COMMUNICATIONS TO THE EDITOR

Handlos and Baron Model: Short Contact Times

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Handlos and Baron have proposed a mathematical model which describes the internal mass transfer mechanism for droplets that have a special type of turbulent internal circulation (*1*). The turbulence is in the form of random radial vibrations superimposed upon internal streamlines which are in the form of a system of tori. These fluid dynamic conditions are most likely to be encountered in the high-droplet Reynolds number-nonoscillating region. A detailed description of the model and the assumptions involved are given elsewhere (*1*, *2*). The model has been compared (with *approximate* solutions used) with experimental data for single droplet systems (*1* to *4*), and its use has also been suggested for multiple droplet systems (*5*, *6*). The object of this work is to obtain a more exact solution to the Handlos and Baron model which is rigorously applicable for *short* (and long) contact times and *finite* continuous phase resistances. This more exact solution (along with various approximate solutions) will then be compared with some of the single droplet extraction data which have been presented in the literature.

Solutions to droplet extraction models can be presented either in terms of droplet extraction efficiencies or in terms of mass transfer coefficients. These two performance indices for extraction are related by the following expression

$$E_m = 1 - \exp\left(-\frac{A}{V}K_d t\right) \tag{1}$$

when there is resistance to mass transfer in both the continuous and dispersed phases. If the continuous phase resistance is negligible ($k_c \to \infty$), K_d equals k_d.

The solution to the Handlos and Baron model may be expressed in terms of an extraction efficiency as

$$E_m = 1 - 2\sum_{h=1}^{\infty} B_n^2 \exp(-\lambda_n bt) \tag{2}$$

where

$$b = \frac{U}{128(1 + \mu_d/\mu_c)d} \tag{3}$$

APPROXIMATIONS FOR LARGE CONTACT TIMES

The approximate solutions of Handlos and Baron (*1*) and Wellek and Skelland (*2*), consider only the first term in the above series summation; thus their solutions are limited to large contact times when only the first term is dominant. Furthermore, in both papers (*1*, *2*) it is tacitly assumed that $2B_1^2$ is equal to unity. Thus, Equations (1) and (2) may be solved for K_d, subject to the above assumptions and, therefore, for large contact times

$$K_d = \frac{\lambda_1 U}{768(1 + \mu_d/\mu_c)} \tag{4}$$

In terms of the droplet extraction efficiency, these large contact time solutions are expressed as

$$E_m = 1 - \exp(-\lambda_1 bt) \tag{5}$$

The Rayleigh-Ritz variational method was used in references *1* and *2* to solve for values of λ_1. Handlos and Baron considered the case of *zero* continuous phase resistance and found $\lambda_1 = 2.88$. Wellek and Skelland considered the effect of various *finite* continuous phase resistances on the value of λ_1 (see Table 1 in reference *2*, where λ_1 is given as a function of a modified continuous phase mass transfer coefficient *h*).

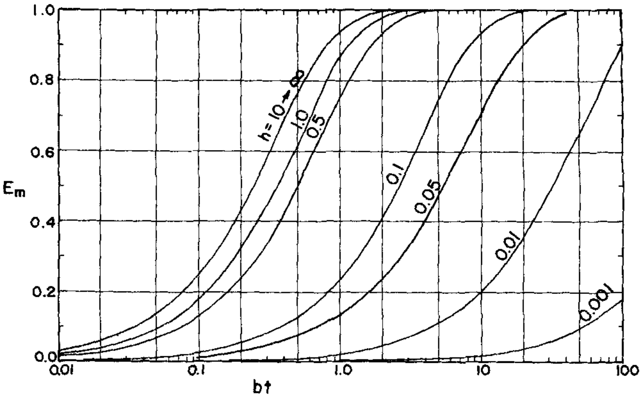

Fig. 1. Fraction extracted vs. dimensionless time. Not to be used for low values of *bt*. Solution by Wellek and Skelland (2).

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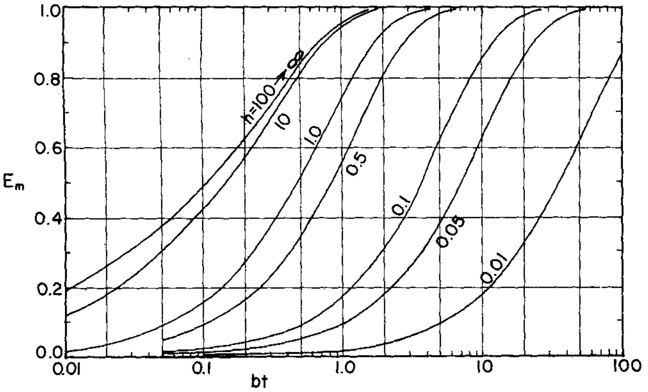

Fig. 2. Fraction extracted vs. dimensionless time. Solution by finite-difference method.

Equation (5) is represented in Figure 1 for various values of the continuous phase resistance parameter h. The extent to which this approximate solution is valid at short contact times is investigated in the next section of this work.

SOLUTION FOR EXTENDED RANGE OF CONTACT TIMES

In this communication the Handlos and Baron model was solved for the extraction efficiency E_m as a function of the dimensionless contact time bt for various values of the modified continuous phase mass transfer coefficient h.

The partial differential equation and boundary conditions describing the Handlos and Baron model with continuous phase resistance [see Equations (10), (11), and (12) of reference 2] were solved by using the implicit finite-difference technique suggested by Crank and Nicholson (*7*). All computational details are given in reference *16*. The results are summarized in Figure 2.

As may be seen from Figures 1 and 2, the continuous phase resistance (which is proportional to the inverse of h) has a significant effect on the extraction efficiency. Olander (*8*) has recently solved this model for the case where h is equal to *infinity*, and the results are in close agreement with the upper curve in Figure 2 of this work. The error introduced by the use of the large contact time solution [that is, Equation (5)] can be seen by comparing Figures 1 and 2.

At values of h greater than about 5.0, the finite-difference solution (Figure 2) indicates values of E_m greater than those predicted when using the large contact time solution (Figure 1). This difference is more pronounced at small values of bt. For values of h less than 5.0, E_m as calculated in this work is less than that predicted by Equation (5). However, as the continuous phase resistance increases (at values of h less than about 0.01), the difference between the large contact time solution and the finite-difference solution becomes negligible; this might be expected since the continuous phase resistance would then dominate the extraction process.

COMPARISONS WITH EXPERIMENTAL DATA

Comparisons of mathematical models with single droplet experimental data will be divided in two sections: data for which virtually all the resistance to mass transfer is in the dispersed phase, and data for which resistance to mass transfer exists in both the dispersed and continuous phase. Conclusions drawn from these comparisons should be considered tentative in view of the relatively small data sample studied and the apparently inevitable scatter exhibited by the mass transfer data for dispersed phases.

NEGLIGIBLE CONTINUOUS PHASE RESISTANCE

The Colburn and Welsh (*13*) technique has been used to study the dispersed phase resistance for the water (droplet)-ethyl acetate system. Thirty data points were available for these high Reynolds number-low interfacial tension data. Some of the droplets were oscillating. Experimentally observed extraction efficiencies are closer to the values of E_m as given in Figure 2 (for $h = \infty$) than with those calculated with the use of the large contact time Handlos and Baron solution [that is, Equation (5) with $\lambda_1 = 2.88$, which is shown in Figure 1 for $h = \infty$]. The average deviation of E_m calculated with Figure 2 from the experimental values of E_m is $+18\%$, whereas the average deviation of E_m calculated with Equation (5) from the experimental values of E_m is -27%.

RESISTANCE IN BOTH PHASES

In this section comparisons will be made with the *overall* mass transfer coefficients used (although extraction efficiencies could also have been used). The data examined are primarily those of Handlos and Baron (see reference *2* for exceptions). The overall mass transfer coefficient K_d will be calculated by using four variations of the Handlos and Baron model which are described as follows:

1. Finite-difference results developed in this work

$$K_d = -\frac{d}{6t}\ln(1-E_m) \tag{6}$$

where E_m is a function of k_c and bt as shown in Figure 2. The continuous phase mass transfer coefficient was estimated by using the following correlation of Garner and Tayeban (*14*):

$$\frac{k_c d}{D_c} = 50 + 0.0085\left(\frac{dU\rho_c}{\mu_c}\right)^{1.0}\left(\frac{\mu_c}{\rho_c D_c}\right)^{0.7} \tag{7}$$

2. Large contact time method

$$K_d = \frac{\lambda_1 U}{768(1+\mu_d/\mu_c)} \tag{4}$$

where λ_1 is a function of k_c as indicated in Table 1 of reference *2*. Here also, k_c is estimated by using Equation (7).

3. Two-resistance method I

$$\frac{1}{K_d} = \frac{m}{(k_c)_{\text{G.T.}}} + \frac{1}{(k_d)_{k_c\to\infty}} \tag{8}$$

where k_c is estimated by Equation (7) and k_d (at $k_c \to \infty$) is given by Equation (4) with $\lambda_1 = 2.87$.

4. Two-resistance method II

$$\frac{1}{K_d} = \frac{m}{(k_c)_{\text{Hig.}}} + \frac{1}{(k_d)_{k_c\to\infty}} \tag{9}$$

where k_c is estimated by the Higbie (*15*) type relation:

$$k_c = \sqrt{\frac{4D_c}{\pi t}} \tag{10}$$

This method is the form originally used by Handlos and Baron (*1*). Four variations have been listed above in what was believed to be, from a theoretical standpoint, decreasing accuracy. The errors associated with the numerical method used to prepare Figure 2 (used in method 1) are believed to be minimal. As described earlier, method 2 assumes that only the first term in the series solution is important and thus should be applicable—primarily for large contact times. Methods 3 and 4 introduce additional deviations from methods 1 and 2 by the assumption of the additivity of resistances and also the use of the Handlos and Baron solution for k_d ($k_c \rightarrow \infty$). The Garner and Tayeban correlation [Equation (7)] is expected to predict k_c with greater accuracy than the Higbie relation [Equation (10)].

[The authors would like to call attention to the fact that k_d in Equations (19) and (20) of reference 2 should have, for generality, been given as K_d, so that Equations (19) and (20) in reference 2 could have applied for all values of continuous phase mass transfer coefficients. Thus, the expressions for k_d in modifications *A* and *B* of reference 2 are not correct, and consequently they have not been considered in this work.]

The data which are compared with the above four methods are the same as those indicated in reference 2. The droplet Reynolds numbers ranged from 569 to 987 and interfacial tensions ranged from 25 to 35. The average absolute and arithmetic percentage deviations of the experimental data from the values of K_d predicted by these four methods are given as follows:

	Absolute	Arithmetic
Method 1	48.6	+41.0
Method 2	101	+96.8
Method 3	25.8	+ 3.8
Method 4	31.8	+27.0

Although one should be cautious about drawing conclusions from the limited data studied, it appears that both methods 3 and 4 (the long contact time-additive resistance methods) are more accurate than the finite-difference results. It should be noted that the droplet extraction data used in this comparison (only seventeen measurements of K_d) did not include experiments for very short drop contact times. Future experimentation, for a wider range of drop contact times and interfacial tensions, may indicate closer agreement with the finite difference results developed in this work.

The Handlos and Baron model is frequently described as applying to turbulent and/or *oscillating* droplets (*2*, *3*, *9*, *10*). However, recent photographic evidence of Rose and Kintner suggests that the circulation patterns in oscillating droplets deviate considerably from those postulated by the Handlos and Baron model (*11*). Fluid motion within fully oscillating drops, for the systems they studied, appeared to be a type of random mixing with only a slight tendency for internal circulation. Thus, it is probably best to consider the Handlos and Baron model (solution given in Figure 2) as applying only for droplets with vigorous internal circulation, that is, in the high Reynolds number-*non*oscillating region.

On the basis of their observations, Rose and Kintner have proposed an extraction model for vigorously oscillating droplets which assumes complete mixing in the droplet for every oscillation cycle and which also considers the effect of interfacial stretch (*11*). Angelo et al. (*12*) have recently presented a generalization of the penetration theory for surface stretch which was also applied to predict mass transfer to oscillating droplets. Both models (*11*, *12*) require estimates of the amplitude and frequency of oscillation. These models are appealing from a physical standpoint, although the calculation of the fraction extracted appears to be slightly complicated.

Despite the fact that these surface stretch models (*11*, *12*) more closely represent the actual phenomena present in an oscillating droplet, comparisons (*12*) with experimental data available (for $k_c = \infty$) indicate that the Handlos and Baron model [essentially Equation (8), in terms of E_m] predicts values of E_m not significantly different from those of the two surface stretch models. More precise and extensive data in the high Reynolds number region are needed to provide a more conclusive test of the available droplet extraction models.

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NOTATION

A = average interfacial area of the droplet

b = $\dfrac{U}{128(1 + \mu_d/\mu_c)d}$

B_n = coefficients in series solution

C_o, C_f, C^* = initial, final, and equilibrium concentrations, respectively

E_m = extraction efficiency during the free fall period = $(C_o - C_f)/(C_o - C^*)$

d = equivalent droplet diameter

D_c = molecular diffusivity in the continuous phase

h = modified continuous phase mass transfer coefficient = $512\ k_c\ (1 + \mu_d/\mu_c)/mU$

k_c, k_d = individual continuous and dispersed phase mass transfer coefficients, respectively

K_d = overall mass transfer coefficient based on dispersed phase concentration units

m = dispersed phase concentration/continuous phase concentration at equilibrium

t = contact time during the free fall period

U = droplet free fall (or rise) slip velocity

V = droplet volume

λ_n = eigenvalues in the series solution

μ_d, μ_c = viscosity of the dispersed and continuous phase, respectively

ρ_c = density of the continuous phase

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