

## HEAT TRANSFER IN PACKED BEDS—A REEVALUATION

CHI-HSIUNG LI and B. A. FINLAYSON

Department of Chemical Engineering, University of Washington, Seattle, WA 98195, U.S.A.

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**Abstract**—Data for heat transfer from packed beds are reexamined in the light of new insights. Much of the data includes a length effect, resulting from a higher heat transfer coefficient near the inlet, making it unsuitable for use in chemical reactor design, where the length is so long that an asymptotic heat transfer coefficient is desired. The data is reexamined in order to exclude studies influenced by the length effect and retaining data giving an asymptotic heat transfer coefficient. The asymptotic coefficient is correlated well over a large range of Reynolds numbers (20–7000). The data also indicate that the Biot number decreases as the Reynolds number increases, but is approximately constant for Reynolds numbers above 500, taking the value

$$\frac{Bi_d \epsilon}{R(1-\epsilon)} = 0.27.$$

### INTRODUCTION

The tubular fixed bed chemical reactor is widely used for exothermic chemical reactions. The energy released by the chemical reaction must be removed by cooling at the walls and this leads to the desirability of having accurate correlations for the heat transfer coefficient from packed beds. Experimental data for the Nusselt number as a function of Reynolds number often show considerable disagreement from one study to the next, with discrepancies as large as 100% being common[1, 2]. The purpose of this report is to reexamine the experimental data to see the cause of the discrepancy and to eliminate it.

Most data is obtained in a Graetz-type experiment. The fluid passes through a packed bed and is cooled at the walls. In an empty pipe, with either laminar or turbulent flow, there is an entry length effect, and the heat transfer coefficient at the bed inlet is infinite, but for positions farther down the pipe the coefficient decreases and eventually approaches an asymptotic value. In a packed bed the same phenomena is much less likely because the heat transfer coefficient depends more on the fluid flow near the wall rather than on the temperature distribution. However, evidence is given below to suggest that the heat transfer coefficient does depend on length, and this idea is the key to unraveling the discrepancies in the data reported in the literature.

### THEORY

The two-dimensional pseudo-homogeneous model for heat transfer in a packed bed is[3]

$$\begin{aligned} GC_p \frac{\partial T'}{\partial z'} &= \frac{k_e}{r'} \frac{\partial}{\partial r'} \left( r' \frac{\partial T'}{\partial r'} \right) \\ T' &= T'_i \quad \text{at} \quad z' = 0 \\ \frac{\partial T'}{\partial r'} &= 0 \quad \text{at} \quad r' = 0 \\ -k_e \frac{\partial T'}{\partial r'} &= h_w (T' - T_w) \quad \text{at} \quad r' = R. \end{aligned} \quad (1)$$

In nondimensional form this is

$$\begin{aligned} \frac{\partial T}{\partial z} &= \frac{\alpha'}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) \\ T &= 1 \quad \text{at} \quad z = 0 \\ \frac{\partial T}{\partial r} &= 0 \quad \text{at} \quad r = 0 \\ -\frac{\partial T}{\partial r} &= BiT \quad \text{at} \quad r = 1. \end{aligned} \quad (2)$$

If the effective thermal conductivity and heat transfer coefficient are taken as constants, this problem has a well-known solution

$$T = 2 \sum_{n=1}^{\infty} \frac{J_0(A_n r) \exp[-\alpha' A_n^2 z]}{A_n J_1(A_n) [(A_n/Bi)^2 + 1]} \quad (3)$$

where  $J_0(A_n r)$  is the zeroth order Bessel function of the first kind, and the  $A_n$  are the roots to

$$A_n J_1(A_n) = Bi J_0(A_n). \quad (4)$$

As the length of the packed bed increases only the first term in the series is needed, but the position at which this approximation is valid depends on the Biot number. Table 1 lists the values of  $\alpha'_{\min}$  as a function of Biot number, where  $\alpha'_{\min}$  is determined such that the second term in the expansion for the centerline temperature is less than 1% of the first term. For lengths beyond  $\alpha'_{\min}$  a plot of  $\log T$  vs  $z$  is a straight line. For simplicity we use the value  $\alpha'_{\min} = 0.2$  for all Biot numbers.

The Biot number can be expressed as a ratio of the resistance to heat transfer through the packed bed,  $R/k_e$ , to that at the wall,  $1/h_w$ . Thus small Biot numbers refer to a large wall resistance, while for large Biot numbers the resistance to heat transfer is largely interior to the bed. The eigenvalue  $A_1$  depends on  $Bi$ , but for  $Bi$  greater than 10,  $A_1$  approaches 2.4048 and  $(A_1/Bi) \ll 1$ .

Table 1. Minimum length for a one-term expansion to be valid in eqn (3)

Bi	0.1	0.3	1.0	3.0	10.	100	$\infty$
$\alpha'_{\min}$	0.08	0.15	0.21	0.23	0.20	0.18	0.14

The asymptotic solution can then be approximated as

$$T = \{2J_0(A_1 r) \exp[-\alpha' A_1^2 z]\} / [A_1 J_1(A_1)] \quad (5)$$

$A_1 \approx 2.4048$

and is independent of  $Bi$ , although it still depends on  $\alpha'$ , and thus  $k_e$ . This suggests that if the Biot number is high, it would be difficult to measure the heat transfer coefficient, since the temperature solution is independent of  $Bi$ . Of course the Biot number under those conditions is less critical. We find below that the  $Bi$  decreases as Reynolds number increases, so that the scatter of data for  $h_w$  and  $Bi$  at low Reynolds numbers (high  $Bi$ ) is not surprising, nor is it of great consequence.

Nearly all the heat transfer data is obtained in the same way, namely the radial temperature profile is measured at several bed depths with the thermocouples placed just above the packing. The difference in studies results from the analysis of the data to determine  $k_e$  and  $h_w$ . Four widely used methods are outlined.

**Method 1.** The effective thermal conductivity is determined by solving eqn (1) for  $k_e$ , and the temperature derivatives are obtained by differentiating the temperature profiles. This local value of  $k_e$  is averaged to obtain a constant  $k_e$  used for the entire bed. The  $h_w$  is found from eqn (4) with  $A_1$  obtained from the slope of the straight line of  $\log T$  vs  $z$ , which is  $-\alpha' A_1^2$ . This method was used by Coberly and Marshall[4].

**Method 2.** First  $A_1$  is found from the radial temperature profile at the exit of the bed

$$\frac{T_m(z=1)}{T(r=0, z=1)} = \frac{2J_1(A_1)}{A_1}. \quad (6)$$

The slope of  $\log T$  vs  $z$  gives  $-\alpha' A_1^2$  as before, and since  $A_1$  is now known,  $k_e$  may be found from  $\alpha'$ .  $Bi$  is found as a function of  $A_1$  from eqn (4). With  $k_e$  known, the  $h_w$  is found from  $Bi$ . This method was used by Maeda[5] and Yagi and Wakao[6] and others listed below.

**Method 3.** The  $k_e$  is found as in Method 1. The  $h_w$  is found from an overall energy balance for the test section

$$h_w = \frac{GRC_p [T'_m(z'_2) - T'_m(z'_1)]}{2(z'_2 - z'_1)(T'_w - T'_R)_m}. \quad (7)$$

The fluid temperature at the wall,  $T'_w$ , which is different from the coolant temperature,  $T'_w$ , is estimated by extrapolating the measured bed temperature to the wall.

$$(T'_w - T'_R)_m = \frac{1}{z'_2 - z'_1} \int_{z'_1}^{z'_2} [T'_w - T'(r=R, z')] dz'. \quad (8)$$

This method was used by Felix[7].

**Method 4.** The  $k_e$  and  $h_w$  are found by comparing calculated results using eqn (3) with the experimental data and adjusting  $k_e$  and  $h_w$  to give the best fit in a least

squares sense. Valstar[8] minimized the errors in temperature values throughout the bed, whereas De Wasch and Froment[9] used only exit profiles.

All of these methods have been extensively discussed in the literature, but usually without mentioning the fact that if  $k_e$  and  $h_w$  depend on length then each method may yield a different value  $k_e$  and  $h_w$  even in the absence of experimental errors. The major emphasis of this paper is that each of these methods of data analysis yields a different heat transfer coefficient, which is not always the asymptotic coefficient, and the data should be compared with caution.

#### LENGTH EFFECT

Experimental evidence confirms the fact that the heat transfer coefficient ( $h_w$ ) and the effective thermal conductivity ( $k_e$ ) depend on length. Figure 1 shows data from three investigations illustrating how the effective thermal conductivity decreases as the length is increased. Most methods of data analysis (Methods 1, 2 and 4) cause an error in  $h_w$  if  $k_e$  is wrong, thus making  $h_w$  depend on length, too. DeWasch and Froment[9] (their Figs. 6 and 7 and our Table 2) and Paterson[11] found that  $k_e$  and  $h_w$  decreased with increasing bed depth. Finally we anticipate the conclusions below: Methods 3 and 4 do not give asymptotic heat transfer coefficients and should give a coefficient above the asymptotic coefficients of Method 2. Data analyzed by Methods 3 and 4 do lie generally above that analyzed by Method 2. Method 1 does not give an asymptotic coefficient, but can be either above or below the asymptotic value. Data analyzed by Method 1 follow this comparison, also.

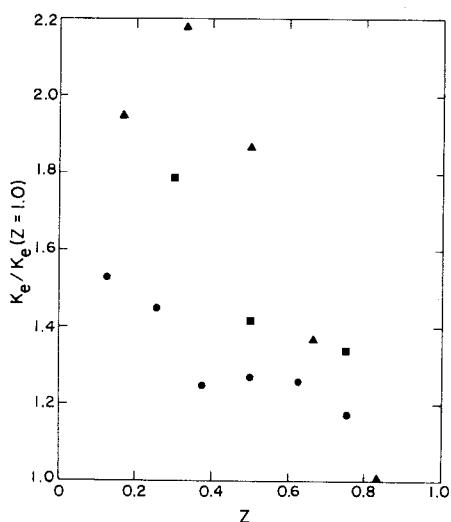


Fig. 1. The effective thermal conductivity vs bed depth. (The  $k_e$  shown in this figure is the average value of the local  $k_e$ 's across the radius of the bed.)

Symbol	Ref.	$\alpha'$	$Re_p$
■	[12]	0.28	48
●	[4]	0.56	506
▲	[13, 14]	0.16	238

Table 2. The  $k_e$  and  $h_w$ 's reported by De Wasch and Froment [9] for different bed depths.  $Re_p = 400$ ,  $d_t = 0.099$  m,  $d_p = 0.0057$  m

L (m)	$k_e$ (kcal/m hr °C)	$h_w$ (kcal/m <sup>2</sup> hr °C)	$\alpha'$	Bi
0.284	1.38	167	0.1267	5.99
0.582	1.28	158	0.2399	6.13
0.875	1.14	152	0.3266	6.60
1.016	1.12	146	0.3695	6.42

To illustrate the errors in  $k_e$  and  $h_w$  introduced by the length effect, let us calculate a temperature profile using the measured  $k_e$  and  $h_w$  for beds of different lengths. These calculations are then used as simulated experimental data, and  $k_e$  and  $h_w$  are deduced using the different methods. Differences in  $k_e$  and  $h_w$  so deduced are due entirely then to the method of data analysis, since the data is identical. The parameters used for the simulated data are reported in Table 2, as read from the graphs of actual experimental data obtained by DeWasch and Froment [9] for four different bed depths.

#### Methods 2 and 4

First analyze the simulated data using Method 4 for a bed depth of 1.016 m. The  $k_e$  and  $h_w$  are determined to give an exact fit of the temperature profile at the bed exit, but they account for the  $k_e$  and  $h_w$  throughout the bed, including the entrance region where  $k_e$  and  $h_w$  depend on length. In Method 2, however, only the shape of the temperature profile at the outlet is used (not the absolute value) to determine  $A_1$  and then  $\alpha'$  (or  $k_e$ ) is found from the slope of the  $\log T$  vs  $z$  curve, which thereby matches the local rate of heat transfer regardless of what happened upstream. Method 2 thus gives asymptotic values of  $k_e$  and  $h_w$ , at least if the bed is long enough for those asymptotic values to hold. Next the values of  $k_e$  and  $h_w$  so determined are used in a calculation of eqn (2), with  $k_e$  and  $h_w$  (hence  $\alpha'$  and  $Bi$ ) constant with length. Figure 2 illustrates the predictions of the two methods. Method 4 matches exactly the outlet average temperature, as it should, whereas Method 2 matches exactly the local rate of heat transfer as determined by the slope of the curve. The temperature at the bed outlet is not predicted correctly by Method 2 because the calculation assumes  $h_w$  and  $k_e$  are constant throughout the bed, while they are not. However, once the asymptotic region is reached, Method 2 correctly predicts the rate of heat transfer, whereas Method 4 does not. If the bed depth were very much longer, so that the region over which  $h_w$  and  $k_e$  were at their constant asymptotic value constituted, say, 95% of the bed, then Method 2 and 4 would give equivalent predictions. The length of bed needed to make negligible the entrance effect is very much larger if the data is analyzed using Method 4 than is the case for Method 2, and seldom are experimental studies done in such long beds. Consequently Method 2 yields an asymptotic heat transfer coefficient, but Method 4 does not.

#### Method 3

Whether or not this method gives an asymptotic heat

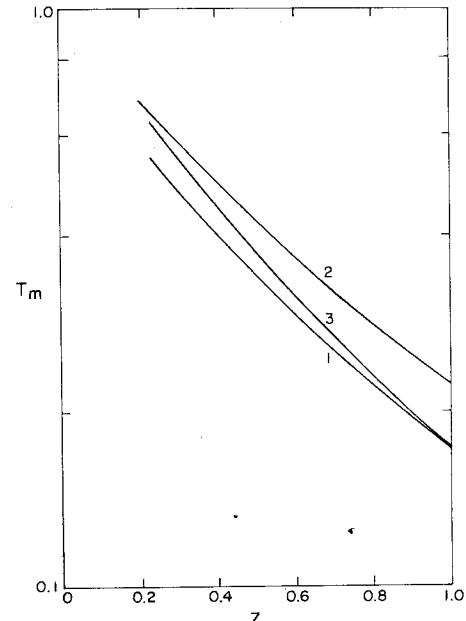


Fig. 2. Comparison of Method 2 and Method 4: 1, the assumed experimental result; 2, predicted by Method 2; 3, predicted by Method 4.

transfer coefficient depends on the location of the test section,  $z_1$  in eqn (7). If this includes the whole bed, the  $h_w$  is not an-asymptotic one, and would tend to be above the asymptotic  $h_w$ . This effect is illustrated in Table 3, where the  $h_w$  clearly depends on the location of the test section, and decreases as the test section is moved downstream.

#### Method 1

This method has an entrance effect because  $k_e$  is the average over the whole bed, and the  $h_w$  depends on the  $k_e$ . For Method 1, a sensitivity analysis based on computing  $d \ln h_w / d \ln k_e$  shows that errors in  $k_e$  give much larger errors in  $h_w$ , and in the opposite direction. For  $Bi = 2$  ( $Bi = 4$ ) the value of  $d \ln h_w / d \ln k_e$  is  $-1.3$  ( $-6.5$ ). A  $k_e$  that is 5% high due to the length effect gives a  $h_w$  in

Table 3. The  $h_w$  determined by method 3 vs bed depth

$z'_1$ (m)	$z'_2$ (m)	$h_w$ (kcal/m <sup>2</sup> hr °C)
0	1.016	146
0.284	1.016	137
0.582	1.016	121
0.875	1.016	122

Method 1 that is about 30% low when  $Bi = 4$ . Thus Method 1 is particularly susceptible to the length effect.

The  $k_e$  and  $h_w$  found by the different methods of data analysis are given in Table 4. We emphasize these differences are entirely due to the method of data analysis, since the simulated data were identical. As predicted, Method 2 gives the asymptotic  $h_w$ , Methods 3 and 4 give higher  $h_w$  and Method 1 gives a lower  $h_w$ . The  $h_w$  given by Method 2 is the least affected or is unaffected by length. It thus gives the best value of asymptotic heat transfer coefficient.

To further support the idea of length dependence, we plot in Fig. 3 data from various sources for spherical packing. The data analyzed with Method 2 (denoted by  $\Delta$  and  $\nabla$  in Fig. 3) give a Nusselt number generally lower than the other data which, because of the conditions of the experiments and method of data analysis are *predicted* to include a length dependence. The same thing holds true for cylindrical packing (Fig. 4) except now the data analyzed by Method 1 by Pogorski lies below the asymptotic coefficient, as predicted.

It is still unclear how long the bed should be for the local  $k_e$  and  $h_w$  to reach the asymptotic values. So far no such experimental results have been recorded. However, several experimental temperature profiles reported in the literature (see Fig. 5) indicate that whenever  $\alpha'z$  is greater than 0.2 the plot of  $\log T$  vs  $z$  is a straight line. This is consistent with the result derived above under the assumption of constant  $k_e$  and  $h_w$ : the temperature

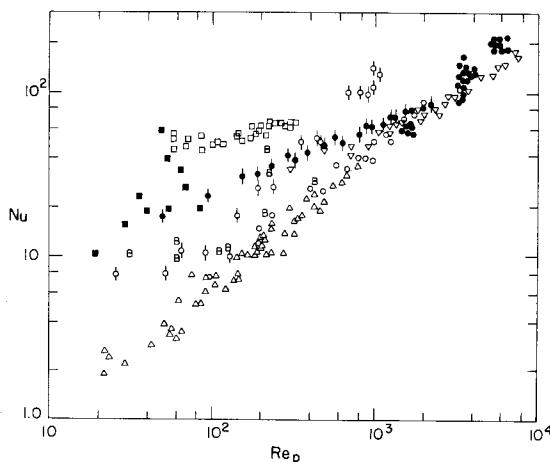


Fig. 3. Experimental data for wall heat transfer coefficient with spherical packing. ( $\Delta$  and  $\nabla$  determined by Method 2.)

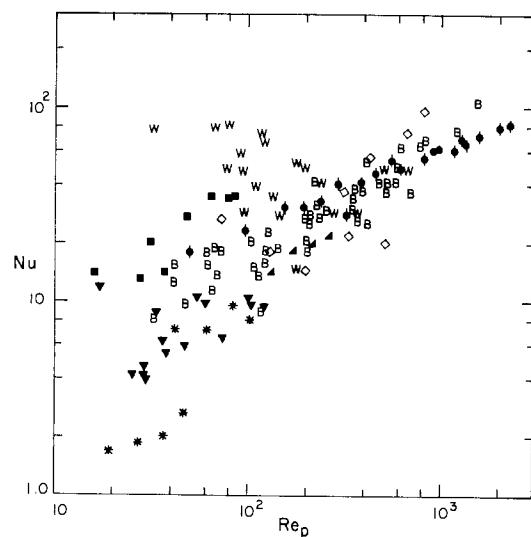


Fig. 4. Experimental data for wall heat transfer coefficient with cylindrical packing.

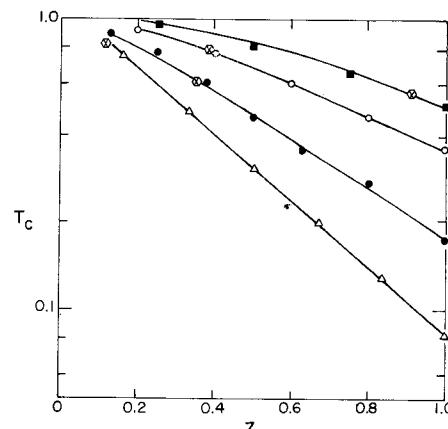


Fig. 5. Axial temperature profiles along the center of the bed ( $\otimes$  marks  $\alpha'z = 0.2$ ).

Symbol	Ref.	$\alpha'$	$Bi$	$Re_p$
■	[12]	0.22	94	
●	[4]	0.56	4.54	506
△	[7]	1.71	0.92	680
○	[6]	0.52	4.28	234

profile can be expressed by a one-term asymptotic solution when  $\alpha'$  is greater than 0.2, and a straight line results on a plotted  $\log T$  vs  $z$ . If the local  $k_e$  and  $h_w$  still change with bed depth for  $\alpha'$  greater than 0.2, the experimental

Table 4. Comparison of the  $k_e$  and  $h_w$  determined by the different calculation methods

Method	$k_e$ (kcal/m hr °C)	$h_w$ (kcal/m <sup>2</sup> hr °C)	Bi
1	1.23*	77.0	3.10
2	0.97	123	6.30
3 <sup>†</sup>	1.23*	137	5.53
4 <sup>++</sup>	1.12	146	6.42

\*Using  $k_e$  = the average of those in Table 2.

<sup>†</sup> $z_1' = 0.284\text{m}$ ,  $z_2' = 1.016\text{m}$

<sup>++</sup> $L = 1.016\text{m}$

Table 5. Key to the experimental data presented in Figs. 3, 4, 6, 9, 10

Symbol	Author	Ref.
◇	Coberly and Marshall	[4]
B	Felix	[7]
▲	DeWasch and Froment	[9]
*	Hashimoto <i>et al.</i>	[15]
●	Hawthorn <i>et al.</i>	[16]
▽	Kunii <i>et al.</i>	[17]
W	Maeda	[5]
▼	Phillips <i>et al.</i>	[23]
○	Plautz and Johnston	[18, 19]
●	Pogorski	[13, 14]
◇	Quinton and Storrow	[20]
■	Valstar	[8]
△	Yagi and Wakao	[6]
□	Ziolkowski	[21]

temperature profile would not yield a straight line. Based on this reasoning we use  $\alpha'$  greater than 0.2 as a criterion to determine when the entrance region does not affect the  $k_e$  and  $h_w$  determined by Method 1 and Method 3.

We next examine the effect of axial dispersion on  $k_e$  and  $h_w$ . If an axial dispersion term,  $-\gamma' \partial^2 T / \partial z^2$ , is added to the left hand side of eqn (2), the equation can be solved assuming  $\alpha'$ ,  $\gamma'$  and  $Bi$  are constant. If we use the solution for  $\alpha' > 0.2$ , when these assumptions are valid, we can determine the error in  $k_e$  and  $h_w$  assumed by analyzing the data ignoring axial dispersion. The detailed analyses are derived elsewhere[22] and show that for methods 1, 2 and 3 the data analysis gives the same result,

$$\frac{\Delta k_e}{k_e} = \frac{\Delta h_w}{h_w} = \frac{1 - a_1}{1 + a_1} \quad (9)$$

$$a_1^2 = 1 + 4\alpha' \gamma' A_1^2 \quad (10)$$

where  $\Delta k_e$  is the error in  $k_e$  due to the neglected axial dispersion effect. Phillips *et al.*[23], have proved eqn (9) for  $k_e$  when using Method 1. For  $a_1$  close to 1 (as assumed) eqn (9) can be further simplified to

$$\frac{\Delta k_e}{k_e} = \frac{\Delta h_w}{h_w} = -\alpha' \gamma' A_1^2. \quad (11)$$

Using the approximation from orthogonal collocation[24] that

$$A_1^2 \approx \frac{6Bi}{Bi + 3} \quad (12)$$

and  $Pe_{z,h} \approx 2$  gives

$$\frac{\Delta k_e}{k_e} = \frac{\Delta h_w}{h_w} = -\frac{1}{Pe_{z,h}} \left( \frac{d_p}{R} \right)^2 \frac{3Bi}{Bi + 3}. \quad (13)$$

This expression can be used to estimate the per cent error caused by axial dispersion. For Method 4 the result was derived by Young[25].

$$\frac{\Delta k_e}{k_e} = \frac{\Delta h_w}{h_w} = -\frac{1}{Pe_{z,h}} \left( \frac{d_p}{R} \right)^2 \frac{3Bi}{Bi + 3} + \frac{d_p}{2L}. \quad (14)$$

We see that axial dispersion causes errors in  $h_w$  measured by all methods, but only in the case of Method 4 are the errors dependent on length. Gunn and Khalid[10] found axial conduction important in their measurements, which were analyzed by Method 4. Equation (14) shows that the length of bed could influence the results. However, in the examination of the data described below, we use eqn (13) to estimate the importance of axial conduction, and seldom is data discarded because of axial conduction.

We next examine the effect of a non-flat temperature profile at the inlet to the bed. Felix[7] derived the solution for the following inlet profile:

$$T = 1 \quad 0 \leq r \leq r_i$$

$$T = T_i + br^2 \quad r_i \leq r \leq 1.$$

If we take  $r_i = 0$  and use this solution, we find that data analyzed by Methods 1, 2 and 3 are unaffected by this problem, provided  $\alpha' z \geq 0.2$ . Method 4 is affected, however, and the error is

$$\frac{\Delta \alpha'}{\alpha'} = \frac{b}{\alpha' z} \left( \frac{3 + Bi}{6Bi} \right) \left( 1 - \frac{4}{A_1^2} + \frac{2}{Bi} \right).$$

For  $b = 0.2$ ,  $\alpha' z = 0.2$  and  $Bi = 1$ , the  $\Delta \alpha' = 0.3\alpha'$ , giving significant error. For  $Bi = 5$  the error is reduced to  $\Delta \alpha' = 0.1\alpha'$ .

We see that the data for  $h_w$  is affected by the length of the bed, and different methods of analysis give different  $h_w$ . We do not have a well-justified explanation of the cause of the length effect, but several possibilities exist. Possibly the developing velocity and temperature profiles influence the results. In a packed bed the velocity profile would be developed within a few particle diameters of the inlet, due to the presence of the packing. In a non-isothermal bed, however, the axial velocity profile can change because of a changing temperature profile (see Schertz and Bischoff[26]), and, because of continuity, anytime the axial velocity profile changes a radial velocity is introduced. This might affect the wall heat transfer coefficient, especially since the radial flow would be more pronounced near the wall. None of these effects can at present be conclusively shown to be the cause of the "length effect", but are possibilities which are consistent with the data. The importance of differentiating between overall coefficients, applicable to the whole bed, and asymptotic coefficients, is, however, clearly established. Method 2 gives an asymptotic  $h_w$ , or at least the best estimate of it, compared to Methods 1, 3 and 4.

Finally we examine the crucial question: which  $k_e$  and  $h_w$  should be used for chemical reactor design? Should the  $h_w$  be one which fits the heat transfer data of the whole bed or only the one representing the asymptotic local heat transfer rate? Table 6 lists typical values of bed lengths, particle diameters and tube diameters for

Table 6. Design parameters for the packed bed reactors operated in the chemical industry

Reaction	L/R	$d_p/R$	$\alpha'$	Reference
Ethylene oxidation	190			
Methanol oxidation	70	0.04 - 0.25*	0.3-6	[2]
Vinylacetate oxidation	110			
Benzene hydrogenation	60			
Naphthalene oxidation	240	0.20	6	[2, 27]
Butane dehydrogenation	106	0.10	1.32	[28]
O-xylene oxidation	200	0.24	6	[29]
Methane reforming	126	0.30	4.7	[30]

\*estimated values

industrial reactors. The beds are so long that  $\alpha' \gg 0.2$  for most cases. This means that if a heat transfer experiment were done in such a bed the asymptotic heat transfer coefficient would be measured if the data were analyzed using any of the methods (within experimental error). To reproduce data in these long beds, the asymptotic heat transfer coefficients would have to be used. When heat effects due to chemical reaction are present, the shape of the temperature profile could be changed so that even the asymptotic value was inappropriate. However, in the absence of information on the effect of chemical reaction on heat transfer coefficient, the best we can do is use the asymptotic heat transfer coefficient.

The asymptotic heat transfer coefficient is lower than any average  $h_w$  for the entire bed. Thus a reactor design based on the asymptotic  $h_w$  is conservative: the actual reactor would be less likely to exhibit a "runaway" situation than the design would indicate.

#### EXAMINATION OF HEAT TRANSFER DATA

Experimental data was examined in detail to decide which data was influenced by length effects or axial conduction. In many cases the original thesis was examined, and in some cases the data was re-analyzed using Method 2. A complete tabulation is available[22]. We are interested in only the asymptotic heat transfer coefficient.

Data obtained by Maeda[5], Phillips *et al.*[23], Yagi and Wakao[6], Hashimoto *et al.*[15], and Kunii *et al.*[17], is accepted since the  $h_w$  and  $k_e$  were determined by Method 2. Data obtained by Felix[7] was analyzed by Method 3, and it is accepted if the testing section did not fall in the entrance region (i.e. if the region over which  $h_w$  is deduced is for  $\alpha'z_i \geq 0.2$ ).<sup>†</sup> Coberly and Marshall[4]

made measurements for  $\frac{1}{4}$  by  $\frac{1}{4}$  in. cylindrical packing, but plots of  $\log T$  vs  $z$  did not reach a straight line, so this data includes a length effect and is not used. Data for  $\frac{3}{8}$  by  $\frac{1}{2}$  in. cylindrical packing was originally analyzed by Method 1. Here it is re-analyzed by Method 2 to eliminate the length effect, and this recalculated data is used below. Hawthorn *et al.*[16], did experiments in very long beds ( $\alpha' > 7$ ) for constant wall heat flux. A mathematical model was used to calculate the radial temperature profile and the calculated temperature drop from the center of the bed to the wall was subtracted from the measured temperature drop from the center of the bed to the cooling medium. These numbers were compared, and the difference was ascribed to the temperature drop very near the wall, which is modeled by the heat transfer coefficient. Due to the length of bed the length effect is negligible. This is one of the few sources of heat transfer data derived from a chemically reacting system at high temperature. We re-analyzed Hawthorn's data using  $Pe_h = 8$  rather than 10, and the revised calculations are presented below (the effect of  $Pe_h$  is small).

Data for annular beds is reported by Yagi and Kunii[31], Baddour and Yoon[32], and Kunii and Suzuki[33]. A constant temperature difference was maintained between the inner and outer surfaces along the entire bed depth. Thus the temperature varied only radially and there is no length effect. The data, however, applies to an annular bed, and for large  $d_p/R$  the packing at the inner surface may be different from that of a cylindrical bed. Thus the data must be put in a separate classification.

Unfortunately a large number of data must be rejected because they include a length effect and thus do not give an asymptotic  $h_w$ . Valstar[8], De Wasch and Froment[9], and Ziolkowski[21] all used Method 4 to analyze their data and it thus is affected by the length. Campbell and Huntington[34, 35] used Method 1 to analyze a constant wall heat flux experiment, even though the Method assumes a constant wall temperature. In some experiments

<sup>†</sup>Felix's testing section usually began at  $z_i = 5$  in. We have applied the stringent criterion that  $\alpha'z_i \geq 0.2$ , but a less stringent criterion could be argued. Unfortunately we have no basis on which to decide, and so have used the number 0.2.

gas was cooled by the natural convection of air in the room, but no correction was made for the heat transfer resistance external to the pipe. In some cases the calculated  $h_w$  is less than the measured overall heat transfer coefficient, which is impossible physically. For these reasons the data by Campbell and Huntington is not included below. We note that their data for  $k_e$  is acceptable since it is determined by differentiating the temperature profile. Plautz and Johnston[18, 19] found  $h_w$  using Method 3 and  $k_e$  by making calculated temperature profiles agree with experimental ones over the whole bed. Both  $h_w$  and  $k_e$  thus include a length effect and are not used below. Calderbank and Pogorski[13, 14] used Method 1 but most of the beds are so short ( $\alpha' < 0.2$ ) that the experiment includes a length effect regardless of the method of data analysis. This data is not used below.

Aerov and Umnik[36] report data which is frequently[1, 2] reported as following the correlation:

$$\frac{h_w d_p}{k_f} = 0.155 \left( \frac{Gd_p}{\mu} \right)^{0.75} Pr^{1/3}. \quad (15)$$

The original Russian report, however, gives the correct correlation as

$$\frac{h_w (4d_p)}{k_f} \left( \frac{\epsilon}{1-\epsilon} \right) = 0.155 \left( \frac{4Gd_p}{6\epsilon(1-\epsilon)\mu} \right)^{0.75} Pr^{1/3} \quad (16)$$

eqn (16) is about 45% higher than Yagi and Wakao's data, whereas eqn (15) is 35% lower. Aerov and Umnik did not measure  $h_w$ ; they determined it from their measured  $k_e$  and correlations for the overall heat transfer coefficient (see below). Unfortunately these correlations include a length effect so that the data is not usable.

Quinton and Storrow[20] performed a careful experiment keeping the wall flux constant. The boundary condition in eqn (2) is replaced by

$$-\frac{\partial T}{\partial r} \Big|_{r=1} = \frac{F_0 R}{k_e T'_i} = Q \quad (17)$$

where now  $T = T'/T'_i$ .

The solution valid for  $\alpha'z > 0.15$

$$T = 1 + 2\alpha'Qz + Q \left( \frac{1}{2} r^2 - \frac{1}{4} \right). \quad (18)$$

The  $\alpha'$ , and hence  $k_e$ , can be found from

$$\alpha' = \frac{1}{4} \frac{\partial T / \partial z}{\partial T / \partial (r^2)}. \quad (19)$$

Quinton and Storrow used exit temperature profiles so that no entrance effect was incurred in finding  $k_e$ . Local overall and wall heat transfer coefficients can be defined as

$$U = \frac{F_0}{T'_w - T'_m}; \quad h_w = \frac{F_0}{T'_w - T'_R}. \quad (20)$$

Using eqn (18) gives

$$\frac{1}{U} = \frac{1}{h_w} + \frac{R}{4k_e}. \quad (21)$$

This relation, which is exact for  $\alpha'z > 0.15$ , was used to calculate  $h_w$  from the  $k_e$  and a  $U$  which was the arithmetic mean of the local overall heat transfer coefficients over the whole bed. Thus  $U$  included a length effect, and so did  $h_w$ . For one case the  $h_w$  so reported is about 25% higher than that calculated from the asymptotic  $U$ . Thus the data for  $h_w$  cannot be used.

#### CORRELATION OF DATA

The data which is accepted as being free from length effects is put to two additional tests. We require that the error due to axial dispersion effects be less than 5%, as calculated by eqn (13). We also discard data for  $Bi > 12$ , since then the temperature profile is insensitive to  $h_w$ , being determined primarily by  $k_e$ . For  $Bi > 12$ , less than 20% of the total thermal resistance exists at the wall, and an experimental error of  $\pm 10\%$  in the total rate of cooling leads to an error in  $h_w$  of  $\pm 50\%$ .

The data for  $h_w$  is correlated in terms of the Nusselt number. Spherical and cylindrical packing give different results and are correlated separately. The best correlation for spherical packings is

$$\frac{h_w d_p}{k_f} = 0.17 \left( \frac{Gd_p}{\mu} \right)^{0.79} \quad (22)$$

$0.05 \leq d_p/d_t \leq 0.3$

$20 \leq Re_p \leq 7600$

Constant wall temperature, spherical packing.

This correlation is compared to the experimental data in Fig. 6 and predicts the data with an average deviation of 14%, and a modified correlation coefficient of  $\bar{R}^2 = 0.98$  from the linear regression analysis. The  $\bar{R}^2$  represents the fraction of the variation of data which can be explained by the correlation. Other forms of correlations tried are listed in Table 7. The form of the correlation

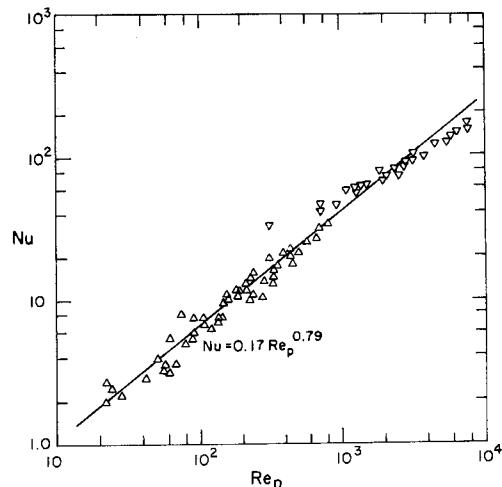


Fig. 6. Accepted data of the asymptotic wall heat transfer coefficient for spherical packing.

Table 7. Correlations tested for heat transfer coefficient

Correlation	$a^{\dagger}$	$b^{\dagger}$	average deviation %	$\bar{R}^2$
$h_w$ , Spherical Packing				
1. $Nu = a Re_p^b$	0.17	0.79	14	0.98
2. $Nu = a+b Re_p Pr$	10.7	0.033	73	0.94
3. $Nu_m = a Re_m^b$	0.029	0.94	21	0.97
4. $Nu = a(Re_p Pr)^{0.33} + b Re_p^{0.8} Pr^{0.4}$	1.33	0.14	38	0.98
$h_w$ , Cylindrical Packing				
5. $Nu = a Re_p^b$	0.16	0.93	33	0.85
6. $Nu = a+b Re_p Pr$	5.21	0.126	62	0.76
7. $Nu_m = a Re_m^b$	0.03	1.06	39	0.85
8. $Nu = a(Re_p Pr)^{0.33} + b Re_p^{0.8} Pr^{0.4}$	0.36	0.38	49	0.79
$U^*$ , Spherical Packing				
9. $U^* d_p / k_f = a+b Re_p Pr$	3.88	0.03	104	0.96
10. $U^* d_t / k_f = a Re_p^b$	7.13	0.47	17	0.92
11. $(U^* d_t / k_f) \exp(4.6 d_p / d_t) = a Re_p^b$	2.72	0.72	18	0.96
12. $(U^* d_t / k_f) \exp(6 d_p / d_t) = a Re_p^b$	2.03	0.80	21	0.96
$U^*$ , Cylindrical Packing				
13. $U^* d_p / k_f = a+b Re_p Pr$	0.76	0.07	63	0.87
14. $U^* d_t / k_f = a Re_p^b$	1.84	0.76	35	0.79
15. $(U^* d_t / k_f) \exp(4.6 d_p / d_t) = a Re_p^b$	1.39	0.90	27	0.88
16. $(U^* d_t / k_f) \exp(6 d_p / d_t) = a Re_p^b$	1.26	0.95	27	0.89

$\dagger$  a and b are found from a linear regression analysis.

preferred by Beek[1], form 4 in Table 7, is less successful in predicting the data, with an average deviation of 38%.

We note that all the experiments were done with air, so there is no Prandtl number dependence. A reasonable extension would be to regard the constant  $0.17 = 0.17 (Pr/0.7)^{1/3}$ , but this is not proved by these data. The correlation (17) is very close to that proposed by Yagi and Wakai[6], but here it correlates their data as well as that by Kunii *et al.*[17]. The data of Felix[7] and Plautz and Johnson[18, 19], which has a slight length dependence at high Reynolds numbers, is slightly above this correlation. At lower Reynolds numbers the Felix and Plautz data have more length dependence and the data is significantly above the correlation. Thus we see again that the reason for the large scatter in heat transfer data is partially due to a mixing of "overall" and asymptotic  $h_w$ .

The best correlation for cylindrical packings is

$$\frac{h_w d_p}{k_f} = 0.16 \left( \frac{G d_p}{\mu} \right)^{0.93}$$

$$20 \leq Re_p \leq 800 \quad (23)$$

$$0.03 \leq d_p/d_t \leq 0.2; \quad d_p = 6 V_p / S_p$$

Constant wall temperature, cylindrical packing.

This correlation gives an average deviation of 33% and  $\bar{R}^2 = 0.85$  and is compared to the data in Fig. 7. There is clearly more scatter in the data for cylindrical packings, and the heat transfer coefficients are larger.

For annular beds there is very little data available for the heat transfer coefficient on the inner wall. Yagi and

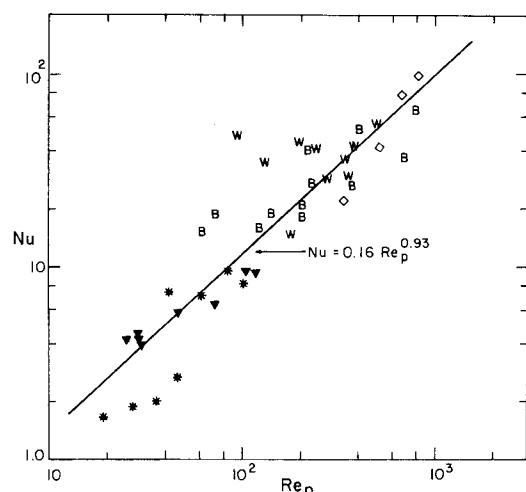


Fig. 7. Accepted data of the asymptotic wall heat transfer coefficient for cylindrical packing.

Kunii[31] correlate their data with the form

$$\frac{h_w d_p}{k_f} = a + 0.041 Re_p Pr \quad (24)$$

and "a" depends on the type of packing material.

The data of Hawthorn *et al.*[16] is the only accepted data for the boundary condition of constant wall flux. Furthermore the experiment has a high average temperature of 800°F and chemical reaction is taking place. This data is compared in Fig. 8 with the correlation for spherical packing, eqn (22). The constant flux  $Nu$  is about 20% higher than the value for constant wall temperature. We note that the  $h_w$  is not actually used in the boundary condition for constant wall flux, eqn (17).

As shown in eqn (2), the variables affecting the rate of heat transfer are not  $k_e$  and  $h_w$  separately, but in the combinations  $\alpha'$  and  $Bi$ . The Biot number, in particular, is the ratio of  $h_w$  to  $k_e/R$ . The data in Fig. 9 indicate that the Biot number decreases as the Reynolds number increases. Figure 10 shows data for the Biot number as a function of Reynolds number. We see that we can use

$$Bi \left( \frac{d_p}{R} \right) \left( \frac{\epsilon}{1-\epsilon} \right) = 0.27 \quad (25)$$

$$0.05 \leq d_p/d_t \leq 0.15$$

$$500 \leq Re_m \leq 6000$$

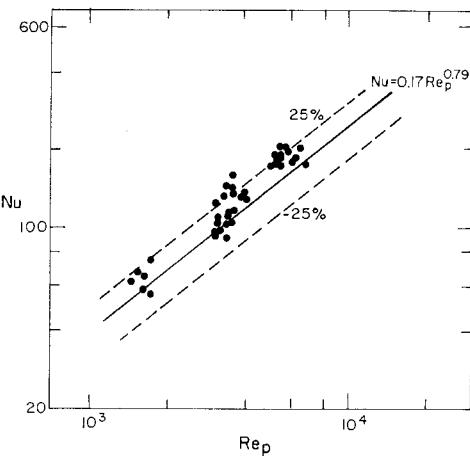


Fig. 8. The wall heat transfer coefficient by Hawthorn *et al.*[16].

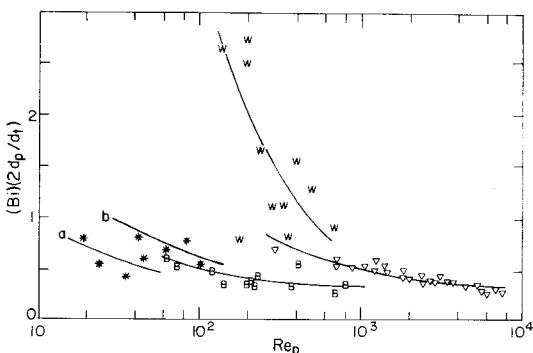


Fig. 9. Biot number vs Reynolds number. (a) Hydrogen; (b) nitrogen.

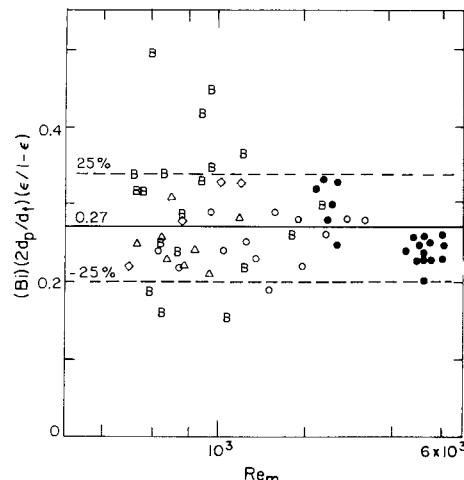


Fig. 10. Biot number vs Reynolds number.

which predicts the data with ±25%. The Biot number is the ratio of two experimentally measured quantities,  $k_e$  and  $h_w$ . If the overall heat transfer rate is measured accurately, but  $k_e$  is, say lower than it should be due to experimental error, then  $h_w$  is larger than it should be. Errors in  $Bi$  ( $= h_w R / k_e$ ) are thus magnified. Thus a scatter of the data of ±25% is not surprising. Some data in Fig. 8 is not included in Fig. 5 or 7; it is included in Fig. 8 because it is just at the limit of acceptability as to the length effect, and it provides the void fraction dependence, which is important. This is also the reason there is no void fraction dependence in the correlation (22): the accepted data did not cover a wide range of  $\epsilon$  values. Equation (25) is thus a new result which can be used along with  $Pe_h = 8-10$  for high Reynolds numbers. We note that a  $Bi$  constructed from  $k_e$  and  $h_w$  has a slight Reynolds number dependence, but at these high Reynolds numbers, the variation of  $Bi$  with  $Re$  is within the scatter of the data.

#### ONE DIMENSIONAL MODEL

The possibility of replacing a two-dimensional model, eqn (2), with a one-dimensional model, eqn (26), is well-known[3].

$$\frac{dT_m}{dz} = -\frac{2UL}{GC_p R} T_m \quad (26)$$

$$T_m = 1 \text{ at } z = 0.$$

The solution is

$$T_m = \exp(-2ULz/(GC_p R)). \quad (27)$$

There is a great scatter in the data for  $U$ , and some of this scatter we attribute to the often-ignored length effect.

The solution to the two-dimensional model, eqn (2), for  $\alpha'z \geq 0.2$  is

$$T_m(z) = \frac{4Bi^2}{A_1^2(A_1^2 + Bi^2)} \exp[-A_1^2 \alpha' z]. \quad (28)$$

Let us define an overall heat transfer coefficient  $\bar{U}$  such that the average temperature out of the packed bed is the same in the one- and two-dimensional models. Then

$$\bar{U} = -\frac{GC_p R}{2L} \ln T_m(1). \quad (29)$$

Putting eqn (28) into eqn (29) gives

$$\bar{U} = \frac{\bar{A}_1^2 \bar{k}_e}{d_t} + \frac{GC_p d_t}{4L} \ln \frac{\bar{A}_1^2 (\bar{A}_1^2 + \bar{B}i^2)}{4\bar{B}i^2}. \quad (30)$$

We have used an overbar on  $\bar{B}i$ ,  $\bar{A}_1$  and  $\bar{k}_e$  to denote the fact that they are determined by Method 4 to exactly reproduce the exit temperature profile. The second term is less than 5% of the first term when

$$\frac{20}{\bar{A}_1^2} \ln \frac{\bar{A}_1^2 (\bar{A}_1^2 + \bar{B}i^2)}{4\bar{B}i^2} \leq \alpha'_{\min} \quad (31)$$

and  $\alpha'_{\min}$  is listed in Table 8 as a function of  $Bi$ . Many packed beds used in experiments have  $\alpha'$  less than this value, so that the  $\bar{U}$  depends on the length of the bed directly, in the second term, as well as through the dependence on length of  $\bar{A}_1^2$  and  $\bar{k}_e$ .

Let us next define another overall heat transfer coefficient such that the asymptotic heat flux is the same in the one- and two-dimensional models.

$$U^*(T'_m - T'_w) = h_w^*(T'_R - T'_w). \quad (32)$$

Then we obtain

$$U^* = \frac{A_1^{*2} k_e^*}{d_t} \quad (33)$$

as derived by Crider and Foss[37]. Using the approximation eqn (12) enables this relation to be expressed as

$$\frac{1}{U^*} = \frac{1}{h_w^*} + \frac{R}{3k_e^*} \quad (34)$$

as derived by Finlayson[24]. Here  $k_e^*$ ,  $h_w^*$  and  $U^*$  are asymptotic values at large  $z$ , such as those derived using Method 2. Equation (34) is the best approximate equation for one-dimensional model only when the wall temperature is kept constant. If the wall heat flux is fixed instead of the temperature, the exact relationship, eqn (21), should be used.

Table 8.  $\alpha'_{\min}$  vs  $Bi$  for one-dimensional model

Bi	$\alpha'_{\min}$
0.1	0.0385
0.5	0.1053
1.0	0.2011
3.0	0.5053
5.0	0.6910
10.0	0.9191

The difference of  $A_1^2 k_e / d_t$  in eqn (30, 33) is due to the fact that  $A_1^2 k_e$  depends on length. The additional term in eqn (30), however, is the effect of the temperature profile being different at different positions, and this effect is less important in long beds, but is usually important in experimental studies.

To illustrate the difference in  $\bar{U}$  and  $U^*$ , consider the example in Table 2. There we have

$\bar{k}_e = 1.12$ kcal/m hr°C	$k_e^* = 0.97$
$\bar{h}_w = 146$ kcal/m <sup>2</sup> hr°C	$h_w^* = 123$
$\bar{A}_1^2 = 4.28$	$A_1^{*2} = 4.24$
$\bar{B}i = 6.42$	$Bi^* = 6.30$
$\bar{U} = 53.2$ kcal/m <sup>2</sup> hr°C	$U^* = 41.5$

The difference between  $\bar{U}$  and  $U^*$  is more than 25% and is due to the length effect. Data summarized in Fig. 11 show that  $\bar{U}$  decreases with increasing bed depth and is always greater than  $U^*$ . Furthermore experimental data for  $\bar{U}$  from Maeda[5], Leva[38], and Vereshoor and Schuit[39] included a length effect and are above data for  $U^*$  given by Yagi and Wakao[6], as expected. Froment[29] noted the  $U$  measured by Yagi and Wakao is lower but gave no reason.

A new set of data for  $U^*$  is obtained by using the data

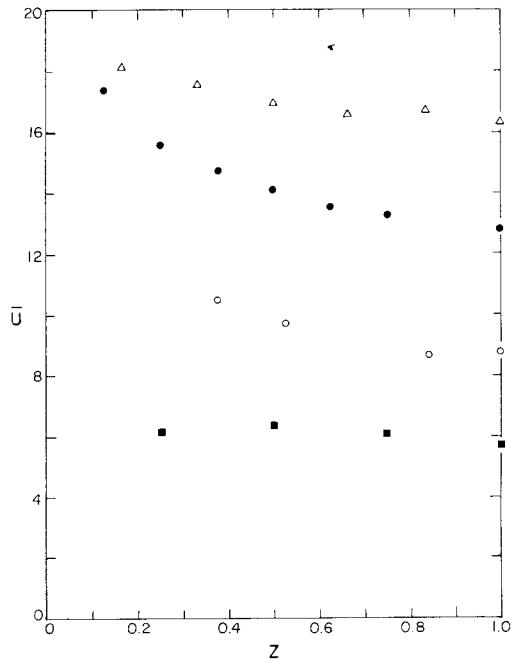


Fig. 11. The overall heat transfer coefficient vs bed depth.

Symbol	Ref.	$Ld_p/R^2$	$\alpha'$	$Re_p$	$\bar{U}_{z=1.0}^*$	$U^*$
■	[1]	1.00	0.28	48	5.63	5.45
●	[4]	2.62	0.56	506	12.83	11.56
△	[7]	5.46	1.71	680	16.37	15.71
○	[9]	2.32		198 <sup>‡</sup>	8.78	6.93

<sup>†</sup>Units of  $U$  are Btu/ft<sup>2</sup> hr°F.

<sup>‡</sup>This data is obtained through private communication. The Reynolds number, 198, is the average value of the Reynolds numbers for four different bed depths, which are 198, 201, 199 and 195.

for  $h^*$  and  $k^*$  in eqn (34). The best correlations are then

$$\frac{Ud_t}{k_f} \exp(6d_p/d_t) = 2.03 Re_p^{0.8} \quad (35)$$

$$20 \leq Re_p \leq 7600$$

$$0.05 \leq d_p/d_t \leq 0.3$$

Spherical packing

and

$$\frac{Ud_t}{k_f} \exp(6d_p/d_t) = 1.26 Re_p^{0.95} \quad (36)$$

$$20 \leq Re_p \leq 800$$

$$0.03 \leq d_p/d_t \leq 0.2$$

Cylindrical packing.

Other correlations tried and the per cent deviation are listed in Table 7.

#### CONCLUSIONS

For both one- and two-dimensional models, the heat transfer coefficients vary along the length of a packed bed. The values need for reactor design are the asymptotic values, which are not affected by the length effect. Analysis by Method 2 gives an asymptotic heat transfer coefficient provided  $\alpha'z \geq 0.2$ . The best correlations of data for asymptotic values are given by eqns (22), (23), (35) and (36).

The Biot number is approximately a constant as the Reynolds number is increased, taking the value given by eqn (25).

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#### NOTATION

$A_n$  eigenvalue of the equation,  $A_n J_1(A_n) = Bi J_0(A_n)$   
 $Bi$  Biot number  $h_w R/k_e$   
 $C_p$  fluid heat capacity  
 $d_p$  particle diameter  
 $d_t$  tube diameter  
 $F_0$  wall heat flux  
 $G$  mass flow rate based on the area of empty tube  
 $h_w$  wall heat transfer coefficient  
 $J_n$   $n$ th order Bessel function of the first kind  
 $k_e$  radial effective thermal conductivity  
 $k_f$  fluid thermal conductivity  
 $k_z$  axial effective thermal conductivity  
 $L$  length of packed bed  
 $Nu$  Nusselt number,  $h_w d_p/k_f$   
 $Nu_m$  modified Nusselt number,  $(h_w d_p/k_f)(\epsilon/1-\epsilon)$   
 $Pe_h$  radial Peclet number,  $C_p G d_p/k_e$   
 $Pe_{z,h}$  axial Peclet number,  $C_p G d_p/k_z$   
 $Pr$  Prandtl number,  $C_p \mu/k_f$   
 $r'$  radial coordinate in bed

$r$  dimensionless radial coordinate,  $r'/R$   
 $R$  radius of bed  
 $Re_m$  modified Reynolds number,  $G d_p / [\mu(1-\epsilon)]$   
 $Re_p$  Reynolds number,  $G d_p / \mu$   
 $S_p$  surface area of packing particle  
 $T'$  temperature  
 $T$  dimensionless temperature,  $(T' - T_w)/(T_i' - T_w)$   
 $T_c$  dimensionless center temperature  
 $T_i'$  inlet temperature  
 $T_m'$  mean temperature  
 $T_m$  dimensionless mean temperature  
 $T_R'$  fluid temperature at the wall  
 $T_w'$  coolant temperature  
 $U$  overall heat transfer coefficient  
 $V_p$  volume of packing particle  
 $z'$  axial coordinate in bed  
 $z$  dimensionless axial coordinate

#### Greek symbols

$\alpha'$   $L d_p (R^2 Pe_h)$   
 $\gamma'$   $k_e / (L G C_p) = (d_p / L) / Pe_{z,h}$   
 $\mu$  viscosity  
 $\epsilon$  void fraction

#### Superscripts

— axial average  
\* local value

#### REFERENCES

- [1] Beek J., *Adv. Chem. Engng* 1962 **3** 223.
- [2] Hlavacek V., *Ind. Engng Chem.* 1970 **62**(7) 8.
- [3] Froment G. F., *Chem. Reaction Engng Adv. Chem. Ser.* 109 1972 1-34.
- [4] Coberly C. A. and Marshall W. R., *Chem. Engng Prog.* 1951 **47** 141.
- [5] Maeda S., *Tech. Rep. Tokoku Univ.* 1952 **16** 1.
- [6] Yagi S. and Wakao N., *A.I.Ch.E.J.* 1959 **5** 79.
- [7] Felix T. R., Ph.D. Thesis, University of Wisconsin (1951).
- [8] Valstar J. M., Ph.D. Thesis, Delft, Netherlands (1969).
- [9] DeWasch A. P. and Froment G. F., *Chem. Engng Sci.* 1972 **27** 567.
- [10] Gunn D. J. and Khalid M., *Chem. Engng Sci.* 1975 **30** 261.
- [11] Paterson W. R., Ph.D. Thesis, University of Edinburgh (1975).
- [12] Bunnel D. G., Irvin H. B., Olson R. W. and Smith J. J., *Ind. Engng Chem.* 1949 **41** 1977.
- [13] Calderbank P. H. and Pogorski L. A., *Trans. Instn. Chem. Engrs.* 1957 **35** 195.
- [14] Pogorski L. A., Ph.D. Thesis, University of Toronto (1957).
- [15] Hashimoto K., Suzuki N., Teramoto M. and Nagata S., *Kagaku Kogaku* 1966 **4** 68.
- [16] Hawthorn R. D., Ackerman G. H. and Nixon A. C., *A.I.Ch.E.J.* 1968 **14** 69.
- [17] Kunii D., Suzuki M. and Ono N., *J. Chem. Engng Japan* 1968 **1** 21.
- [18] Plautz D. A. and Johnston H. F., *A.I.Ch.E.J.* 1955 **1** 193.
- [19] Plautz D. A., Ph.D. Thesis, University of Illinois (1953).
- [20] Quinton J. H. and Storrow J. A., *Chem. Engng Sci.* 1956 **6** 245.
- [21] Ziolkowski D., *Bull. Acad. Pol. Sci., Ser. Sci. Chim.* 1970 **18**(4) 221.
- [22] Li C. H., M.S. Thesis, University of Washington (1974).
- [23] Phillips B. D., Leavitt F. W. and Yoon C. Y., *Chem. Engng Prog. Symp. Ser.* 1960 **56**(30) 219.
- [24] Finlayson B. A., *Chem. Engng Sci.* 1971 **26** 1081.
- [25] Young L. C., Private communication 1975.

- [26] Schertz W. W. and Bischoff, *A.I.Ch.E.J.* 1969 **15** 597.
- [27] Carberry J. J. and White D., *Ind. Engng Chem.* 1969 **61**(7) 35.
- [28] Hougen O. A. and Watson K. M., *Chemical Process Principles*, Vol. 3, 1st Edn, p. 1007. John Wiley, New York 1949.
- [29] Froment G. F., *Ind. Engng Chem.* 1967 **59**(2) 18.
- [30] Golebiowski A. and Wasala T., *Intern. Chem. Engng* 1973 **13**(1) 133.
- [31] Yagi S. and Kunii D., *A.I.Ch.E.J.* 1960 **6** 97.
- [32] Baddour R. F. and Yoon C. Y., *Chem. Engng Prog. Symp. Ser.* 1961 **57**(32) 35.
- [33] Kunii D. and Suzuki M., *Intern. Heat Trans. Conf.* 1966 **V** 344.
- [34] Campbell J. M. and Huntington R. L., *Petrol. Ref.* 1952 **31** 123.
- [35] Campbell J. M., Ph.D. Thesis, University of Oklahoma (1951).
- [36] Aerov M. E. and Umnik N. N., *J. Tech. Phys.* 1951 **21** 1351.
- [37] Crider J. E. and Foss A. S., *A.I.Ch.E.J.* 1965 **11** 102.
- [38] Leva M., Weintraub M., Grummer M. and Clark E. L., *Ind. Engng Chem.* 1948 **40** 747.
- [39] Verschoor H. and Schuit G., *Appl. Sci. Res.* 1952 **A2** 97.