Int. J. Heat Mass Transfer. Vol. 30, No. 12, pp. 2683–2685, 1987
Printed in Great Britain

0017–9310/87 $3.00+0.00

TECHNICAL NOTES

On ceramic particle–wall heat transfer in fixed beds

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(*Received* 1 *December* 1986 *and in final form* 11 *May* 1987)

INTRODUCTION

The mathematical descriptions of both fixed and moving bed heterogeneously solid catalyzed reactors were set forth in the seminal work of Amundson [1] which inspired, in the fullness of time, fruitful experimental work devoted to measurement and correlation of those parameters defined by Amundson.

The fixed bed catalytic reactor quite naturally attracted both experimental and theoretical attention. Levels of fixed bed model sophistication became defined [2, 3], i.e. a hierarchy of complexity emerged ranging from the pseudo-homogeneous steady-state one-dimensional (plug flow [3]) model to a two-dimensional (axial and radial) two-phase (fluid–solid) heterogeneous model [4].

For the most primitive of solid catalyzed reactions, a sophisticated model of the fixed bed requires a mathematical description which includes (a) a fluid phase and (b) a solid-bed phase continuity equation phrased in terms of concentrations and temperatures (fluid–solid phases) as a function of axial and radial coordinates [1–5]. Consequently the boundary condition(s) at the wall at which heat is transferred become key elements in the total resolution of the model [3–5].

For the non-isothermal, non-adiabatic fixed bed catalytic reactor, we have, at the solid catalyst–wall interface

$$-k_{\mathrm{r}}^{0}\frac{\mathrm{d}T}{\mathrm{d}r} = h_{\mathrm{p\text{-}w}}(T-T_{\mathrm{w}}) \tag{1}$$

where T_{w} is the wall temperature.

Equation (1) is gainfully rephrased in dimensionless form ($\gamma = r/R_0$; $t = T/T_0$) to yield the wall–solid particle (catalyst pellet–extrudate) Biot number, *based upon tube radius*, R_0

$$\left(\frac{h_{\mathrm{p\text{-}w}}R_0}{k_{\mathrm{r}}^{0}}\right) = (Bi)_R = \frac{\mathrm{d}t/\mathrm{d}\gamma}{t-t_{\mathrm{w}}} \equiv \frac{\text{core}}{\text{wall}}\Delta T. \tag{2}$$

That is, $(Bi)_R$, is proportional to the bed center (core) temperature gradient relative to that at the wall region.

ANALYSIS

Biot numbers, $(Bi)_R$, for particle–wall heat transport, based upon the packed bed *radius*, have been measured and reported by Melanson and Dixon [6] for $D_{\mathrm{T}}/d_{\mathrm{p}}$ ratio from 5 to 12—a range of aspect ratios of signal import in fixed bed catalytic reaction technology.

Reference [6] provides data for $(Bi)_R$ between $D_{\mathrm{T}}/d_{\mathrm{p}}$ of 5 and 12 for packed beds of diverse core thermal conductivities. The data base [6] includes metal and polymer particles. Only the data for ceramic and nylon packing are relevant here. The stagnant surrounding fluid (air) is of conductivity, k_{f}, of 0.03 W m^{-1} K^{-1}. A direct proportionality between $(Bi)_R$ and $D_{\mathrm{T}}/d_{\mathrm{p}}$ is evident [6]. The relevant data are displayed in Table 1 for ceramic packing typical of fixed bed catalytic reactors, as well as data for nylon spheres.

As the authors note [6], their empirical data seem not to yield to a theoretical rationale. We submit, however, that this unique body of data [6] does lend itself to a legitimate rationalization.

For ceramic and nylon particles the data of Table 1 [6] teach that

$$(Bi)_R = \frac{h_{\mathrm{p\text{-}w}}R_0}{k_{\mathrm{r}}^{0}} = \text{constant}\cdot(D_{\mathrm{T}}/d_{\mathrm{p}})^{a}. \tag{3}$$

In fact $(Bi)_R$ varies with $D_{\mathrm{T}}/d_{\mathrm{p}}$ in a second-order manner, as is indicated in Table 2, where

$$C_R = (Bi)_R\bigg/\left(\frac{D_{\mathrm{T}}}{d_{\mathrm{p}}}\right)^2 \cong \text{constant}.$$

Why a $D_{\mathrm{T}}/d_{\mathrm{p}}$ dependency with respect to $(Bi)_R$? Actually a linear dependency is predictable since [3, 7]

$$(Bi)_R = \frac{h_{\mathrm{p\text{-}w}}R_0}{k_{\mathrm{r}}^{0}} = \left(\frac{h_{\mathrm{w\text{-}s}}d_{\mathrm{p}}}{k_{\mathrm{r}}^{0}}\right)\frac{D_{\mathrm{T}}}{2d_{\mathrm{p}}} \tag{4}$$

or

$$(Bi)_{\mathrm{p\text{-}w}} = 2(Bi)_R/(D_{\mathrm{T}}/d_{\mathrm{p}}) \quad \text{(see Table 1)}.$$

The Melanson–Dixon data are $D_{\mathrm{T}}/d_{\mathrm{p}}$ dependent and the said dependency would be linear *if and only if*

$$(Bi)_{\mathrm{p\text{-}w}} = \left(\frac{h_{\mathrm{p\text{-}w}}d_{\mathrm{p}}}{k_{\mathrm{r}}^{0}}\right) = \text{constant}. \tag{5}$$

Now $h_{\mathrm{p\text{-}w}}$ (the stagnant particle–wall heat transfer coefficient) must depend upon fluid phase conductivity, k_{f}.

Equation (5) is gainfully rephrased

$$(Bi)_{\mathrm{p\text{-}w}} = \frac{k_{\mathrm{f}}}{k_{\mathrm{r}}^{0}}\left(\frac{h_{\mathrm{p\text{-}w}}d_{\mathrm{p}}}{k_{\mathrm{f}}}\right) = \frac{k_{\mathrm{f}}}{k_{\mathrm{r}}^{0}}(Nu)_{\mathrm{p\text{-}w}} \tag{6}$$

which simply declares that

$$(Bi)_{\mathrm{p\text{-}w}} = \frac{k_{\mathrm{f}}}{k_{\mathrm{r}}^{0}}(\text{constant}) \tag{7}$$

if and only if $(Nu)_{\mathrm{p\text{-}w}}$ is constant: it surely is not constant, however.

Equation (7) is a rationale for two suggested formulae for $(Bi)_{\mathrm{p\text{-}w}}$: that of Sullivan and Sabersky [7] and Schlunder's modification of ref. [7] as well as his own forecast which contains a radiation term [8].

Sullivan and Sabersky offer [7]

$$(Bi)_{\mathrm{p\text{-}w}} = \frac{k_{\mathrm{f}}}{k_{\mathrm{r}}^{0}}(11.7). \tag{8}$$

Schlunder's modification is

$$(Bi)_{\mathrm{p\text{-}w}} = \frac{k_{\mathrm{f}}}{k_{\mathrm{r}}^{0}}(6.7) \tag{9}$$

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NOMENCLATURE

$(Bi)_R$	Biot number based on tube radius, $h_{p\text{-}w}R_0/k_r^0$
$(Bi)_{p\text{-}w}$	particle–wall Biot number, $h_{p\text{-}w}d_p/k_r^0$
d_p	particle diameter
D_T	tube diameter
$h_{p\text{-}w}$	particle–wall heat transfer coefficient [W m^{-2} K^{-1}]
k_r^0	bed center (core) stagnant thermal conductivity [W m^{-1} K^{-1}]
k_f	fluid molecular thermal conductivity [W m^{-1} K^{-1}]
$(Nu^0)_{p\text{-}w}$	particle–wall point Nusselt number, $h_{p\text{-}w}d_p/k_f$
R_0	tube radius
t	reduced temperature, T/T_0.

Greek symbol

γ	r/R_0.

Table 1. Data of Melanson and Dixon [6]—ceramic and nylon packing (D_T = 75 mm)

Packing	d_p	D_T/d_p	$(Bi)_R$	$(Bi)_{p\text{-}w}$†
Ceramic				
(A) Glass hollow cylinders (6 × 6 × 4 mm)	6.87	11	14	2.54
(B) Glass hollow cylinders (8 × 8 × 6 mm)	9.1	8.2	7	1.7
(C) Porous ceramic spheres (9.53 mm)	9.53	8	6.65	1.6
(D) Porous ceramic spheres (12.7 mm)	12.7	6	2.48	0.83
(E) Porous ceramic cylinders (9.5 mm)	11	7	5.26	1.5
(F) Ceramic hollow cylinders (7.9 × 7.9 × 4 mm)	9.1	8.3	7.8	1.54
(G) Ceramic hollow cylinders (13 × 13 × 7 mm)	15	5	2.34	0.94
Nylon				
(H) Nylon spheres	6.35	12	14.2	2.36
(I) Nylon spheres	7.94	9.3	7.76	1.67
(J) Nylon spheres	9.53	7.9	5.35	1.35
(K) Nylon spheres	12.7	6	3.52	1.17

† $(Bi)_{p\text{-}w} = 2(Bi)_R/(D_T/d_p)$.

Table 2. Second-order behavior of $(Bi)_R$ vs D_T/d_p derived from the data of ref. [6]

Packing		$(Bi)_R$	D_T/d_p	$C_R = (Bi)_R\Big/\left(\frac{D_T}{d_p}\right)^2$
A	Ceramic	14	11	0.116
B	Ceramic	7	8.2	0.104
C	Ceramic	6.65	8	0.104
D	Ceramic	2.48	6	0.07
E	Ceramic	5.26	7	0.107
F	Ceramic	7.8	8.3	0.113
G	Ceramic	2.34	5	0.094
H	Nylon	14.2	12	0.0986
I	Nylon	7.76	9.3	0.09
J	Nylon	5.35	7.9	0.086
K	Nylon	3.52	6	0.098

while his own [8] elegant derivation reduces, in the absence of a considerable radiation contribution, to

$$(Bi)_{p\text{-}w} \cong \frac{k_f}{k_r^0}(9). \qquad (10)$$

In Table 1 there are set forth the Melanson–Dixon data rephrased in terms of $(Bi)_{p\text{-}w}$. From the derived $(Bi)_{p\text{-}w}$ data of Table 1 it is manifest that $(Bi)_{p\text{-}w}$ is not a constant. As shown in Fig. 1 the said data are linear in D_T/d_p. Which is to state that $(Nu)_{p\text{-}w}$ in equation (6) is not independent of D_T/d_p, in spite of previous forecasts [7, 8].

We argue that $(Nu)_{p\text{-}w}$ must be dependent upon the number of particle–wall contact points.

The number of particle–wall contact points is proportional to the number of particle diameters per perimeter, i.e. D_T/d_p, so

$$(Nu)_{p\text{-}w} = (Nu^0)_{p\text{-}w}\cdot D_T/d_p$$

therefore equation (6) becomes

$$(Bi)_{p\text{-}w} = \frac{k_f}{k_r^0}(Nu^0)_{p\text{-}w}\left(\frac{D_T}{d_p}\right) \qquad (11)$$

where $(Nu^0)_{p\text{-}w}$ is the Nusselt number per contact point.

Derived values of $(Nu^0)_{p\text{-}w}$ are shown in Table 3. Reasonable constancy is evident, particularly given uncertainties in values of k_r^0 and the understandable scatter in the $(Bi)_R$ data. Values of the center bed (core) stagnant conductivities are those of Melanson and Dixon [6]. For air $k_f = 0.03$ W m^{-1} K^{-1} is used to obtain k_f/k_r^0, from which we compute

$$(Nu^0)_{p\text{-}w} = (Bi)_{p\text{-}w}\Big/\left(\frac{k_f}{k_r^0}\cdot\frac{D_T}{d_p}\right). \qquad (12)$$

Figure 1 ($(Bi)_{p\text{-}w}$ vs (D_T/d_p)) is in accord with our analysis (equation (11)). The result is rather persuasive support of our interpretation of the data base [6]. For D_T/d_p between 5

Table 3. $(Nu^0)_{p\text{-}w}$ values derived from data of ref. [6]

Packing		k_r^0 (W m^{-1} K^{-1})†	D_T/d_p†	$(Bi)_{p\text{-}w}$‡	$(Nu^0)_{p\text{-}w}$§
A	Ceramic	0.303	11	2.54	2.3
B	Ceramic	0.280	8.2	1.7	1.93
C	Ceramic	0.313	8	1.66	2.16
D	Ceramic	0.452	6	0.83	2.1
E	Ceramic	0.390	7	1.5	2.8
F	Ceramic	0.489	8.3	1.54	3.1
G	Ceramic	0.469	5	0.94	2.9
H	Nylon	0.214	12	2.36	1.4
I	Nylon	0.209	9.3	1.67	1.26
J	Nylon	0.240	7.9	1.35	1.37
K	Nylon	0.279	6	1.17	1.82

† Table 1 of ref. [6].

‡ $(Bi)_{p\text{-}w} = 2(Bi)_R/(D_T/d_p)$ (see Table 1).

§ $(Nu^0)_{p\text{-}w} = (Bi)_{p\text{-}w}/((k_f/k_r^0)\cdot(D_T/d_p))$: $k_f = 0.03$ W m^{-1} K^{-1}.

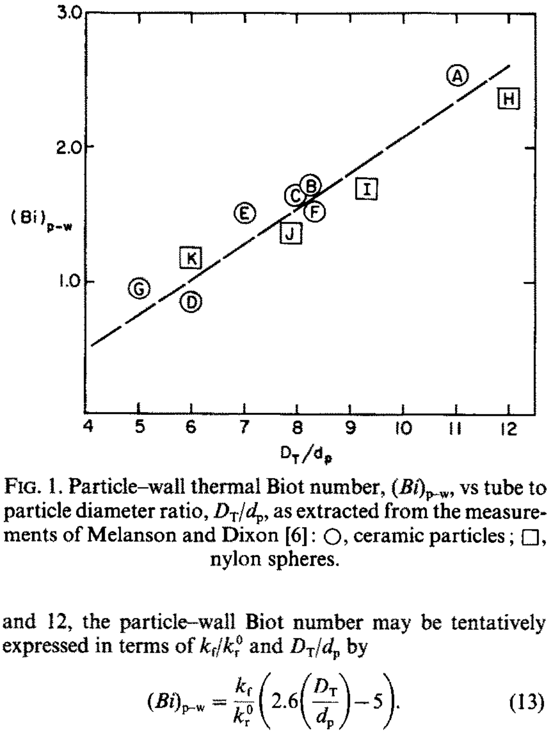

Fig. 1. Particle–wall thermal Biot number, $(Bi)_{p\text{-}w}$, vs tube to particle diameter ratio, D_T/d_p, as extracted from the measurements of Melanson and Dixon [6]: ○, ceramic particles; □, nylon spheres.

and 12, the particle–wall Biot number may be tentatively expressed in terms of k_f/k_r^0 and D_T/d_p by

$$(Bi)_{p\text{-}w} = \frac{k_f}{k_r^0}\left(2.6\left(\frac{D_T}{d_p}\right)-5\right). \tag{13}$$

DISCUSSION AND CONCLUSIONS

It is to be noted that in the range of D_T/d_p of 5–10, equation (13) predicts

$$(Bi)_{p\text{-}w} = \frac{k_f}{k_s^0}(8 \text{ to } 20)$$

which embraces correlations suggested by Sullivan and Sabersky [7] and Schlunder [8]. Equation (13) also reduces to Olbrich's [9] predicted lower limit of

$$(Bi)_{p\text{-}w} = 2.12$$

for $k_f/k_r^0 = 0.1$ and $D_T/d_p = 10$.

The correlation of Specchia *et al.* [10], unlike other forecasts [7–9], does contain a D_T/d_p dependency, but one which predicts an *increase* in $(Bi)_{p\text{-}w}$ with a *decrease* in D_T/d_p—contrary to the data base [6] and the rationale presented here.

It might be argued that the $(Bi)_{p\text{-}w}$–D_T/d_p dependency (Fig. 1) is due to a linear dependence of k_r^0 upon D_T/d_p. But inspection of the Melanson–Dixon data [6], Table 3 of this note, grants little support for such a contention. Variations in packing techniques may well account for variations in k_r^0, e.g. packing B and F at the same value of D_T/d_p (Table 3).

While the data base [6] and our analysis are limited to D_T/d_p values of 5–12, it must be recalled that this aspect ratio embraces those typically employed in highly exothermic fixed bed processes, e.g. xylene, naphthalene oxidation [3, 5]. Further, for $D_T/d_p > 12$, the wall Biot number assumes less importance since the major radial temperature gradient lies not at the wall but within the core of the fixed bed [3]. For $D_T/d_p < 5$, it is questionable as to whether a continuity concept can be invoked in a heat transfer study, no less heat transfer with catalytic reaction.

Acknowledgement—We are grateful to Exxon, Mobile Oil, duPont and Sun Oil for their support which sustains our programs in Catalysis and Catalytic Reaction Engineering.

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