Int. J. Heat Mass Transfer. Vol. 24, No. 7, pp. 1221–1228, 1981
Printed in Great Britain

0017-9310/81/071221-08 $02.00/0
Pergamon Press Ltd.

MASS TRANSFER BETWEEN THE PARTICLES AND THE FLUID IN FLUIDIZED BEDS OF LARGE PARTICLES

D. K. PANDEY* and S. N. UPADHYAY†
Department of Energy Engineering, College of Engineering,
University of Illinois at Chicago Circle, Box 4348, Chicago, IL 60680, U.S.A.
and
SURENDRA KUMAR
Department of Chemical Engineering and Technology,
Institute of Technology, Banaras Hindu University, Varanasi 221005, India

(*Received* 15 *July* 1980 *and in revised form* 29 *January* 1981)

Abstract — Generalized empirical correlations available in the literature for particle to fluid mass transfer in fluidized beds are tested for their effectiveness in predicting the experimental mass transfer data for fluidized beds of large particles. Rowe's modification of Nelson and Galloway's theory of particle to fluid mass transfer in dense systems of fine particles is also re-examined and found to agree fairly well with large particle mass transfer data for liquid fluidized beds. A simplified correlation, which is shown to be an approximation of the Nelson–Galloway–Rowe asymptotic expression, is proposed for predicting the mass transfer rate.

NOMENCLATURE

a, total interfacial area of packing $[L^2/L^3]$;
A_p, particle surface area $[L^2]$;
b, a constant defined by equation (4a);
D, molecular diffusivity $[L^2/t]$;
D_e, equivalent diameter of channel $2\varepsilon D_p/3(1-\varepsilon)$ [L];
D_p, particle diameter [L];
f, area availability factor used by Sengupta and Thodos [8] [—];
Fs, Frössling number; $Sh/(Re^{1/2}Sc^{1/3})$ [—];
G, superficial mass flow velocity $[M/L^2t]$
J_d, mass transfer factor; $(k_c/u)Sc^{2/3}$ [—];
k_c, mass transfer coefficient [L/t];
L, bed depth [L];
L_e, effective fluid path in the bed [L];
m, arbitrary exponent on void fraction [—];
Re, Reynolds number; D_pG/μ [—];
S, renewal frequency [1/t];
Sc, Schmidt number; $\mu/\rho D$ [—];
Sh, Sherwood number; k_cD_p/D [—];
T_D, mass transfer factor defined by Yeh [5]; $ShSc^{-1/3}\{[(u+u_t)/\mu][(\varepsilon-\varepsilon_0)/\varepsilon_0]\varepsilon\}^{-1}$ [—];
u, superficial flow velocity [L/t];
u_e, effective flow velocity; $(u/\varepsilon)(L_e/L)$ [L/t];
u_t, terminal velocity [L/t].

Greek symbols

α, proportionality constant in equation (23) [—];
ε, void fraction [—];
ε_0, maximum void fraction in fixed bed [—];
ξ, Nelson–Galloway's parameter defined by equation (25) [—];
μ, absolute viscosity [M/Lt];
ν, kinematic viscosity $[L^2/t]$;
ρ density of fluid $[M/L^3]$;
ϕ, arrangement exponent [—].

*On leave from Department of Mechanical Engineering, H.B. Technological Institute, Kanpur-208002, India.
†On leave from Department of Chemical Engineering, Institute of Technology, Banaras Hindu University, Varanasi-221005, India.

INTRODUCTION

A QUANTITATIVE knowledge of driving force for mass transfer between fluid and particles and resistance to it is required in designing a fluidized bed reactor and also in evaluating the kinetic data from measured conversions in fluidized beds. Quite a substantial volume of published information is available in the literature on this problem [1]. The conclusions, correlations and statements emerging out of most of these studies are quite chaotic in nature since no standard procedure has been followed in analyzing the data and in reporting the results. For example, many investigators have not been able to include the influence of bed voidage in their correlations because the bed expansion has not been measured while acquiring their mass transfer data. Prompted by such inconsistencies many workers have attempted to re-analyze the then available data and have come up with different expressions and recommendations for design purposes. Most of these relations are basically the same and differ only in their numerical constants and operating ranges. This is mainly due to the difference in the ranges of the experimental data which have been used to evolve these relations. These marginal differences and availability of a large number of relations make it difficult to select a suitable design equation.

The object of this paper is to re-examine the suitability of these correlations in predicting the largest possible cross-section of the available experimental data for large particle systems. An attempt is also made to propose a theoretically justified simple correlation which can be used for predicting the mass transfer rate for large particle beds with a fair degree of accuracy.

DISCUSSION

A summary of the various generalized correlations available for predicting the fluidized bed fluid–particle mass transfer rate along with their operating ranges is given in Table 1. The close similarity between most of these relations is clearly evident from this table.

The effectiveness with which these correlations can be used is made complex by the fact that the fluid flows through the bed by two different paths giving rise to two different types of fluidized bed behaviors, the particulate and aggregative types. These behaviors not only depend upon the nature of the fluidizing phase but also on the particle size and size distribution. Particulate fluidization, a characteristic of liquid fluidized beds, is also exhibited by gas fluidized beds when the particle size is large and bed height is much larger than the bed diameter. The laboratory sized fluidized beds used in obtaining liquid-phase data invariably exhibit particulate fluidization. In the gas fluidized beds this behavior is obtained only when the particle size is large. Since most of the gas-phase data have been obtained with small sized beds with particles of comparatively larger size, the assumption of a particulate behavior is justified to some extent. Under all such cases the 'plug flow' assumption is valid and almost all of the investigators have used it in analyzing their data.

The mass transfer data used in the present analysis are those obtained with large particles only. A summary of the major operating parameters covered by the various investigators in acquiring such data is listed in Table 2. Particle to fluid mass transfer in beds of fine particles is a subject of great controversy because the experimental results are much lower than those for large particles and those predicted by various theories, hence these are not included in the present analysis.

The deviations of the experimental data from the various correlations are listed in Table 3. A comparison of these deviations indicate that majority of the relations predict the experimental data with approximately the same accuracy. The least deviation, however, is obtained with equation (20) proposed by Dwivedi and Upadhyay [14]. This equation is based on both fixed and fluidized bed data. The only limitation with it, however, is its purely empirical nature. Theoretical considerations suggest that the fluidized bed mass transfer coefficient in the intermediate Reynolds number range may be expressed as

$$Sh \propto f(Re^{1/2}). \qquad (21)$$

Accordingly a simplified correlation for large particle fluidized bed mass transfer has been also attempted and by regression analysis a relation

$$\varepsilon Sh Sc^{-1/3} = 0.95\, Re^{1/2} \qquad (22)$$

has been obtained which correlates all the data considered in this analysis with a standard deviation of 20.2%. Equation (21) is compared with the gas and liquid-phase mass transfer data in Fig. 1. This deviation is slightly higher than that of equation (20), however, considering the accuracy of the experimental measurements and closeness of equation (22) to theory, its use for design purposes may be preferred over that of equation (20).

The abnormal heat and mass transfer behavior of beds of fine particles was explained by Nelson and Galloway [29] who recognized that at low Reynolds numbers, the single sphere boundary condition ($Sh = 2$) does not apply. Imposing a finite radius condition where the radial transport vanishes by symmetry, they advanced a surface renewal model which adequately explains the observed low Reynolds number behavior.

Nelson and Galloway defined a surface renewal frequency in such a way that

$$S = \alpha^2 D Re Sc^{2/3}/D_p^2 \qquad (23)$$

and obtained

$$Sh = 2\xi + \left\{\frac{2\xi^2(1-\varepsilon)^{1/3}}{[1-(1-\varepsilon)^{1/3}]^2} - 2\right\}\tanh\xi \Big/ [\xi/1-(1-\varepsilon)^{1/3}] - \tanh\xi \qquad (24)$$

where

$$\xi = \left[\frac{1}{(1-\varepsilon)^{1/3}} - 1\right]\frac{\alpha}{2} Re^{1/2} Sc^{1/3}. \qquad (25)$$

Rowe [30] extended this model to liquid fluidized beds by replacing ξ in equation (24) by (ξ/ε^m).

It would be interesting to point it out that at large values of ξ, Nelson–Galloway's asymptotic expression for Sherwood number given by equation (24) reduces to

$$Sh \approx \frac{2\xi(1-\varepsilon)^{1/3}}{1-(1-\varepsilon)^{1/3}}, \quad \xi \gg 1. \qquad (26)$$

Using Rowe's modification and equation (25), on gets

$$\varepsilon Sh \approx \alpha Re^{1/2} Sc^{1/3}, \quad \xi \gg 1. \qquad (27)$$

The conditions which lead to large values of ξ are high Reynolds number, high Schmidt number, and/or void fraction near unity. It is this region where we are focusing our attention in this paper. Thus equation (22), which is identical to (27), turns out to be a

Table 1. Range of published correlations for fluidized bed mass transfer

Ref.	Void fraction	Sc	Re	Correlation	Equation
[2, 3]	0.25–0.97	0.6–10000	$1 < [D_pG/\mu(1-\varepsilon)] < 30$	$J_d = 5.7[D_pG/\mu(1-\varepsilon)]^{-0.78}$	(1)
			$30 < [D_pG/\mu(1-\varepsilon)] < 10^4$	$J_d = 1.77[D_pG/\mu(1-\varepsilon)]^{-0.44}$	(2)
[4]	0.25–0.97	0.6–1500	$1 < (G/a\varepsilon\mu) < 100$	$\varepsilon^{\phi b}J_d = 0.895[G/a\varepsilon\mu]^{-0.59}$	(3)
			$100 < (G/a\varepsilon\mu)$	$\varepsilon^{\phi b}J_d = 0.38[G/a\varepsilon\mu]^{-0.39}$	(4a)
				$\phi = 1.1,\ b = 1.75[G/a\varepsilon\mu]^{-0.07}$	(4b)
[5]	0.35–1.0	1–2000	$(D_pG/\mu) < 10$	$T_D = 1.91 \times 10^{0.04[D_pG/\mu]}$	(5)
			$50 < (D_pG/\mu) < 3000$	$T_D = 0.8[D_pG/\mu]^{0.65}$	(6)
[6]	0.25–0.97	0.6–10000	$1 < [D_pG/\mu(1-\varepsilon)] < 80$	$J_d = 5.0[D_pG/\mu(1-\varepsilon)]^{-0.70}$	(7)
			$80 < [D_pG/\mu(1-\varepsilon)] < 10^4$	$J_d = 1.45[D_pG/\mu(1-\varepsilon)]^{-0.43}$	(8)
[7]	0.287–0.921	0.6–13200	$[G\sqrt{(A_p)}/\mu] > 50$	$\varepsilon J_d/f = 0.30/\{[G\sqrt{(A_p)}/\mu]^{0.35} - 1.90\}$	(9)
				where $f = 0.865 - 1.34$	
[8]	0.346–0.921	0.6–13200	$(D_pG/\mu) > 1$	$J_d = 0.010 + 0.863/[(D_pG/\mu)^{0.58} - 0.483]$	(10)
[9]	0.416–0.9535	0.6–1326	$60 < [D_pG/\mu(1-\varepsilon)] < 8000$	$J_d = 1.127/\{[D_pG/\mu(1-\varepsilon)]^{0.41} - 1.52\}$	(11)
[10]	0.373–0.9592	0.6–1500	$D_pG/\mu > 20$	$\varepsilon J_d = 0.589[D_pG/\mu]^{-0.427}$	(12)
[11]	0.43–0.75	0.6–2000	$5 < (D_pG/\mu) < 500$	$\varepsilon J_d = (0.81 \pm 0.05)[D_pG/\mu]^{-0.5}$	(13)
			$50 < (D_pG/\mu) < 2000$	$\varepsilon J_d = (0.6 \pm 0.10)[D_pG/\mu]^{-0.43}$	(14)
[12]	0.3415–0.9535	0.6–10000	$100 < (D_eU_e/\nu) < 6000$	$(k_cD_e/D) = 0.20[D_eU_e/\nu]^{3/4}Sc^{1/3}$	(15)
[13]	0.2698–0.9653	572–70000	$[D_pG/\mu(1-\varepsilon)] < 20$	$J_d = 3.8155[D_pG/\mu(1-\varepsilon)]^{-0.7913}$	(16)
			$[D_pG/\mu(1-\varepsilon)] \geq 20$	$J_d = 1.6218[D_pG/\mu(1-\varepsilon)]^{-0.4447}$	(17)
[14]	0.2698–0.9653	0.6–70000	$(D_pG/\mu) < 10$	$\varepsilon J_d = 1.1068[D_pG/\mu]^{-0.72}$	(18)
			$(D_pG/\mu) > 10$	$\varepsilon J_d = 0.4548[D_pG/\mu]^{-0.4069}$	(19)
			$0.01 < (D_pG/\mu) < 15000$	$\varepsilon J_d = \frac{0.765}{(D_pG/\mu)^{0.82}} + \frac{0.365}{(D_pG/\mu)^{0.386}}$	(20)

Table 2. Ranges of major operating variables covered in experimental studies

Ref.	Particle shape	Particle dia. (cm)	Void fraction	Sc	Re
Gas-phase mass transfer					
[2]	Various	0.0711–1.375	0.373–0.9592	2.57	14.7–4130
[15]	Cylinders, spheres	0.635–1.27	0.499–0.84	2.57	581–3466
[16]	Spheres	0.0922–0.2967	0.573–0.811	0.6–2.24	71.4–1225
[17]	Spheres	0.1831–0.9398	0.53–0.70	0.605–5.45	108–492
[10]	Spheres	0.2605–0.309	0.508–0.640	2.402	170–263
[18]	Spheres	0.1831–0.3086	0.526–0.688	3.72	123–359
Liquid-phase mass transfer					
[19]	Spherical pellets	0.319–0.638	0.511–0.9539	1204–1326	32.9–666
[20]	Granular	0.0558–0.211	0.47–0.91	991–1113	1.53–71.6
[21]*	Granular	0.0795–0.21	0.65–0.90	1020–1540	5–130
[22]	Spheres	0.0494–0.4913	0.441–0.989	1047–1173	0.711–1345
[23]	Crystalline	0.00236–0.00824	0.69–0.84	6250	0.8–3.92
[24]	Spheres	0.635–1.27	0.41–0.64	1360–1430	572–1342
[25]	Spheres	—	0.608–0.677	1125	6.9–10.5
[26]	Spheres	0.495–0.607	0.496–0.703	1535–1810	113–288
[13]	Cylindrical pellets	0.596–1.21	0.473–0.8973	572–1350	149–1186.5
[27, 28]†	Cylindrical pellets	0.5838–1.1431	0.4531–0.9411	767–44 745	0.1113–14.78

*Semifluidized beds.
†Includes data obtained with 1.0% aq. carboxyl methyl cellulose solution.

simplified form of Nelson–Galloway–Rowe asymptotic expression for Sherwood number with $\alpha = 0.95$.

Nelson and Galloway used $\alpha = 0.6$, a value established from the single sphere ($\varepsilon = 1$) mass transfer results of Frössling [31, 32] and Ranz [33], and obtained good agreement between theory and experiment. A rigorous and critical testing of Rowe's modification is not possible due to the lack of appropriate experimental data. Low Reynolds number gas-phase data are anomalous due to inherent errors in the measurement of driving force and are irrelevant for testing of the model. The liquid-fluidized bed data, on the other hand, are obtained with large particles and are thus at higher Reynolds number. The zero concentration gradient effect becomes important only in the lower Reynolds number region where adequate data is difficult to obtain. Rowe, however, tested his modification by choosing $m = 1$, and $\alpha = 0.7$, a value established earlier [34] from mass transfer results for an isolated benzoic acid sphere dissolving in water ($Sc = 1400$) and observed a fair agreement between theory and experiment in spite of the inadequacy of the available experimental data.

A comparison of the experimental and predicted Sh values for liquid fluidized beds using $\alpha = 0.7$ and $\xi = (\xi/\varepsilon)$, as shown in Fig. 2, indicates that equation (24) under predicts the Sh values for most of the workers. The deviation becomes quite large for data obtained at higher Reynolds numbers. A comparatively better agreement between the experimental and theoretical Sh values is obtained when α is chosen equal to 0.95, a value equal to the proportionality constant obtained in equation (22) and closer to that (0.9095) established for mass transfer from isolated ($\varepsilon = 1$) spherical and non-spherical particles for systems of widely varying Sc numbers (960–117 000) [27]. Experimental and predicted Sh values for this case are compared in Fig. 3. Most of the predicted Sh values are within $\pm 20\%$ of the experimental Sh. This, therefore, indicates that Rowe's modification of Nelson–Galloway's theory holds good even for large particle systems provided an appropriate value of α is chosen. It is also possible to put forth a theoretical justification in favor of such a larger value of α. From the Galloway–Sage turbulent boundary layer model [35] for transport in multi-particle systems one can easily see that α is identical to Frössling number, Fs, which depends strongly on particle size. For fluidized beds of large particles with Re up to say 500 to 1000 and void fractions around 0.8 to 0.9, Galloway–Sage model predicts turbulence levels around 20 to 30% and thus Fs (or α) around 0.8 to 0.9. It is evident, therefore, that $\alpha = 0.95$, as obtained from the regression analysis of the experimental data, does have a theoretical basis and is not merely an empirical constant.

The larger deviation between the experiment and theory in some cases as observed in both Figs. 2 and 3, is not only due to the inadequacy of the experimental data obtained at higher Reynolds numbers, but to some extent is also due to inherent inaccuracies in the reported bed void fraction. Bed void fraction is an

Table 3. Deviation of experimental data from various correlations

Ref.	Equation	Std Dev., %	No. Data points
[2, 3]	(1), (2)	25.3	642
[4]	(3), (4)	67.7	637
[5]	(5), (6)	28.2	572
[6]	(7), (8)	22.1	642
[7]	(9)	17.2	670
[8]	(10)	17.7	497
[9]	(11)	15.9	505
[10]	(12)	20.1	515
[11]	(13), (14)	20.4	592
[12]	(15)	18.6	452
[13]	(16), (17)	17.2	678
[14]	(18), (19)	18.4	678
	(20)	15.4	678
This study	(22)	20.2	678

important parameter in fluidized bed mass and heat transfer and for a critical testing of a theoretical model like one mentioned above, its value should be as accurately determined as possible. Unfortunately this is not the case with many of the reported experimental studies.

The identical deviations between the experimental Sherwood numbers and those predicted from Nelson–Galloway–Rowe expression and equation (22) as shown in Figs. 3 and 4, respectively, reveal that the extra complexity of the Nelson–Galloway–Rowe expression does not seem to help in improving the correlation of the data. Equation (22) should, therefore, be preferred for predicting the mass transfer data because of its simplicity.

CONCLUSIONS

(1) Most of the published empirical correlations predict the experimental fluidized bed mass transfer data with a comparable degree of accuracy. The least deviation is, however, obtained with equation (20), a purely empirical relation developed on the basis of both fixed and fluidized bed data.

(2) Rowe's modification of Nelson–Galloway's theory gives better prediction of Sh values even for large particle systems provided $\alpha = 0.7$ as proposed by Rowe, is replaced by a larger value 0.95.

(3) An analysis of the gas and liquid-phase fluidized bed mass transfer data, as per requirements of the theory, gives

$$\varepsilon Sh = 0.95\, Re^{1/2}\, Sh^{1/3} \qquad (22)$$

which predicts the experimental Sh values with a standard deviation of 20.2%. This deviation is identical to that obtained for Nelson–Galloway–Rowe expression. Equation (22), therefore, should be preferred for predictions.

Acknowledgements — The authors are indebted to the University of Illinois at Chicago Circle for providing the computational facility.

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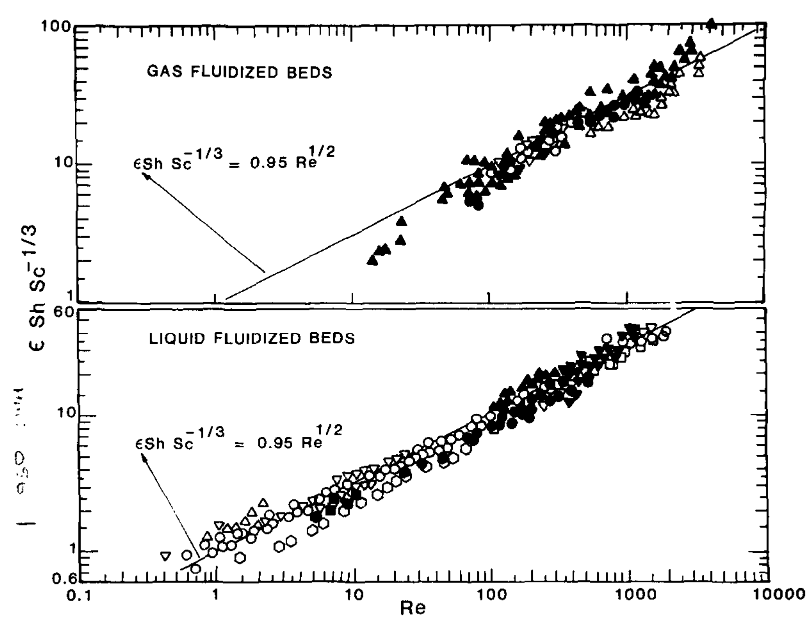

FIG. 1. Mass transfer in fluidized beds: $\varepsilon Sh Sc^{-1/3}$ vs Re. Gas-phase: ▲ [2]; △ [15]; ● [16]; ○ [17]; ▼ [10]; ▽ [18]. Liquid-phase: ● [19]; ⬡ [20]; ⬢ [21]; ○ [22]; △ [23]; □ [24]; ■ [25]; ▲ [26]; ▼ [13]; ▽ [27, 28].

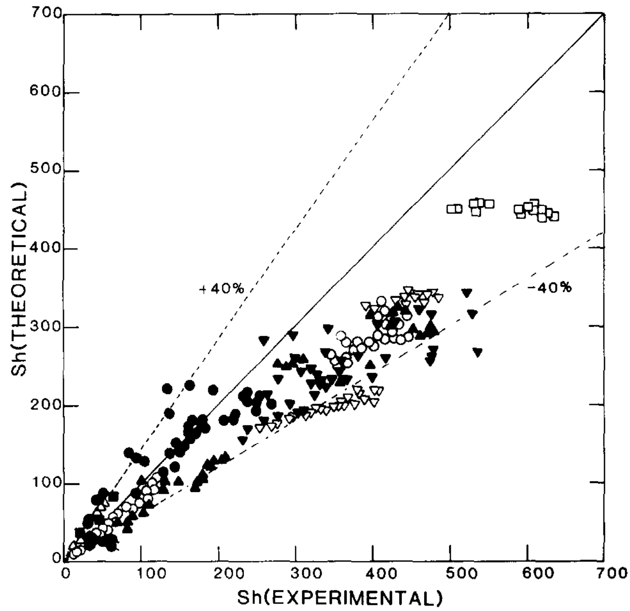

FIG. 2. Comparison of experimental and theoretical *Sh* for liquid fluidized beds ($\alpha = 0.70, m = 1$): ● [19], △ [20], ■ [21], ○ [22], ◥ [23], □ [24], ⬢ [25], ▽ [26], ▼ [13], ▲ [27, 28].

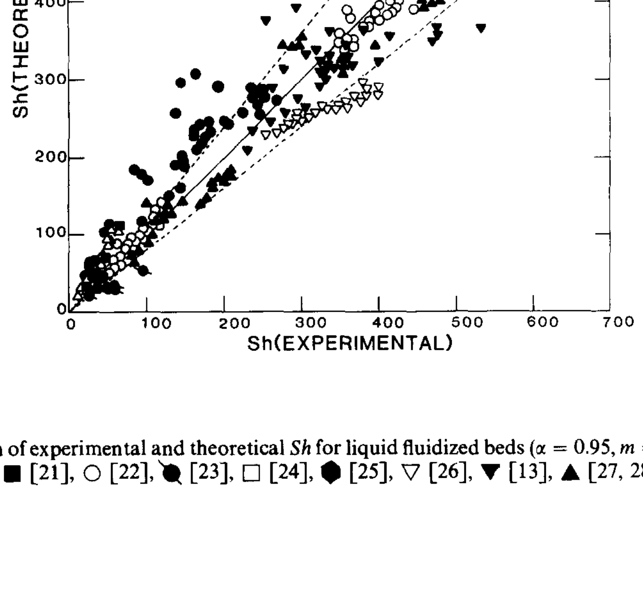

FIG. 3. Comparison of experimental and theoretical *Sh* for liquid fluidized beds ($\alpha = 0.95, m = 1$): ● [19], △ [20], ■ [21], ○ [22], ◥ [23], □ [24], ⬢ [25], ▽ [26], ▼ [13], ▲ [27, 28].

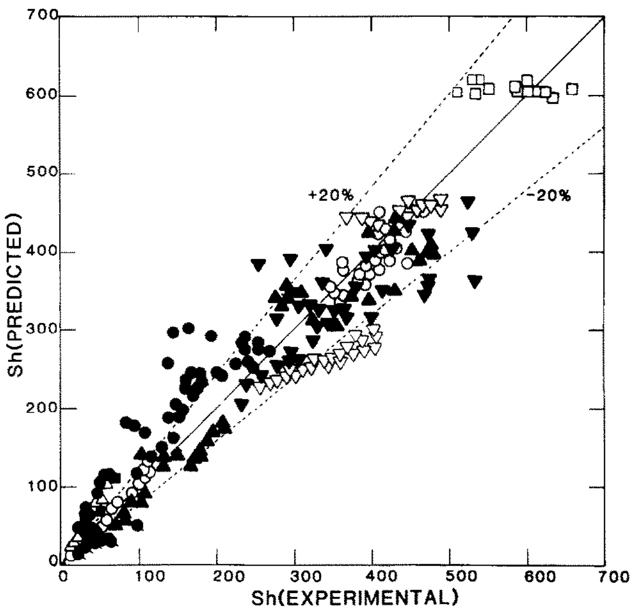

FIG. 4. Comparison of experimental *Sh* with *Sh* predicted from equation (22) for liquid fluidized beds: ● [19], △ [20], ■ [21], ○ [22], ◖ [23], □ [24], ◆ [25], ▽ [26], ▼ [13], ▲ [27, 28].

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TRANSFERT MASSIQUE ENTRE DES PARTICULES ET LE FLUIDE DANS UN LIT FLUIDISE DE GROSSES PARTICULES

Résumé—Des formules générales empiriques pour le transfert massique entre particule et fluide dans des lits fluidisés sont éprouvées pour juger de leur efficacité à prévoir le transfert massique dans des lits fluidisés de grosses particules. La modification par Rowe de la théorie de Nelson et Galloway pour le transfert massique entre particule et fluide dans des systèmes denses de particules fines est réexaminée et on trouve qu'elle s'accorde très bien avec le cas des grosses particules dans des lits fluidisés par liquide. Une formule simplifiée qui est une expression asymptotique de l'approximation de Nelson–Galloway–Rowe est proposée pour évaluer les flux de transfert massique.

STOFFÜBERGANG ZWISCHEN PARTIKELN UND FLUID IN FLIESSBETTEN MIT GROSSEN PARTIKELN

Zusammenfassung—Allgemeine empirische Beziehungen, die in der Literatur für den Stoffübergang zwischen Partikeln und Fluid in Fließbetten zur Verfügung stehen, werden auf ihre Tauglichkeit zur Wiedergabe experimentell gemessener Stoffübergangswerte in Fließbetten mit großen Partikeln untersucht. Ebenso wird die Modifikation von Rowe der Theorie von Nelson und Galloway über den Stoffübergang zwischen Partikeln und Fluid in dichten Systemen feiner Partikel überprüft, wobei sich eine recht gute Übereinstimmung mit den Stoffübergangsdaten in flüssigen Fließbetten mit großen Partikeln ergibt. Zur Berechnung des Stoffstroms wird eine einfache Korrelation vorgeschlagen, die eine Näherung des asymptotischen Ausdrucks nach Nelson–Galloway–Rowe darstellt.

МАССОПЕРЕНОС МЕЖДУ ЧАСТИЦАМИ И ЖИДКОСТЬЮ В ПСЕВДООЖИЖЕННЫХ СЛОЯХ ЧАСТИЦ БОЛЬШОГО РАЗМЕРА

Аннотация — Путем сравнения опытных и расчетных данных проведена проверка возможности использования известных из литературы обобщенных эмпирических соотношений, описывающих перенос массы от частиц к жидкости в псевдоожиженных слоях, для определения массопереноса в псевдоожиженных слоях частиц большого размера. Также дан анализ предложенного Роу модифицированного соотношения Нельсона и Галловея для переноса массы от частиц к жидкости в плотных слоях мелких частиц и найдено довольно хорошее соответствие с данными по переносу массы в псевдоожиженных жидкостью слоях крупных частиц. Предложено упрощенное соотношение для расчета интенсивности переноса массы, которое является аппроксимацией асимптотического выражения Нельсона-Галловея-Роу.