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Interphase fluid-particle mass transport at low Reynolds numbers

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Industrial scale fixed bed processes are largely conducted at modest to high values of the particle Reynolds number. Laboratory fluid–solid fixed bed studies, including catalyst rate measurements, are carried out often at $Re_p < 0.1$, a flow regime largely devoid of fluid-particle transport data and correlations. From a study of methyl iodide adsorption over charcoal it is shown that it is possible to use the transient breakthrough behaviour of a well-mixed recycle absorber to analyse gas-particle film coefficients where standard experimental models are not effective.

Keywords: mass transport; low Reynolds numbers; interphase fluid mechanics

Transport of heat and mass between flowing fluids and particles or pellets in fixed beds has been a topic of considerable interest for many years given the central role occupied by fixed beds in diverse chemical and separation processes. The magnitude of research activity in this area is evident from a review [1] which cites 309 references. It would seem that in the particle Reynolds number range of industrial interest (Re_p of 10–1000) correlations abound although there is significant scatter in the results obtained by different workers. Recent studies [2,3] have established some semblance of order in the region of modest to high Reynolds numbers. The careful study of Hsiung and Thodos [2] for values of Re_p of 0.1–100 suggests for a spherical particle

$$J_D = 1.33/Re_p^{0.4}, \tag{1}$$

where

$$J_D = \frac{kg}{u_0}(Sc)^{2/3} \quad \text{and} \quad Re_p = \frac{d_p u_0}{\nu} \tag{2}$$

(for nomenclature, see table 1).

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Table 1

Nomenclature		
a	external surface area of particles per unit volume	m^{-1}
c	concentration	kg/m^3
d_p	particle diameter	m
D_e	effective diffusity	m^2/s
J_D	mass transfer J factor	
kg	interphase mass transfer coefficient	m/s
K_a	adsorption rate coefficient	m^3/kg s
K_e	adsorption equilibrium constant	m^3/kg
q	adsorbed species concentration	kg/kg
r	radial co-ordinate	m
Re_p	particle Reynolds number, for empty bed	$d_p u_0/\nu$
Re_p^*	particle Reynolds number, for fixed bed	$d_p u_0/\nu\varepsilon$
Sc	Schmidt number	
t	time	s
ε	particle porosity	
θ	reactor residence time	s
ν	kinetic viscosity	m^2/s
u_0	superficial velocity	m/s
ρ_p	particle density	kg/m^3
subscripts		
0	inlet	
r	radial position	
R	radius of particle	

When we extend analysis to include fixed beds, the Reynolds number, Re_p, changes to Re_p^* defined as

$$Re_p^* = \frac{d_p u_0}{\nu\varepsilon} .$$

In terms of the original Reynolds number,

$$\text{(empty) } Re_p = Re_p^* \varepsilon .$$

Theory has been of some value [4], insofar as it predicts, how J_D varies with the bed Reynolds number

$$J_D = \frac{1.15}{(Re_p^*)^{0.5} \varepsilon^{0.5}} \tag{2a}$$

and

$$\varepsilon J_D = \frac{1.15\sqrt{\varepsilon}}{(Re_p^*)^{0.5}} , \tag{2b}$$

for $\varepsilon = 0.4$,

$$\varepsilon J_\mathrm{D} = \frac{0.727}{(\mathrm{Re}_\mathrm{p}^*)^{0.5}} \tag{2c}$$

which compares rather favourably with a recent experimental correlation [3],

$$\varepsilon J_\mathrm{D} = \frac{0.72}{(\mathrm{Re}_\mathrm{p}^*)^{0.51}} \;:\; 10 < \mathrm{Re} < 800\,. \tag{3}$$

While industrial scale fixed bed processes are largely conducted at modest to high values of the particle Reynolds number, laboratory fluid–solid fixed bed studies, including catalyst rate measurements are often carried out at $\mathrm{Re}_\mathrm{p} < 0.1$ – a flow regime largely devoid of fluid-particle transport data and correlations. As the precision of kinetic studies improves, it is desirable to ensure that the criteria used to verify the absence of mass transfer effects contain parameters which are accurately known. In a study primarily concerned with methyl iodide adsorption in fixed beds of charcoal [5], analysis of the governing continuity equations revealed the possibility of extracting values of gas-particle film coefficients in this low and relatively unexplored range of Reynolds numbers where standard experimental methods are not effective. The experimental technique involved measuring the transient breakthrough behaviour of a well mixed recycle adsorber when the methyl iodide concentration of the input stream was changed.

For such a gradientless (recycle) adsorber

$$\frac{\mathrm{d}c}{\mathrm{d}t} = \frac{c_0 - c}{\theta} - \mathrm{kg}a(c - c_R)\,, \tag{4}$$

where c_0 is the constant feed composition, c the time dependent effluent concentration and c_R concentration at particle external surface. For a particle of charcoal, the dynamic mass balance of adsorbate on the particle is given by

$$\varepsilon_\mathrm{p}\frac{\partial C_r}{\partial t} = D_\mathrm{e}\left(\frac{\partial^2 C_r}{\partial r^2} + \frac{2}{r}\frac{\partial C_r}{\partial r}\right) - \rho_\mathrm{p}\frac{\partial q}{\partial t}\,. \tag{5}$$

C_r is the gas phase concentration at position r within the particle and q the concentration of the adsorbed methyl iodide within the particle; assuming a first-order reversible adsorption process

$$\frac{\partial q}{\partial t} = K_\mathrm{a}\left(C_r - \frac{q}{K_\mathrm{e}}\right). \tag{6}$$

The boundary conditions corresponding to the experiment are

$$D_\mathrm{e}\left.\frac{\partial c_r}{\partial r}\right|_{r=R} = \mathrm{kg}(c - c_R)\,, \tag{7}$$

$$\frac{\partial c}{\partial r} = 0 \quad \text{at } t = 0 \text{ and } r = 0\,,$$

Table 2
Derived values of kg (m/s) as a function of particle size (d_p) and fluid velocity (m/s)

Run	T (°C)	d_p (m) $\times 10^4$	$Re_p \times 10^2$	Sc	u_0 (m/s) $\times 10^2$	kg (m/s) $\times 10^2$	J_D
1	110	3.9	1.36	133.3	7.64	2.6	8.8
2	110	3.9	2.72	133.3	15.28	3.6	6.2
3	110	3.9	4.1	133.3	22.9	4.8	5.3
4	130	8.6	7.8	128.5	19.86	4	5.13
5	150	8.6	7.8	117.5	19.86	4	5.3
6	110	3.9	3.54	133.3	19.86	4.5	6.04
7	130	3.9	3.54	128.6	19.86	5	6.4
8	150	3.9	3.54	117.3	19.86	5	5.91

$$c = c_r = q = 0 \quad \text{at } t = 0,$$

$$C_0 = \text{constant at } t > 0.$$

Experiments show that the adsorption rate is virtually instantaneous, and so the adsorption rate data reflect a mass transport controlled process. Solution of the above equations by NAG standard computer routines generates predicted c versus t curves which when matched to experimental data may be used to give values of kg. The results are set forth in table 2.

For $0.01 < Re < 0.1$

$$J_D = 1.26\, Re_p^{-0.45}. \tag{8}$$

For completeness and comparison, other published correlations are given in table 3.

Table 3
Comparison of this data and Re_p with other (steady state) data

Author	Re_p	Correlation	Ref.
Williamson et al.	$0.08 < Re_p < 125$	$J_D = 2.40 Re_p^{-0.66}$	[6]
Hougen	$0.01 < Re < 50$	$J_D = 0.84 Re_p^{-0.51}$	[7]
Evans and Gerald	$1.6 < Re_p < 70$	$J_D = 1.48 (Re_p')^{-0.52}$	[8]
Hsiung and Thodos	$0.1 < Re_p < 100$	$J_D = 0.48 Re_p^{-0.39}$	[2]
Dwivedi and Upadhyay	general correlation	$J_D = 1.11 Re_p^{-0.72}$	[9] [a]
Carberry	boundary layer model	$J_D = 1.15 Re_p^{-0.5}$	[4]
this work	$0.01 < Re < 0.04$	$J_D = 1.26 Re_p^{-0.45}$	

[a] $Re_p < 10$.

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