how small, will show internal circulation if the system is sufficiently free of surface-active contaminants. But when even a trace of surfactant is present, the motion tends to be damped out first at the rear of drop [3]. Elimination of internal circulation causes increasing the drag and, as a result, reducing overall mass transfer rate significantly. In this system which exhibits high interfacial tension (water/non-polar liquid) CCl₄ is free of surfactant when it is first injected into spray tower extraction, but internal circulation and the velocity of fall of CCl₄ drop decreases with time as concentration of SLS (sodium lauryl sulphate) molecules increases at interface. According to the above mentioned reasons, the measured values for K_Ra and K_R by different researchers may disagree.

Deformation and breaking up of drops

In Fig. 3 of said paper, $K_R a$, K_R and a have been shown as functions of the concentration of SLS in solution. $K_{\rm R}a$ and $K_{\rm R}$ are obtained experimentally but *a* is calculated by substituting the values of K_R into $K_R a$. The value of $K_R a$ decreases rapidly as the concentration of SLS increases and reaches a minimum at about 10 ppm, and then increases monotonically as the concentration is further increased. They concluded that below 10 ppm of SLS, the effect of surfactant on the decrease in $K_{\rm R}$ is more important than that on the increase in interfacial area but with further addition of SLS, the increase in $K_{\rm R}a$ is caused mainly by the increase in interfacial area. They inferred that the rapid increase in the mass transfer area is caused by the inhibition of drop coalescence by the dynamic surface effect. This reason cannot explain well this strange sharp change in mass transfer area because very low concentration of SLS (say 0.5 ppm) has a little effect on the inhibition of drop coalescence, in addition, coalescence and drop size are interrelated. We should note that coalescence is also a function of frequency (population) of drops. In fact, the size distribution of drops is naturally determined by the dynamics of break up and coalescence.

I think an interesting phenomenon is occurring in low concentration of SLS if experimental data are accurate and modified for end effects.

Surfactant molecules affect mass transfer via hydrodynamic and molecular effect. Hydrodynamics effects include two phenomena, which act in opposition. In the absence of mass transfer, surfactant decreases the mobility of the interface. In the presence of mass transfer, motion at interface may be enhanced through the action of local surface tension gradients caused by small differences in concentration along the interface. This enhancement of surface motion is called Marangoni effect.

The molecular effects are interfacial resistances to mass transfer, which may arise from the interaction of SLS molecules with the acetic acid molecules being transferred.

Two other concepts have also been neglected in interpreting the mass transfer data:

1. When the size of droplet decreases, not only the transfer area is increased, but the mass transfer coefficient is also increased because of decrease in diffusion length.

2. The spherical drop has a minimum surface area but in this system the drops may deform by adding surfactant and increasing Eotvos number which is defined by

$$Eo = \frac{g \ \Delta \rho d^2}{\sigma}$$

If we analyze the mass transfer rate by film theory, then we can write

$$\frac{1}{K_{\rm R}} = \frac{1}{k_{\rm R}} + \frac{H}{k_{\rm E}} + R_{\rm m}$$

where $R_{\rm m}$ is the mass transfer resistance during acetic acid crossing the interfacial film, and $k_{\rm R}$ and $k_{\rm E}$ are the individual mass transfer coefficients in raffinate and extract phases, respectively. For pure system, i.e., absence of surfactant, $R_{\rm m} = 0$ and $k_{\rm R}$ has its enhanced value because of internal circulation. By adding SLS molecules, internal circulation is dampened and interfacial film resistance grows up. According to the said paper data, when SLS concentration approaches to 20 ppm, internal circulation was completely prevented and interfacial film resistance reached to its highest value. Fig. 3 in said paper, shows that $K_{\rm R}$ remains constant when surfactant is being added up to 400 ppm. This behavior may indicate that accumulation of SLS around each drop remains monolayer in spite of increasing SLS concentration. We can conclude that beyond 20 ppm, all SLS are consumed solely for deformation, breaking up of drops and generation of new interfacial area without any effect on mass transfer flux.

Effect of bubble size distribution

It is obvious that there will be a wide distribution of drop sizes in any operating spray tower extraction column. The drop size distribution will change with position depending on the influence of the inlet distributor, column internals and degree of agitation. The dispersed phase is assumed to consist of different fractions, each with its mean drop diameter, d_i . We must make a distinction between the relative volume passing through the column and the relative volume inside the column. There will be a large fraction of the smaller drops in the column than passing through the column, because of their lower velocities. It should be noted that, although it will be the static volume distribution of drop sizes that is seen, or photographed, or otherwise measured, it is the kinetic distribution that is important from a mass transfer standpoint.

Acknowledgements

We should thank Dr. Lee who has made a valuable and praiseworthy research work. I think a logical extension of this research is to study the effect of surfactant in either CCl₄ or both phases on mass transfer.

References

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