θ_s = dimensionless temperature in the pellet, $(T - T_s)/T_s$
λ_g = geometry factor [Equation (51)]
λ_s = geometry factor [Equation (47)]
ξ = dimensionless distance in the micropore, z/L; $\xi = 0$ at pore mouth, $\xi = 1$ at end of pore
ρ = apparent or bulk density of the catalyst material
φ_1 = macro mass transfer parameter, defined by Equation (42)
φ_2 = macro heat transfer parameter, defined by Equation (43)
Ψ_1 = micro mass transfer parameter, defined by Equation (13)
$\overline{\Psi}_1$ = Thiele number, value of Ψ_1 when $r = \overline{r}$
Ψ_2 = micro heat transfer parameter, defined by Equation (14)
Ψ_3 = $\Psi_2/\Psi_1^{1/2}$
Λ = $(\varphi_1 E_c)^{1/2}$, defined by Equation (57)

Subscripts

e = effective
K = Knudsen
l = linear case
n = order of reaction
o = conditions at the mouth of the micropore, that is conditions at the surface of the microporous particle
s = conditions at the surface of the macropellet

Superscripts

G = Gaussian
l = linear case
M = Maxwellian
T = Thiele

Calculus

Prime = differentiation
(j) = evaluation at value of independent variable $= j$

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Effective Thermal Conductivity in Packed Beds

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Values of the effective thermal conductivity *Ke* for liquids in packed beds have been obtained from heat transfer studies in such beds. The method applied by Yagi and Kunii for gases is clarified, their model is extended and is then applied to liquids. Experimental data for three different packings, glass, steel, and aluminum, with spheres of D_P/D_T ratio from 0.07 to 0.33 have been correlated. A brief comparison of the results from this analysis with previously published results from similar studies is presented.

The process of heat transfer into and through packed beds and tubes has been the subject of numerous experimental studies. Heat transfer studies in packed beds have resulted in publication of many different correlations for the effective thermal conductivity in terms of the physical and operational parameters of the systems (*1, 2, 5, 8, 15, 17, 18*). Such correlations are useful for design purposes where the conditions of the given problem are within the stated limitations of the correlation but do not generally contribute to our understanding of the physics of this process. Recently Yagi and Kunii (*14*) proposed a model for heat transfer to gases in packed beds and derived an equation which contributes considerably to an understanding of the physical processes which are of importance in determining the effective conductivity. This model is used as a starting point in the present paper. This paper presents, first, some experimental values of effective conductivity obtained for an extended range of particle to tube diameter ratios and for various liquids flowing through beds of spheres composed of either glass, steel, or aluminum. In addition a further clarification and extension of the Yagi-Kunii physical model is shown.

In the past two different types of studies in this field have been made. In the first of these effective conductivities were measured from mixed mean average temperatures inlet and outlet, known constant wall temperatures or a known constant wall heat flux rate, and the operating character-

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istics of the system. The second type of procedure is to measure temperature profiles in the exit gases from the bed and from such data to obtain values for the effective conductivity in terms of the contribution from the solids conduction within the bed, the fluid conduction within the voids of the bed, and a convection coefficient, often expressed in terms of an eddy diffusivity or, perhaps, a modified Peclet number. In this latter technique the contribution from the wall of the bed is neglected. The work of Argo and Smith (*1*) is illustrative of the second class of experiments, whereas the work of Leva (*9*) and that of Singer and Wilhelm (*17*) are examples of work done which includes the effect of the wall transfer rates and resistances.

In this present paper the results will be comparable to those of Leva and to those of Singer and Wilhelm but will not be comparable to the results of Argo and Smith.

THE YAGI-KUNII MODEL

The general equation developed by Yagi and Kunii is

$$\frac{k_e}{k_f} = \frac{k_e^\circ}{k_f} + \alpha \beta N_{Pr} N'_{Re} \quad (1)$$

where

$$\frac{k_e^\circ}{k_f} = \delta \frac{k_s}{k_f} + \frac{(1-\epsilon-\delta)\beta}{\gamma\left(\frac{k_f}{k_s}\right) + \frac{1}{\frac{1}{\varphi} + \frac{D_p h_{rs}}{k_f}}} + \frac{\epsilon \beta D_p h_{rv}}{k_f} \quad (2)$$

The following assumptions were made in its derivation:

1. k_e can be separated into two terms, one of which is independent of fluid flow and the other dependent on the lateral mixing of the fluid in packed beds with the following result:

$$k_e = k_e^\circ + (k_e)_t \quad (3)$$

2. The equation proposed by Ranz is applicable; that is

$$(k_e)_t = \alpha \frac{C_p G}{N} \quad (4)$$

where the value of N is assumed to be equal to $1/\beta D_p$.

In addition the equations are applied only to flow of gases through fixed beds, and no reference is made to their possible use for liquid flows. Finally it is possible that a natural convection process may contribute significantly at low flow rates. Contribution of natural convection has not been taken into account in the model proposed by Yagi and Kunii.

TABLE 1. SAMPLE OF EXPERIMENTAL DATA AND CALCULATED VALUES OF EFFECTIVE THERMAL CONDUCTIVITY

(a) Glass spheres: $D_p = 0.157$ in. $k_s = 0.50$ $D_T = 2.25$ in. Liquid = Nitrobenzene $L = 4$ ft.

No.	G	h_m	k_e	k_e/k_e	N_{Pr}	$N_{Re}N_{Pr}$	N_{Nu}
1.	94,000	513.7	12.38	135	9.27	63,221	1050.0
3.	67,600	385.0	9.64	106	8.56	45,796	793.0
5.	40,600	240.7	6.00	66	8.54	27,687	495.5
7.	28,000	188.1	4.83	53	8.69	18,927	386.7
9.	16,300	117.8	3.10	34	8.80	11,009	242.0

(b) Glass spheres: $D_p = 0.671$ in. $k_s = 0.50$ $D_T = 2.25$ in. Liquid = (Toluene) $L = 4$ ft.

No.	G	h_m	k_e	k_e/k_e	N_{Pr}	$N_{Re}N_{Pr}$	N_{Nu}
10.	37,000	228.5	5.15	60.2	4.85	36,000	505.0
11.	30,500	196.1	4.48	52.4	4.83	29,600	430.0
12.	24,600	166.7	3.92	45.8	4.81	23,900	365.5
13.	20,100	146.6	3.52	41.3	4.77	19,600	321.0
14.	14,600	116.0	2.86	33.6	4.77	14,250	254.5

(c) Steel spheres: $D_p = 0.125$ in. $k_s = 34.8$ $D_T = 1$ in. Liquid = Nitrobenzene $L = 2$ ft.

No.	G	h_m	k_e	k_e/k_e	N_{Pr}	$N_{Re}N_{Pr}$	N_{Nu}
15.	2,13,000	1136	12.50	136	9.84	63,500	1025
17.	1,69,000	944	10.55	115	9.35	50,500	857
19.	1,37,500	769	8.62	94	9.12	41,000	699
21.	92,500	568	6.50	71	9.05	27,600	517
23.	62,200	405	4.70	52	8.96	18,700	369

(d) Steel spheres: $D_p = 0.25$ in. $k_s = 34.8$ $D_T = 1$ in. Liquid = 41% Aqueous glycerine $L = 2$ ft.

No.	G	h_m	k_e	k_e/k_e	N_{Pr}	$N_{Re}N_{Pr}$	N_{Nu}
24.	1,84,000	2863	31.0	114.0	12.98	57,500	879
26.	1,54,500	2491	27.4	101.0	12.98	48,250	762
28.	1,27,000	2016	21.9	80.5	12.82	40,000	616
30.	63,250	1263	14.6	53.5	11.72	19,800	384

(e) Aluminum spheres: $D_p = 0.50$ in. $k_s = 118$ $D_T = 2.25$ in. Liquid = 40% Aqueous glycerine $L = 4$ ft.

No.	G	h_m	k_e	k_e/k_e	N_{Pr}	$N_{Re}N_{Pr}$	N_{Nu}
31.	1,17,000	2150	52.5	190.0	10.50	80,000	1460
33.	85,000	1625	41.3	149.0	10.52	58,234	1095
35.	46,000	900	22.6	82.0	10.60	31,600	680
37.	37,200	730	18.3	67.0	10.70	26,400	495
40.	21,900	540	14.0	50.0	10.60	15,700	368

PRESENT WORK

The process of heat transfer from a uniformly heated wall to a fluid flowing through packed beds and packed tubes has been under intensive investigation for some time. Chennakesavan (*3*) and Raghavan and Laddha (*9*) have presented considerable data on this process for a wide range of operating and equipment variables. For the present study some further data were accumulated to extend the

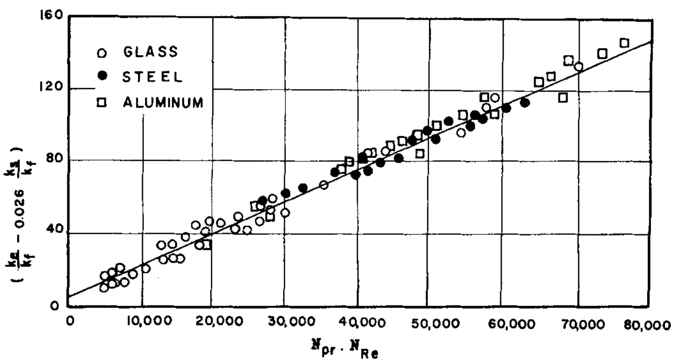

Fig. 1. Final correlation for effective thermal conductivity in packed beds.

range of variables covered to include the following:

Liquids used:
1. Nitrobenzene
2. Aqueous glycerine solutions
3. Toluene

Tube to particle diameter ratio:

$$3 < \frac{D_T}{D_p} < 14$$

Reynolds number (modified):
$50 < N'_{Re} < 2{,}100$

Values for effective thermal conductivity were obtained by standard procedures (*10*). The experimental equipment and procedure used in this investigation has been described by Raghavan and Laddha (*11, 12*) and by Chennakesavan (*4*). The wall temperature was measured, the average or bulk mean temperature of the inlet and exit fluids were measured, and the operating conditions, notably the fluid flow rate, and bed geometries were obtained. Standard procedures described by McAdams (*10*) were used to obtain values for the effective conductivity. Such values then obviously included the contribution from the heat transfer resistance at the wall of the column and the fluid-fluid, solid-solid conduction in the bed, and the various convective mechanisms which are of importance. Table 1 presents some sample results. Detailed experimental data are available elsewhere (*12*). The packings used were spheres made of glass, steel, and aluminum. Plots of k_e/k_f vs. N'_{Re} were made in accordance with Equation (1), and in all cases excellent straight lines were obtained with a positive slope and intercept as required. At this point a further study of the processes which determine the slope and intercept was made.

EXTENSION OF THE YAGI-KUNII MODEL

The above model is subject to questioning on at least two points. First the value of φ seems from its definition to be necessarily a function of flow rate which would then make the simple linear plots described in a previous paragraph unlikely. Second one might suspect that as the flow rate goes to zero in the system, a natural convection process would continue to be operative and would increase the value of $k_e°/k_f$ above that dictated by Equation (2). These two points are discussed below.

It seems evident φ varies not only with flow rate but also on a point-to-point basis throughout the bed. This variation has been discussed by Hoelscher (*6*) in a paper dealing with reactor design, and a proposed distribution in accordance with a Beta distribution function is described. It is evident that φ should be written as φ_m, that is an average or perhaps most probable value in the bed. This value may be obtained either by some suitable averaging procedure, perhaps by integration, or by taking the value of φ_m at which the Beta distribution function goes through its maximum. Following the procedure outlined by Hoelscher (*7*) one may write

$$\frac{1}{\varphi_m} = \frac{D_p}{\delta_m} = N'_{Nu} = \left(\frac{h_m D_p}{k_f}\right) = a\, N'_{Re}{}^{(b+1)}\, N_{Pr}{}^{1/3} \quad (5)$$

Values of the constant a and b (defined in the usual heat transfer *J*-factor relationship) are obtainable from the literature. Using the values of a and b suggested by Baumeister and Bennet (*6*) and the extreme ranges of variables used in the experimental work described above one may show

$$0.0005 < \varphi_m < 0.02$$

Furthermore it seems reasonable to assume that the radiation transfer mechanism is of no importance at the low temperatures used throughout the work done to date and that the terms involving both the solid-to-solid radiation coefficient and the solid-to-void radiation coefficient may be dropped from the expression for $k_e°/k_f$ proposed by Yagi and Kunii (*14*) for Equation (2). Doing so one obtains the following:

$$\frac{k_e°}{k_f} = \delta\frac{k_s}{k_f} + \frac{(1-\epsilon-\delta)\beta}{\gamma\dfrac{k_f}{k_s} + \varphi_m} \quad (6)$$

For the range of solids and fluids used in this experimental work the value of k_f/k_s is

$$0.0017 < \frac{k_f}{k_s} < 0.33$$

Hence (with some uncertainty) one may propose to test the assumption that φ_m may be neglected by comparison to k_f/k_s and write

$$\frac{k_e°}{k_f} = \frac{k_s}{k_f}\left[\delta + \frac{\beta(1-\epsilon-\delta)}{\gamma}\right] = \delta'\frac{k_s}{k_f} \quad (7)$$

where δ' is now a function of the physical shape of the packing only (that is whether it is spherical, cylindrical, etc.) and not a function of the material.

The second doubt raised about the Yagi-Kunii model concerned the absence of a natural convection term to take care of the zero flow condition. This may, following the form of the other terms in this paper, be introduced as an additive term of the form $\epsilon\,\beta\,h_c\,D_p/k_f$ where the convection coefficient may be written in conventional notation as

$$\frac{h_c D_p}{k_f} = C'_o\,[N'_{Gr}\,N_{Pr}]^m \quad (8)$$

One may then combine Equation (8) with Equation (7) and introduce the result into Equation (1) to obtain finally the following result:

$$\frac{k_e}{k_f} = \delta'\frac{k_s}{k_f} + C'_o\,[N'_{Gr}\,N_{Pr}]^m + \alpha\,\beta\,N_{Pr}\,N'_{Re} \quad (9)$$

The physical interpretation of the two constants α and β may now be considered. The constant $\beta = l_p/D_p$ is considered by Yagi and Kunii to be essentially unity. This seems implausible to the present authors, since the value of l_p seems likely to depend directly on the particle diameter but, simultaneously, inversely on the number of particles available for heat transfer in the radial direction. Thus one might more reasonably postulate that β is proportional to D_T/D_p. The constant α is stated to be the ratio of flow in the direction of transfer to the flow based on the free cross-sectional area of the tube. Hence it must be proportional in some manner to the thickness of the boundary layer surrounding any particle. It may then be expected to depend upon the fluid properties. Writing $\beta = \beta_o \; (D_T/D_p)$ but retaining α without modification for the present one obtains from Equation (9)

$$\frac{k_e}{k_f} - \delta' \frac{k_s}{k_f} = C'_o \, [N'_{Gr} \, N_{Pr}]^m + \alpha \, \beta_o \, N_{Pr} \, N_{Re} \quad (10)$$

where the Reynolds number is now based on the tube diameter. An extensive analysis of data taken and presented by previous workers in this field over the range of variables indicated earlier in this paper resulted in the following values for the constants:

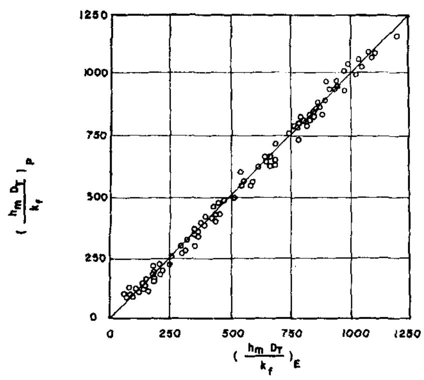

Fig. 2. Correlation for heat transfer, data taken from this work.

$$m = 0.00$$
$$\delta' = 0.026$$
$$\alpha \, \beta_o = 0.00176$$
$$C'_o = 6.00$$

The final form of the correlation is

$$\frac{k_e}{k_f} - 0.026 \frac{k_s}{k_f} = 6 + 0.00176 \, N_{Pr} \, N_{Re} \quad (11)$$

The result is shown in Figure 1.

One may now combine Equation (11) with the equation for the wall heat transfer coefficient in terms of effective conductivity as presented below (*10*):

$$h_m = 5.79 \frac{k_e}{D_T} + 0.0912 \frac{C_p \, G_o \, D_T}{L} \quad (12)$$

The following result is obtained:

$$N_{Nu} = \left(\frac{h_m \, D_T}{k_f} \right) = \left(0.151 \frac{k_s}{k_f} + 34.7 \right) + \left(0.0102 + 0.0912 \frac{D_T}{L} \right) N_{Pr} \, N_{Re} \quad (13)$$

Equation (13) was used to predict values for the Nusselt group for the same conditions as those used in the experiment referred to earlier. The result is shown on Figure 2. The ordinate is the value of the Nusselt group calculated from Equation (13) for selected values of the solid and fluid conductivity, tube diameter and length, and the operating conditions needed to specify the Reynolds number. The abscissa is the value of the Nusselt number obtained from the experiment under the same conditions. The result is shown as

$$\left(\frac{h_m \, D_T}{k_f} \right)_P \text{vs.} \left(\frac{h_m \, D_T}{k_f} \right)_E$$

in Figure 2.

The result shown as Figure 1 indicates an agreement between the value of effective conductivity predicted by Equation (11) and the values reported from experiment to within an average deviation of ± 7.5% and maximum deviation of 16.5%. The result shown in Figure 2 indicates an average deviation of ± 8.5% and a maximum deviation of 17%.

The final equation of this paper was applied with apparent equal success to three different fluids flowing through packings of three widely differing thermal conductivities. As indicated earlier the experimental data for this study involved the use of nitrobenzene, aqueous clycerol solutions, and toluene. Equation (13) has also been tested against the experimental data of Leva (*9*) for air flowing through beds of glass spheres where the tube to particle diameter is between 5 and 12. These data are shown on Figure 3. The maximum deviation from the 45-deg. line for the data of Leva is ± 10% with an average deviation of ± 4%. It would appear therefore that the final correlations for effective thermal conductivities presented in this present paper are equally applicable to both gases and liquids over the range of variables investigated. It should be noted however that it is not possible to compare the results of this study with those presented by Singer and Wilhelm. The latter authors plotted the values of the modified Peclet group obtained from heat transfer data in packed tubes against the modified Reynolds number and indicated that at low values of the particle-to-tube diameter ratio (of the order of 0.04) the modified Peclet group becomes constant. They did not present any relationship between the modified Peclet group and particle-to-tube diameter ratio. The present investigation includes particle-to-tube diameter ratios from 0.07 to 0.33, values which are considerably above those used by others.

Assumptions Made in Addition to Those Of Yagi-Kunii

The following comments relative to the derivation of the final equation seem pertinent.

1. It is assumed that radiation has no effect at the low temperatures employed in this work and with fluids of high absorptivity.
2. The value of φ is taken as φ_m (the most probable value) rather than the average; this assumption yields a safe (low) value of φ_m and tends to support the transition from Equation (6) to Equation (7).
3. φ_m is neglected by comparison to $\gamma \, k_f/k_s$ in the range of variables studied.
4. The convection contribution proposed seems dubious and unreal in its present form. Further experimental work is needed.

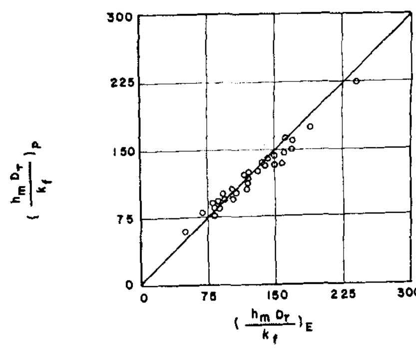

Fig. 3. Test of Equation (13), data from Leva.

The Physical Interpretation of δ'

One further consideration regarding δ' is of interest. From Equation (7)

$$\delta' = \delta + \frac{\beta}{\gamma} [1 - \epsilon - \delta]$$

From the definitions of Yagi and Kunii

$$\frac{\beta}{\gamma} = \frac{l_p}{l_s}$$

and $1/l_p = n/D_T$ (number of particles radially/unit tower diameter).

To get the over-all heat transfer rate, Δt should be the difference between the wall temperature and the mean fluid temperature. Then l_s should be proportional to $D_T/2$

$$\beta/\gamma \sim \frac{2}{n}$$

$$\delta' \sim \delta + \frac{2(1 - \epsilon - \delta)}{n}$$

Taking the average value of ϵ as 0.45 one obtains

$$\delta' = \delta + \frac{1}{n} - \frac{2\delta}{n}$$

or

$$\delta' = \delta \text{ when } n \text{ is large}$$

Analysis of k_e/k_f: Data From Other Studies

The Yagi Kunii equation is

$$\frac{k_e}{k_f} = \frac{k_e^\circ}{k_f} + \alpha \beta N_{Pr} N'_{Re}$$

and was applied only to data involving heat transfer to gases. The equation presented for liquids in this paper is

$$\frac{k_e}{k_f} = \left(\delta' \frac{k_s}{k_f} + C'_o \right) + \alpha \beta_o \frac{D_T}{D_p} N_{Pr} N'_{Re}$$

In the case of the plots made by Yagi and Kunii $\alpha \beta N_{Pr}$ is the slope, whereas the slope of a corresponding plot from Equation (11) is equal to $\alpha \beta_o$. Therefore by comparison of the two equations one gets

$$\alpha \beta_o N_{Pr}(\text{liquid}) \frac{D_T}{D_p} = \alpha \beta N_{Pr}(\text{gas})$$

or

$$\alpha \beta_o = \frac{\alpha \beta N_{Pr}(\text{gas})}{N_{Pr}(\text{liquid})} \frac{D_p}{D_T}$$

The value of $\alpha \beta N_{Pr(\text{gas})}$ for glass spheres–air and insulating firebrick–air for an average particle size of 0.15 is given as 0.12 (*14*):

$$\alpha \beta_o = \frac{0.12 \times 0.15}{10} = .0018$$

This number is in very good agreement with the results presented in this paper.

SUMMARY

1. Values of effective thermal conductivity k_e have been reported for the case of liquids flowing through packed beds.
2. In order to correlate such values the model proposed by Yagi and Kunii is clarified and extended to the case of liquids.
3. Some assumptions were made which seem to be reasonable within the experimental range employed in the author's experiments.
4. A final correlation is obtained for glass, steel, and aluminum spheres. It it found that the same equation can be applied in the case of gases also.
5. The resulting correlation indicates that the effective thermal conductivity and hence the values of the film coefficient of heat transfer are independent of particle diameter over the range of variables studied. This result is in agreement with the prediction of Schumacher (*16*) for this same range of operating conditions.

ACKNOWLEDGMENT

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NOTATION

a = constant defined in Equation (5)
b = constant defined in Equation (5)
C_p = specific heat, B.t.u./lb. °F.
C_o' = constant defined in Equation (8)
D_p = diameter of the particle, ft.
D_T = diameter of the tube, ft.
G = mass velocity of fluid, lb./(sq. ft.) (hr.)
h_c = heat transfer coefficient for convection, B.t.u./(hr.) (sq. ft.) °F.
h_{rs} = heat transfer coefficient for radiation, solid to solid
h_{rv} = heat transfer coefficient for radiation, void to void
h_m = heat transfer coefficient of the liquid film, B.t.u./(hr.) (sq. ft.) °F.
k_e = effective thermal conductivity of the core of the packed tube, B.t.u./(hr.) (ft.) (°F.)
k_e° = effective thermal conductivity of the packed tube with motionless fluid, B.t.u./(hr.) (ft.) (°F.)
$(k_e)_t$ = effective thermal conductivity due to lateral mixing, B.t.u./(hr.) (ft.) (°F.)
k_f = thermal conductivity of the fluid, B.t.u./(hr.) (ft.) (°F.)
k_s = thermal conductivity of the solid, B.t.u./(hr.) (ft.) (°F.)
L = length of the packed bed
l_p = effective length between centers of the particles
l_v = effective thickness of fluid film adjacent to contact surface of two solid particles
m = constant defined in Equation (8)
n = number of particles radially
N = constant defined in Equation (4)
N'_{Gr} = modified Grashof number, dimensionless
N_{Nu} = modified Nusselt number, dimensionless
N_{Pr} = Prandtl number, dimensionless
N'_{Re} = modified Reynolds number, dimensionless
N_{Re} = Reynolds number based on the tube diameter, dimensionless
Δt = temperature difference

Subscripts

P = value predicted from the derived equation
E = value obtained from experiment

Greek Letters

β_o = constant used in Equation (10)
β = l_p/D_p = effective length between the centers of the particles and D_p = particle diameter
ϵ = fraction void
α = the ratio of mass velocity of fluid flowing in the direction of heat or mass transfer to the total mass velocity of fluid based on the cross-sectional area of empty tube in the direction of fluid flowing
δ = total area of perfect contact surface as solid phase
δ' = a constant defined in Equation (7)
δ_m = the most probable value of δ
γ = length of solid affected by thermal conductivity/mean diameter of the solid = l_s/D_p; l_s = effective length of solid relating to thermal conduction
φ = thickness of fluid film in the voids effective for thermal conduction/mean diameter of solid = l_v/D_p
φ_m = the most probable value of φ

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