Studies on Heat Transfer Near Wall Surface in Packed Beds

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From studies of annular packed beds wherein the heat flows purely radially, the authors obtained the coefficients of heat transfer on the inner tube surface, as well as the mean effective thermal conductivities of bed. The inner and outer diameters of the annular packed bed were 22 and 70 mm. respectively, and the packings shown in Table 1 were used. The wall film coefficients obtained with air flowing axially through the bed were correlated for $N_{ReM} < 600$ by means of Equation (14).

The coefficients of heat transfer for cylindrical packed beds reported previously by other observers were correlated also by Equation (14), with 0.054 used for values of α_w in the range $N_{ReM} < 2{,}000$.

Consideration of Equation (14) in terms of a theoretical model of heat transfer showed that it was reasonable to apply it for the prediction of wall film coefficient, especially for low Reynolds numbers.

In the development of the analytical methods for the design of catalytic reactors many measurements have been made of effective thermal conductivities in cylindrical packed beds. Hatta and Maeda (*6*) analyzed their experimental data of heat transfer in packed beds with their theoretical formula, which includes the wall film coefficient of heat transfer.

The coefficient h_w, which is important for the heat transfer mechanism in packed beds, has been measured by Coberly and Marshall (*3*), Campbell and Huntington (*2*), Felix (*4*), Plautz and Johnstone (*8*), Quinton and Storrow (*9*), and Calderbank and Pogorski (*1*).

With respect to the usual packed beds of cylindrical packings Hanratty (*5*) has presented the following equation which is based on the correlation for the range $D_pG/\epsilon\mu$ of 80 to 500:

$$\frac{h_w D_p}{k_g} = 0.95 \sim 1.44\left(\frac{D_p G}{\epsilon\mu}\right)^{0.5} \qquad (1)$$

Yagi and Wakao (*13*) measured the coefficients for beds of spherical packings and correlated their data with those of Felix (*4*) and Plautz and Johnstone (*8*) by

$$\frac{h_w D_p}{k_g} = 0.18\left(\frac{D_p G}{\mu}\right)^{0.8} \qquad (2)$$

for the range $D_pG/\mu = 20 \sim 2{,}000$

Equations (1) and (2) for the wall film coefficient of heat transfer cannot be used for flow conditions of small Reynolds numbers, as they make h_wD_p/k_g become zero when $N_{ReM} = 0$.

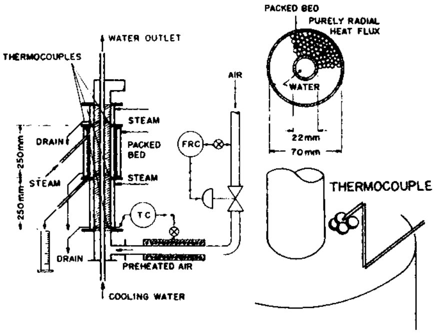

Fig. 1. Experimental annular packed bed and arrangement of copper-constantan thermocouples.

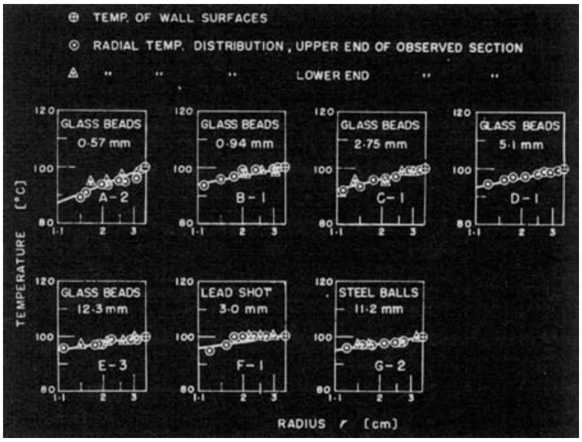

Fig. 2. Radial temperature distribution in annular packed bed, in blank tests without water flow.

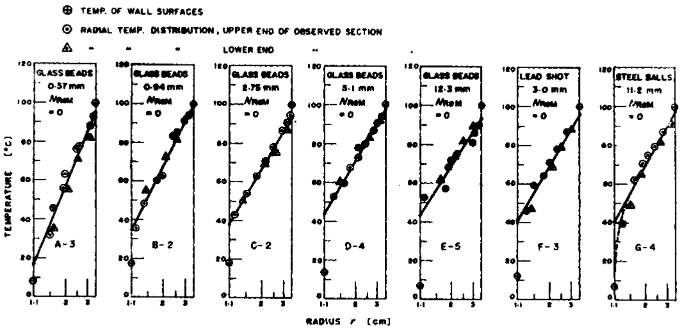

Fig. 3. Radial temperature distribution in annular packed beds without air flow; that is $N_{ReM} = 0$.

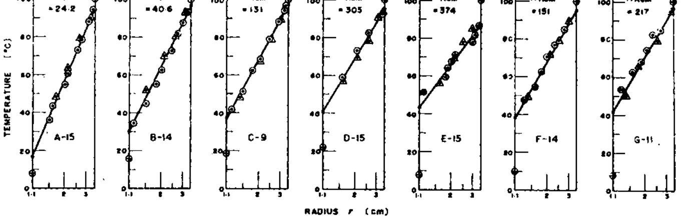

Fig. 4. Radial temperature distribution in annular packed beds with air flowing; $N_{ReM} > 0$.

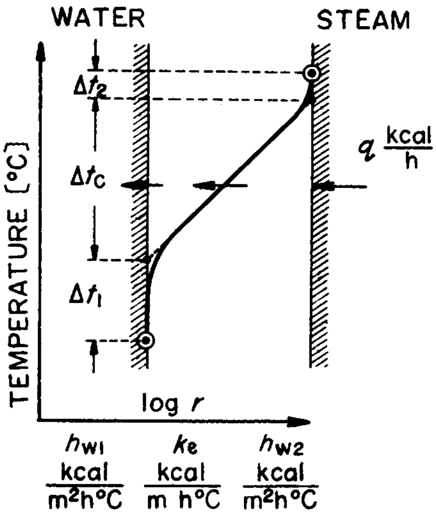

Fig. 5. Model of heat transfer system in annular packed bed.

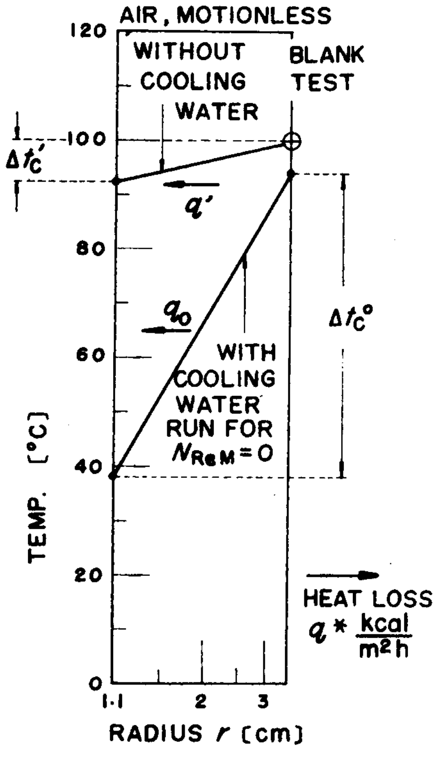

Fig. 6. Schematic illustrations of temperature distributions in annular packed bed, in both cases of blank test and of run with stationary air.

Catalytic reactors in practice have flow conditions corresponding to small Reynolds numbers, due to small catalyst diameters and low linear velocities. Therefore it is necessary for reasonable design calculations of catalytic reactors to obtain correct values of h_w for small Reynolds numbers and further to know the heat transfer mechanisms near the wall surface of packed beds.

However it is impossible to obtain the reliable data for h_w at small Reynolds numbers if the experimental method applied is similar to the previous studies. In these studies the flowing gas was heated or cooled in cylindrical packed beds, and the radial temperature distributions approached the constant wall temperature for small Reynolds numbers. This leads to inaccurate measurement. Hence the authors have made an experimental heater with an annular packed bed, wherein the heat flows purely radially, and have obtained new data for the wall film coefficient of heat transfer on the inner tube surface and mean effective thermal conductivity of the annular packed beds. The results for the wall film coefficient have provided the new correlations which have been compared with the previous experimental data from cylindrical packed beds.

EXPERIMENTAL METHODS

A diagram of the experimental packed bed is shown in Figure 1. The main part was constructed from three steam jackets and a steel pipe cooled by water, with solid particles packed in the annular space between them. The preheated air, the temperature of which is kept constant by a temperature controller, is blown upward into the bed, after being controlled at constant flow rate by a flow-recording controller. The packed bed is kept motionless by the two sheets of screen clamped on the bottom and top of the bed. The test section, the middle steam jacket, is covered by another steam jacket for the thermal insulation. Since the steam jacket of the test section was kept at atmospheric pressure by releasing the excess steam to the air, the saturated temperature of the steam was measured as 100°C. The heat transferred from the steam jacket to the cooling water was calculated from the measured rate of steam condensation, with the heat loss as obtained from preliminary blank tests taken into account. The radial temperature distributions were measured with fine and bare copper-constantan thermocouples arranged in the packed bed, (Figure 1).

To get purely radial heat flux a long calming section of the same packed bed was added below the measuring section, and the inlet air temperature was controlled to be nearly equal to the mean temperature of the outlet air. The calming section proved adequate in ensuring that radial temperature distributions were identical at inlet and outlet planes of the test section. With these conditions the experimental data could be analyzed on the basis that the air was neither heated nor cooled in flowing through any position in the test section of the bed.

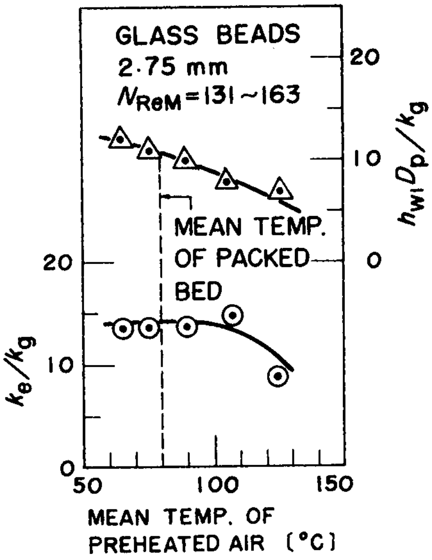

Fig. 7. Effects of preheated air temperature.

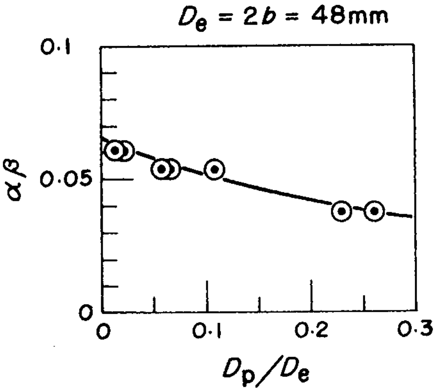

Fig. 8. Factor ($\alpha\beta$) in annular packed beds used.

Before final measurements were made, blank tests were done without cooling water to measure the heat losses in each packed bed. Figures 2, 3, and 4 show the radial temperature distributions for the blank tests without water flow, for runs without air flow (that is $N_{ReM} = 0$). Table for runs with air flow ($N_{ReM} > 0$). Table 2 includes some examples of the experimental data, from series A to G.*

EXPERIMENTAL RESULTS

In the annular packed bed with the heat flowing purely radially, theoretically the temperature distributions

TABLE 1. PACKINGS USED IN EXPERIMENTS

	Glass beads and balls					Lead shot	Steel balls
Diameter, (mm.)	0.57	0.94	2.75	5.1	12.3	3.0	11.2
Density, (g./cc.)	2.48	2.53	2.47	2.50	2.52	10.9	7.98
Void fraction (—)	0.355	0.372	0.352	0.375	0.427	0.382	0.461

* Tabular material has been deposited as document No. 6136 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or for 35-mm. microfilm.

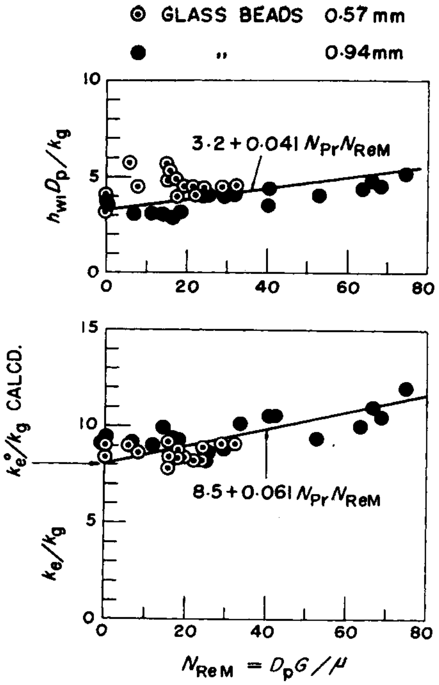

Fig. 9. Data obtained from annular packed beds, glass beads: 0.57-mm., 0.94-mm. diameter.

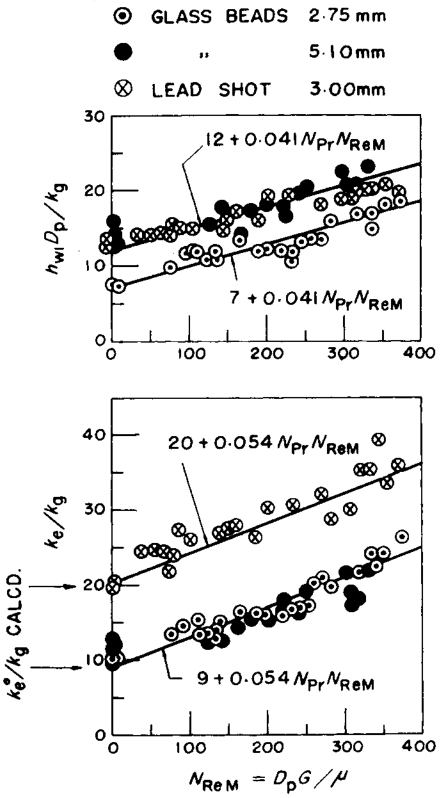

Fig. 10. Data obtained from annular packed beds, glass beads: 2.75-mm., 5.10-mm. diameter; lead shot: 3.0-mm. diameter.

should be straight on the semilogarithmic coordinates, as shown in Figure 5, if the effective thermal conductivity is assumed constant in the packed bed. From the results shown in Figures 3 and 4, it seems reasonable to think that there are thermal resistances on both the external and internal wall surfaces. The data were analyzed by applying the following equations, based on the typical model shown in Figure 5.

$$q = A_{w1} \cdot h_{w1} \cdot \Delta t_1 = A_{avg} \cdot (k_e/b)\ \Delta t_c = A_{w2} \cdot h_{w2} \cdot \Delta t_2 \qquad (3)$$

where

$$A_{avg} = (A_{w2} - A_{w1})/2.3 \log_{10}(A_{w2}/A_{w1})$$

$$A_{w1} = \pi(0.022)(0.250)m^2 = 0.01728\ m^2$$

$$A_{w2} = \pi(0.070)(0.250)m^2 = 0.05498\ m^2$$

$$A_{avg} = 0.03254\ m^2$$

$$b = (D_2 - D_1)/2 = (0.070\ m - 0.022\ m)/2 = 0.024\ m$$

When one substitutes the above values into Equation (3), the effective thermal conductivity and the wall film coefficient of heat transfer on the inner, and outer surfaces can be calculated from the following equations respectively:

$$k_e = 0.0740\ (q/\Delta t_c) \qquad (4)$$

$$h_{w1}' = 58.0\ (q/\Delta t_1) \qquad (5)$$

$$h_{w2} = 18.2\ (q/\Delta t_2) \qquad (6)$$

The values of the radial heat flow rate are evaluated from the rates of water-vapor condensation as follows:

$$q = V\gamma - q^* \qquad (7)$$

In the blank test for finding the heat loss from the test section the rate of water-vapor condensation would give the heat loss immediately, if the temperature at all points of the packed bed equals the wall surface temperature. Since there were small deviations of the radial temperature distribution for the blank tests as shown in Figure 2, the actual outward heat loss from the test section must be corrected. In Figure 6 the values for the blank test and for the run with stationary air are shown schematically.

In the blank test

$$q^* + q' = \gamma V' \qquad (8)$$

For the runs with the stationary air and with the cooling water

$$q^* + q_o = \gamma V_o \qquad (9)$$

The effective thermal conductivity for both cases should be the same; therefore

$$q'/q_o = \Delta t'_c/\Delta t^\circ_c \qquad (10)$$

Using Equations (8), (9) and (10), one can easily obtain the value of q^* from

$$q^* = \gamma \frac{V' - (\Delta t'_c/\Delta t^\circ_c)V_o}{1 - (\Delta t'_c/\Delta t^\circ_t)} \qquad (11)$$

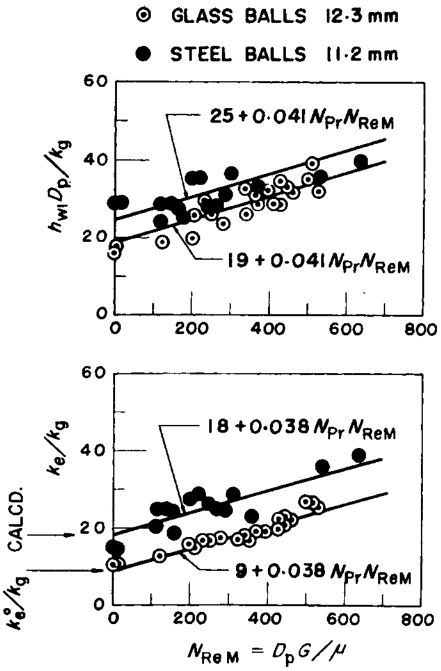

Fig. 11. Data obtained from annular packed beds, glass balls: 12.3-mm. diameter; steel balls: 11.2-mm. diameter.

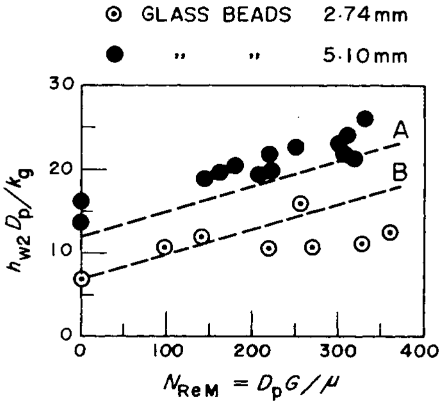

Fig. 12. Wall film coefficient on outer-tube surface in annular packed beds.

The following is a sample of the calculation for 2.75-mm. glass beads in Table 2, series C:

$$V' = 30.5\ \text{g./hr.} = (30.5)10^{-3}\ \text{kg./hr.}$$

$$V_o = 63.5\ \text{g./hr.} = (63.5)10^{-3}\ \text{kg./hr.}$$

$$\Delta t'_c = 6.5°C.$$

$$\Delta t^\circ_c = 56.0°C.$$

$$\gamma = 539.3\ \text{kg.-cal./kg.}$$

$$q' = (539.3)\frac{(30.5)10^{-3} - (6.5/56.0)(63.5)10^{-3}}{1 - (6.5/56.0)} = 14.1\ \text{kg.-cal./hr.}$$

TABLE 2. EXAMPLES OF EXPERIMENTAL DATA

Experiment No.	Packing	Diam., mm.	Inlet air Flow rate NTP, cc./sec.	Inlet air Mean temp., °C.	Bed mean temp., °C.	Vapor condensation rate, g./sec.	Cooling water flow rate, g./sec.	Cooling water mean temp., °C.	Reynolds Number	Temp. Differential Δt_c, °C.	Temp. Differential Δt_1, °C.	k_e kg.-cal./(m.)(hr.)(°C.)	h_{w1} kg.-cal./(sq. m.)(hr.)(°C.)	k_e/k_g	$h_{w1} D_p/k_g$
A-2	Glass	0.57	0	—	97	37.6	0	—	—	14	—		Blank test for heat loss		
4	beads		0	—	76	74.4	321	7	0	83	12	0.216	121	8.6	3.2
6			760	85	76	74.0	316	8	7.8	80	9	0.221	170	8.6	4.5
11			1720	85	75	74.0	316	8	17.6	81	8	0.221	185	8.6	4.9
17			3160	85	75	76.0	262	8	32.4	79	9	0.237	170	9.2	4.5
B-1	Glass	0.94	0	—	98.5	29.3	0	—	—	6	—		Blank test for heat loss		
3	beads		0	—	83	66.0	331	17	0	67	15.5	0.237	82.5	9.22	3.52
6			850	75	82.5	67.6	310	16	14.3	65	19	0.254	70.0	9.88	3.00
12			1900	79	80	69.0	310	17	32.2	65.6	14.5	0.261	95.5	10.2	4.08
19			4450	63	80	81.0	310	16	75.3	70.5	12.5	0.310	142	12.0	5.13
C-1	Glass	2.75	0	—	98	30.5	0	—	—	6.5	—		Blank test for heat loss		
3	beads		0	—	81	63.5	363	19	0	56	19.5	0.266	61.0	10.4	7.62
4			1500	80	81	75.2	297	22	74	56.5	19	0.346	82.7	13.5	10.3
9			2650	90	81	77.3	215	19	131	59.5	19	0.338	85.5	13.2	10.7
20			5400	75	78	103	286	15	266	56	21	0.548	107	21.3	13.4
26			7550	75	78	122	286	15	372	56.5	21.5	0.676	145	26.4	18.1
D-1	Glass	5.1	0	—	98.5	33.0	0	—	—	6.5	—		Blank test for heat loss		
5	beads		0	—	83	74.0	286	14	0	56	28	0.334	55.4	13.0	12.8
9			1950	79	80	84.2	271	13.5	178	57.5	24	0.395	75	15.4	17.6
13			2400	79	80	89.0	271	13.5	236	59	23	0.417	85.9	16.2	19.8
19			3650	65	80	108	245	13.5	332	57.5	26	0.560	99.5	21.8	23.0
E-1	Glass	12.3	0	—	98.5	37.2	0	—	—	4	—		Blank test for heat loss		
5	balls		0	—	81	66.4	335	7	0	48	6	0.285	28.4	11.0	15.8
6			530	85	78	70.8	366	8	116	45	36	0.343	34.0	13.3	18.9
10			1130	85	75	84.4	352	7	249	48	31	0.434	50.9	16.9	28.3
19			1930	80	77	96.3	361	7	425	49	33	0.603	61.9	23.5	34.6
24			2400	85	78	101.8	366	8	528	42	38	0.661	57.9	25.7	32.2
F-2	Lead	3.0	0	—	99.5	36.6	0	—	—	2.5	—		Blank test for heat loss		
5	shot		0	—	81.5	110.8	233	11.5	0	60.5	27	0.511	92.1	19.9	12.5
6			670	80	83.0	125	229	10.5	36.1	58	29.5	0.630	100	24.5	13.6
15			3000	80	81.5	140	311	11	161.5	59	27.5	0.720	124	28.0	16.9
21			5650	85	79.0	145.2	211	10	304	57.5	26	0.777	135	30.2	18.5
25			6550	85	78.5	160.6	273	10	352	59	26.5	0.860	151	33.5	20.6
G-1	Steel	11.2	0	—	99	33.6	0	—	—	5	—		Blank test for heat loss		
4	balls		0	—	84	80.6	393	7	0	56	29	0.371	57.0	14.8	29.1
5			520	85	85	84.8	333	8	115	54	37	0.490	56.6	25.8	28.9
11			1080	85	84	95.0	333	8	217	56	34	0.548	70.6	29.7	36.0
18			3150	80	83	125.6	290	7	634	42	44	1.015	78.3	39.5	39.9

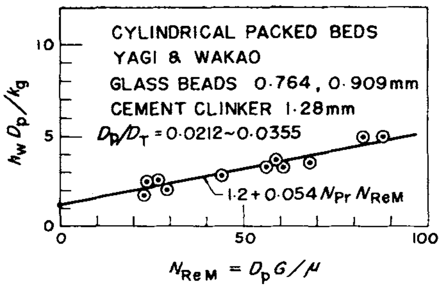

Fig. 13. Correlation of data observed by Yagi and Wakao (*13*) in cylindrical packed beds.

This value was used to calculate q for all of the data in series C of Table 2.

The linear velocity of the flowing water was greater than 1 m./sec., and the temperature rise of the water was less than 1°C. through the packed bed; therefore the mean temperature of water was taken as the temperature of the heat sink as shown in Figures 2, 3, and 4. The resistances for thermal conduction through the metal walls are negligible compared with those of the wall film in the packed bed. However the resistance of boundary film of the cooling water must be considered, even though the necessary corrections are a few per cent for every case. The heat transfer coefficient of the cooling water under the conditions in the experiments was estimated to be 3,670 kg.-cal./(sq.m.)(hr.)(°C.) from the Dittus-Boelter equation. A corrected value for the wall film coefficient of heat transfer on the inner tube surface was calculated from

$$\frac{1}{h_{w1}} = \frac{1}{h_{w1}'} - \frac{1}{3{,}670} \qquad (12)$$

The values obtained in this way from series A to G are shown in Table 2 and correlated in Figures 9, 10, and 11, where the data for the wall film coefficient of heat transfer on the outer surface are omitted.

For the over-all heat transfer system in the annular packed bed shown in Figure 5, the temperature difference becomes too small to determine h_{w2} accurately, because both the surface area and the thermal conductivity of the air for the outer tube surface are larger than those of the inner tube surface, the ratio of their effects being $A_{w1}\cdot k_{g1}/A_{w2}\cdot k_{g2} = 0.247$.

Even though the data for Δt_2 are thus not so reliable as those for Δt_1, the examples of calculated h_{w2} are shown in Figure 12, where the two lines A and B are the correlations for $h_{w1}\cdot D_p/k_g$ on the inner tube surface for the same packings respectively.

To indicate the deviations of the observed value from the correct one when there is considerable difference between the temperatures of inlet and outlet air, the effects of the temperature of preheated air are shown in Figure 7. Since the mean temperature of the annular packed bed, which is the same as that of the outlet air, is about 80°C. for the runs in Figure 7, the correct values of k_e/k_g and $h_{w1}\cdot D_p/k_g$ should be the values corresponding to 80°C. on the abscissa. The deviations from the correct data do not exceed 10% if the inlet temperature is kept within 80° ± 15°C.

DISCUSSION

As for the values in the common cylindrical packed beds, the data of k_e/k_g obtained here are correlated by the following equation, which was described theoretically in a previous paper (*12*):

$$\frac{k_e}{k_g} = \frac{k_e^\circ}{k_g} + (\alpha\beta) N_{Pr} N_{ReM} \qquad (13)$$

The values of $(\alpha\beta)$ are correlated with the ratio (D_p/D_e) in Figure 8. Values of k°_e/k_g calculated by Equation (15) of the authors' previous paper (*12*) nearly coincide with the experimental data as shown in Figures 9, 10, and 11 of this paper. The effects of the thermal conductivity of the packings seem to be considerable, especially under flow conditions of low Reynolds number.

Figure 3 shows the existence of the thermal resistances at the tube surface, or the wall film coefficients of heat transfer, when $N_{ReM} = 0$. In Figure 3 the temperature differences on both tube surfaces are too small to get the accurate data in the case of 0.57-mm. glass beads. On the other hand for large ratios of the packing diameter to the thickness of the annular space of the packed bed, 0.467 and 0.515 for the cases of 11.2-mm. steel balls and 12.3-mm. glass balls respectively, temperature measurements scatter as shown in Figures 3 and 4. For the other packings the temperature differences were determined easily, and so there are comparatively small deviations from correlations for these cases, Figures 9 and 10.

The following formula for heat transfer coefficient was obtained here. It is similar to that for effective thermal conductivity but differs very much from previously reported equations for h_w:

$$\frac{h_{w1}\cdot D_p}{k_g} = \frac{h_{w1}^\circ\cdot D_p}{k_g} + \alpha_w N_{Pr} N_{ReM} \qquad (14)$$

The value of (α_w) suitable for Figures 9, 10, and 11 is 0.041 in these experiments on the annular packed beds, and the values of $h_w^\circ \cdot D_p/k_g$ are shown in Table 3.

With reference to the wall film coefficients on the outer tube surface, there are few reliable data in these experiments, as mentioned above. However comparing Figure 12 with Figure 10, one can reasonably consider that there are no fundamental differences between the two correlations for both the wall film coefficients.

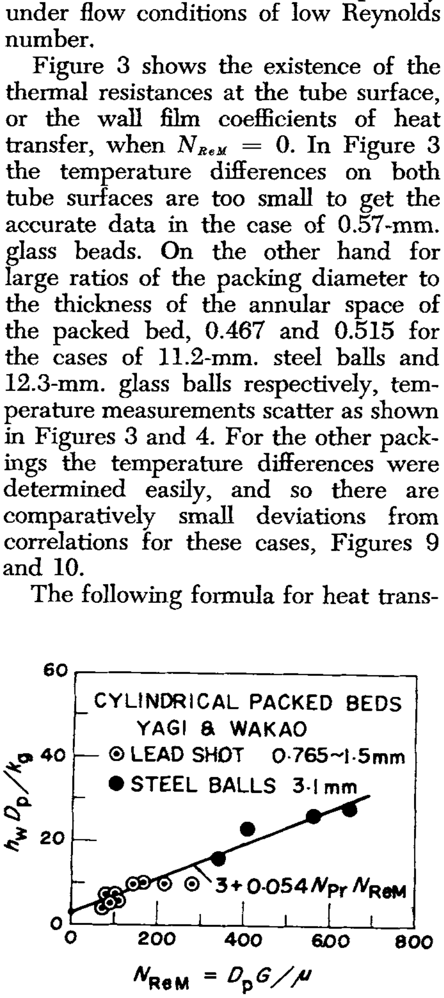

Fig. 14. Correlation of data observed by Yagi and Wakao (*13*) in cylindrical packed beds.

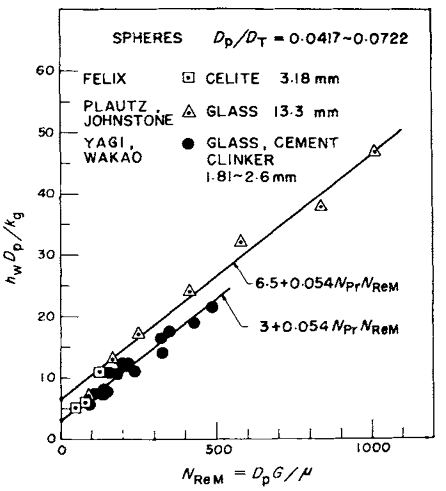

Fig. 15. Correlation of data by Felix (*4*), Plautz and Johnstone (*8*), and Yagi and Wakao (*13*) in cylindrical packed beds.

APPLICATION OF NEW EQUATION TO CYLINDRICAL PACKED BEDS

If one assumes that there are no fundamental differences between the heat transfer mechanisms in cylindrical and annular packed beds, the recent and reliable data directly measured by Felix (*4*), Plautz and Johnstone (*8*), and Yagi and Wakao (*13*) may be correlated with the modified Reynolds number $D_p\cdot G/\mu$ as shown in Figures 13 to 16 for each value of D_p/D_T. Thus Equation (14) can be applied to the experimental values of h_w obtained previously by other observers for the common cylindrical packed beds of spherical packings. However the values of (α_w) in Equation (14) suitable for Figures 13 to 16 is 0.054, which is larger than the value 0.041 in the annular packed bed used here. Table 3 shows the values of $h_w^\circ\cdot D_p/k_g$. The difference between the two numerical values of (α_w) may be attributed to the difference in the packing states for

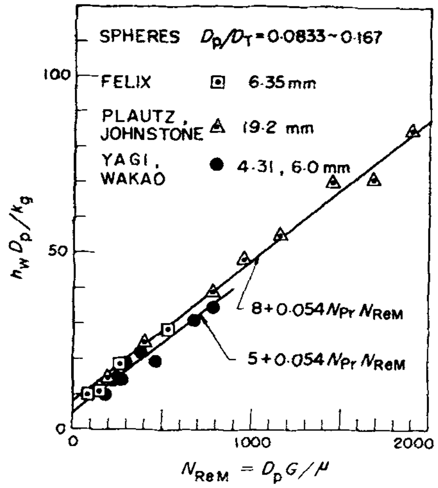

Fig. 16. Correlation of data by Felix (*4*), Plautz and Johnstone (*8*), and Yagi and Wakao (*13*) in cylindrical packed beds.

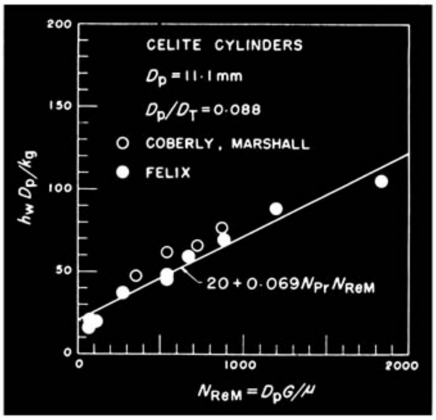

Fig. 17. Correlation of data by Coberly and Marshall (3) and Felix (*4*) in cylindrical beds packed with cylindrical solids.

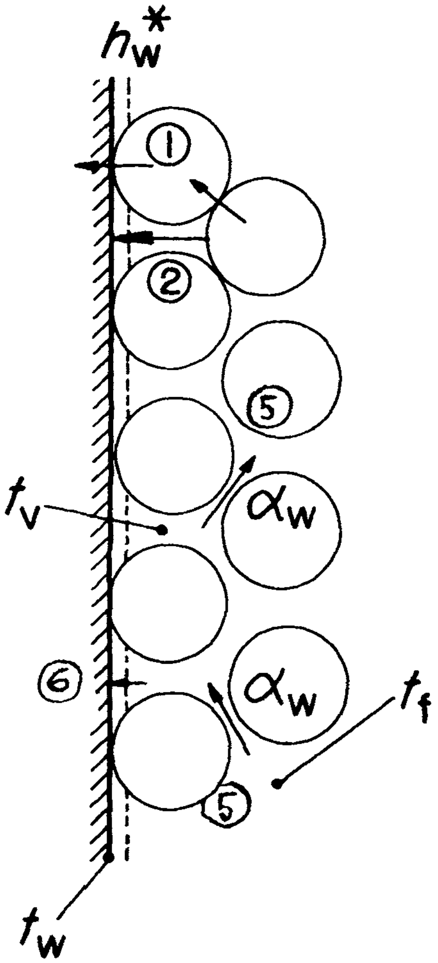

Fig. 18. Model of heat transfer near wall surface of packed bed.

particles in annular and cylindrical packed beds. Thus the fundamental *mechanisms of heat transfer on the wall* surfaces must be considered similar for both cylindrical packed beds and annular beds.

For cylindrical packings Coberly and Marshall (*3*) and Felix (*4*) have presented experimental data of h_w which are too scattered to be correlated by a simple equation. However if the data of h_w are plotted for some narrow range of D_p/D_T as shown in Figure 17, it can be assumed that the correlation similar to Equation (14) is applicable for the cylindrical packings, even though the numerical values of constants may not necessarily be the same as those for spherical packings.

THEORETICAL CONSIDERATIONS ON NEW CORRELATION

The correlation obtained here is so different from previously presented correlations, that is Equations (1) or (2), that neither the penetration theory (*5*) nor boundary-film theory explains the real heat transfer phenomena near the wall surface in the packed beds. Therefore another model of heat transfer must be applied.

Previously Kwong and Smith (*7*) reported the data for the radial distribution of the effective thermal conductivities in packed beds and showed the sharp decrease near the wall surface. These phenomena are considered most important in this paper in the analysis of the thermal resistance near the wall surface of the packed beds, especially under the flow conditions of low Reynolds numbers.

TABLE 3. $h_{w1}{}^{\circ} D_p/k_g$ FOR ANNULAR PACKED BEDS AND FOR CYLINDRICAL PACKED BEDS

Annular packed beds, D_e = 48 mm.

Packing	D_p/D_e	D_p, mm.	$h_{w1}{}^{\circ} D_p/k_g$
Glass beads	0.0196	0.94	3.2
Glass beads	0.0572	2.75	7
Glass beads	0.106	5.1	12
Glass beads	0.256	12.3	19
Lead shot	0.0625	3.0	12
Steel balls	0.233	11.2	25

Cylindrical packed beds

Packing	D_p/D_T	D_p, mm.	$h_w{}^{\circ} D_p/k_g$
Glass beads and cement clinkers	0.021-0.036	0.76-0.91	1.2
Celite balls and glass beads	0.0417-0.0722	1.81-3.18 13.3	3 6.5
Celite balls, glass beads and cement clinkers	0.0833-0.167	4.31-6.35 19.2	5 8
Lead shot	0.021-0.042	0.77-1.5	3

Schwartz and Smith (*11*) showed that the fraction void near the wall surface is larger than that in the core portion of the packed bed. It is reasonable to assume that the lower degree of lateral mixing is the main cause of the sharp decrease in the effective thermal conductivity near the wall surface.

In cases of analysis for wall film coefficients of heat transfer the average values of the effective thermal conductivities in packed beds are determined by all observers. Thus the sharp decrease of the effective thermal conductivities near the wall surface looks like the existence of a definite thermal resistance at that point, which may be ascribed to a fluid boundary film on the wall.

In Figure 18, which is the authors' model of heat transfer mechanisms near the wall surface in the packed bed, the following mechanisms are assumed, with the real film coefficient of heat transfer on the wall surface considered to be caused only by the true fluid boundary layer at the wall surface.

a. Heat transfer through solid phases
1. Heat transfer through the thin

TABLE 4. COMPARISON OF α_w WITH α

		Mean value in* packed bed	Near the wall surface
Cylindrical packed bed	$D_p/D_T = 0.15$	$\alpha = 0.11$†	$\alpha_w = 0.054$
Annular packed bed	$D_p/D_e = 0.15$	$\alpha = 0.078$	$\alpha_w = 0.041$

* Where β is assumed to be 0.9.
† Calculated from the correlation obtained in authors' previous paper (12).

gas film near contact points
2. Radiant heat transfer from solid surface to solid surface (Mechanisms 1 and 2 are parallel.)

b. Heat transfer in void spaces, independent of fluid flow
3. Molecular thermal conduction
4. Radiant heat transfer from void to void (Mechanisms 3 and 4 are parallel.)

c. Heat transfer in void spaces, dependent on fluid flow
5. Heat transfer caused by the lateral mixing of the flowing fluid
6. Heat transfer through the true boundary film (Mechanisms 5 and 6 are in series.)

As shown in Figure 18 the mechanisms *a*, *b*, and *c* can be considered in parallel. The heat flow by mechanism 5, namely by the lateral mixing of flowing fluids, is defined here as Q kg.-cal./(sq.cm.)(hr.), which can be presented with $Q = GC_p\alpha_w t_f - GC_p\alpha_w t_v = GC_p\alpha_w(t_f - t_v)$.

Therefore the heat transfer coefficient by lateral mixing can be calculated as

$$(h_w)_l = \frac{Q}{t_f - t_v} = \alpha_w GC_p \quad (15)$$

Then

$$\frac{(h_w)_l D_p}{k_g} = \alpha_w (C_p\mu/k_g)(D_pG/\mu) = \alpha_w N_{Pr} N_{ReM} \quad (16)$$

The mechanisms *a* and *b* can be assumed independent of the fluid flow, the constant term h_w° presumably originating from the above mechanisms. Therefore, considering the heat transfer mechanisms mentioned above, one can give the apparent wall film coefficient of heat transfer by

$$h_w = h_w^\circ + (h_w)_f \quad (17)$$

$$\frac{1}{(h_w)_f} = \frac{1}{h_w^*} + \frac{1}{(h_w)_l} \quad (18)$$

Therefore for all ranges of N_{ReM}

$$\frac{h_wD_p}{k_g} = \frac{h_w^\circ D_p}{k_g} + \frac{1}{\dfrac{1}{h_w^* D_p/k_g} + \dfrac{1}{\alpha_w N_{Pr} N_{ReM}}} \quad (19)$$

Since there are no reliable data which can present the correct relationship between the real wall film coefficient of heat transfer and the Reynolds number, an exact quantitative discussion is impossible here. However it seems quite probable that the numerical values of h_w° predominate under conditions of low Reynolds numbers compared with h_w^* and that in turbulent regions h_w^* is larger than $(h_w)_l$ for Reynolds numbers less than 2,000. Thus α_w in the experimental Equation (14) is approximately equal to the ratio of the lateral mixing of flowing fluid, which was defined by Ranz (10) as

$$\alpha_w = \frac{\text{Mass velocity of fluid flowing in the direction of heat and mass transfer (near the wall surface)}}{\text{Mass velocity of fluid based on sectional area of empty tube in the direction of fluid flowing}}$$

The numerical values of α_w are compared with those of α for the mean value of the effective thermal conductivities in Table 4. The ratios of α_w to α are almost ½ for both cases. Therefore it can be assumed that the degree of lateral mixing decreases near the wall surface to about half of that in the core.

CONCLUSIONS

The data for the wall film coefficient for the inner tube surface can be correlated with the new formula, namely Equation (14). The data previously presented for the wall film coefficient of heat transfer in the ordinary cylindrical packed beds by the other observers are replotted on Cartesian coordinates, and it is found that the above correlation, that is Equation (14), can be applied for these cases as well. The new correlation obtained by the authors is so different from the correlations previously presented that a new model of heat transfer near the wall surface in the packed beds must be applied in order to explain it.

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NOTATION

A_w = surface area of tube, sq. m.
b = $(D_2 - D_1)/2$ = clearance of annular space, m.
C_p = specific heat of fluid, kg.-cal./(kg.)(°C.)
D_e = $2b = D_2 - D_1$ = effective diameter of annular packed bed, m.
D_1 = diameter of inner tube, m.
D_2 = diameter of outer tube, m.
D_p = average diameter of packings, m.
D_T = diameter of cylindrical packed bed, m.
G = mass velocity of fluid, kg./(sq. m.)(hr.)
h_w = wall film coefficient of heat transfer in cylindrical packed bed, kg.-cal./(sq.m.)(hr.)(°C.)
h_w° = wall film coefficient of heat transfer in packed bed with stationary fluid, kg.-cal./(sq. m.)(hr.)(°C.)
h_{w1} = wall film coefficient of heat transfer on surface of inner tube in annular packed bed, kg.-cal./(sq.m.)(hr.)(°C.);
h_{w1}° = that with stationary gas.
h_{w1}' = uncorrected value of wall film coefficient of heat transfer on surface of inner tube in annular packed bed, kg.-cal./(sq. m.)(hr.)(°C.)
h_{w2} = wall film coefficient of heat transfer on surface of outer tube in annular packed bed, kg.-cal./(sq. m.)(hr.)(°C.)
h_w^* = wall film coefficient of heat transfer caused from boundary layer on wall surface, kg.-cal./(sq. m.)(hr.)(°C.)
$(h_w)_f$ = heat transfer coefficient concerning heat flux in void space of packed bed, kg.-cal./(sq. m.)(hr.)(°C.)
$(h_w)_l$ = heat transfer coefficient caused from lateral mixing of fluid, kg.-cal./(sq. m.)(hr.)(°C.)
k_e = mean effective thermal conductivity in packed bed, kg.-cal./(sq. m.)(hr.)(°C./m.)
k_e° = mean effective thermal conductivity in packed bed with stationary fluid, kg.-cal./(sq. m.)(hr.)(°C./m.)
k_g = thermal conductivity of fluid flowing in packed bed, kg.-cal./(sq. m.)(hr.)(°C./m.)
N_{Pr} = $C_p\mu/k_g$ = Prandtl number
N_{ReM} = D_pG/μ = Reynolds number
Q = lateral heat flux by mixing

through void space in packed bed, kg.-cal./(sq. m.) (hr.)

q = purely radial heat flow in annular packed bed, kg.-cal./(hr.)

q^* = heat loss from test section of annular packed bed, kg.-cal./(hr.)

q' = purely radial heat flow in annular packed bed for blank test, kg.-cal./(hr.)

q_o = purely radial heat flow in annular packed bed with stationary fluid, kg.-cal./(hr.)

t_f = mean temperature of fluid in void space, °C.

t_v = mean temperature of fluid in void space nearest to wall surface, °C.

Δt_o = apparent temperature difference in annular packed bed, °C.

$\Delta t_o'$ = apparent temperature difference in annular packed bed for blank test, °C.

Δt_o^* = apparent temperature difference in annular packed bed with stationary fluid, °C.

Δt_1 = apparent temperature difference on inner tube surface of annular packed bed, °C.

Δt_2 = apparent temperature difference on outer tube surface of annular packed bed, °C.

V = rate of vapor condensation in annular packed bed, kg./(hr.)

V_o = rate of vapor condensation in annular packed bed with stationary flund, kg./(hr.)

V' = rate of vapor condensation in annular packed bed for blank test, kg./(hr.)

Greek letters

α = (mass velocity of fluid flowing in direction of heat or mass transfer)/(mass velocity of fluid based on sectional area of empty tube in direction of fluid flowing)

α_w = above value near wall surface in packed bed

β = (effective length between centers of two neighboring solids in direction of heat flow)/(average diameter of solid)

γ = latent heat of vapor condensation, kg.-cal./kg.

ϵ = void fraction of packed bed

μ = viscosity of fluid, kg./(m.)(hr.)

LITERATURE CITED

1. Calderbank, P. H., and L. A. Pogorski, *Trans. Inst. Chem. Engrs.* (*London*), **35**, 195 (1957).
2. Campbell, T. M., and R. L. Huntington, *Petrol. Refiner*, **31**, 123 (1952).
3. Coberly, C. A., and W. R. Marshall, Jr., *Chem. Eng. Progr.*, **47**, 141 (1951).
4. Felix, T. R., Ph.D. thesis, Univ. of Wisconsin, Madison (June, 1951).
5. Hanratty, T. J., *Chem. Eng. Sci.*, **3**, 209 (1954).
6. Hatta, S, and S. Maeda, *Chem. Eng.* (*Japan*), **12**, 56 (1948); **13**, 79 (1949).
7. Kwong, S. S., and J. M. Smith, *Ind. Eng. Chem.*, **49**, 894 (1957).
8. Plautz, D. A., and H. F. Johnstone, *A.I.Ch.E. Journal*, **1**, 193 (1955).
9. Quinton, J. H., and J. A. Storrow, *Chem. Eng. Sci.*, **5**, 245 (1956).
10. Ranz, W. E., *Chem. Eng. Progr.*, **48**, 248 (1952).
11. Schwartz, C. E., and J. M. Smith, *Ind. Eng. Chem.*, **45**, 1209 (1953).
12. Yagi, Sakae, and Daizo Kunii, *A.I.Ch.E. Journal*, **3**, 373 (1957); *Chem. Eng.* (*Japan*), **18**, 576 (1954).
13. Yagi, Sakae, and Noriaki Wakao, *A.I.Ch.E. Journal*, **5**, 79 (1959).

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Thermodynamic Consistency of Binary Liquid-Vapor Equilibrium Data When One Component Is Above Its Critical Temperature

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Methods are developed for testing binary system phase-equilibrium data for thermodynamic consistency when the more volatile component is above its critical temperature. The isothermal case with varying pressure and the isobaric case with varying temperature are considered individually. The rigorous form of the Duhem equation is employed rather than the simplified one, which for binary systems is inconsistent with the phase rule. Although derived for the specific case given in the title, the relationships developed are applicable to all conditions of binary systems. Sample calculations illustrating the methods are included.

As the design procedures originally applied to petroleum plant distillation problems have spread to the chemical and petrochemical fields, there has been an increasing need to use laboratory-obtained phase-equilibrium data. With this has developed the need for establishing the thermodynamic consistency of such information. Considerable attention has been given to this subject in the literature (*1*, *4* to *8*, *13*, *17*).

All methods published for making the necessary thermodynamic tests for reliability are based on some form of the Duhem equation. Some authors (*7*, *17*) have allowed for the fact that a binary system cannot change in composition when both temperature and pressure remain constant; others have not. A few have included a correction for the nonideality of the vapor phase above the solution. Most of the methods

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