

$\rho$  = effective specific resistance of electrolyte in electrode pores

#### LITERATURE CITED

1. de Levie, R., "Advances in Electrochemistry and Electrochemical Engineering," P. Delahay, ed., Vol. 6, Interscience, New York (1967).
2. Austin, L. G., "Handbook of Fuel Cell Technology," C. Berger, Ed., p. 178, Prentice-Hall, Englewood Cliffs, N. J. (1968).
3. *Ibid.*, p. 138.
4. ———, *Trans. Faraday Soc.*, **60** (499), 1319 (1964).
5. ———, *Electrochim. Acta*, **14** (7), 639 (1969).
6. Gagnon, E. G. and L. G. Austin, *J. Electrochem. Soc.*, **118**, 479 (1971).
7. ———, "Handbook of Fuel Cell Technology," C. Berger, Ed., p. 141, Prentice-Hall, Englewood Cliffs, N. J. (1968).
8. Gagnon, E. G., Ph.D. thesis, Pennsylvania State Univ., University Park (1970).

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# Relative Velocities and Pressure Drops in Clouds of Drops, Bubbles, or Solid Particles

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Relative velocities of assemblages of drops or bubbles with respect to the continuous phase depend on many parameters. Generally, these include the densities and viscosities of both phases, the local acceleration, the size and shape of the particles, wall effects, concentration of particles and concentration of the practically ever-present surfactant impurities. The effect of the last two parameters on viscous flow in assemblages consisting of many spherical drops or bubbles (moving in an overall uniform velocity field) has been analyzed recently by Gal-Or and Waslo (2). Employing a free-surface cell model, they expressed their results in terms of the "ensemble" (ens.) relative velocity

$$U_{\text{ens.}} = \frac{3}{2} U_s \left( \Phi^{1/3} - \frac{Y}{W} \right) \quad (1)$$

where

$$W = 3 \left( 1 + \frac{2}{3} \beta \right) + 2\Phi^{5/3} (1 - \beta) \quad (2)$$

$$Y = 2(1 + \beta) + 3\Phi^{5/3} \left( 1 - \frac{2}{3} \beta \right) \quad (3)$$

$$\beta = \frac{\mu^c}{\mu^d + \gamma} = \frac{\mu^c}{\mu^d - \frac{1}{3} K(\partial\sigma/\partial C)_{eq}} \quad (4)$$

and

$$K = \frac{2\Gamma_0 a}{2D_i s + \alpha^* D_b s a^2 / [D_b s + \alpha^* \delta(\partial\Gamma/\partial C)_{eq}]} \quad (5)$$

Here  $\Phi$  is the volumetric fraction occupied by the particles and  $\gamma$  an interfacial viscosity due to surfactant impurities.

More recently, a zero-vorticity cell model has been employed by Gal-Or (3) to express ensemble relative velocity as

$$U_{\text{ens.}} = -\frac{3}{2} U_s \left( W'\Phi - Y' + \frac{6}{5} \Phi^{1/3} \right) \quad (6)$$

where

$$W' = \frac{1}{3 + 2\beta} \left[ \frac{2}{5} \Phi(1 - \beta) - 1 \right] \quad (7)$$

and

$$Y' = \frac{1}{3 + 2\beta} (\Phi + 2\beta + 2) \quad (8)$$

The aim of this paper is to evaluate an alternative definition of relative velocity and then to check which one may better characterize experimental data on viscous flow at low Reynolds numbers in dispersed systems. Employing the aforementioned models, we derive new formulations for the average relative velocity and pressure drop which may be widely applicable in characterizing viscous flows of drops or bubbles.

#### AVERAGE RELATIVE VELOCITY

The capability of Equations (1) and (6) to predict ensemble velocities is shown in Figure 1. Curves 1 to 4 are based on Equations (6), (1), (15), and (12), respectively; curves 6 and 7, on semitheoretical relations of Oliver (12) and Hawksley (7), respectively; curves 5 and 9 on experimental data of Adler and Happel (1) and Happel and Epstein (5), respectively; and curve 8 on experimental data of Hanratty and Bandukualka (4), Le Clair (8), and Lewis et al. (10). The theoretical predictions are, in general, lower than the experimental data.

Actual experimental data of this type are generally reported in terms of average relative velocity between the two phases involved (sometimes called the "slip" velocity). Comparison between experimental data and theoretical relations should, therefore, be carried out on this basis, rather than on the basis of  $U_{\text{ens.}}$ . The average relative velocity for a spherical cell model is defined by

$$\bar{U}_{\text{rel.}} = \frac{\int_{\phi=0}^{2\pi} \int_{\eta=0}^{\Phi^{-1/3}} [V_{\theta}^c]_{\theta=\pi/2} a^2 \eta d\eta d\phi}{\pi a^2 (\Phi^{-2/3} - 1)}$$

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$$\frac{\int_{\phi=0}^{2\pi} \int_{\eta=0}^1 [V_{\theta}^d]_{\theta=\pi/2} a^2 \eta d\eta d\phi}{\pi a^2} \quad (9)$$

This definition may at first sight seem to be inconsistent with some of the early uses of the cell models (6), where the inner sphere in the cell was assumed to move in a straight line with the ensemble velocity. However, such an assumption has not been used in the evaluation of the velocity components  $V_{\theta}$  as introduced in Equation (10). Hence this new definition of the average relative velocity is consistent with the general concept of a cell model which is so useful in the evaluation of simultaneous momentum, heat, and mass transfer in concentrated particulate systems (14).

Here one must also bear in mind that the velocity components given below represent a statistical expectation (3) of the velocity field in ensembles consisting of many drops or bubbles. They do not represent the velocity field as may be measured around a given particle in the ensemble. Therefore,  $\bar{U}_{rel.}$  is an expected statistical property of the entire ensemble. Employing in Equation (9) the tangential velocities

$$V_{\theta}^c = \frac{3}{2} \frac{U_s}{W} \left( -\frac{1}{2\eta^3} - \frac{1}{2\eta} W + Y - 2\Phi^{5/3}\eta^2 \right) \sin \theta \quad (10)$$

$$V_{\theta}^d = \frac{3}{2} \frac{U_s}{W} \beta (1 - \Phi^{5/3}) (2\eta^2 - 1) \sin \theta \quad (11)$$

from the Gal-Or and Waslo solution (2), and noting that the second integral in Equation (9) must vanish, the average relative velocity is expressed by

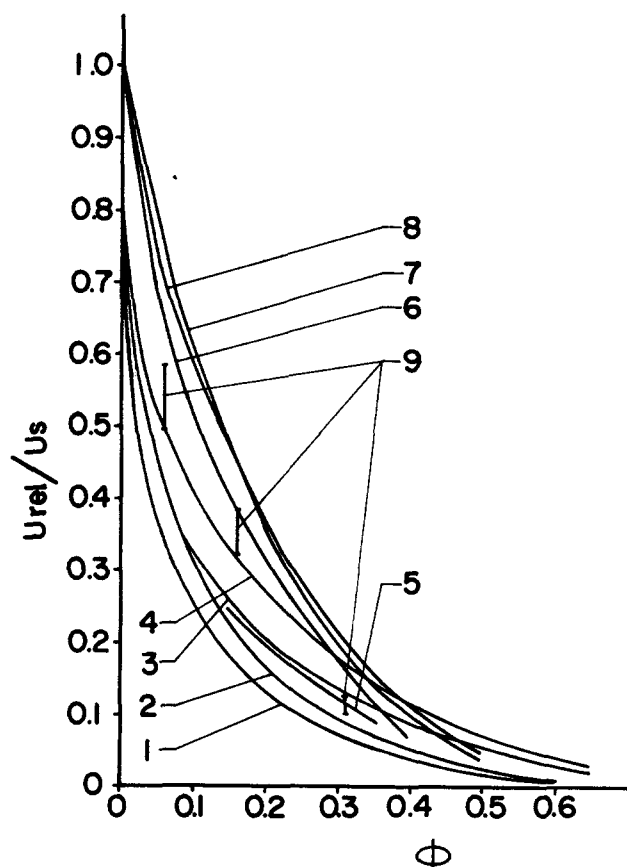


Fig. 1. Comparison of theory and experimental data for flow through beds of fluidized solid particles. Data adapted in part from Le Clair and Hamielec (9).

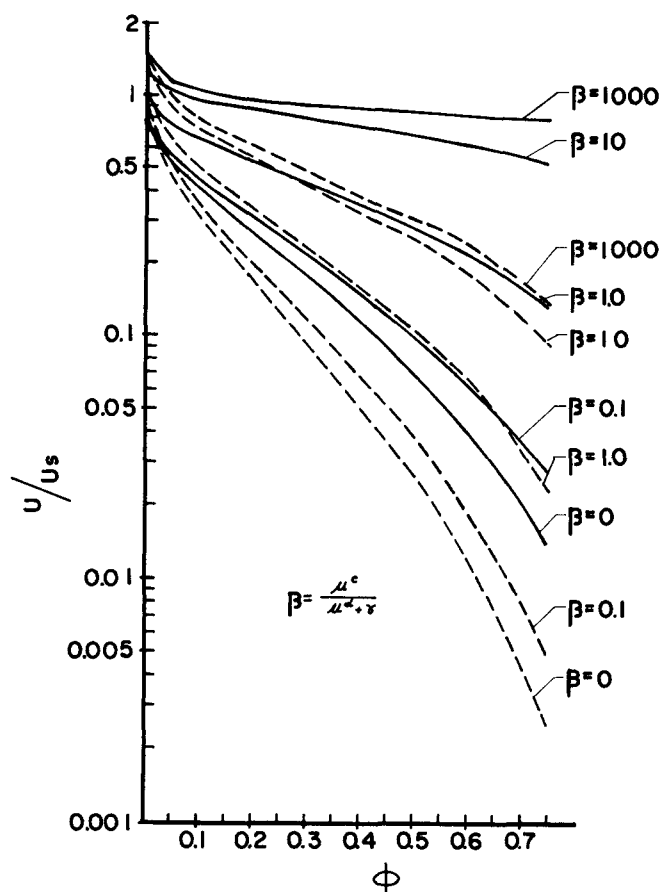


Fig. 2. Dependence of average relative velocity (solid lines) and of ensemble velocity (dashed lines) on the void fraction and the viscosity ratio  $\mu^c/(\mu^d + \gamma)$ , based on the free-surface model.

$$\bar{U}_{rel.} = -\frac{3}{2} U_s \left[ \frac{\Phi^{-1/3} - 1}{\Phi^{-2/3} - 1} - \frac{\Phi^{5/3} - 1}{W(\Phi^{-2/3} - 1)} - \frac{Y}{W} \right] \quad (12)$$

Similarly for the zero-vorticity model (6)

$$V_{\theta}^c = \frac{3}{2} U_s \left( -\frac{1}{2\eta} + \frac{1}{2\eta^3} W' + Y' - \frac{2}{5} \Phi \eta^2 \right) \sin \theta \quad (13)$$

$$V_{\theta}^d = \frac{3}{2} \frac{U_s \beta}{3 + 2\beta} (1 - \Phi) (1 - 2\eta^2) \sin \theta \quad (14)$$

whereby the average relative velocity becomes

$$\bar{U}_{rel.} = -\frac{3}{2} U_s \left[ \frac{W'(\Phi^{1/3} - 1)}{\Phi^{-2/3} - 1} - \frac{\frac{1}{5} \Phi + \frac{6}{5} \Phi^{-1/3} + 1}{\Phi^{-2/3} - 1} - Y' \right] \quad (15)$$

Figure 1 shows that this new definition of average relative velocity is in much better agreement with empirical data than the ensemble relative velocity. Equations (12) and (15) can therefore be used as a reliable guide in correlating experimental data to a first approximation. Figure 1 demonstrates also that Equations (10), (11), (13), and (14) can be employed in analytical evaluations (to a first approximation) of convective heat and mass transfers (15, 16).

Comparison of  $U_{rel.}/U_s$  with  $U_{ens.}/U_s$  for various values of the viscosity parameters  $\beta$  is shown in Figures 2 and 3

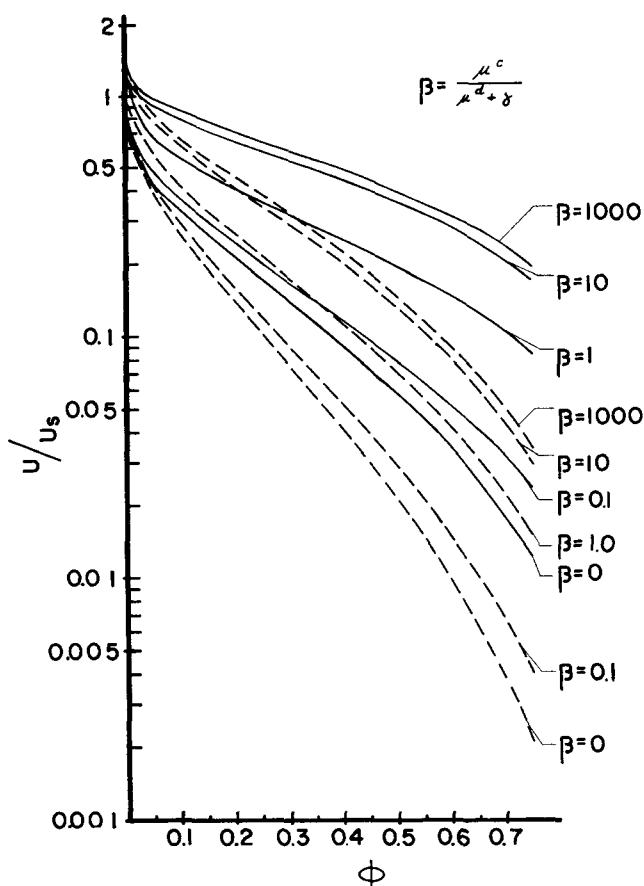


Fig. 3. Dependence of average relative velocity (solid lines) and of ensemble velocity (dashed lines) on the void fraction and viscosity ratio, based on the zero-vorticity model.

from which the following conclusions are deduced:

1. Both  $U_{rel.}/U_s$  and  $\bar{U}_{ens.}/U_s$  are decreased by two basic effects:

a. For a given value of  $\Phi$ , both velocity ratios decrease as  $\beta$  decreases. For a given fluid-fluid system, it means that the relative velocity of the assemblage is decreased as the interfacial viscosity  $\gamma$  increases.

b. At constant  $\beta$  values, the velocity ratios decrease due to increase in particle concentration. This effect is due to hindering interactions among adjacent particles.

2. The relative velocity is highly sensitive to traces of surfactant impurities (through  $\gamma$  and  $\beta$ ), whose effects are much more pronounced at high concentration of droplets. This behavior is in agreement with experimental results (11, 14).

3.  $\bar{U}_{rel.}/U_s$  are higher than  $U_{ens.}/U_s$  for the same value of  $\Phi$  and  $\beta$ .

4. As one approaches the limiting case of a single solid sphere (i.e., when  $\beta \rightarrow 0$ ,  $\Phi \rightarrow 0$ ),  $\bar{U}_{rel.}$  (or  $U_{ens.}$ ) tends to the Stokes velocity.

5. Viscous flow through assemblages of solid particles, drops, or bubbles is governed by the same principles.

For fluid-fluid systems in the absence of an a priori method to estimate  $\gamma$  [cf. Equations (4) and (5)], one may resort to an empirical comparison, such as presented in Figure 4, which shows the data of Weaver et al. (13) for methyl isobutyl ketone and isobutanol drops rising through a quiescent water phase in a 1 7/8-in., Elgin-type spray column. The organic phase was dispersed through 1/8-in. and 1/16-in. diam. nozzles. The slip velocity of Weaver et al. is actually the average relative velocity de-

fined by Equation (9). In their system the dispersed phase was not of uniform size.

Nevertheless, as will be shown, one may disregard the size-distribution effect and consider the dispersion to be composed of uniform droplets with an equivalent radius equal to the mean volume radius of the size-distributed dispersion. Then, using Equation (12), one finds that the theoretical curves for a given value of  $\beta$  follow the general trend of the experimental data. However, agreement cannot be considered satisfactory, particularly for the isobutanol-water system. For the latter, the ratio  $\mu^c/\mu^d$  is 0.234. Even if one completely neglects the retardation effect of possibly present impurities, one should expect the theoretical curve conforming to the experimental data to be of a lower value than that found to best represent the data. Therefore, a large number of experimental tests with various fluid-fluid systems should be carried out before one can regard Equation (12) as furnishing enough justification for full confidence in predicting terminal relative velocities in fluid-fluid particulate systems.

The applicability of the above equations is limited by the requirement that the Reynolds number be small. However, the critical Reynolds number for flow separation is increased considerably as  $\Phi$  increases. At  $\Phi = 0.35$ , for example, separation may not exist even at  $N_{Re} = 500$  (9).

#### PRESSURE DROP IN ENSEMBLES OF DROPS OR BUBBLES

The pressure drop per unit length of a uniform ensemble can be expressed by

$$\frac{\Delta p}{l} = -\frac{F_z N}{V} \quad (16)$$

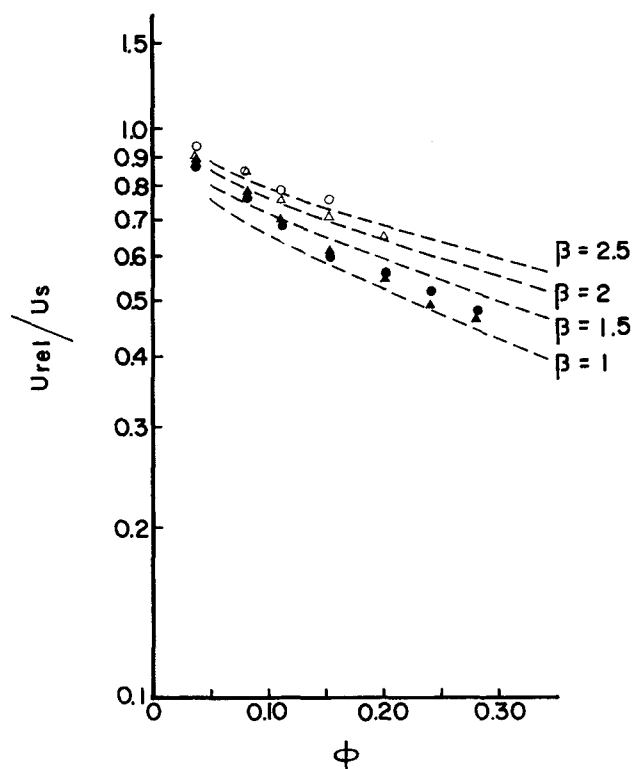


Fig. 4. Comparison of theory and experimental results for flow through spray columns. Data of Weaver et al. (13) for flow of methyl isobutyl ketone (open symbols) and isobutanol (closed symbols) through quiescent water in Elgin-type spray column. Organic phase dispersed by 1/8-in. (circles) and 1/16-in. (triangles) diameter nozzles. Dashed lines— $\bar{U}_{rel.}/U_s$  from free-surface model.

where

$$N = \frac{V\Phi}{\frac{4}{3}\pi a^3} \quad (17)$$

In our treatment of ensembles of uniform drops or bubbles, one finds that the drag on a single typical particle in the ensemble is the same as that on a single particle in an infinite unbounded fluid (17). Thus

$$\frac{\Delta p}{l} = -\frac{F_z \Phi}{\frac{4}{3}\pi a^3} = -(\rho^d - \rho^c)g\Phi = -\frac{9}{2} \frac{\mu^c U_s \Phi}{a^2} \quad (18)$$

However, actual dispersions are seldomly composed of particles of uniform size. Let us consider therefore, the pressure drop over a group of spherical drops or bubbles having a radius between  $a$  and  $(a + da)$  and belonging to a large population of particles characterized by a normalized size distribution  $f(a)$  expressed in terms of number density (14). Employing here the subreactor concept (18)

$$d\left(\frac{\Delta p}{l}\right) = -\frac{F_z N}{V} f(a) da \quad (19)$$

where

$$N = \frac{V\Phi}{\frac{4}{3}\pi(\bar{a}_3)^3} \quad (20)$$

$$\bar{a}_3 = \left[ \int_0^\infty a^3 f(a) da \right]^{1/3} \quad (21)$$

Introducing Equations (20) and (21) into (19), and integrating from 0 to  $\infty$ , one obtains

$$\frac{\Delta p}{l} = -(\rho^d - \rho^c)g\Phi \quad (22)$$

which is identical to Equation (18) for an ensemble of uniform spheres. Using these models one finds therefore that pressure gradients over ensembles of spheres with size distributions are similar to those over ensembles of uniform spheres. This conclusion is general as it is independent of the nature of the size-distribution function.

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## NOTATION

|             |   |
|-------------|---|
| $a$         | = radius of a spherical bubble, drop, or solid particle |
| $\bar{a}_3$ | = mean volume radius defined by Equation (21)           |
| $c$         | = concentration of surfactants                          |
| $D^s$       | = diffusivity of surfactants                            |
| $f(a)$      | = a normalized particle-size distribution function      |
| $F_z$       | = drag force  |
| $g$         | = local acceleration                                    |
| $K$         | = retardation coefficient defined by Equation (5)       |
| $l$         | = length of particulate system                          |
| $N$         | = number of particles in system                         |
| $p$         | = pressure  |
| $r$         | = radius  |

|                   |  |
|-------------------|--|
| $U_{\text{ens.}}$ | = velocity of ensemble of drops, bubbles, or solid particles, Equation (1) or (6)            |
| $U_{\text{rel.}}$ | = average relative velocity between dispersed and continuous phase, defined by Equation (9). |
| $U_s$             | = Stokes' velocity   |
| $V$               | = volume of system   |
| $V_\theta$        | = tangential component of the velocity vector  |
| $W$               | = parameter defined by Equation (2)  |
| $W'$              | = parameter defined by Equation (7)  |
| $Y$               | = parameter defined by Equation (3)  |
| $Y'$              | = parameter defined by Equation (8)  |

## Greek Symbols

|            |  |
|------------|--|
| $\alpha^*$ | = overall desorption rate constant   |
| $\beta$    | = viscosity parameter defined by Equation (4)  |
| $\gamma$   | = interfacial viscosity due to surfactants   |
| $\Gamma$   | = surface concentration of surfactants   |
| $\Gamma_0$ | = equilibrium surface concentration of surfactants   |
| $\delta$   | = thickness of Nernst diffusion boundary layer   |
| $\eta$     | = dimensionless radius, $\eta = r/a$   |
| $\theta$   | = cone angle between the radius vector and the vertical directed in the sense of particle motion |
| $\mu$      | = viscosity  |
| $\rho$     | = density  |
| $\sigma$   | = surface tension  |
| $\phi$     | = polar angle in spherical coordinates   |
| $\Phi$     | = volumetric dispersed-phase holdup fraction   |

## Superscripts

|     |                                  |
|-----|----------------------------------|
| $c$ | = refers to the continuous phase |
| $d$ | = refers to the dispersed phase  |

## Subscripts

|     |                               |
|-----|-------------------------------|
| $b$ | = refers to the bulk solution |
| $i$ | = refers to the interface     |

## LITERATURE CITED

- Adler, I. L., and J. Happel, *Chem. Eng. Progr. Symp. Ser.*, No. 38, 58, 98 (1962).
- Gal-Or, B., and S. Waslo, *Chem. Eng. Sci.*, **23**, 1431 (1968).
- Gal-Or, B., *Can. J. Chem. Eng.*, **48**, 526 (1970).
- Hanratty, T. J., and A. Bandukuala, *AIChE J.*, **3**, 293 (1959).
- Happel, J., and N. Epstein, *Ind. Eng. Chem.*, **46**, 1187 (1954).
- Happel, J., *AIChE J.*, **4**, 197 (1958).
- Hawksley, P. G. W., paper 7 in "Some Aspects of Fluid Flow," Edward Arnold, London (1951).
- Le Clair, B. P., M.A.Sc. thesis, Univ. British Columbia, Vancouver (1964).
- Le Clair, B. P., and A. E. Hamielec, *Ind. Eng. Chem. Fundamentals*, **7**, 542 (1968).
- Lewis, W. K., E. R. Gilliland, and W. C. Bauer, *Ind. Eng. Chem.*, **41**, 1104 (1949).
- Lindland, K. P., and S. G. Terjesen, *Chem. Eng. Sci.*, **5**, 1 (1956).
- Oliver, D. R., *Chem. Eng. Sci.*, **15**, 320 (1961).
- Weaver, R. E. C., L. Lapidus, and J. C. Elgin, *AIChE J.*, **5**, 533 (1959).
- Gal-Or, B., *Intern. J. Heat Mass Transfer*, **11**, 551 (1968); see also *Chem. Eng. Sci.* **19**, 653 (1964); **23**, 631 (1968); **22**, 553 (1969), and *Advances Chem. Eng.* **7**, 295-395 (1968).
- Yaron, I., and B. Gal-Or, *Intern. J. Heat Mass Transfer*, **14**, 727 (1971).
- Waslo, S., and B. Gal-Or, *Chem. Eng. Sci.*, **26**, 829 (1971).
- Yaron, I., and B. Gal-Or, to be published.
- Gal-Or, B., and H. E. Hoelscher, *AIChE J.*, **12**, 499 (1966).

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