R & D NOTES

Prediction of High Peclet Number Mass Transfer in Granular Beds Using The Constricted Tube Model

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Granular beds are widely used in the chemical industry as contacting devices. Hence, mass transfer in granular beds has occupied an important place in chemical engineering literature. Karabellas et al. (1971) list in their review over 30 major works on this topic. Since then the number has at least doubled.

The classical view of mass transfer in a granular bed is to generalize the results obtained for single spheres to an assemblage of spheres comprising a bed. Using this approach, we showed (Tardos et al., 1976, Tardos, 1977) that mass transfer to an active sphere situated in a homogeneous granular bed of identical elements, can be computed from the expression

$$Sh_s = 0.997\, g(\epsilon)\, Pe_s^{1/3} \qquad (1)$$

where $g(\epsilon)$ is a porosity dependent function. This function has different expressions when different flow models are used for the fluid motion in the bed and, is given by

$$g(\epsilon) = \sqrt[3]{\frac{2}{3\sin^2\theta} \cdot \left| \frac{\partial^2\psi(1, \epsilon, \theta)}{\partial R^2} \right|} \qquad (2)$$

$\psi(R, \epsilon, \theta)$ being the characteristic stream function of the fluid flow. Expressions (1) and (2) are valid for high Peclet numbers ($Pe_s > 100$), low Reynolds numbers and nonslip condition on the active granule. Values for the function $g(\epsilon)$, as obtained from theoretical considerations using Equation (2), as well as from mass transfer experiments are given in Table 1. As can be seen, the agreement between computed and measured values of $g(\epsilon)$ is very good for the case of regularly packed spheres and less satisfactory for random beds.

It may be argued at this point that a randomly packed bed of granules can not be characterized completely by porosity only. Therefore, for an actual bed of granules, more information is required. One cannot help but note that computed values of $g(\epsilon)$, i.e., the mass transfer rate, in randomly packed beds are always higher than the ones from actual mass transfer experiments. In retrospect, one may explain the lower experimental values by the existence of dead zones at the points of contact between adjacent granules. These zones are not accounted for in the various "single sphere" theories resulting in inflated mass transfer rates.

Recently, a different approach in describing the granular bed has been proposed. Rather than viewing the particles comprising the bed, one considers the interstices between them. This automatically eliminates the dead zones from consideration and hence, holds considerable promise for improving the prediction of mass transfer rates in random beds.

Experimenters using this method describe the fluid passing through a portion of the bed as if it passed through tubes of varying cross section. Fedkiw and Newman (1977) assumed that the tubes are sinusoidal periodically constricted and solved the pertinent mass transfer problem. Unfortunately, at present there is no way that one could apply their results to an actual bed of, say, given porosity and grain diameter.

The constricted tube model of Payatakes and Neira (1977) goes a step further, describing actual beds by means of 4 measured characteristics. This model predicts accurately the Darcy constant and the pressure drop through the bed from the knowledge of bed porosity, ϵ, mean granule diameter, $\langle d_s \rangle$, and the initial drainage curve. This curve yields the minimum pore diameter, d_c, and the irreducible saturation value, S_{wi}. The granular bed is supposed to be composed of randomly oriented constricted tubes, the dimensions of which are given in Figure 1. The main features of the model are given in Table 2.

In this article, the high Peclet number mass transfer problem pertinent to the above model is solved, and the

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TABLE 1. VALUES OF CORRECTION FACTOR $g(\epsilon)$ FOR MASS TRANSFER IN A GRANULAR BED

	Author	Flow field used	$g(\epsilon)$	Range	Remarks
1.	Pfeffer (1964) Cookson (1970)	Happel (1958)	$\left[\frac{2[1-(1-\epsilon)^{5/3}]}{2-3(1-\epsilon)^{1/3}+3(1-\epsilon)^{5/3}-2(1-\epsilon)^2}\right]^{1/3}$	$Re_0 < 0.01$ $Pe \geq 1000$	Theoretical
2.	Tardos et al. (1976)	Kuwabara (1959)	$\left(\frac{\epsilon}{2-\epsilon-\frac{9}{5}(1-\epsilon)^{1/3}-\frac{1}{5}(1-\epsilon)^2}\right)^{1/3}$	$Re_0 < 0.01$ $Pe \geq 1000$	Theoretical
3.	Sirkar (1974, 1975)	Tam (1969)	$\left[\frac{2+1.5(1-\epsilon)+1.5[8(1-\epsilon)-3(1-\epsilon)^2]^{1/2}}{\epsilon[2-3(1-\epsilon)]}\right]^{1/3}$	$Re_0 \leq 1$ $\epsilon > 0.33$ $Pe \geq 1000$	Theoretical
4.	Tardos et al. (1976)	Neale & Nader (1974)	$\left\{\frac{6[-4\beta^6-14\beta^5-30\beta^4-30\beta^3+\beta^4\alpha^2-5\beta^3\alpha^3+10\beta^3\alpha^2+5\beta^2\alpha^3-\beta\alpha^5-\alpha^5]}{-4\beta^6-24\beta^5-180\beta^4-180\beta^3+9\beta^5\alpha+45\beta^4\alpha-10\beta^3\alpha^3+180\beta^3\alpha-30\beta^2\alpha^3+9\beta\alpha^5-\beta\alpha^6+9\alpha^5}\right\}^{1/3}$	$\beta = \frac{\alpha}{(1-\epsilon)^{1/3}}$ $Re_0 \leq 10$ $Pe \geq 1000$	Can be approximated by: $g(\epsilon) = 1.31/\epsilon$ $0.3 \leq \epsilon \leq 0.7$
5.	Tardos et al. (1976)	Kuwabara (1959)	$1.0 + 3.43(1-\epsilon)$	$Re_0 < 0.01$ $Pe \geq 1000$	Numerical solution
6.	Tan et al. (1975)	—	$1.1/\epsilon$	$Re_0 < 1$ $0.35 < \epsilon < 0.7$	Experimental
7.	Wilson and Geankoplis (1966)	—	$1.09/\epsilon$	$Re_0 < 10$ $0.35 < \epsilon < 0.7$	Experimental
8.	Thoenes & Kramers (1958)	—	$1.448/\epsilon$	$Re_0 < 10$ $\epsilon = 0.476$	Experimental (regular packing)
9.	Karabellas et al. (1971)	—	$1.19/\epsilon$	$Re_0 < 10$ $\epsilon = 0.26$	Experimental (regular packing)
10.	Sorensen & Stewart (1974, IV)	Sorensen & Stewart (1974, II)	$1.104/\epsilon$ $1.17/\epsilon$	$\epsilon = 0.476$ $\epsilon = 0.26$	Theoretical (regular packing)

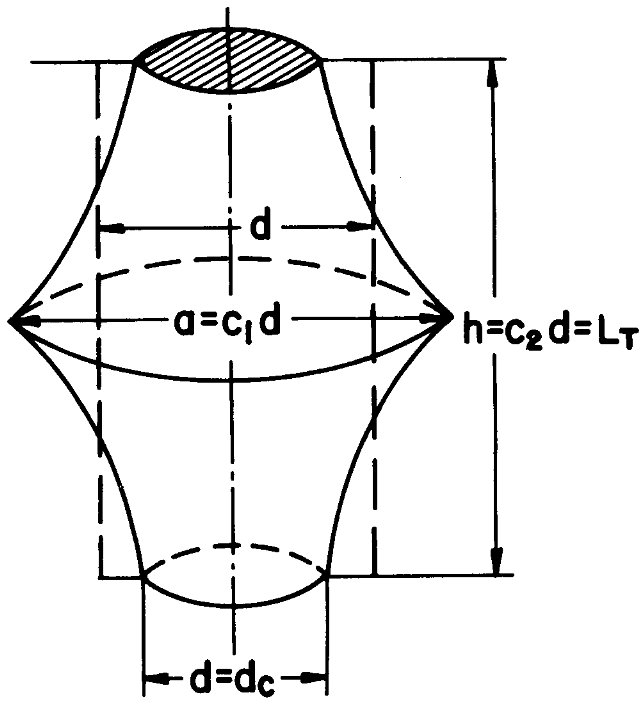

Figure 1. Dimensionless constricted tube (after Payatakes & Neira 1977).

results are compared to those obtained from experiments as well as from theories using the "single sphere" model. The restriction to high Peclet numbers ($Pe_T > 100$) applies to a wide variety of mass transfer processes (Levich 1962) as well as to the problem of diffusional filtration of submicron particles (Tardos et al. 1976).

Mass transfer from the fluid to the walls of the constricted tube (Figure 1) is not influenced by the curvature of the walls, if the transfer takes place at high Peclet numbers (Levich 1962). Therefore, a more simple problem is obtained when mass transfer in an equal volume cylinder is studied. The diameter of such a cylinder is readily obtained as

$$\bar{d} = \sqrt{\frac{4V_p}{\pi h}} = \left[\frac{8 + 4c_1 + 3c_1^2}{15} \cdot d_c^2\right]^{1/2} \quad (3)$$

Here, c_1 is a complicated function of the bed characteristics and is given in Table 2 (after Payatakes & Neira 1977).

Heat and/or mass transfer in a tube of limited length was initially solved by Graetz (1885), Leveque (1928) and more recently by Sorensen and Stewart (1947,I). The overall Sherwood number for the active tube portion may be expressed as

$$Sh_T = 2.035\sqrt[3]{\frac{d_T}{2L_T}}\, Pe_T^{1/3} - 1.23 \quad (4)$$

Equation (4) is correct for finite L_T/d_T, $Pe_T > 50$ and $Re_0 < 1$. Applying (4) to the equal volume tube one obtains

$$\frac{d_T}{2L_T} = \frac{\bar{d}}{2h} = \frac{1}{2}\left[\frac{8 + 4c_1 + 3c_1^2}{15c_2^2}\right]^{1/2} \quad (5)$$

and substitution back into Equation (4) yields

$$Sh_T = 1.62\left(\frac{8 + 4c_1 + 3c_1^2}{15c_2^2}\right)^{1/6} Pe_T^{1/3} - 1.23 \quad (6)$$

In Equation (6), Pe_T and Sh_T are computed relative to the tube diameter and to the tube surface involved in mass transfer. Simple geometric considerations and a mass transfer balance over an active volume in the bed show (Appendix):

TABLE 2. CHARACTERISTICS OF THE CONSTRICTED TUBE FLOW MODEL (AFTER PAYATAKES & NEIRA 1977)

Experimental Quantities	Notation	Expression
Porosity	ϵ	
Sieve analysis (mean granule diameter)	$<d_S>$	
Irreducible saturation value	S_{wi}	
Constriction diameter	$<d_c>$	
Calculated quantities		
Constant	c_1	$\left[\frac{\epsilon(1 - S_{wi})}{1 - \epsilon}\frac{<d_S^3>}{<d_c^3>}\right]^{1/3}$
Constant	c_2	$\frac{<d_S>}{<d_c>}$
Number of pores per unit volume of bed	N_p	$\frac{60\epsilon}{\pi c_2(8 + 4c_1 + 3c_1^2)}\frac{1}{<d_c^3>}$
Volume of pore	V_p	$\frac{\pi c_2}{60}(8 + 4c_1 + 3c_1^2)<d_c^3>$
Darcy constant	k	$\frac{5\epsilon(1 + \cos\alpha + \cos^2\alpha)\, c_2^2 <d_c^5>^*}{(8 + 4c_1 + 3c_1^2)<d_c^3>(-\Delta P_1^*)}$

* The angle $\alpha \cong 72.37°$.
$(-\Delta P_1^*) = 111\text{-}128$ (computed constant).

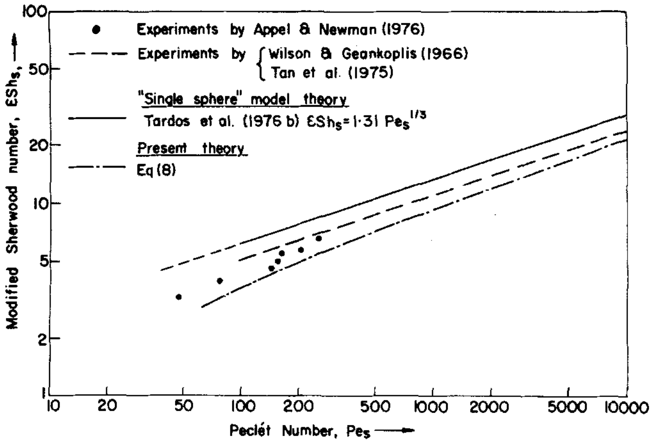

Figure 2. Comparison between experimental and theoretical mass transfer data.

$$\left.\begin{aligned} Pe_T &= Pe_s\left(\frac{8+4c_1+3c_1^2}{15c_2^2}\right)^{1/2} \\ Sh_T &= Sh_s\frac{1-\epsilon}{\epsilon}\left(\frac{8+4c_1+3c_1^2}{10c_2^2}\right) \end{aligned}\right\} \qquad (7)$$

Using these values in Equation (6), one obtains the Sherwood number for a sphere as

$$Sh_s = \frac{\epsilon}{1-\epsilon}\left[\frac{16.2}{\sqrt[3]{15}}\left(\frac{c_2^2}{8+4c_1+3c_1^2}\right)^{2/3} Pe_s^{1/3} - \frac{12.3c_2^2}{8+4c_1+3c_1^2}\right] \qquad (8)$$

At very high Peclet numbers ($Pe_s > 1000$) the last term may be neglected, resulting in

$$Sh_s \cong \frac{6.58}{1-\epsilon}\left(\frac{c_2^2}{8+4c_1+3c_1^2}\right)^{2/3} Pe_s^{1/3} \qquad (9)$$

Computations for a sand bed of $\epsilon = 0.47$, $d_s = 0.714$ mm, $d_c = 0.26$ mm and $S_{wi} = 0.127$ were carried out by Payatakes and Neira (1977). Using their results, Equation (9) becomes

$$Sh_s \cong \frac{1.01}{\epsilon} Pe_s^{1/3} \qquad (10)$$

which is in remarkable agreement with the experimental relationship obtained by Wilson and Geankoplis (1966) and is given in Table 1.

In Figure 2, the present solution, Equation (8), is plotted together with experimental results of Appel and Newman (1976), the experimental correlation of Wilson and Geankoplis (1966) and the theoretical solution based on the "single sphere" model as obtained by Tardos et al. (1976*b*). The "single sphere" theory provides an upper bound on mass transfer rates, while the present theory provides a lower bound on the experimental data.

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NOTATION

a = constricted tube dimension (Figure 1)
A = total mass transfer area, Equation (A-5)
c_1, c_2 = constants defined in Table 2
Δc = concentration gradient
d = diameter
$\bar{d}$ = diameter of the equal volume cylinder, Equation (3)
d_e = equivalent diameter, Equation (A-6)
D = diffusion coefficient
$g(\epsilon)$ = porosity dependent function, Equation (2)
h = constricted tube dimension (Figure 1)
k = Darcy constant (Table 2)
K_L = mass transfer coefficient
L_T = active tube length
Q = overall mass transfer rate, Equation (A-4)
n_p, n_s = number of pores and number of spheres in volume $1 \times 1 \times h$ respectively, Equations (A-7-A-8)
N_p = number of pores per unit volume of bed (Table 2)
Pe = Peclet number, Equation (A-1)
R, θ = cylindrical polar coordinates, dimensionless
Re_0 = Reynolds number, $Re_0 = dU_0/\nu$
S_{wi} = irreducible saturation value (Table 2)
Sh = Sherwood number, Equation (A-4)
U_0 = superficial fluid velocity
V_p = volume of pore (Table 2)

Greek Letters

ψ = stream function, dimensionless
ϵ = bed porosity
ν = fluid kinematic viscosity

Indices

c = constriction
p = pore
S = sphere
T = tube

LITERATURE CITED

Appel, P. W. and Y. Newman, Application of the Limit Current Method to Mass Transfer in Packed Beds at Very Low Reynolds Numbers, *AIChE J.*, **22** (6), 979 (1976).

Cookson, J. T., Removal of Submicron Particles in Packed Beds, *Environ. Sci. Technol.*, **4**, 128 (1970).

Fedkiw, P. and J. Newman, Mass Transfer at High Peclet Numbers for Creeping Flow in a Packed-bed Reactor, *AIChE J.*, **23** (3), 255 (May, 1977).

Graetz, L., Über die Warmeleitfähigkeit von Flüssigkeiten, *Ann. Phys.*, **25**, 337 (1885).

Happel, J., Viscous Flow in Multiparticle Systems, Slow Motion of Fluids Relative to Beds of Spherical Particles, *AIChE J.*, **4**, 197 (1958).

Karabellas, A. J., T. H. Wegner, and T. J. Hanratty, Use of Asymptotic Relations to Correlate Mass Transfer Data in Packed Beds, *Chem. Eng. Sci.*, **26**, 1581 (1971).

Kuwabara, S., The Forces Experienced by Randomly Distributed Parallel Circular Cylinders or Spheres in a Viscous Fluid at Small Reynolds Numbers, *J. Phys. Soc. Japan*, **14**, 527 (1959).

Levêque, M. A., Les lois de la transmission de la chaleur par convection, *Ann. mines*, **13**, pp. 201, 305, 381 (1928).

Levich, V. G., *Physicochemical Hydrodynamics*, Prentice-Hall, Englewood Cliffs, NJ (1962).

Neale, H. N. and W. K. Nader, Prediction of Transport Processes Within Porous Media: Creeping Flow Relative to a Fixed Swarm of Spherical Particles, *AIChE J.*, **20**, 530 (1974).

Payatakes, A. C. and M. A. Neira, Model of the Constricted Unit Cell Type for Isotropic Granular Porous Media, *AIChE J.*, **23** (6), 922 (November, 1977).

Pfeffer, R., Heat and Mass Transfer in Multiparticle Systems, *Ind. Eng. Chem. Fundam.*, **3**, 380 (1964).

Sirkar, K. K., Creeping Flow Mass Transfer to a Single Active Sphere in a Random Spherical Inactive Particle Cloud at High Schmidt Numbers, *Chem. Eng. Sci.*, **29**, 863 (1974).
Sirkar, K. K., Transport in Packed Beds at Intermediate Reynolds Numbers, *Ind. Eng. Chem. Fundam.*, **14**, 73 (1975).
Sorensen, J. P. and W. E. Stewart, Computation of Forced Convection in Slow Flow Through Ducts and Packed Beds—I, II, III, IV, *Chem. Eng. Sci.*, **29**, 819 (1974).
Tam, C. K. W., The Drag on a Cloud of Spherical Particles in Low Reynolds Number Flow, *J. Fluid Mech.*, **38**, 537 (1969).
Tan, A. Y., B. D. Prasher, J. A. Guin, Mass Transfer in Nonuniform Packing, *AIChE J.*, **21** (2), 396 (March, 1975).
Tardos, G., N. Abuaf, and C. Gutfinger, Diffusional Filtration of Dust in a Fluidized Bed Filter, *Atmos. Environ.*, **10**, 389 (1976*a*).
Tardos, G. J., C. Gutfinger, and N. Abuaf, High Peclet Number Mass Transfer to a Sphere in a Fixed or Fluidized Bed, *AIChE J.*, **22** (6), 1147 (1976*b*).
Tardos, G. I., Granular Bed Filters—Theory and Experiments, Ph.D. Dissertation, Technion Israel Institute of Technology, Haifa, Israel, 1977.
Thoenes, D. and H. Kramers, Mass Transfer from a Sphere in Various Regular Packings to a Flowing Fluid, *Chem. Eng. Sci.*, **8**, 271 (1958).
Wilson, E. J. and C. J. Geankoplis, Liquid Mass Transfer at Very Low Reynolds Numbers in Packed Beds, *Ind. Eng. Chem. Fundam.*, **5**, 9 (1966).

APPENDIX

CORRELATIONS BETWEEN SHERWOOD, *Sh*, AND PECLET, *Pe*, NUMBERS FOR A SPHERE AND FOR A TUBE SITUATED IN A GRANULAR BED

The Peclet number for a tube and for a sphere are defined respectively as

$$Pe_T = \frac{d_T U_0}{D} = \frac{\bar{d} U_0}{D}; \quad Pe_S = \frac{d_s U_0}{D} \tag{A-1}$$

Hence they may be related by

$$Pe_T = Pe_S \frac{\bar{d}}{d_S} = Pe_S \sqrt{\frac{8 + 4c_1 + 3c_2}{15} \cdot \frac{d_c^2}{d_S^2}} \tag{A-2}$$

Introducing c_2 into Equation (A-2) one obtains

$$Pe_T = Pe_S \left[\frac{8 + 4c_1 + 3c_1^2}{15c_2^2} \right]^{1/2} \tag{A-3}$$

The overall mass transfer rate may be expressed as

$$Q = K_L A \Delta c = \frac{K_L d_e}{D} \frac{D}{d_e} A \Delta c = Sh \frac{D}{d_e} A \Delta c \tag{A-4}$$

Here, d_e is the equivalent diameter and A, the total transfer area. Equation (A-4) for a sphere, becomes

$$Q_S = Sh_S \frac{D}{d_s} A_s \Delta c = Sh_S D(\pi d_S) \Delta c \tag{A-5}$$

while for a tube it reads

$$Q_T = Sh_T \frac{D}{d_T} A_T \Delta c = Sh_T D(\pi L_T) \Delta c \tag{A-6}$$

A mass transfer balance is now performed on a volume of the granular bed of dimensions $(1 \times 1 \times h)$, where $h = c_2 d_c$. The number of pores, n_p, in such a volume is

$$n_p = N_p \cdot c_2 d_c \cdot 1 \cdot 1 \tag{A-7}$$

whereas the number of spheres in the same volume is

$$n_s = \frac{c_2 d_c \cdot 1 \cdot 1 \cdot (1 - \epsilon)}{\frac{\pi d_s^3}{6}} \tag{A-8}$$

Using Equations (A-5 to A-8), the balance yields

$$Q_T \cdot n_p = Q_S \cdot n_s \tag{A-9}$$

and finally, after some algebraic manipulations, one obtains

$$Sh_T = Sh_s \frac{1 - \epsilon}{\epsilon} \cdot \left[\frac{8 + 4c_1 + 3c_1^2}{10c_2^2} \right] \tag{A-10}$$

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Entrance Region (Lévêquelike) Mass Transfer Coefficients in Packed Bed Reactors

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Calculations for the high Peclet number, entrance region (Lévêquelike) packed bed, mass transfer coefficient using a sinusoidal periodically constricted tube model for the void structure of the bed are presented. An inverse cube root dependence of the mass transfer coefficient on the bed depth is predicted. This length dependence is anticipated only at very low Reynolds numbers. Calculations which assume a mixing region between successive periods are also presented. No bed length dependence is anticipated in these coefficients.

The periodically constricted tube model for porous media constitutes a useful model for mass transfer in two-phase, packed-bed reactors. This model was developed by Payatakes and co-workers (1973, 1977) to predict the permeability of a nonconsolidated packed bed. They envisioned the bed as cell structures made of segments of parabolic periodically constricted tubes. A sinusoidal periodically constricted tube (PCT) is used in this work to model the void structure in a bed in order to predict the mass transfer coefficient. The fluid is assumed to be in the viscous flow regime, and the reactant conversion is assumed to be controlled by mass transfer from the

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