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TRANSITIONAL MASS-TRANSFER BEHAVIOR FROM PACKED-BED CONDITIONS TO ONSET OF FLUIDIZATION

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Abstract—A critical review of mass-transfer data for both packed and fluidized beds in which axial and radial dispersion contributions have been eliminated shows the existence of specific patterns unique to each type of bed. For packed beds, the j_d vs Re behavior of gas–solid and liquid–solid systems exhibits different dependences which gradually approach each other and become essentially the same above $Re = 50$.

For the fluidized state of gas–solid systems, the correlation of $\log j_d$ vs $\log Re$ produces linear relationships of slope -1 that are unique to the particular size and density of particles being fluidized. With decreasing Re, these relationships terminate at Re_{mf} to yield j_d values that extend beyond the corresponding gas–solid relationship for packed beds. This unexpected behavior is most pronounced in the low Reynolds number region ($Re < 10$) and decreases in intensity with increasing Re values. Explanations are advanced to account for this disparity which disappears above $Re = 50$.

NOMENCLATURE

D_p,	average particle size [cm];
D_v,	molecular diffusivity [$\text{cm}^2\,\text{s}^{-1}$];
E,	axial dispersion coefficient [$\text{cm}^2\,\text{s}^{-1}$];
F,	axial mixing factor for packed beds, equation (3);
G,	superficial mass velocity of flowing fluid [$\text{g}\,\text{s}^{-1}\,\text{cm}^{-2}$];
k_g,	mass-transfer coefficient [$\text{g-mol}\,\text{s}^{-1}\,\text{cm}^{-2}\,\text{atm}^{-1}$];
L,	height of packed bed [cm];
j_d,	mass transfer factor, $(k_g p_{gf} M/G)(\mu/\rho D_v)_f^{2/3}$;
$(j_d)_{mf}$,	mass-transfer factor at minimum fluidization conditions;
M,	average molecular weight of flowing fluid;
n,	number of perfect mixing cells in series, $(L/D_p)/(Pe/2)$;
Pe,	Peclet number, $D_p u/\varepsilon E$;
p_{gf},	partial pressure of nontransferable component [atm];
R,	ratio of inlet to outlet driving potentials;
Re,	Reynolds number, $D_p G/\mu$;
Re_{mf},	Reynolds number at minimum fluidization conditions;
u,	superficial velocity of fluid through packed bed [$\text{cm}\,\text{s}^{-1}$].

Greek symbols

ε,	void fraction of bed;
μ,	absolute viscosity [$\text{g}\,\text{s}^{-1}\,\text{cm}^{-1}$];
ρ,	density [$\text{g}\,\text{cm}^{-3}$].

THE CONTINUING introduction of mass-transfer data in the literature and the disparity existing between packed-bed and fluidized-bed behavior prompted the initiation of this study in order to explore the possible existence of a continuity between these two modes of operation. To eliminate extraneous information inherently present from axial and radial dispersion contributions, the selection of data for the major involvement of this study was restricted to experimental measurements obtained from actual concentration profiles. Consequently, the mass-transfer measurements of Hsiung and Thodos [1, 2] available for both packed beds and fluidized beds were utilized and critically evaluated in order to establish any interlocking pattern of behavior possible between these two modes of mass-transfer operation.

Over the last thirty years extensive experimental information has been presented in the literature for the mass-transfer behavior of both gas–solid and liquid–solid systems. These studies include investigations on both packed beds and fluidized beds for which the log-mean driving force has been assumed to apply to both types of mass transfer. Initial attempts to generalize these modes of mass transfer have shown that the dependence of the product εj_d on Re produces a satisfactory correlation for both packed and fluidized beds [3]. However, because of the prevailing scatter of data, and particularly in the low Reynolds region, this method of correlation does not provide a satisfactory conclusion and therefore it became necessary to re-examine them in view of the presence of other extraneous factors that could influence the actual behavior of both mass-transfer processes.

The unavoidable presence of axial dispersion and radial dispersion in mass-transfer operations through packed beds introduces a distortion to the log-mean driving force that has hitherto been applied for the analysis of such data. In general, this distortion is difficult to predict and particularly for shallow beds when this effect becomes most pronounced. From the comprehensive analysis of this subject, McHenry and Wilhelm [4] conclude that the contribution due to axial dispersion is approximately six times larger than the corresponding radial contribution. To account for axial dispersion in packed beds, Epstein [5] applies the tanks-in-series model and presents a correction factor to be applied to the log-mean driving force.

TREATMENT OF DATA OBTAINED FROM ACTUAL CONCENTRATION PROFILES

To avoid the complex involvement associated with both axial and radial dispersion phenomena, Hsiung and Thodos [1, 2] present measurements associated with the sublimation of naphthalene into air and account for the actual concentration profiles existing within both packed and fluidized bed systems. Their approach utilized the flow of air over beds of naphthalene spheres dispersed within a matrix of divinylbenzene copolymer spheres. These copolymer beads acted as inert diluents to the active naphthalene spheres and thus avoided a state of saturation of naphthalene vapors in the stream of air leaving the bed. These beads were of the same density as naphthalene ($1.04\,\mathrm{g\,cm^{-3}}$) and were used only after they were found to be inert to the adsorption and desorption of naphthalene.

The equipment used in the studies of Hsiung and Thodos [1, 2] included the involvement of three reactors. The main reactor was constructed of Lucite tubing, i.d. = 6.59 cm (2.59 in), 0.318 cm (1/8 in) thick, and 38.2 cm (15 in) in length and was provided with an entrance calming section made up of a bed of brass spheres, 0.95 cm (3/8 in) in diameter. Two additional reactors, i.d. = 4.68 and 9.37 cm were also used by them in order to investigate the possible effect of reactor diameter on mass transfer. The top closure of the reactor accommodated two travelling thermocouples and a sampling probe. These thermocouples and the sampling probe could be moved up and down for the measurement of temperatures and for the withdrawal of air samples from different positions within the bed. One of the thermocouples measured in the course of a run, the temperature of the air leaving the bed while the other accounted for the temperature of the solid particles by surrounding this thermocouple junction with a bead of naphthalene which was melted directly on it.

In the course of a run, a gas sample was continuously withdrawn through the sampling probe which was made to traverse the entire height of either the fixed bed or the fluidized bed. This gas sample was then made to pass through a solenoid sampling valve which when actuated, diverted the gas sample into a hydrocarbon analyzer of a flame ionization type which used nitrogen as a carrier gas. The amplified signal from the analyzer was transmitted to a recorder and the resulting response curve was integrated to obtain the naphthalene content of each air sample. The results of this approach permitted the establishment of the concentration profile of naphthalene in air for each run. Altogether seven sizes of naphthalene spheres with average diameters ranging from 0.02480 to 0.2000 cm were investigated.

The concentration profiles obtained from these studies [1, 2] deviated considerably from their corresponding log-mean profiles. These actual profiles were found to depend not only on particle size, but on other system parameters [1, 2]. Mass transfer coefficients, k_g, obtained from the driving forces resulting from actual concentration profiles were used to establish j_d, the corresponding mass-transfer factors. The resulting values of j_d were related to their corresponding Reynolds numbers for both packed bed and fluidized bed systems. Figure 1 presents the dependence of j_d on Reynolds number for packed beds of the gas–solid system, naphthalene–air, expressed on a log–log coordinate system. The data for all the particle sizes investigated by Hsiung and Thodos [1] can be represented by a linear relationship on log–log coordinates and can be expressed in equation form as follows:

$$j_d = \frac{1.33}{Re^{0.40}}, \quad 0.1 < Re < 100. \tag{1}$$

For fluidized beds the relationship of j_d on Reynolds number exhibits a definite particle size dependence as shown in Fig. 2. The relationship for each particle size can be represented linearly with a slope of -1 on log–log coordinates and implies that the product $j_d Re$ is a constant that is particle size dependent for this type of fluidization. For comparison, the packed bed relationship of Fig. 1 expressed through equation (1) is also included in Fig. 2. A careful examination of the fluidized bed data included in this figure indicates that a significant number of them extend above the relationship representing the packed bed state condition. This behavior, which is more pronounced in the low Reynolds region, is somewhat unexpected and is best exhibited by particle sizes of $D_p = 0.02480$, 0.03446 and 0.04895 cm. For particle sizes of $D_p \geqslant 0.07260$ cm, the transition from packed-bed to fluidized-bed behavior appears to follow a normal trend with the fluidized state points all existing below the packed-bed relationship of equation (1) and exhibiting linear relationships of $\log j_d$ versus Re of slope -1.

TRANSITIONAL BEHAVIOR IN LOW REYNOLDS REGION ($Re < 50$)

A closer examination of the points representing the three sizes $D_p = 0.02480$, 0.03446 and 0.04895 cm shows that their actual behavior as they approach

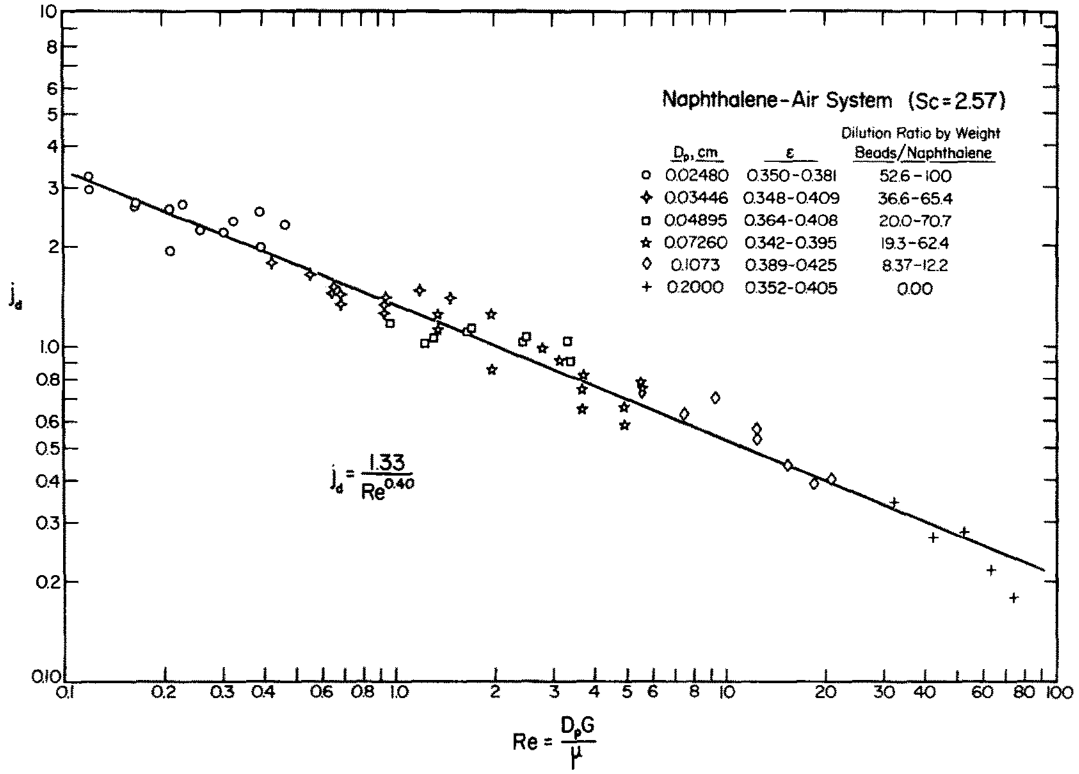

FIG. 1. Relationship between j_d and Re for packed beds established from driving forces involving actual concentration profiles.

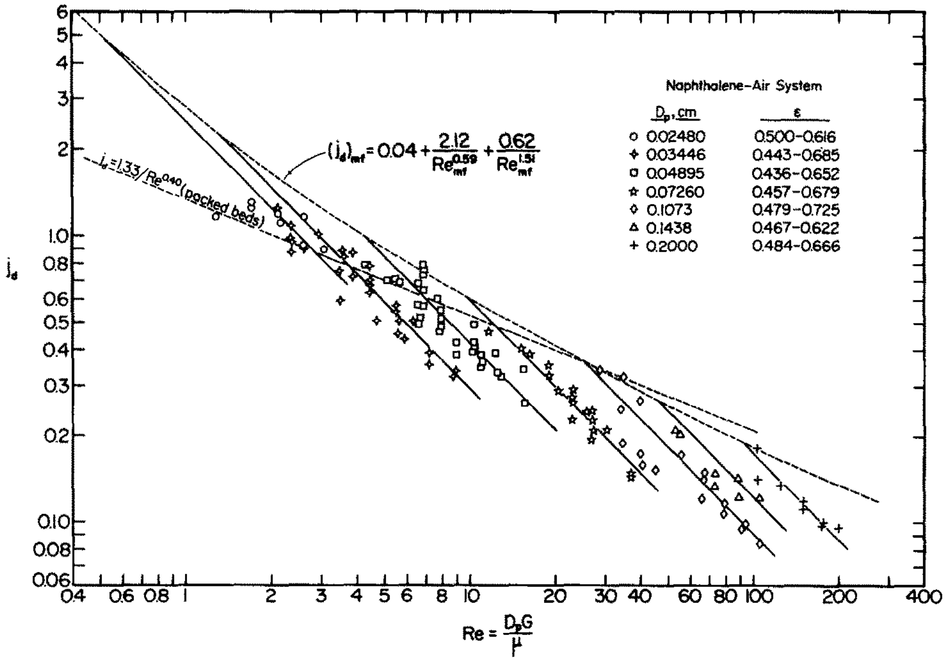

FIG. 2. Relationships between j_d and Re established from the measurement of actual concentration profiles for fluidized beds.

packed bed state conditions is not linear, but instead they exhibit a scatter with a trend that follows a curve to the left to approach the packed bed relationship. This pattern of behavior indicates the existence of a transitional state in this low Reynolds region that has not heretofore been identified. This phenomenon, although interesting, does not help resolve the generalized behavior of fluidized beds in this transitional region, unless considerations are included to define this region. To resolve the difficulties associated with this region, the following additional approach was adopted. For each particle size, the relationship of log j_d vs log Re for fluidized beds was extended linearly to its corresponding minimum fluidization value, Re_{mf}. These minimum fluidization conditions were also experimentally verified from relationships expressing the pressure drop across the bed vs mass velocity. For the general case, the minimum fluidization velocity can be estimated from a modified Ergun relationship [6]. Both measured and calculated values for minimum fluidization velocity and corresponding Reynolds numbers calculated from them are presented along with the measured void fractions as follows:

		u_{mf}, cm s^{-1}		Re_{mf}	
D_p, cm	ε_{mf}	exptl	calc'd	exptl	calc'd
0.02480	0.408	2.87	3.02	0.443	0.446
0.03446	0.411	5.77	5.09	1.27	1.12
0.04895	0.436	13.4	12.1	4.12	3.72
0.07260	0.429	22.6	22.3	10.3	10.2
0.1073	0.422	37.4	36.7	25.2	24.7
0.1438	0.425	50.9	51.9	45.5	46.4
0.2000	0.425	70.2	70.0	90.0	89.7

These values of Re_{mf} can be considered to be the hypothetical starting points for fluidized behavior. The locus of these hypothetical minimum fluidization points for the seven particle sizes generates a relationship that represents a functional dependence describing the onset of fluidization. This limiting relationship can be represented in equation form as follows:

$$(j_d)_{mf} = 0.040 + \frac{2.12}{Re_{mf}0.59} + \frac{0.62}{Re_{mf}1.51}$$

$$0.4 < Re_{mf} < 1000 \qquad (2)$$

and can be utilized to establish the uppermost value of j_d possible for Re_{mf} representing the conditions existing at minimum fluidization conditions. For conditions of $Re > Re_{mf}$, the linear behavior log j_d versus log Re applies with a slope of -1. Equation (2) has been developed from the experimental measurements of Hsiung and Thodos [2] from fluidized bed data and represents a limiting state that in reality may not be approached in the lower Reynolds region when $Re \rightarrow Re_{mf}$. In this region, it is conceivable that for a particular particle size, the relationship of log j_d versus log Re tends to the left in this transitional region, and approaches in the limit the packed bed relationship at Re_{mf}, without actually reaching the value of j_d predicted by equation (2). This behavior suggests that a packed bed follows normal behavior as depicted by equation (1) and upon reaching conditions equivalent to minimum fluidization conditions, a situation arises wherein the packed-bed structure begins to become unabridged with the production of a high degree of instability that manifests itself by attempting to approach in the low Reynolds region the relationship defined by equation (2). In this region, because of the changing pattern of instability, it is not likely that this limiting case is reached, but instead, the actual response follows a curved behavior to join up the linear relationship of slope -1 associated with the particular particle size. The final resolution of this transitional involvement requires further experimental work to supplement the results presented by Hsiung and Thodos [1, 2], with particular emphasis placed on the fluidized-bed behavior in the low Reynolds region and particularly that associated with j_d factors exceeding those possible for packed bed systems.

ADDITIONAL MASS TRANSFER DATA FOR PACKED BEDS

It is of interest to consider at this stage the experimental results of Wilson and Geankoplis [7] in the low Reynolds region for the benzoic acid–water system and those of Gamson, Thodos and Hougen [8] in the high Reynolds region associated with the evaporation of water into air. To render the data of Gamson *et al.* [8] internally consistent with those of Hsiung and Thodos [1], their results were corrected for axial dispersion effects. This approach required the introduction of the correction factor proposed by Epstein [5] who postulated the tanks-in-series model and developed the relationship,

$$F = \frac{\ln R}{n[R^{1/n} - 1]}, \qquad (3)$$

where R = the ratio of inlet to outlet driving potentials and n = the number of perfect mixing cells in series. McHenry and Wilhelm (4) show that the number of perfect mixing cells in a fixed bed can be obtained from the expression,

$$n = \frac{L}{D_p}\frac{Pe}{2}, \qquad (4)$$

where L = the height of packed bed, D_p = average particle diameter and Pe = Peclet number, $D_p u/\varepsilon E$. The correction factor F, when applied to the log-mean driving force values utilized in 1943 by Gamson *et al.* [8] transformed their packed-bed data and made them comparable to the basis utilized by Hsiung and Thodos [1]. This correction became insignificant for the solid–liquid system (benzoic acid–water) of Wilson and Geankoplis [7]. Figure 3 includes the corrected values of Gamson *et al.* [8] and those of

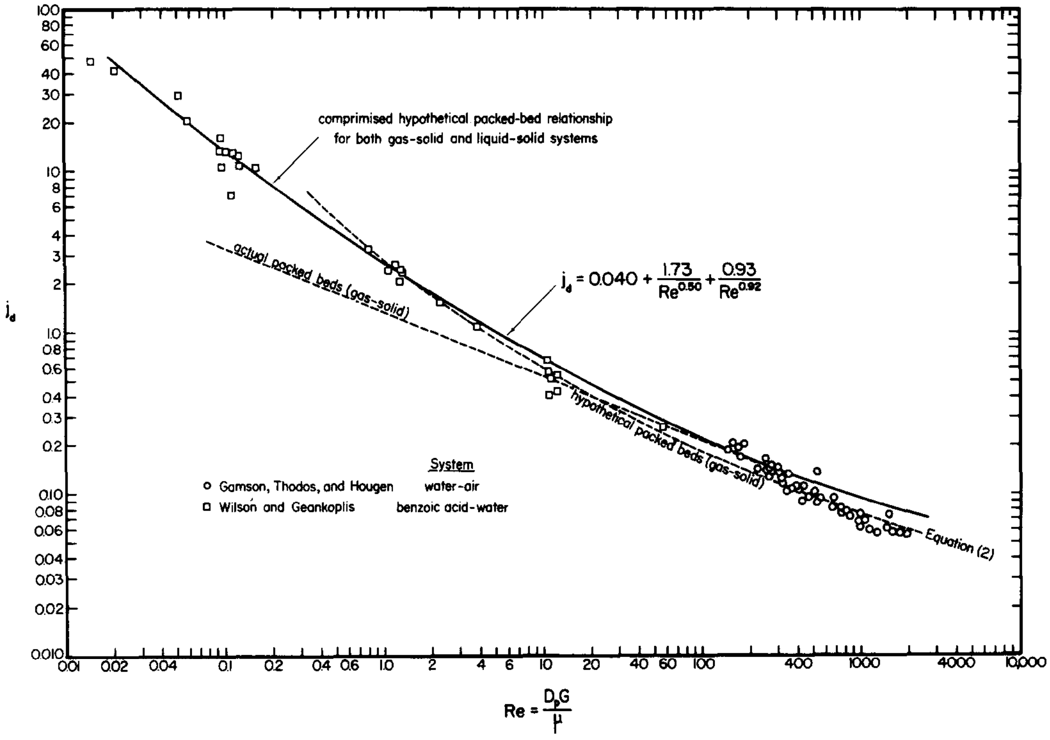

FIG. 3. Comparison of j_d vs Re relationships for packed beds.

Wilson and Geankoplis [7] as well as the relationships resulting from equations (1) and (2). It is of interest to note that the data of Gamson *et al.*, corrected for axial dispersion, fall in the vicinity of the relationship representing the gas–solid data of Hsiung and Thodos [1] for packed beds of equation (2) representing the onset of fluidization while the liquid–solid data of Wilson and Geankoplis [7] represent an extension of this hypothetical fluidized-bed relationship. An overall compromise suggests that this relationship can be represented as,

$$j_d = 0.040 + \frac{1.73}{Re^{0.50}} + \frac{0.93}{Re^{0.92}}. \qquad (5)$$

Equation (5) represents the combined contributions of Wilson and Geankoplis [7] in the low Reynolds region, Hsiung and Thodos [1, 2] in the intermediate region, and those of Gamson *et al.* [8] in the high Reynolds region for packed beds. The relationship of Equation (5) is uniquely different in the low Reynolds region from that represented by equation (1) for packed beds of gas–solid systems and as such the direct involvement of liquid–solid systems must be associated with equation (5) while that of gas–solid systems must be restricted to the use of equation (1) at low Reynolds numbers. A disparity appears to exist between gas–solid and liquid–solid systems in this low Reynolds region. For Reynolds numbers, $Re \geqslant 50$, equations (1) and (5) yield essentially identical results; however, the use of these relationships for the $Re < 50$ must be discriminated depending on the nature of the packed bed system.

CONCLUSIONS

The results of this study indicate that the relationship for packed-beds,

$$j_d = \frac{1.33}{Re^{0.40}}, \quad 0.1 < Re < 100, \qquad (1)$$

represents the actual mass transfer behavior between j_d and Reynolds number for gas–solid systems in which axial and radial dispersion contributions are eliminated. The hypothetical state representing the onset of fluidization can be established by the relationship,

$$j_d = 0.040 + \frac{1.73}{Re^{0.50}} + \frac{0.93}{Re^{0.92}} \qquad (5)$$

where $Re = Re_{mf}$, the Reynolds number existing at minimum fluidization conditions. This relationship also serves to define in an unrestricted sense the j_d vs Re dependence of liquid–solid systems for $Re < 50$ and for both liquid–solid and gas–solid systems for $Re \geqslant 50$. The relationships represented by equations (1) and (5) become essentially the same when $Re \geqslant 50$. For $Re < 50$, these relationships diverge and account for the unexpected noncomformity existing in this region between gas–solid and liquid–solid systems. However, equation (5), over the complete range, should prove acceptable for representing the limiting case defining the onset of fluidization of gas–solid systems initiated when Re approaches Re_{mf}.

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COMPORTEMENT TRANSITOIRE DU TRANSFERT MASSIQUE LORS DE L'APPARITION DE LA FLUIDISATION DE LITS FIXES

Résumé—Une revue critique des données de transfert massique, à la fois pour les lits fixes et les lits fluidisés avec des contributions de dispersion axiale et radiale, éliminées, montre l'existence de configurations spécifiques à chaque type de lit. Pour les lits fixes, j_d variant en fonction de Re pour les systèmes gaz–solide et liquide–solide, il existe différentes lois de dépendance qui s'approchent graduellement les unes des autres et qui deviennent identiques au dessus de $Re = 50$.

Pour le système fluidisé gas–solide, la relation entre log j_d et log Re est linéaire avec une pente—1, unique pour chaque taille et chaque densité de particules en fluidisation. Lorsque Re décroît, ces relations finissent par donner à Re_{mf} des valeurs de j_d qui prolongent la relation des lits fixes gaz–solide. Ce comportement inespéré est plus prononcé dans la région des petits nombres de Reynolds ($Re < 10$) et il décroit en intensité lorsque les valeurs de Re augmentent. On avance des explications pour cette disparité qui disparait au dessus de $Re = 50$.

DAS STOFFÜBERGANGSVERHALTEN IM ÜBERGANGSBEREICH ZWISCHEN FESTBETT- UND FLIESSBETT-BEDINGUNGEN

Zusammenfassung—Eine kritische Durchsicht von Stoffübergangsdaten für Fest- und Fließbetten, in denen der Einfluß axialer und radialer Dispersion eliminiert wurde, zeigt die Existenz von spezifischen, für jeden dieser beiden Bett-Typen eigentümlichen Verhaltensmustern. Bei Festbetten ergeben sich für den Verlauf von j_d über Re von Gas/Feststoff- und Flüssigkeit/Feststoff-Systemen unterschiedliche Abhängigkeiten, die sich einander nähern und oberhalb $Re = 50$ im wesentlichen gleich werden. Für den fluiden Zustand von Gas/Feststoff-Systemen ergibt die Auftragung von log j_d über log Re lineare Beziehungen mit der Steigung -1, die von Größe und Dichte der schwebend gehaltenen Partikel abhängen. Mit fallender Reynolds–Zahl enden diese Beziehungen bei Re_{mf}, wo sie Werte für j_d liefern, die höher liegen, als sich aus der entsprechenden Beziehung für gasdurchströmte Festbetten ergibt. Dieses unerwartete Verhalten ist besonders ausgeprägt im Gebiet niedriger Reynolds–Zahlen ($Re < 10$) und tritt mit zunehmenden Werten der Reynolds–Zahl weniger in Erscheinung. Erklärungen für diese Ungleichheit, die oberhalb $Re = 50$ verschwindet, werden gegeben.

ПЕРЕНОС МАССЫ В ПЕРЕХОДНОМ РЕЖИМЕ ОТ НЕПОДВИЖНОГО СЛОЯ ДО НАЧАЛА ПСЕВДООЖИЖЕНИЯ

Аннотация — Критический анализ данных по переносу массы как в режиме неподвижного, так и псевдоожиженного слоёв при отсутствии влияния аксиальной и радиальной дисперсий выявил наличие отличительных особенностей, характерных для каждого из режимов. В режиме неподвижного слоя величина *jd* по-разному зависит от числа *Re* для систем газ-твердое тело и жидкость-твердое тело. Это различие постепенно сглаживается и при *Re* выше 50 почти исчезает. В режиме псевдоожиженного слоя имеет место линейная зависимость величины log *jd* от log *Re* (с наклоном -1), которая является характерной для определенного размера частиц и плотности их упаковки. С уменьшением значения *Re* линейный закон нарушается при Re_{mf}, и получаемые при этом значения *jd* намного превышают значения из соответствующего соотношения для системы газ-твердые частицы в режиме неподвижного слоя. Это различие проявляется наиболее отчётливо в области низких значений числа Рейнольдса ($Re < 10$) и уменьшается с ростом *Re*. Высказан ряд предположений, объясняющих это различие, которое исчезает при значениях *Re* выше 50.