# Effects of Surfactants on Mass Transfer During Spray Drying

A study was conducted to investigate the effects of added surfactants on mass transfer during spray drying. Measurements were made of retention of volatile components and amount of water evaporation at various points near the atomizer inside a spray dryer of pilot plant scale. Experimental data indicate that surfactants can have a substantial effect on transport processes when certain types of liquid solutions are spray dried. Experimental results are interpreted in terms of transport mechanisms and are compared with relevant previous studies on non-spray systems.

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## SCOPE

Spray drying is widely used for dehydration of liquid foods such as coffee and tea extracts, milk concentrates, and purees. Because of the economic importance of these products, and because of numerous other industrial applications of spray drying, there is growing interest in a better understanding of the mechanisms of mass transfer during spray drying. This knowledge should aid optimization of these processes for minimum cost, for better retention of volatile aroma components, and/or for improving other product quality factors.

In spite of the large number of fundamental mass transfer studies to be described below, most previous work dealing with added surfactants has been on idealized systems and does not take into account the following complexities which are characteristic of industrial spray drying:

1. Small drop sizes and wide distribution of drop sizes
2. High initial velocity and subsequent deceleration of drops
3. Turbulence and oscillations in drops caused by high-pressure atomization
4. Effects of drying on the internal motions of drops
5. Formation of a surface crust or a selective-diffusion barrier

Notwithstanding the relatively small amount of past work in this area, it is important to understand the effects of surfactants upon spray drying. Many or all liquid foods contain natural surfactants. In situations such as foam-spray drying, it may be necessary to add surfactants to the liquid feed to obtain the desired processing effect. Finally, addition of surfactants may be a convenient means to affect and control mass-transfer rates during spray drying or in other types of spray contacting.

For these reasons, an experimental and theoretical investigation was undertaken of the spray drying of surfactant-containing aqueous sucrose and coffee solutions, with acetates as model aroma components. The principal aims of this investigation were to determine the effects of various surfactants on the retentions of highly volatile aromalike components and on the rate of water evaporation in regions close to the nozzle. An aerodynamic model of spray behavior was used to interpret the experimental results.

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## CONCLUSIONS AND SIGNIFICANCE

This study indicates that surfactants can have a large effect on volatiles retention during spray drying. The primary effect of a surfactant appears to be on droplet hydrodynamics and particularly on oscillations, turbulence, and internal circulation in drops. In solutions of high solids content where volatiles losses from drops are small, surfactants have only a minor effect since mass transfer during atomization is not substantially affected. In solutions, such as coffee extract, that contain large levels of natural surfactants, added surfactants also had relatively little effect. Most of the evaporation in aqueous sucrose and coffee solutions takes place in the constant-activity period for the regions of spray monitored in this study. Since gas phase mass transfer controls evaporation rates in this period, added surfactants did not substantially affect the percent water evaporation at a given axial distance.

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## Introduction

### *Mass transfer and effects of surfactants in spray drying*

Only a few past studies address the mechanism of mass transfer within particles and/or the effects of surfactants during spray drying. Kieckbusch and King (1980, 1981) and Zakarian and King (1982) have compared measured and calculated retentions of volatile acetates at various positions near the atomizer within a spray dryer fed with aqueous sucrose solutions. Their results indicate that mass transfer rates are larger than those calculated assuming that the drops are stagnant, noncirculating spheres. Kerkhof and Thijssen (1977) have compared measured final retentions of model aroma components during spray drying with predictions from the selective-diffusion theory. From this comparison it was concluded that the losses of the aroma components were greater than could be accounted for by simple diffusion within a stagnant medium. The discrepancy was attributed in part to internal circulation or other motions inside drops.

Very little work has been done to investigate the effects of surfactants on mass transfer during spray drying. Kerkhof and Schoeber (1974) have performed theoretical calculations which suggest that internal motions in drops exist in regions close to the atomizer. These authors also suggested that surfactants can reduce mass transfer rates through their effect on internal motions. Sivetz and Desrosier (1979) state that the final retentions of aroma components in coffee are increased when surfactants are added, attributing the effect to a reduced average drop size. However, no data are presented.

### *Mass transfer inside drops*

Mass transfer inside liquid drops has been extensively studied and covered in several reviews (Pritchard and Biswas, 1967; Gal-Or et al., 1969; Tavlarides et al., 1970). The approach to mass transfer equilibrium for a spherical stagnant drop of constant radius and diffusivity, with no external mass transfer resistance, was first given by Newman (1931)

$$E_a = 1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(\frac{-n^2 D\pi^2 t}{R^2}\right) \qquad (1)$$

Kronig and Brink (1950) have derived a similar expression applicable to spherical drops with internal circulation described by Hadamard (1911) flow patterns:

$$E_a = 1 - \frac{3}{8}\sum_{n=1}^{\infty}\beta_n^2\exp\left(-\lambda_n\frac{16Dt}{R^2}\right) \qquad (2)$$

Values for the quantities $\beta_n$ and $\lambda_n$ are given in various references (Kronig and Brink, 1950; Johnson and Hamielec, 1960). Equation 2 predicts mass transfer rates that are approximately 1.5 times greater than those predicted by Eq. 1 at similar values of $Dt/R^2$. Although Kronig and Brink derived Eq. 2 for the Stokes flow regime ($Re < 1$), several workers have suggested that this equation may describe mass transfer rates at $Re > 1$ (Hamielec and Johnson, 1962; Danckwerts, 1951).

Skelland and Wellek (1964), Shroeder and Kintner (1965), and Pritchard and Biswas (1967) have reported the onset of oscillations in droplets to occur at Reynolds numbers between 200 and 600 depending on the droplet viscosity. Several mass transfer models have been proposed that include the effects of oscillations and turbulence (Handlos and Baron, 1957; Patel and Wellek, 1967; Angelo et al., 1966). These models generally predict mass transfer rates that are 5 to 10 times as great as the rates predicted by Eq. 1.

Many experimental studies have been performed to investigate mass transfer rates in liquid drops. For many of these studies, covering both gas-liquid and liquid-liquid systems, mass transfer rates are significantly larger than predicted by Eq. 1 or Eq. 2 (Johnson and Hamielec, 1960; Skelland and Wellek, 1964; Hughes and Gilliland, 1955; Whitman et al., 1926; Streicher and Schügerl, 1977; Garner and Lane, 1959; Boye-Christensen and Terjesen, 1959). These high mass transfer rates are generally attributed to oscillations and/or turbulence produced during drop formation or as a result of drop motion through the continuous medium. In several instances (Johnson and Hamielec, 1960; Skelland and Wellek, 1964) high mass transfer rates were measured, but no drop oscillations were observed.

### *Effects of surfactants on mass transfer*

The effects of surfactants on mass transfer in liquid drops can be divided into two major categories: hydrodynamic effects and interfacial resistance effects. Hydrodynamic effects include changes in drop size and velocity, and effects on internal circulation, oscillations, and turbulence. Interfacial resistance effects include surface blockage and solute-surfactant interactions. The reduction of water evaporation rates by an insoluble monolayer is usually attributed to the formation of a surfactant barrier at the air-water interface (La Mer, 1962). Insoluble surfactant monolayers forming solid-type surfactant films have also been reported to produce interfacial diffusional barriers for various solutes in aqueous solution (Burnett and Himmelblau, 1970; Plevan and Quinn, 1966). Soluble surfactants forming Gibbs monolayers generally do not exhibit barrier effects, at least for small diffusing species, although there is some disagreement on this point (Plevan and Quinn, 1966; Springer and Pigford, 1970; Mudge and Heideger, 1970).

Effects of surfactants on hydrodynamics have been dicussed by many authors (Davies, 1963; Gal-Or and Waslo, 1968). The ability of surfactants to affect mass transfer by reducing internal circulation, oscillations, turbulence, and interfacial movement in drops has been used to explain the results of several experimental studies (Garner and Hale, 1953; Lewis, 1954). In many cases it is difficult to determine whether hydrodynamic effects or barrier effects are responsible for changes in mass transfer rates. For Reynolds numbers greater than unity, there is currently no well-accepted method for predicting whether internal circulation will occur (Kerkhof and Schoeber, 1974).

## Experimental

### *Equipment and operating conditions*

The equipment used in this study was the spray dryer and solvent collection system described by Etzel and King (1984). Cyclones were added to the air exit line (Frey, 1984). Samples were withdrawn axially for analysis at various distances from the atomizer. The analytical equipment was also identical to that used by Etzel and King. The experiments in this study, unless otherwise noted, were performed at 200°C inlet air temperature, 50°C liquid feed temperature, 7.00 MPa atomization pressure, $8.33 \times 10^{-2}$ $m^3/s$ airflow rate, and ambient column pressure. A Spraying Systems Co. model 950017-TC flat-spray nozzle was used in the experiments. This nozzle gave liquid flow rates of approximately $3.9 \times 10^{-6}$ $m^3/s$ at 7.00 MPa pressure. Details of the experimental equipment and procedures are given by Frey.

### *Materials*

The sucrose used was the grade commercially marketed by C&H Sugar Co.; ordinary tap water was used as the solvent. Ethyl, *n*-propyl, and *n*-butyl acetates were reagent grade (Mallinkrodt Co.). The surfactants used were sodium lauryl sulfate (Matheson, Coleman and Bell), sodium lauryl benzene sulfate (Orion Chemical Co.), and Brij 35 [$HO(C_2H_4O)_{22} \cdot C_2H_4OC_{12}H_{25}$] (Chem Services Inc.). All surfactants were used at feed concentration levels of 500 ppm w/w. Unagglomerated spray-dried coffee used to make coffee solutions was supplied by Proctor and Gamble Corporation.

## Results and Discussion

### *Volatiles loss and water evaporation without added surfactants*

Results for aqueous sucrose solutions with no added surfactants and in the region close to the atomizer are shown in Figure 1. These are similar to those reported by Etzel and King (1984) (see also Kieckbusch and King, 1980, 1981; Zakarian and King, 1982). A large loss of volatile acetates takes place within 10 cm of the atomizer. This loss has been attributed to the relatively large amount of mass transfer that takes place in the thinning sheet from which drops are formed (Simpson and Lynn, 1977; Kieckbusch and King). The order of retention of the various acetates for 50 and 60% sucrose solutions indicates that the loss process is controlled primarily by liquid phase diffusion. For external mass transfer control butyl acetate will show the greatest loss because of its larger volatility, whereas for internal mass transfer control ethyl acetate will show the greatest loss because of its larger liquid phase diffusion coefficient (Kieckbusch and King).

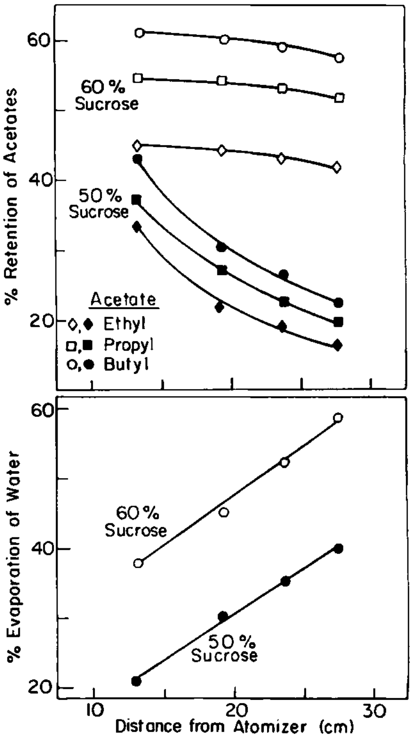

**Figure 1. Volatiles retention and percentage evaporation of water for spray drying of 50 and 60% sucrose solutions (200°C air temp., 7.00 MPa atomization pres.).**

However, the order of the percent evaporation curves in Figure 1 indicates that the majority of the water is being evaporated in the constant-activity period, where the rate of evaporation is controlled by external mass transfer (Etzel and King). Similar trends in percent evaporation were observed for solutions of coffee extract (Frey, 1984). Particularly striking is the comparison between the large losses of acetates for the 50% sucrose solutions and the relatively small amount of loss for the 60% sucrose solutions during this period. Etzel and King attributed this effect to the relatively small diffusion coefficients in 60% sucrose solutions, and the formation of a high-solids layer at the outer surface when the drop enters the falling-rate period of drying. According to the selective-diffusion theory of Thijssen (1971), at sufficiently low moisture content this surface layer allows transport of water from the drop with little or no simultaneous diffusion of trace volatile organics.

### *Volatiles loss and water evaporation with added surfactants*

The effect of added surfactants on the rate of water evaporation from aqueous sucrose and coffee solutions is shown in Figure 2. For these solutions, little or no effect can be observed. The scatter in the results for 50% sucrose and 15% coffee solutions seems to be primarily due to run-to-run irreproducibilities and the difficulty in measuring small percentage evaporations.

Particle sizes for the dried product from experiments with and without added surfactants were measured from scanning electron micrographs. On the basis of counting and measuring 600 particles for each case, it was determined that the Sauter mean diameter was approximately 0.040 cm. The addition of a surfactant caused only a minor change (i.e., less than 20%) in the Sauter mean diameter. At least part of this change is due to a greater degree of expansion in the late stages of drying in droplets with added surfactant (Frey, 1984). Although theoretical and empirical studies of atomization indicate that changes in surface tension can result in a changed drop size distribution (Hasson and Mizrahi, 1961), qualitative arguments by Dombrowski and Munday (1968) indicate that there is insufficient time for an appreciable amount of surfactant adsorption during high-pressure atomization. In addition, a substantial change in the drop size distribution should alter the percent evaporation of water at a given axial distance as compared to experiments without a surfactant. For example, a smaller drop size would result in an increased evaporation rate and an increase in drop residence time at a given axial distance. Also, high-speed photography indicates that the surfactant does not affect the mechanism of atomization (Frey, 1984).

The lack of any major effect of added surfactant upon percent evaporation appears to be in agreement with the mass transfer studies previously described. The soluble surfactants used in this study would not be expected to form diffusional barriers for water transport, as have been attributed to insoluble surfactant monolayers (Springer and Pigford, 1970). In addition, although the surfactants in this study may influence drop hydrodynamics, these effects would have major influences on the water evaporation rate only when drying is predominantly controlled by liquid phase mass transfer. Since, as indicated previously, the drying near the atomizer in these experiments seems to take place largely in the constant-rate period, surfactant effects on hydrodynamics should not influence the fraction of water evaporated at a given axial distance.

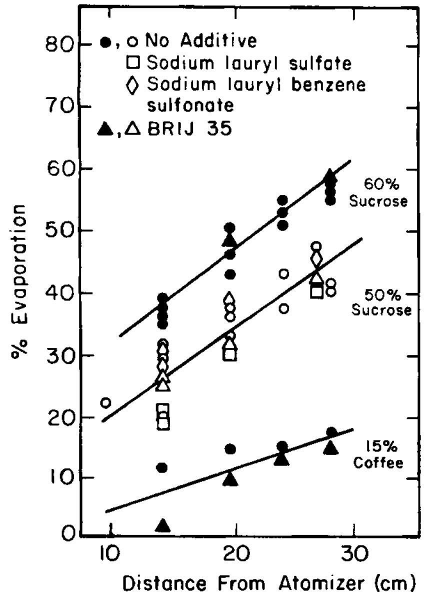

**Figure 2. Effect of surfactants on the evaporation of water during spray drying of sucrose and coffee solutions (200°C air temp., 7.00 MPa atomization pres., coffee solutions atomized at 3.55 MPa).**

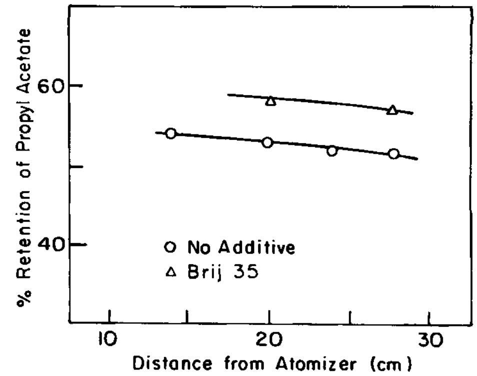

**Figure 3. Effect of a surfactant on retention of *n*-propyl acetate during spray drying of 60% sucrose (200°C air temp., 7.00 MPa atomization pres.).**

Figure 3 displays the effects of surfactants on *n*-propyl acetate losses in the nozzle region during spray drying of 60% sucrose solutions. Only a minor effect is apparent. For 60% sucrose solutions, the acetate retention is primarily determined by losses from the expanding sheet, and acetate losses from droplets are small. Since surfactants do not seem to affect the atomization process significantly, their effect on acetate retention in 60% sucrose is relatively small.

The retention of *n*-propyl acetate for spraying 50% sucrose

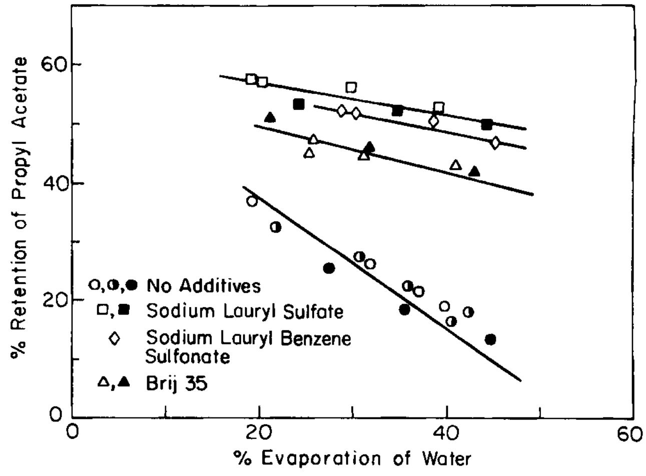

**Figure 4. Effect of surfactants on retention of *n*-propyl acetate during spray drying of 50% sucrose (200°C air temp., 7.00 MPa atomization pres.).**

solutions is shown in Figure 4. In this figure, *n*-propyl acetate retention is plotted against percent evaporation for experiments without added surfactants and for several experiments with added surfactants. It was shown theoretically by Zakarian and King (1982) and experimentally by Etzel (1983) that plotting data in this way largely eliminates effects caused by changes in drop size distribution. For example, Etzel used this method to plot results from two spray drying experiments in which the Sauter mean drop diameter differed by a factor of 3. It was found that the results from both experiments lay on the same curve. For these reasons, it can be concluded that the effect of any change in drop size distribution caused by the addition of a surfactant is greatly minimized or eliminated in Figure 4.

In contrast to the results for 60% sucrose solutions, Figure 4 indicates that surfactants in 50% solutions have a considerable effect on the amount of acetate retention. It should be noted that the experimental curves in Figure 4 extrapolate approximately to a common value at 0% evaporation. Therefore, as was the case for 60% sucrose solutions, surfactants do not seem to influence the loss of acetates during atomization. Instead, the primary effect of the surfactant seems to be on the rate of mass transfer from the droplets. It should be noted that small amounts of surfactant impurities are probably present in runs without added surfactants. Also, the acetates themselves have some degree of surface activity. This suggests that it takes greater than trace amounts of strongly surface-active materials to cause the effects shown in Figure 4.

The primary effect of the soluble surfactants used in this study should be on hydrodynamics, and particularly on motions inside drops rather than through surface barriers to transport. Chromatographic analysis of the gas phase in contact with the surfactant-containing liquid solutions used in this study indicates that surfactants have a very small effect on acetate vapor pressure. For this reason, it appears that surfactant-solute interactions are not a significant factor in these experiments. It would therefore appear that for 50% sucrose solutions without added surfactants there is a significant enhancement of mass transfer by motions inside drops. The presence of surfactants seems to reduce these motions and thereby increase the retention of acetates. The higher retention of volatile acetates measured in the nozzle region also produces a correspondingly higher retention in the dried product. 50% sucrose solutions without surfactants produce dried material with 8% retention, while surfactant-containing 50% solutions have final retentions of 32% (Frey, 1984).

The effect of an added surfactant on the retention of *n*-propyl acetate in 15% coffee solutions is shown in Figure 5. In contrast to sucrose solutions, no effect is evident. The difference would seem to be due to the fact that coffee extract solutions contain large amounts of natural surfactants. The major types of naturally occurring surfactants in coffee extract are described by Sivetz and Desrosier (1979).

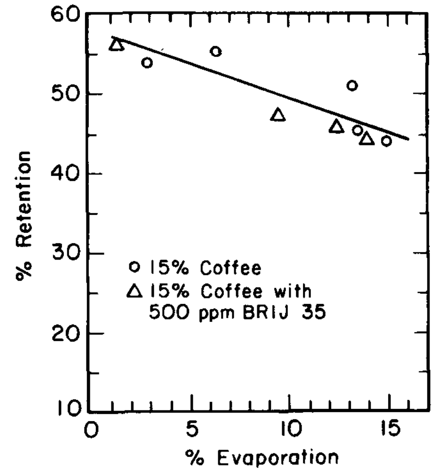

**Figure 5. Effect of a surfactant on retention of *n*-propyl acetate during spray drying of 15% coffee solutions (200°C air temp., 3.55 MPa atomization pres.).**

### *Modeling of a simplified spray system*

Theoretical calculations of the retention of volatile components during spray drying are difficult to perform for a number of reasons. Simultaneous drying causes steep gradients in water concentration and correspondingly large variations in the diffusion coefficient for a volatile component. Drop temperatures increase as the droplets enter the falling-rate period of drying, again affecting diffusion coefficients. Also, it is usually difficult to predict temperatures, and humidities inside sprays. Although attempts at modeling under these conditions have been undertaken (Zakarian and King, 1982; Kerkof and Schoeber, 1974), the extent to which the above factors can be accounted for is unclear.

To avoid some of these modeling difficulties, simplified experiments were performed in which water containing volatile acetates was sprayed under ambient conditions (drying air temperature = 22°C) with the liquid feed at its wet-bulb temperature. Experiments with a tracer of nonvolatile material (sodium chloride) indicate that the amount of water evaporated in these experiments was less than 1%. These experiments therefore avoid the majority of the modeling difficulties that occur when drying and volatiles transport occur simultaneously.

Since the Biot number for mass transfer is relatively large for droplets in this study (Zakarian, 1979), acetate mass transfer should be liquid-phase controlled. Acetate retentions can then be predicted using Eqs. 1 and 2 if the quantity $Dt/R^2$ at various axial positions is known. Diffusion coefficients of the acetates in aqueous solutions are given by Frey and King (1982). The times of flight for various drop sizes can be calculated using the aerodynamic model of Rothe and Block (1977), as modified by Zakarian and King (1982) and Etzel (1983). Numerical results of the aerodynamic model were compared to experimental data by Keickbusch (1978). Good agreement was found. The measured distribution of spray flux in the radial direction was approximated as a step function (Frey, 1984). It was observed that the addition of a surfactant has little effect on the spray flux distribution for fan-spray nozzles.

The distribution of drop sizes produced when spraying water from the nozzle used in these experiments is reported by Etzel (1983). The measurement of size distributions for droplets in flight requires relatively sophisticated equipment that was not available during this study. For this reason, and because changes in drop size distribution did not appear to be a major factor in explaining the experimentally observed effects of surfactants in sucrose-containing solutions, it was assumed that the addition of a surfactant did not affect the drop size distribution during the spraying of water. Thus the primary objective of the theoretical and experimental work described in this section is to show that the difference in acetate retentions calculated using Eqs. 1 and 2 under the assumption of a fixed drop size distribution can account for the experimental results.

Experimental results and theoretical calculations for the spray experiments are shown in Figure 6. The results are similar to those observed for the experiments with 50% sucrose. The experiment with 500 ppm of sodium lauryl sulfate shows a marked increase in the retention of *n*-propyl acetate at any given distance. As was the case for 50% sucrose, the experiments with and without added surfactants extrapolate approximately to a common value at zero axial distance. This extrapolated value can be used as an approximation for the acetate retention of drops just after formation. The order of the retentions for the three acetates indicates that liquid-phase mass transfer resistance predominates although the relative difference in retentions between the acetates is less in these experiments than in experiments where the sprayed liquid contains dissolved solids (Frey, 1984).

Calculations of *n*-propyl acetate retention in drops using Eq. 1 (stagnant spheres) and Eq. 2 (internal circulation) are shown in Figure 6. There appears to be reasonable agreement between the results calculated assuming stagnant spheres and those measured experimentally with added surfactant. The experimental results without added surfactant lie below the calculation assuming internal circulation in drops. These results suggest that without added surfactants, internal motions in drops accelerate mass transfer beyond the rate calculated by Eq. 2. It therefore appears that oscillations and turbulence, in addition to circulation, contribute to mass transfer in drops during spraying with zero or low levels of surfactants. The effect of a surfactant is to reduce these internal motions, so that the experimental results with surfactants tend to agree better with Eq. 1. In addition, minor changes in drop size distribution caused by the addition of the surfactant may be contributing to the difference in measured retentions in Figure 6. It should be noted that these conclusions are sensitive to the precision of the drop size distribution measurement.

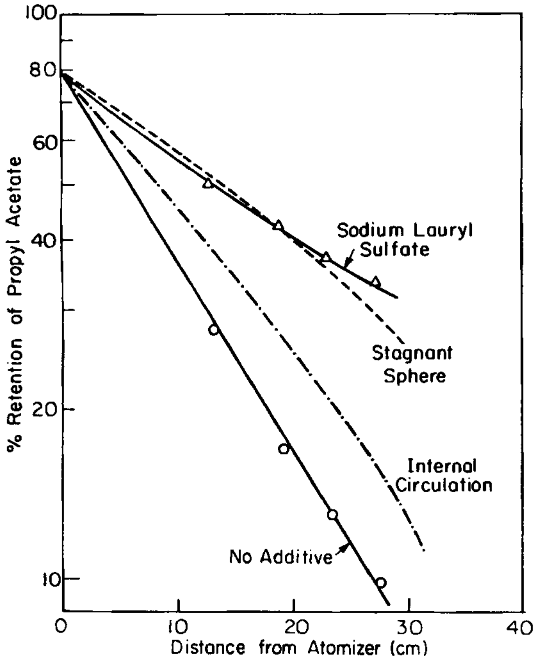

**Figure 6. Comparison of experimental and theoretically calculated retentions of *n*-propyl acetate (22°C air temp., 7.00 MPa atomization pres.).**

The presence of droplet oscillations, turbulence, and internal circulation is also consistent with the high droplet velocities that exist in most spray situations with pressure atomizers. The Reynolds number for an average size drop near the atomizer in these experiments is approximately 150. These results are therefore similar to the many studies on liquid-drop mass transfer in which mass transfer rates are larger than calculated by Eqs. 1 and 2.

These conclusions derived from experimental and theoretical results for 0% sucrose solutions are consistent with experimental results for solutions of higher sucrose content. An important difference in the two cases is that the viscosities of sucrose-containing aqueous solutions are significantly larger than the viscosity of water. For example, a 50% sucrose solution at 50°C has a viscosity of $5.1 \times 10^{-3}$ Pa · s while the viscosity of water at 17°C is $1.0 \times 10^{-3}$ Pa · s (Perry et al., 1984). Although the fluid viscosity has no effect on the mass transfer rate obtained from the simple Kronig and Brink model, more general models indicate that fluid viscosity can have a significant effect on mass transfer rates (Angelo et al., 1966). Under most conditions, the higher viscosity of sucrose-containing solutions would cause internal movements in droplets to be more rapidly damped by viscous dissipation, and therefore the intensity of internal motions would be lessened. For this reason, it might be expected that the addition of surfactants would have a smaller effect on mass transfer rates in sucrose-containing solutions than in solutions without sucrose. However, it is also true that internal circulation in these drops would accelerate mass transfer of volatile components more than given by the difference between constant-diffusivity Eqs. 1 and 2, since internal circulation would hamper the development of water concentration gradients within drops.

## Acknowledgment

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## Notation

$a$ = drop radius
$D$ = diffusion coefficient
$E$ = fraction loss of a component
$n$ = integer
$t$ = time

### *Greek letters*

$\lambda_n$ = constant, Eq. 2
$\beta_n$ = constant, Eq. 1

## Literature Cited

Angelo, J. B., E. N. Lightfoot, and D. W. Howard, "Generalization of the Penetration Theory for Surface Stretch: Application to Forming and Oscillating Drops," *AIChE J.*, **12**, 751 (1966).

Boye-Christensen, G., and S. G. Terjesen, "On the Action of Surface-Active Agents on Mass Transfer in Liquid-Liquid Extraction," *Chem. Eng. Sci.*, **9**, 225 (1959).

Burnett, J. C., and D. M. Himmelblau, "The Effect of Surface-Active Agents on Interphase Mass Transfer," *AIChE J.*, **16**(2), 185 (1970).

Danckwerts, P. V., "Absorption by Simultaneous Diffusion and Chemical Reaction into Particles of Various Shapes and into Falling Drops," *Trans. Farad. Soc.*, **47**, 1,014 (1951).

Davies, J. T., *Advances in Chemical Engineering*, Academic Press, New York, **4**, 1 (1963).

Dombrowski, N., and G. Munday, "Spray Drying," in *Biochemical and Biological Engineering Science*, N. Blakebrough, ed., Academic Press, New York, **1** (1968).

Etzel, M. R., and C. J. King, "Loss of Volatile Trace Organics During Spray Drying," *Ind. Eng. Chem. Proc. Des. Dev.*, **23**(4), 705 (1984).

Etzel, M. R., "Loss of Volatile Trace Organics During Spray Drying," Ph.D. Diss., Univ. of California, Berkeley (1983).

Frey, D. D., "Experimental and Theoretical Investigation of Foam Spray Drying," Ph.D. Diss., Univ. of California, Berkeley (1984).

Frey, D. D., and C. J. King, "Diffusion Coefficients of Acetates in Aqueous Sucrose Solutions," *J. Chem. Eng. Data*, **27**, 419 (1982).

Gal-Or, B., G. E. Klinzing, and L. L. Tavlarides, "Bubble and Drop Phenomena," *Ind. Eng. Chem.*, **61**(2), 21 (1969).

Gal-Or, B., and S. Waslo, "Hydrodynamics of an Ensemble of Drops (or Bubbles) in the Presence or Absence of Surfactants," *Chem. Eng. Sci.*, **23**, 1,431 (1968).

Garner, F. H., and A. R. Hale, "The Effect of Surface-Active Agents in Liquid Extraction Processes," *Chem. Eng. Sci.*, **2**, 157 (1953).

Garner, F. H., and J. J. Lane, "Mass Transfer to Drops of Liquid Suspended in a Gas Stream. II: Experimental Work and Results," *Trans. Inst. Chem. Eng.*, **37**, 162 (1959).

Hadamard, J., *Compt. Rend.*, **152**, 1,735 (1911). Results reported in English in Happel, J., and H. Brenner, *Low Reynolds Number Hydrodynamics*, 2nd ed., Noordhoff, Leyden, The Netherlands (1973).

Hamielec, A. C., and A. I. Johnson, "Viscous Flow Around Fluid Spheres at Intermediate Reynolds Numbers," *Can. J. Chem. Eng.*, **4**, 41 (1962).

Handlos, A. E., and T. Baron, "Mass and Heat Transfer from Drops in Liquid-Liquid Extraction," *AIChE J.*, **3**, 127 (1957).

Hasson, D., and J. Mizrahi, "The Drop Size of Fan-Spray Nozzles: Measurements by the Solidfying Wax Method Compared with Those Obtained by Other Sizing Techniques," *J. Trans. Inst. Chem. Eng.*, **39**, 415 (1961).

Hughes, R. R., and E. R. Gilliland, "Mass Transfer Inside Drops," *Chem. Eng. Prog. Symp. Ser.*, **51**(16), 101 (1955).

Johnson, A. I., and A. E. Hamielec, "Mass Transfer Inside Drops," *AIChE J.*, **6**(1), 145 (1960).

Kerkhof, P. J. A. M., and H. A. C. Thijssen, "Quantitative Study of the Effects of Process Variables on Aroma Retention During the Drying of Liquid Foods," *AIChE Symp. Ser.*, **73**(163), 33 (1977).

Kerkhof, P. J. A. M., and W. J. A. H. Schoeber, "Theoretical Modeling of the Drying Behavior of Droplets in Spray Drying," in *Advances in Preconcentration and Dehydration of Foods*, A. Spicer, ed., Applied Sci. Publ., London, 349 (1974).

Kieckbusch, T. G., and C. J. King, "Volatiles Loss During Atomization in Spray Drying," *AIChE J.*, **26**, 718 (1980); *ibid.*, **27**, 528 (1981).

Kieckbusch, T. G., "Volatiles Losses in the Nozzle Zone During Spray-Drying of Liquid Foods," Ph.D. Diss., Univ. of California, Berkeley (1978).

Kronig, R., and J. C. Brink, "The Theory of Extraction from Falling Droplets," *Appl. Sci. Res.*, **A-2**, 142 (1950).

La Mer, V. K., *Retardation of Evaporation by Monolayers*, Academic Press, New York (1962).

Lewis, J. B., "The Mechanism of Mass Transfer of Solutes Across Liquid-Liquid Interfaces. I: The Determination of Individual Transfer Coefficients for Binary Systems, *Chem. Eng. Sci.*, **3**, 248 (1954).

Mudge, L. K., and W. J. Heideger, "The Effect of Surface-Active Agents on Liquid-Liquid Mass Transfer Rates," *AIChE J.*, **16**(4), 602 (1970).

Newman, A. B., "The Drying of Porous Solids: Diffusion and Surface Emission Equations," *Trans. Am. Inst. Chem. Eng.*, **27**, 203 (1931).

Patel, J. M., and R. M. Wellek, "Handlos and Baron Model: Short Contact Times," *AIChE J.*, **13**(2), 384 (1967).

Perry, R. H., D. W. Green, and J. O. Maloney, *Perry's Chemical Engineers' Handbook*, 6th ed., McGraw-Hill, New York (1984).

Plevan, R. E., and J. A. Quinn, "The Effect of Monomolecular Films on the Rate of Gas Absorption into a Quiescent Liquid," *AIChE J.*, **12**(5), 894 (1966).

Pritchard, C. L., and S. K. Biswas, "Mass Transfer from Drops in Forced Convection," *Brit. Chem. Eng.*, **12**(6), 879 (1967).

Rothe, P. H., and J. A. Block, "Aerodynamic Behavior of Liquid Sprays," *Int. J. Multiphase Flow*, **3**, 263 (1977).

Sada, E., and D. M. Himmelblau, "Transport of Gases through Insoluble Monolayers," *AIChE J.*, **13**, 860 (1967).

Simpson, S. G., and S. Lynn, "Vacuum-Spray Stripping of Sparingly Soluble Gases from Aqueous Solutions," *AIChE J.*, **23**, 666 (1977).

Sivetz, M., and N. W. Desrosier, *Coffee Technology*, AVI Pub. Co., Westport, CN (1979).

Skelland, A. H. P., and R. M. Wellek, "Resistance to Mass Transfer Inside Droplets," *AIChE J.*, **10**(4), 491 (1964).

Springer, T. G., and R. L. Pigford, "Influence of Surface Turbulence and Surfactants on Gas Transport through Liquid Interfaces," *Ind. Eng. Chem. Fundam.*, **9**(3), 458 (1970).

Streicher, R., and K. Schügerl, "Interchange of Fluid Mechanics and Mass Exchange in Droplets," *Chem. Eng. Sci.*, **32**, 23 (1977).

Tavlarides, L. L., et al., "Bubble and Drop Phenomena," *Ind. Eng. Chem.*, **62**(11), 6 (1970).

Thijssen, H. A. C., "Flavor Retention in Drying Preconcentrated Food Liquids," *J. Appl. Chem. Biotechnol.*, **21**, 372 (1971).

Whitman, W. G., L. Long, and H. Y. Wang, "Absorption of Gases by a Liquid Drop," *Ind. Eng. Chem.*, **18**, 363 (1926).

Zakarian, J. A., and C. J. King, "Volatile Loss in the Nozzle Zone During Spray-Drying of Emulsions," *Ind. Eng. Chem. Proc. Des. Dev.*, **21**, 107 (1982).

Zakarian, J. A., "Volatile Loss in the Nozzle Zone During Spray-Drying of Emulsions," Ph.D. Thesis, Univ. of California, Berkeley, (1979).

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