R & D NOTES

Mass Transfer During Drop Formation

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In an earlier paper (Rajan and Heideger, 1971), we reported measurements of mass transfer coefficients determined photographically for extraction from single drops during formation. The system employed was continuous phase resistance controlled, and yet the mass transfer was strongly affected by circulation within the drop, especially in the early formation period. The reported coefficients generally decayed to a nearly steady value long before the drop detached from the forming nozzle, consistent with the subsequent observation (Humphrey et al., 1974) that internal circulation during drop formation can effectively cease before drop detachment. All of the results were correlated as

$$\text{Cumulative mass transfer} = \text{const.}\ (\text{formation time})^n$$

in which n varied from 0.6 to 2.4 with variation in nozzle diameter and dispersed phase flow rate.

No single correlation was obtained for either instantaneous or cumulative mass transfer, because of the large number of variables affecting the exchange between phases. However, in going back to that data, we have observed that the total formation time for any drop is a good correlating parameter for the average mass transfer coefficient integrated over the formation period. Although the linear infusion velocity of the dispersed phase fluid should obviously be important in establishing internal circulation, it appears that total formation time effectively incorporates that with nozzle size and drop diameter to represent all of the observations. Figure 1 shows this result, in which even the measurements made at different continuous phase velocities are well correlated by a single line.

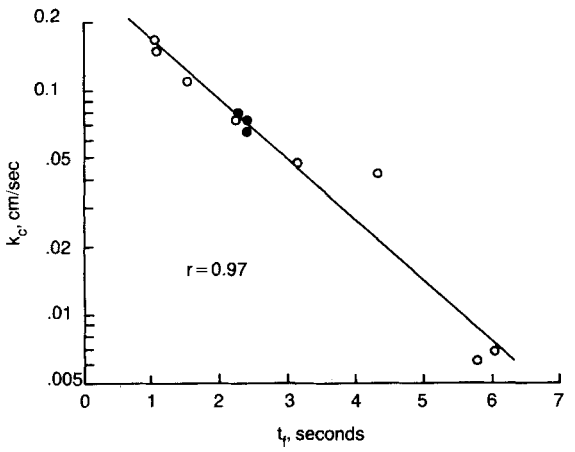

Figure 1. Correlation of average mass transfer coefficient with formation time, continuous phase controlled (Rajan and Heideger, 1971). Solid circles represent measurements taken at different continuous phase flow rates.

Since circulation was obviously so important even in this continuous phase resistance system, we decided to make the comparable study for a system controlled by the dispersed phase resistance. Photographic measurements similar to those already reported were made, except that in the present study drops of water were formed in a stationary continuous organic phase consisting of 2-ethoxyethyl acetate. The organic was presaturated with water so that no mass transfer occurred within the continuous phase. Thus mass was transferred only from the interface into the forming drop and the total resistance to mass transfer at all times resided in the dispersed phase. Again, the instantaneous drop volume was determined by careful measurements on cine photographs and the mass transferred was evaluated by difference between infused volume and measured volume.

RESULTS

As in the previous study, the instantaneous mass transfer coefficients were generally high at the onset of drop formation, decreased rapidly early in the formation period, and then either held constant or increased only slightly until final detachment of the drop from the nozzle. Absolute magnitudes of the coefficients are different from those determined for the continuous phase but, since the system's physical properties (especially diffusivity, dispersed phase viscosity, and interfacial tension) are different, no attempt was made to compare values for the coefficients.

Five different volumetric flow rates and five different nozzle diameters were employed, resulting in approximately a tenfold variation in total formation time. Again, no overall correlation of the results could be obtained in terms of the dispersed phase infusion velocity, though it obviously had a strong influence on the

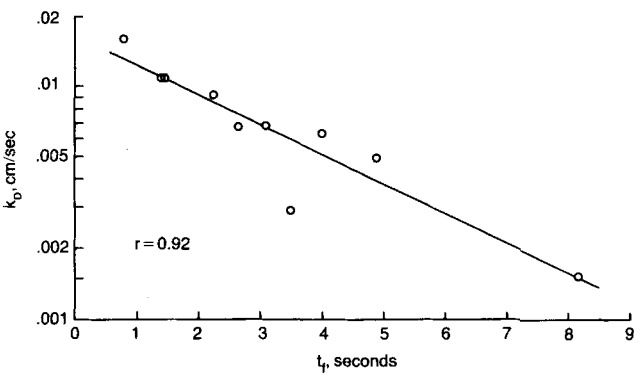

Figure 2. Correlation of average mass transfer coefficient with formation time, disperse phase controlled.

magnitude of the coefficients. However, when the average mass transfer coefficient was matched to the total drop formation time, a linear correlation was again obtained on semilogarithmic coordinates. Figure 2 shows this result for the present study of dispersed phase resistance.

DISCUSSION

Mathematical models for mass transfer during drop formation, summarized by Popovich et al. (1964) and more recently by Walia and Vir (1976), are generally based on a solution to the diffusion equation in the continuous phase and do not account for circulation within the forming drop. Zimmermann et al. (1980) report short formation time mass transfer results strongly influenced by internal convection. A study of water extraction by isobutanol drops (Heertjes et al., 1954) showed marked circulation for rapid formations, typical drops having a Reynolds number on the order of 0.008. Drop Reynolds numbers in the present project were in the range from 0.05 to 0.5; it therefore seems reasonable to expect rapid circulation in all these drops and a strong dependence on formation time.

The most interesting observation of this study would appear to be the correlation obtainable between average mass transfer coefficient and total drop formation time. This is evident for systems controlled both by the continuous phase resistance (Figure 1) and by the dispersed phase resistance (Figure 2). The lower slope of the line in Figure 1 is consistent with the expectation that internal movement of the drop should not be as significant in determining mass transfer rates at the interface when the resistance lies in the continuous phase. Nevertheless, shear transfers across the interface and generates convection in the continuous phase as well. Thus it appears that total formation time may be the most important parameter in setting the mass transfer coefficient during generation of a dispersed phase.

LITERATURE CITED

Heertjes, P. M., W. A. Holve, and H. Talsma, "Mass Transfer Between Isobutanol and Water in a Spray-Column," *Chem. Eng. Sci.*, **3**, 122 (1954).

Humphrey, J. A. C., R. L. Hummel, and J. W. Smith, "Note on the Mass Transfer Enhancement Due to Circulation in Growing Drops," *Chem. Eng. Sci.*, **29**, 1496 (1974).

Popovich, A. T., R. E. Jervis, and O. Trass, "Mass Transfer During Single Drop Formation," *Chem. Eng. Sci.*, **19**, 357 (1964).

Rajan, S. M., and W. J. Heideger, "Drop Formation Mass Transfer," *AIChE J.*, **17**, 202 (1971).

Walia, D. S., and D. Vir, "Interphase Mass Transfer During Drop or Bubble Formation," *Chem. Eng. Sci.*, **31**, 525 (1976).

Zimmermann, V., W. Halwachs, and K. Schugerl, "Mass Transfer Investigations During Droplet Formation by Means of a Modified Liquid Scintillation Technique," *Chem. Eng. Commun.*, **7**, 95 (1980).

Controller Tuning Using Optimization to Meet Multiple Closed-Loop Criteria

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INTRODUCTION

The PID controller family and its variations continue to be widely used in the process industries, both in analog and digital applications. Tuning or controller design techniques include the methods of Ziegler and Nichols (1942), Cohen and Coon (1953), Harriott (1964), Chidambara (1970), Lopez et al. (1969), and Yuwana and Seborg (1982).

Although the available controller design relations are convenient and easy to use once a simple model is known, they do not really yield standard system performance results from case to case. In particular, the resonant peaks and gain margins vary widely, and in some cases the controllers may be quite unsatisfactory—the response to a disturbance would be much too oscillatory. With some techniques, e.g., Ziegler-Nichols, an unstable system occasionally is obtained. Graphical methods, such as the one recently presented by Edgar et al. (1981) based on the interactive use of a graphics terminal, yield much superior results in terms of system performance, but can be quite time consuming to use.

In general, tuning methods that are based on just one particular input signal or on one particular closed-loop characteristic, such as the decay ratio, have been criticized. This paper suggests a new technique for controller tuning based on several characteristics of the closed-loop process and not on any particular type of input signal. Bollinger et al. (1979) have taken a related approach in the control of a synchronous generator using a lag-lead element. They use the method of inequalities (with moving boundaries) presented by Zakian and Al-Naib (1973). Several functionals describing the desired system behavior in terms of inequalities are defined. Controller settings are then found that meet the inequalities. Zakian and Al-Naib (1973) perform a similar design for a PI controller. The results are superior to standard design methods; however, they may not be consistent from case to case since inequalities are used.

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