# Extraction with Single Turbulent Droplets

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A dispersed phase of liquid droplets is present in many types of liquid extraction contactors. One of the major design problems is to predict the rate of mass transfer to these droplets. Much of this work on mass transfer is reviewed in references (*4*), (*5*), (*9*), (*15*), and (*18*).

The present work is concerned with the mathematical model of Handlos and Baron (*8*), which describes the unsteady state mass transfer mechanism for droplets possessing a special type of turbulent internal circulation. The Handlos and Baron turbulence model for circulating and/or oscillating droplets has been modified in this paper by considering the effect of a finite continuous phase resistance in the boundary condition of the original differential equation. A solution to the problem is obtained for various values of the continuous phase resistance by the Rayleigh-Ritz variational technique, using a polynomial approximation for the concentration distribution inside the droplet.

Major problems that will not be discussed here are the prediction of interfacial area, mass transfer during formation, coalescence and breakup of droplets, and the effect of gross flow patterns in the contactor.

## PREVIOUS THEORETICAL CONSIDERATIONS

Including the Handlos and Baron model, there are four popular mathematical representations of mass transfer in the dispersed phase (*2, 9, 12, 18*).

### Stagnant Spherical Droplets

Newman's (*16*) expression for spherical droplets with no internal circulation was derived from the usual unsteady state differential equation for molecular diffusion in solid spheres assuming no resistance to mass transfer in the continuous phase. In terms of a dispersed phase mass transfer coefficient

$$k_d = -\frac{d}{6t}\ln\left[\frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-n^2\pi^2\frac{4D_L t}{d^2}\right)\right] \quad (1)$$

Groeber (*6*) is credited with considering the effect of a finite continuous phase resistance for the rigid sphere in the following expression:

$$k_d = -\frac{d}{6t}\ln\left[6\sum_{n=1}^{\infty}A_n\exp\left(-\lambda_n^2\frac{4D_L t}{d^2}\right)\right] \quad (2)$$

where $A_n$ and $\lambda_n$ are functions of $k_c$ (*2*).

### Laminar Circulation in Spherical Droplets

Kronig and Brink (*13*) derived a relation for droplets with internal circulation described by the Hadamard-Rybczinski (*7, 17*) flow patterns. These flow patterns were derived from the equations of motion simplified for the Stokes flow regime ($N_{Re} < 1$). It was assumed that solute diffusion is only in a direction perpendicular to the internal streamlines and that continuous phase resistance is negligible.

$$k_d = -\frac{d}{6t}\ln\left[\frac{3}{8}\sum_{n=1}^{\infty}B_n^2\exp\left(-\lambda_n 64\frac{D_L t}{d^2}\right)\right] \quad (3)$$

$B_n$ and $\lambda_n$ are given by Heertjes et al. (*10*) for $1 \leq n \leq 7$. The relation predicts mass transfer coefficients about 1½ times greater than the Newman expression.

Elzinga and Banchero (*2*) extended the Kronig and Brink solution by considering the effect of a finite continuous phase resistance. Their final expression is the same as the expression of Kronig and Brink, except that $B_n$ and $\lambda_n$ are functions of the continuous phase resistance.

Calderbank and Korchinski (*1*) analyzed heat and mass transfer to the droplet phase by substituting an effective diffusivity in place of the molecular diffusivity appearing in the Groeber expression. This led to the use of a dimensionless correlation factor $R$ ($= D_{eff}/D_L$) for analyzing experimental results (*1, 12*). For mass transfer described by the Kronig and Brink model, $R$ is approximately 2.5.

### Turbulent Circulation in Spherical Droplets

The Handlos and Baron (*8*) mechanism for oscillating and/or vigorously circulating droplets was derived assuming negligible resistance to transfer in the continuous phase, and is expressed below in terms of the dispersed phase mass transfer coefficient by considering just the first term in a series solution.

$$k_d = \frac{\lambda_1 U}{768(1+\mu_d/\mu_c)},\ \lambda_1 = 2.88 \quad (4)$$

Handlos and Baron recommended that when resistance to mass transfer exists in the continuous phase, the Higbie (*11*) relation should be assumed for $k_c$.

$$k_c = \sqrt{\frac{4}{\pi}\frac{D_c}{t_c}} \quad (5)$$

$k_c$ is combined with $k_d$ to obtain an overall mass transfer coefficient by means of the two-resistance theory.

### Completely Mixed Droplet Model

In this model it is assumed that the diffusivity and turbulence of the fluid inside the droplet are large enough so that it can be considered completely mixed (*2*). There is no resistance to mass transfer inside the droplet, and hence $k_d$ may be considered to be infinitely large. One might assume oscillating droplets to approach this mechanism.

It is the purpose of the remainder of this paper to derive a relation identical to Equation (4) expressing, however, $\lambda_1$ as a function of a finite continuous phase resistance, proportional to $1/k_c$.

## OUTLINE OF HANDLOS AND BARON MECHANISM

The development of this eddy diffusion model is devoid of any parameters that must be obtained from experimental measurements. However, it is assumed that the droplet fall velocity, continuous phase viscosity, and dispersed phase viscosity are known.

The model is based on the assumption that internal circulation is fully developed and that the circulation pattern is a system of circular tori. (See

Figure 1 in reference 8.) It is further assumed that *random radial vibrations* are superimposed upon the streamlines. It is the presence of these vibrations that provides mixing between streamlines, and hence is the key to the eddy diffusion mechanism. While the cause of these vibrations is not stated, droplet oscillation is one likely source. The entire transfer process is assumed to take place within the outer surface of the torus.

The differential mass balance on the system within the torus is

$$\frac{\partial C}{\partial t} = \nabla \cdot (\bar{E} \nabla C) \qquad (6)$$

The problem faced by Handlos and Baron was to devise a relationship for the effective diffusivity, $\bar{E}$. It is assumed that the Einstein diffusion equation for two dimensions applies

$$\bar{E} = \frac{\overline{Z}^2}{4\bar{t}} \qquad (7)$$

where $\overline{Z}^2$ *is the mean square displacement of an element of fluid during the average* circulation time of the element, $\bar{t}$.

Handlos and Baron assume that $\bar{t}$ for their model can be approximated by the value of $\bar{t}$ for Hadamard-Rybczinski internal circulation velocities and pattern for laminar circulation. Thus

$$\bar{t} \cong \frac{16}{3} \frac{d}{U} (1 + \mu_d/\mu_c) \qquad (8)$$

By assuming the *limiting* case of complete mixing of an element of fluid in one circulation period (inside the droplet), the following expression was obtained for the mean square displacement of the element for many complete circuits:

$$\overline{Z}^2 = \frac{d^2}{96} (6r^2 - 8r + 3) \qquad (9)$$

If mass transfer is only in the $r$ direction, and if the effect of the torus curvature is negligible, Equations (6) through (9) may be combined to give

$$\frac{\partial C}{\partial t} = \frac{b}{1-y} \frac{\partial}{\partial y} \left[ (1 - 5y + 10y^2 - 6y^3) \frac{\partial C}{\partial y} \right] \qquad (10)$$

where

$$b = \frac{U}{128(1 + \mu_d/\mu_c)} \qquad (11)$$

and $r = 1 - y$.

Handlos and Baron solved Equation (10) for the case of a zero continuous phase resistance. In order to account for a finite continuous phase resistance, their Equation (25) is replaced in this work by the following initial and boundary conditions:

$$C = C_o \qquad 0 \leq y \leq 1 \qquad t = 0 \qquad (12a)$$

$$C \text{ is finite} \qquad y = 1 \qquad t > 0 \qquad (12b)$$

$$\frac{\partial C}{\partial y} = hC \qquad y = 0 \qquad t > 0 \qquad (12c)$$

where

$$h = \frac{512\, k_o (1 + \mu_d/\mu_c)}{mU} \qquad (13)$$

Boundary condition (12c) at the surface of the droplet is obtained as follows, using the nomenclature of Handlos and Baron as much as possible:

$$-\bar{E}(\rho) \frac{\partial C}{\partial t} = \frac{k_c}{m} C \qquad \rho = \frac{d}{4} \qquad t > 0 \qquad (14a)$$

$$-\bar{E}(r) \frac{4}{d} \frac{\partial C}{\partial r} = \frac{k_o}{m} C \qquad r = 1 \qquad t > 0 \qquad (14b)$$

$$-\bar{E}(y) \frac{4}{d} \left( -\frac{\partial C}{\partial y} \right) = \frac{k_o}{m} C \qquad y = 0 \qquad t > 0 \qquad (14c)$$

$$\frac{\partial C}{\partial y} = \frac{k_o}{m} \frac{d}{4} \left[ \frac{2048(1 + \mu_d/\mu_o)}{Ud} \right] C \qquad y = 0 \qquad t > 0 \qquad (14d)$$

$$\therefore \frac{\partial C}{\partial y} = hC \qquad y = 0 \qquad t > 0 \qquad (12c)$$

Boundary condition (12c) assumes that the continuous phase mass transfer coefficient $k_c$ and the distribution coefficient $m$ are uniform over the entire droplet interface. This may not be a precise physical description, but it is probably good enough to describe many situations. These assumptions are also necessary for the Groeber (6) and Elzinga and Banchero (2) modifications mentioned earlier in this communication. Although relations for the variation of $k_c$ with position are available for solid spheres, there apparently is no similar relation for circulating liquid droplets, taking into account the presence of a wake, which could have been used in this work. This boundary condition also assumes that the concentration of solute in the bulk of the continuous phase is negligible during the contact period. In some applications, for example, in extraction studies with swarms of droplets, some countercurrent flow geometries, and certain single droplet systems where $m$ is small, the continuous phase concentration could be high, and a modification of the model would be necessary.

## SOLUTION OF EQUATIONS (10) AND (12)

The method of solution is similar to that used by Kronig and Brink (13) and Handlos and Baron (8). A solution of the following form is obtained:

$$C(y,t) = C_o \sum_{n=1}^{\infty} B_n Y_n \exp(-\lambda_n bt) \qquad (15)$$

which may be transformed by methods described in the above references to

$$E_m = 1 - \frac{M(t)}{M(o)} = 1 - 2 \sum_{n=1}^{\infty} B_n^2 \exp(-\lambda_n bt) \qquad (16)$$

In order to determine $\lambda_n$, the eigenfunction $Y_n(y)$ is approximated by a fourth order polynomial

$$Y_n(y) = a_0 + a_1 y + a_2 y^2 + a_3 y^3 + a_4 y^4 \qquad (17)$$

where

$$a_0 = \frac{a_1}{h}$$

Applying the Rayleigh-Ritz method (19), one solves for $\lambda_n$ from the characteristic equation obtained from the determinant of the coefficients $a_1$, $a_2$, $a_3$, and $a_4$. The determinant is obtained by successive partial differentiation of

$$0 = K \equiv \int_0^1 \frac{d}{dy} \left[ (1 - 5y + 10y^2 - 6y^3) \frac{dY_n}{dy} \right] - \lambda \int_0^1 (1 - y)\, Y_n^2 dy \qquad (18)$$

with respect to $a_1$, $a_2$, $a_3$, and $a_4$. The characteristic equation was obtained using the Hessenberg algorithm (14).

The calculations for $\lambda_n$ were repeated for various values of the continuous phase resistance parameter $h$, and the results are presented in Table 1. The calculations were performed with the aid of an IBM 1620 digital computer. Sixteen significant digits were carried in the intermediate calculations to minimize round off errors.

The dispersed phase mass transfer coefficient is defined as

$$k_d = -\frac{d}{6t} \ln (1 - E_m) \qquad (19)$$

Considering Equations (16) and (19) and limiting attention to the first eigenvalue—as done by Handlos and Baron—leads to the following formulation of the mass transfer coefficient:

$$k_d = \frac{\lambda_1 U}{768(1 + \mu_d/\mu_c)} \qquad (20)$$

where $\lambda_1$ is a function of $k_c$. The extent to which this procedure is justified may be inferred by comparison with experiment. The second (and higher) eigenvalues may be useful in interpreting droplet transfer data for experiments with very short contact periods.

## COMPARISON WITH EXPERIMENTAL DATA

The overall mass transfer coefficients, $K_d$, experimentally observed by Handlos and Baron (*8*) and Garner and Skelland (*3*) for droplets in the high $N_{Re}$ range ($> 500$) were compared with those predicted by the following models.

Original Handlos and Baron

$$\left[\frac{1}{K_d}\right]_{\text{calc.}} = \frac{m}{(k_c)_{\text{Higbie}}} + \frac{1}{(k_d)_{h\to\infty\,(k_c\to\infty)}} \quad (21)$$

Modification A

$$\left[\frac{1}{K_d}\right]_{\text{calc.}} = \frac{m}{(k_c)_{\text{Higbie}}} + \frac{1}{(k_d)_{h=f(k_c,\,\text{Higbie})}} \quad (22)$$

Modification B

$$\left[\frac{1}{K_d}\right]_{\text{calc.}} = \frac{m}{(k_c)_{\text{Garner}}} + \frac{1}{(k_d)_{h=f(k_c,\,\text{Garner})}} \quad (23)$$

Equation (20) is used to predict $k_d$ in each of the above models. Handlos and Baron considered $k_c$ to be infinite (negligible continuous phase resistance)

TABLE 1. EIGENVALUES FOR HANDLOS AND BARON MODEL

| $h$ | $\lambda_1$ | $\lambda_2$ | $\lambda_3$ | $\lambda_4$ |
|---|---|---|---|---|
| 0.001 | 0.002 | 7.46 | 39.0 | 300 |
| 0.01 | 0.022 | 7.49 | 39.1 | 300 |
| 0.1 | 0.270 | 7.85 | 39.3 | 300 |
| 0.5 | 1.377 | 10.71 | 41.6 | 302 |
| 1.0 | 1.980 | 14.27 | 45.4 | 304 |
| 2.5 | 2.445 | 19.54 | 55.9 | 311 |
| 5.0 | 2.674 | 23.42 | 77.2 | 322 |
| 7.0 | 2.708 | 23.76 | 80.0 | 327 |
| 10.0 | 2.731 | 23.72 | 78.6 | 332 |
| 25.0 | 2.821 | 25.26 | 97.4 | 355 |
| 50.0 | 2.847 | 25.54 | 99.5 | 366 |
| 70.0 | 2.850 | 25.24 | 91.2 | 365 |
| 100.0 | 2.852 | 24.88 | 83.5 | 364 |
| 250.0 | 2.861 | 24.85 | 80.9 | 367 |
| 500.0 | 2.864 | 24.76 | 78.8 | 367 |
| 700.0 | 2.864 | 24.64 | 77.0 | 366 |
| 1000.0 | 2.865 | 24.56 | 75.8 | 366 |
| $\infty$ | 2.866 | 24.58 | 75.6 | 367 |

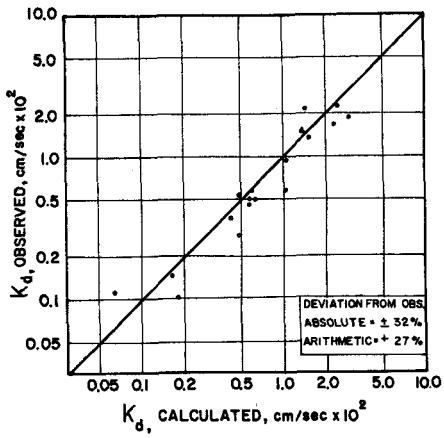


**Fig. 1. Comparison of experimental data of Garner and Skelland (2) and Handlos and Baron (8) with the original Handlos and Baron model.**

and hence $\lambda_1 = 2.88$. In modification *A* and *B* presented in this work, $k_d$ was considered to be a function of $k_c$ by virtue of the dependence of $\lambda_1$ on $k_c$ as indicated in Table 1.

Two relationships for the continuous phase mass transfer coefficient $k_c$ were investigated. The Higbie (*11*) type relation was used for modification *A*

$$k_c = \sqrt{\frac{4}{\pi}\frac{D_c}{t_c}} \quad (5)$$

The use of this relation tacitly ignores the effect of the wake outside the droplet and assumes the maximum possible interfacial velocity. The correlation of Garner and Tayeban (*4*) for circulating and oscillating droplets was used for modification *B*:

$$k_c = \frac{D_c}{d}\left[50.0 + 0.0085\left(\frac{dU\rho_c}{\mu_c}\right)^{1.0}\left(\frac{\mu_c}{\rho_c D_c}\right)^{0.7}\right] \quad (24)$$

The Higbie relation predicts greater continuous phase coefficients than the Garner and Tayeban correlation.

The average percentage deviation of $K_d$ predicted by the three models from the experimental data (17 systems) is presented as follows:

| | Absolute* | Arithmetic† |
|---|---|---|
| Original Handlos and Baron | ±31.8 | +27.0 |
| Modification *A* | ±24.8 | +15.2 |
| Modification *B* | ±31.0 | +16.7 |

* Absolute % deviation

$$= \frac{1}{17}\left[\sum_{n=1}^{17}\left|\frac{(K_d)\text{calc.} - (K_d)\text{exp.}}{(K_d)\text{exp.}}\right|\right](100)$$

† Arithmetic % deviation

$$= \frac{1}{17}\left[\sum_{n=1}^{17}\frac{(K_d)\text{calc.} - (K_d)\text{exp.}}{(K_d)\text{exp.}}\right](100)$$

Modification *A* is considered to be an improvement over the original Handlos and Baron model. Modification *B* is a negligible improvement. One should not necessarily conclude that the Higbie relation is better than the Garner and Tayeban correlation. The observed and calculated values of $K_d$ for the original Handlos and Baron model and modification *A* are presented graphically in Figures 1 and 2.

Although the original experimental data (*3*, *8*) used in this comparison will not be reproduced in this work, the range of the prominent variables is presented as follows:

| | | |
|---|---|---|
| $d$, cm. | : | 0.41 to 0.61 |
| $U$, cm./sec. | : | 10.8 to 16.6 |
| $\mu_c$, centipoise | : | 0.61 to 1.05 |
| $\mu_d$, centipoise | : | 0.61 to 1.75 |
| $m$ | : | 0.023 to 37.7 |
| $\sigma$, dyne/cm. | : | 25 to 35 |
| $N_{Re}$ | : | 569 to 987 |
| $(N_{Sc})_c$ | : | 211 to 1140 |
| $K_d$, cm./sec. | : | $0.101 \times 10^{-2}$ to $2.22 \times 10^{-2}$ |
| $h$ | : | 0.054 to 60.0 |
| $\lambda_1$ | : | 0.0145 to 2.87 |

All data in Table 2 of reference (*8*) were included in this comparison, except two systems in which 100% of the resistance was estimated to be inside the droplet. These latter two systems would have not been a test of the modification presented in this work. It should not be emphasized that the droplet Reynolds number was in all cases greater than 500.

It is possible that with the appropriate changes in nomenclature, the development presented in this work may describe the analogous single droplet heat transfer process; however, a comparison with experimental heat transfer data was not attempted.

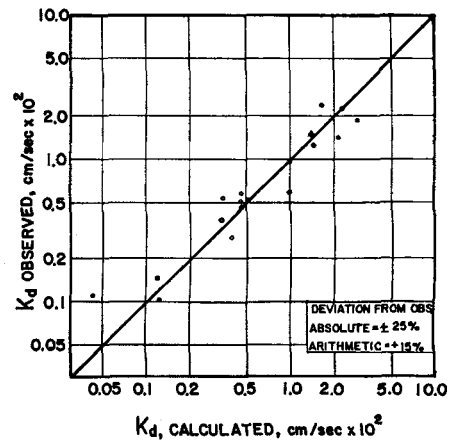


**Fig. 2. Comparison of experimental data of Garner and Skelland (2) and Handlos and Baron (8) with Equation (22).**

While modification A is an improvement over the original method of Handlos and Baron, the absolute percentage deviation of 25 is still high. It is hoped that future work, both theoretical and experimental, will result in a more accurate description of the droplet mass transfer mechanism at high Reynolds numbers. For example, at very high $N_{Re}$ when oscillation of droplet shape is encountered, it is rather doubtful that the circulation patterns as assumed in this model will actually be observed.

## NOTATION

$A_n$ = coefficients in Equation (2), dimensionless

$a_i$ = coefficients in Equation (17), dimensionless; $i$ = 0, 1, 2, 3, 4

$B_n$ = coefficients in Equations (3) and (16), dimensionless

$b$ = a constant defined by Equation (11)

$C$ = concentration of solute in dispersed phase

$C_o$, $C_f$, $C^*$ = initial, final, and equilibrium concentrations, respectively, (initial and final are uniform concentrations)

$D_L$, $D_e$ = molecular diffusivity of the solute in the dispersed and continuous phase, respectively

$D_{eff}$ = experimental effective mass diffusivity of the solute in droplet

$d$ = droplet diameter

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

$\overline{E}$ = effective diffusivity predicted by Equation (7)

$h$ = modified continuous phase mass transfer coefficient, defined by Equation (13)

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

$K_d$ = overall mass transfer coefficient, dispersed phase, cm./sec.

$K$ = variation constant defined by Equation (18), dimensionless

$m$ = dispersed phase concentration/continuous phase concentration at equilibrium

$M(t)$ = mass of solute in dispersed phase at time $t$

$n$ = an integer number

$N_{Re}$ = droplet Reynolds number = $\frac{dU\rho_c}{\mu_c}$

$(N_{Sc})_c$ = continuous-phase Schmidt number = $\frac{\mu_c}{\rho_c D_c}$

$r$ = $4\rho/d$ = torus radius, dimensionless

$t$ = time during free fall period

$\overline{t}$ = average circulation time in droplet

$t_c$ = approximate continuous film contact time = $d/U$

$U$ = droplet free fall (or rise) velocity

$y$ = $1 - r$ = distance in from surface of torus, dimensionless

$Y_n(y)$ = a function of $y$ alone (eigenfunction)

$\overline{Z}$ = average displacement of fluid

### Greek Letters

$\lambda_n$ = an eigenvalue, dimensionless

$\mu_d$, $\mu_c$ = viscosity of dispersed and continuous phase, respectively

$\rho$ = torus radius

$\rho_c$ = density of continuous phase

$\sigma$ = interfacial tension

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# Incipient Bubble Destruction and Particulate Fluidization


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On the basis of recent studies of single-particle drag and of bed-shearing forces in horizontal flow, a simple mechanism is proposed for the formation or destruction of bubbles in fluidized beds.

## A MODEL OF INCIPIENT BUBBLE DESTRUCTION

Figure 1 schematically illustrates the well demonstrated flow patterns in and around a rising bubble (5). The interstitial fluidizing velocity field is designated by $V_{mf}$, the *shell* of dense bed surrounding the bubble, through which there exist the streams of recirculating fluid, is outlined by the dashed lines and its thickness designated by $y$; the horizontally directed surface velocity of the circulating fluid along the bottom of the bubble is denoted as $U$. Over the major portion of the bubble's lower surface there exists a velocity gradient penetrating the shell, which in terms of Figure 1 can be denoted by $dU/dy$. This velocity gradient creates a shearing stress tending to dilate the bed within the depth $y$. If this dilation is sufficient in magnitude to swell the bottom surface to within a few particle diameters of the upper surface then the bubble is destroyed or rather its incipient formation is thwarted. This is true, not only because it becomes "flooded" by the swelling bottom surface, but also because the upper surface particles will collapse onto the particles approaching from beneath when their distance of separation is small enough to disturb the drag forces on the upper particles (6).

To evaluate this mechanism of bubble formation, even if only qualitatively, requires means of calculating $U$, $y$, and the bed-dilating *dispersive grain pressures* resulting from $dU/dy$.

## VELOCITY GRADIENT AND DISPERSIVE GRAIN PRESSURE

A rather unique measurement of what he chose to call dispersive grain pressures, a measure of bed dilation forces, was reported by R. A. Bagnold (2) in 1954. His apparatus consisted of a pair of concentric drums. The solid outer drum could be rotated at