EFFECTIVE THERMAL CONDUCTIVITY FOR GRANULAR BEDS OF VARIOUS BINARY MIXTURES

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The effective thermal conductivity was investigated for granular beds of various binary mixtures. Samples used were two or three sizes of glass beads, resin particles and copper spheres respectively. A unit cell model was developed after due consideration of particle diameter ratio and it was assumed that the coordination number was a function of diameter ratio and volume fraction of components. Predicted values coincided with experimental ones, in which the ratio of diameters was 1–2.8 and the ratio of solid conductivities was 1–5.8, within 6% deviation.

Introduction

The effective thermal conductivity of a granular bed is very important to various calculations for many operations, such as catalyst reaction of packed bed, solid-gas reaction and drying. In particular, it is one of the basic physical properties in the drying process. A number of experimental and theoretical investigations^{2–6,8–10} have been carried out in this field.

Most of them treat granular beds as homogeneous particles, that is, particles having the same diameters and same conductivities. On the other hand, we can see that it is not so rare in actual granular beds that there is a considerably wide range of particle diameter and that the different components are mixed. Until now, only Bauer and Schlünder^{2,3)} made measurement and predictions for such cases, using diameter ratios 1.85, 2.95 and 6.15. Their prediction formula involves parameters decided from experiments and is fairly complicated.

In this work, the authors (I) measured the effective thermal conductivities of granular beds with dry binary mixtures having a specified diameter ratio and solid thermal conductivity ratio, (II) proposed a modified unit cell model after consideration of particle diameter ratio and of change of coordination numbers, which is calculated from void fractions, and (III) compared the predicted value to the experimental data.

1. Experiments

1.1 Experimental apparatus

The authors used 'Quick Thermal Conductivity

Measurement Apparatus QTM-D1 (Showa Denko Co.)' to measure effective thermal conductivities of granular beds. This apparatus is based on the principle of the Hot Wire Method, which is a kind of transient method. The measuring technique is very easy and the measuring time required for one point is within 5 minutes.

1.2 Samples

Samples used for experiments were various sizes of glass beads (G.B.), acrylic resin particles (A.P.) and copper spheres (C.S.). Their properties are tabulated in Table 1. It is difficult in practice to measure the solid thermal conductivities λ_s of samples using a large lump. Therefore, we measure the effective thermal conductivities, λ_e , of homogeneous particles and calculate backward to λ_s from measured λ_e by our prediction formula, determining the solid thermal conductivity λ_s as a kind of experimental parameter. The values of λ_s decided by the above method are shown in Table 1.

1.3 Experimental results

1) Mixtures with same diameter but different solid

Table 1 Properties of samples

Sample	Particle diameter D [μm]	Solid density ρ_s [kg/m^3]	Solid conductivity λ_s [$\text{W}/(\text{m} \cdot \text{K})$]
Glass beads (G.B.)	125–149 (A) 74–88 (B) 44–53 (C)	2480	1.57
Acrylic resin particles (A.P.)	125–149 (A) 74–88 (B)	1190	0.357
Copper spheres (C.S.)	125–149 (A) 44–53 (C)	8800	9.09* 5.66*

* The variation in λ_s with particle diameter range seems to depend on the thin film of copper oxide, with low thermal conductivity, that covers these particles.

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Fig. 1 Effective thermal conductivities of beds of binary mixtures

Fig. 2 Unit cell model of packed bed

Fig. 3 Three cases of contacting two particles

- thermal conductivities (Run 1–4)
- Mixtures with different diameters but same solid thermal conductivity (Run 5–8)
- Mixtures with different diameters and different solid thermal conductivities (Run 9–14)

For the above mixtures we measured the effective thermal conductivities of granular beds, changing the volume fractions every 20%. The void fractions of granular beds were measured by the ordinary method. These results are tabulated in Table 2 and typical cases are shown in Fig. 1. In the case of Run 8, the maximum point of effective thermal conductivity exists at the center part of volume fraction because of decreasing void fraction caused by mixing of different-

radius particles.

2. Theory

2.1 Bed of particles of same diameter and same conductivity

For the case of same diameter and same conductivity the authors reported previously⁶⁾, so here we'll summarize briefly.

The coordination number N of a particle is given for the arbitrary void fraction by Ridgway and Tarbuck⁷⁾.

$$\epsilon = 1.072 - 0.1193N + 0.00431N^2 \quad (\text{for } \epsilon \geq 0.259) \quad (1)$$

or

$$N = 13.84 - \sqrt{232.0\epsilon - 57.18} \quad (2)$$

The effective number of contact points on a hemisphere n based on the contribution of heat transfer is given by Eq. (3).

$$n = N/6 \quad (3)$$

The authors introduced the unit cell model of Fig. 2, which consists of a solid part and a macro void part. The fractional areas of the solid part and the macro void part across which heat flows are given respectively as follows.

$$\begin{aligned} \epsilon \geq 1/3 & \quad \epsilon < 1/3 \\ A_s = 3(1 - \epsilon)/2 & \quad 1 \\ A_v = (3\epsilon - 1)/2 & \quad 0 \end{aligned} \quad (4)$$

The apparent thermal conductivity of solid part λ_{es} is deduced by a process similar to that of Kunii and Smith⁵⁾.

$$\frac{\lambda_{es}}{\lambda_f} = 2n \left(\frac{\kappa}{\kappa - 1} \right)^2 [\ln \{ \kappa - (\kappa - 1) \cos \theta_0 \} - \left(\frac{\kappa - 1}{\kappa} \right) (1 - \cos \theta_0)] \quad (5)$$

where

$$\kappa = \lambda_s / \lambda_f \quad (6)$$

$$\theta_0 = \sin^{-1} \sqrt{1/n} \quad (7)$$

Then the effective thermal conductivity λ_e of granular beds is given by Eq. (8).

$$\lambda_e / \lambda_s = A_v + A_s \lambda_{es} / \lambda_f \quad (8)$$

2.2 Bed of particles of same diameter and different conductivities

When two kinds of particles of same diameter but different conductivity are mixed, three contacting cases exist, as shown in Fig. 3. Using volume fractions of components X_1 and X_2 ($=1 - X_1$), one can present the probability of the cases (1), (2) and (3) as X_1^2 , X_2^2 and $2X_1X_2$ respectively. λ_{es1} and λ_{es2} are exactly the same as in 2.1 and are calculated by Eqs. (1)–(8). For λ_{es3} we consider the heat flow like λ_{es1} , λ_{es2} and get Eq. (9), as shown in Appendix 1.

Table 2 Observed and predicted conductivities

Run No.	Component 1	Component 2	X_1 [—]	ε [—]	$\lambda_{e,obs}$ [W/m·K]	$\lambda_{e,pred}$ [W/m·K]	Run No.	Component 1	Component 2	X_1 [—]	ε [—]	$\lambda_{e,obs}$ [W/m·K]	$\lambda_{e,pred}$ [W/m·K]
1	G.B. -A	A.P. -A	0.0	0.390	0.120	0.116	8	G.B. -A	G.B. -C	0.0	0.427	0.172	0.173
			0.202	0.389	0.128	0.127				0.216	0.375	0.187	0.196
			0.402	0.386	0.140	0.139				0.423	0.351	0.203	0.211
			0.601	0.376	0.157	0.163				0.623	0.331	0.219	0.221
			0.799	0.365	0.179	0.189				0.815	0.335	0.213	0.218
2	G.B. -A	C.S. -A	1.0	0.375	0.206	0.205	9	C.S. -A	C.S. -C	1.0	0.375	0.208	0.205
			0.0	0.363	0.216	0.214				0.0	0.347	0.298	0.317
			0.103	0.348	0.224	0.233				0.202	0.330	0.316	0.331
			0.206	0.336	0.237	0.252				0.403	0.315	0.331	0.349
			1.0	0.340	0.358	0.361				0.603	0.307	0.346	0.345
3	G.B. -B	A.P. -B	0.0	0.403	0.113	0.112	10	G.B. -A	A.P. -B	0.0	0.403	0.112	0.112
			0.193	0.375	0.129	0.132				0.204	0.379	0.129	0.134
			0.389	0.373	0.142	0.147				0.406	0.368	0.147	0.147
			0.589	0.395	0.151	0.153				0.606	0.396	0.156	0.153
			0.794	0.407	0.176	0.165				0.805	0.408	0.169	0.161
4	G.B. -C	C.S. -C	1.0	0.403	0.198	0.182	11	A.P. -A	G.B. -B	1.0	0.375	0.203	0.205
			0.0	0.415	0.181	0.180				0.0	0.403	0.188	0.187
			0.218	0.389	0.205	0.209				0.209	0.394	0.173	0.176
			0.426	0.397	0.231	0.217				0.413	0.379	0.154	0.163
			0.626	0.365	0.246	0.259				0.612	0.373	0.136	0.151
5	G.B. -A	G.B. -B	0.817	0.351	0.267	0.292	12	C.S. -A	G.B. -B	0.808	0.386	0.126	0.133
			1.0	0.347	0.325	0.317				1.0	0.390	0.114	0.116
			0.0	0.427	0.187	0.173				0.0	0.399	0.185	0.190
			0.211	0.391	0.198	0.193				0.214	0.363	0.235	0.222
			0.416	0.371	0.206	0.204				0.420	0.363	0.259	0.235
6	A.P. -A	A.P. -B	0.616	0.367	0.208	0.206	13	A.P. -B	G.B. -C	0.619	0.360	0.279	0.256
			0.811	0.371	0.210	0.206				0.813	0.354	0.305	0.292
			1.0	0.375	0.206	0.205				1.0	0.340	0.358	0.361
			0.0	0.403	0.114	0.112				0.0	0.427	0.171	0.173
			0.202	0.383	0.117	0.118				0.212	0.403	0.159	0.171
7	G.B. -B	G.B. -C	0.403	0.368	0.120	0.121	14	A.P. -A	G.B. -C	0.418	0.412	0.146	0.150
			0.603	0.374	0.125	0.118				0.617	0.405	0.136	0.138
			0.803	0.388	0.118	0.114				0.811	0.393	0.124	0.128
			1.0	0.390	0.114	0.116				1.0	0.403	0.113	0.112
			0.0	0.427	0.166	0.173				0.0	0.427	0.176	0.173
			0.205	0.415	0.177	0.178				0.215	0.405	0.170	0.170
			0.407	0.387	0.185	0.194				0.420	0.387	0.156	0.162
			0.607	0.379	0.193	0.198				0.620	0.407	0.138	0.137
			0.805	0.387	0.193	0.194				0.813	0.406	0.126	0.123
			1.0	0.403	0.191	0.187				1.0	0.390	0.113	0.116

$\lambda_f=0.0259$ W/m·K

$$\frac{\lambda_{es3}}{\lambda_f} = 2n \left(\frac{2\kappa_1}{2\kappa_1 - 1 - c} \right)^2 \times \left[\ln \left\{ \frac{2\kappa_1 - (2\kappa_1 - 1 - c) \cos \theta_0}{1 + c} \right\} - \left(\frac{2\kappa_1 - 1 - c}{2\kappa_1} \right) (1 - \cos \theta_0) \right] \quad (9)$$

where

$$c = \lambda_{s1} / \lambda_{s2} \quad (10)$$

$$\kappa_1 = \lambda_{s1} / \lambda_f \quad (11)$$

Approximating the heat transfer resistance of case (3) as the summation of heat transfer resistances for upper and lower hemispheres, one can express λ_{es3} by Eq. (12).

$$2/\lambda_{es3} = 1/\lambda_{s1} + 1/\lambda_{s2} \quad (12)$$

The values of λ_{es3} by Eqs. (9) and (12) are evidently equal in the case of $c=1.0$. If the value of c changes

within the range 0.3–3.0, the deviations of the two values of λ_{es3} are about 2% for a system with $\kappa_1=50$ and $\theta_0=\pi/3$.

Consequently, the effective thermal conductivity of solid part λ_{esm} of the mixed granular bed is

$$\lambda_{esm} = X_1^2 \lambda_{es1} + X_2^2 \lambda_{es2} + 2X_1 X_2 \lambda_{es3} \quad (13)$$

We can predict the thermal conductivity of a granular bed by substituting λ_{esm} into Eq. (8) instead of λ_{es} .

2.3 Bed of particles of different diameters and same or different conductivities

When particles of large and small diameter are mixed, the number of contact points might be different from the case where all are of the same diameter. The coordination numbers in a granular bed of mixed coarse and fine particles were referred to by Akao,

Fig. 4 Geometry of contacting two spheres

Fig. 5 Heat transfer model of different-diameter particles

Noda, Toyama and Inoue¹⁾, who proposed a model for the system with diameter ratio above 10.

In this paper, we deal with the system with smaller ratios, 1–3, to which their model cannot be applied.

The geometry of contacting two spheres with radius R_1 and R_2 has to be considered. As shown in Fig. 4, a tangent is drawn from sphere with radius R_1 to neighbouring sphere with radius R_2 . θ_1' is the angle made by the tangent from sphere 1 to sphere 2 and the line between the centers of spheres, where

$$\sin \theta_1' = R_2 / (R_1 + R_2) = r_1 / (1 + r_1) \quad (14)$$

$$r_1 = R_2 / R_1 \quad (15)$$

The next relation is evident from Fig. 4.

$$R_1 \sin \theta_1' = R_2 \sin \theta_2' \quad (16)$$

When the diameters of two adjacent spheres are the same, $r_1 = 1$, the value of $\sin \theta_1'$ is $1/2$. $\sin \theta_1'$ is assumed to be a measure of the area occupied by contacting sphere.

We introduce an assumption as follows.

Assumption (1): When a sphere with radius R_1 contacts one with radius R_2 , the occupying area corresponding to one contact point is proportional to the value of $\sin \theta_1'$. Then the ratio of the area with diameter ratio r_1 and the area with same diameter becomes $2r_1/(1+r_1)$.

On the other hand, for granular beds of binary mixtures, the total number of contact points on the sphere of R_1 with the sphere of R_2 must be equal to

the total number of contact points on the sphere of R_2 with the sphere of R_1 . The number of spheres with R_1 and R_2 are presented as $X_1/(4\pi R_1^3/3)$ and $X_2/(4\pi R_2^3/3)$ respectively. So, we get the next relation.

$$n_{12} X_1 / R_1^3 = n_{21} X_2 / R_2^3 \quad (17)$$

n_{12} means the number of effective contact points on a sphere of R_1 with spheres of R_2 .

Now another assumption is introduced.

Assumption (2): When the sphere of radius R_1 contacts the spheres of R_1 and R_2 , the numbers of contact points are proportional to the volume fractions X_1 , X_2 and the inverses of R_1^k , R_2^k respectively. From Assumptions (1) and (2), n_{11} and n_{12} are given as follows.

$$n_{11}/n_{12} = (X_1/R_1^k) / [(X_2/R_2^k)(1+r_1)/(2r_1)] \quad (18)$$

Similarly, the next equation is given.

$$n_{22}/n_{21} = (X_2/R_2^k) / [(X_1/R_1^k)(1+r_2)/(2r_2)]; \quad r_2 = R_1/R_2 \quad (19)$$

From Assumptions (1) and (2), Eq. (20) is also given.

$$n_{11}/n_{22} = (X_1/R_1^k) / (X_2/R_2^k) \quad (20)$$

n_{12} and n_{21} must satisfy Eq. (17), so the next relation is given.

$$k=2 \quad (21)$$

It is evident that the summation of n_{11} and n_{12} becomes n when the diameters are the same, $r_1 = 1$.

Consequently, the effective coordination numbers of arbitrary cases are predicted based on those of homogeneous spherical particles, n .

$$n_{11} = \frac{X_1/R_1^2}{X_1/R_1^2 + X_2/R_2^2} n, \quad n_{21} = \frac{X_1/R_1^2}{X_1/R_1^2 + X_2/R_2^2} \frac{R_1 + R_2}{2R_1} n \quad (22)$$

$$n_{12} = \frac{X_2/R_2^2}{X_1/R_1^2 + X_2/R_2^2} \frac{R_1 + R_2}{2R_2} n, \quad n_{22} = \frac{X_2/R_2^2}{X_1/R_1^2 + X_2/R_2^2} n$$

The heat flow corresponding to one contact point is assumed to be parallel with the macroscopic temperature gradient within each particle according to the conception of Kunii and Smith⁵⁾. In the region of fluid between particles, the area across which heat flows varies and the direction of heat flow may slant, as shown in Fig. 5. The apparent effective thermal conductivity for one contact point is given by Eq. (23) and its derivation is given in Appendix 2.

$$\lambda_{e12}/\lambda_f = \int_0^{\sqrt{2r_1/(1+r_1)} \sin \theta_0} 2(1+r_1)l dl / [\sqrt{1-l^2}/\kappa_1 + \sqrt{r_1^2 - h^2 l^2}/h^2 \kappa_2 + (1+r_1 - \sqrt{1-l^2}) - \sqrt{r_1^2 - h^2 l^2}/h] \quad (23)$$

where

$$h = R_2'/R_1' \quad (24)$$

$$r_1 = R_2/R_1, \quad r_2 = 1/r_1 \quad (25)$$

$$\kappa_1 = \lambda_{s1}/\lambda_f, \quad \kappa_2 = \lambda_{s2}/\lambda_f \quad (26)$$

$$R'_1 = R_1 \sqrt{2r_1/(1+r_1)} \sin \theta_0 \quad (R'_1 \leq R_1) \quad (27)$$

$$R'_2 = R_2 \sqrt{2r_2/(1+r_2)} \sin \theta_0 \quad (R'_2 \leq R_2) \quad (28)$$

where $\sin \theta_0$ is given by Eq. (7) (Appendix 3).

R'_1 , R'_2 are the radii of area possessed by one contact point for heat transfer, as calculated by Eqs. (27), (28), and they cannot exceed the original particle radii R_1 , R_2 respectively.

Integrating Eq. (23), we get the apparent effective thermal conductivity corresponding to one effective contact point in the system of different-size particles. Since Eq. (23) includes many constants, we integrated Eq. (23) by numerical method in this work.

For granular beds of different-diameter particles the thermal conductivity of solid part λ_{es} is calculated by Eq. (29) with effective coordination numbers and effective thermal conductivities of one contact point mentioned above.

$$\lambda_{es} = (n_{11}\lambda_{e11} + n_{12}\lambda_{e12})X_1 + (n_{21}\lambda_{e21} + n_{22}\lambda_{e22})X_2 \quad (29)$$

λ_{e11} and λ_{e22} are the effective thermal conductivities between like particles and are calculated by Eqs. (30) and (31), similar to Eq. (5).

$$\frac{\lambda_{e11}}{\lambda_f} = 2 \left(\frac{\kappa_1}{\kappa_1 - 1} \right)^2 \left[\ln \{ \kappa_1 - (\kappa_1 - 1) \cos \theta_0 \} - \left(\frac{\kappa_1 - 1}{\kappa_1} \right) (1 - \cos \theta_0) \right] \quad (30)$$

$$\frac{\lambda_{e22}}{\lambda_f} = 2 \left(\frac{\kappa_2}{\kappa_2 - 1} \right)^2 \left[\ln \{ \kappa_2 - (\kappa_2 - 1) \cos \theta_0 \} - \left(\frac{\kappa_2 - 1}{\kappa_2} \right) (1 - \cos \theta_0) \right] \quad (31)$$

Substituting λ_{es} and fractional area of solid part A_s and macro void part A_v into Eq. (8), the effective thermal conductivity can be presented.

3. Comparison of Experimental and Predicted Values

We calculated the effective thermal conductivities of each volume fraction for various binary mixtures according to the prediction method described in section 2 and presented as dotted lines in Fig. 1. The deviations between these lines and measured values were calculated and average values for each run are shown in Table 3. The deviations for all runs were 2–6%, and the predicted values coincided well with experimental ones. The standard deviations between observed and predicted conductivities by Eq. (8) and by prediction formulas of Schumann and Voss⁸⁾, Kunii and Smith⁵⁾, Krupiczka⁴⁾, Zehner and Schlünder¹⁰⁾, Okazaki *et al.*⁶⁾ and Bauer and Schlünder^{2,3)} are listed in Table 4. These formulas are for homogeneous particles, so we calculated each solid conductivity from measured λ_s , and in the case of mixture we decided λ_s as volume fraction average value from each solid conductivity.

Bauer and Schlünder proposed a prediction formula

Table 3 Observed and predicted conductivities

Run No.	Component		m (R_1/R_2)	c ($\lambda_{s1}/\lambda_{s2}$)	E^* [%]
1	G.B.-A	A.P.-A	1.0	4.4	3.19
2	G.B.-A	C.S.-A	1.0	5.8	3.69
3	G.B.-B	A.P.-B	1.0	4.4	3.90
4	G.B.-C	C.S.-C	1.0	3.6	5.14
5	G.B.-A	G.B.-B	1.7	1.0	3.41
6	A.P.-A	A.P.-B	1.7	1.0	2.76
7	G.B.-B	G.B.-C	1.7	1.0	2.95
8	G.B.-A	G.B.-C	2.8	1.0	2.75
9	C.S.-A	C.S.-C	2.8	1.6	4.36
10	G.B.-A	A.P.-B	1.7	4.4	2.57
11	A.P.-A	G.B.-B	1.7	4.4	5.85
12	C.S.-A	G.B.-B	1.7	5.8	5.84
13	A.P.-B	G.B.-C	1.7	4.4	3.59
14	A.P.-A	G.B.-C	2.8	4.4	2.27

$$* E = \sqrt{\frac{1}{k} \sum_{i=1}^k |(\lambda_{e, \text{pred}} - \lambda_{e, \text{obs}}) / \lambda_{e, \text{obs}}|^2}$$

Table 4 Standard deviations of prediction error

Schumann and Voss ⁸⁾	12.60%
Kunii and Smith ⁵⁾	9.95%
Krupiczka ⁴⁾	8.24%
Zehner and Schlünder ¹⁰⁾	8.07%
Okazaki <i>et al.</i> ⁶⁾	8.79%
Bauer and Schlünder ^{2,3)}	36.52%
Present work	3.90%

for packed beds with binary mixtures of spheres, but the values predicted by their method are much larger than our observed values. On the other hand, the predicted values that we calculated for their data by the present method are much smaller than the observed ones. They determined the effective thermal conductivities of the packing in motionless gas by graphic extrapolation of the measured values at different flow rates. However, the effective thermal conductivities without gas flow were much smaller than the terms for convective transport even at the smallest gas flow rate. Further, this would become more evident as the number of contact points of each packed particle increases because the convective term strongly depends on the number of contact points. On the other hand, when the through flow rate becomes smaller, natural convection begins to occur and larger conductivity must be obtained. The larger the particles used, the greater is the effect of natural convection. In the case of binary mixtures of particles the total number of contact points increases relatively because the void fraction decreases and the total number of particles increases, so the effect of natural convection becomes more evident. From the considerations above it can be said that their data may be considerably overestimated and cannot be considered with confidence.

In Fig. 6, the effect of ratio of particle diameters and solid conductivities for predicting deviations are shown partially. In the present work the deviations

Fig. 6 The effect of ratios of particle diameters or solid conductivities on deviations

are generally constant, but by other methods they increase in proportion to the ratios of particle diameters or the ratios of solid conductivities. From the comparison above, it is shown that the prediction formula proposed in this work gives better values of mixtures of granular beds than do others for homogeneous granular beds.

The defect of the present work is that Eq. (23) must be integrated by numerical method expressing the heat flow between smaller and larger particles. We must introduce an approximation formula to simplify the calculation.

We proposed one equation to predict the number of contacts between different-diameter particles. To date neither observations nor simulations have been achieved for three-dimensional random-packed beds. Therefore we have been unable to investigate the appropriateness of Eq. (22), but hope to do so in the future.

Conclusion

For prediction of the effective thermal conductivity of a granular bed of binary mixtures, the authors proposed a new cell model derived from the prediction formula for granular beds of homogeneous particles. In this model the variation of particle sizes and solid conductivities are considered and a prediction formula of coordination numbers of granular beds with two kinds of particles at a voluntary void fraction is presented. Effective thermal conductivities for various granular beds of binary mixtures were observed and compared with predicted ones calculated from void fraction, solid and fluid thermal conductivities, particle diameters and volume fractions of each component. The predicted values coincided with observed ones within 2–6% deviation.

Fig. A-1 Heat transfer model near a contact point

Appendix

1. Derivation of Eq. (9)

The heat flux for one contact point, q , is presented by Eq. (A-1), similar to that of Kunii and Smith⁶⁾.

$$q = \int_0^{\theta_0} dq \quad (\text{A-1})$$

where dq is presented by the next equation from Fig. A-1.

$$dq = 2\pi R^2 \sin \theta \cos \theta d\theta \times \Delta t \left\{ \frac{R \cos \theta}{\lambda_{s1}} + \frac{2R(1 - \cos \theta)}{\lambda_f} + \frac{R \cos \theta}{\lambda_{s2}} \right\} \quad (\text{A-2})$$

Substituting Eq. (A-2) to Eq. (A-1) and introducing c and κ_1 :

$$c = \lambda_{s1}/\lambda_{s2} \quad (\text{A-3})$$

$$\kappa_1 = \lambda_{s1}/\lambda_f \quad (\text{A-4})$$

$$\begin{aligned} q &= 2\pi R \Delta t \lambda_f \int_0^{\theta_0} \frac{\kappa_1 \sin \theta \cos \theta d\theta}{\cos \theta (1+c) + 2(1-\cos \theta)\kappa_1} \\ &= 2\pi R \Delta t \lambda_f \int_0^{\theta_0} \left\{ \frac{-\kappa_1}{2\kappa_1 - 1 - c} \sin \theta \right. \\ &\quad \left. + \frac{2\kappa_1^2 \sin \theta / (2\kappa_1 - 1 - c)}{2\kappa_1 - (2\kappa_1 - 1 - c) \cos \theta} \right\} d\theta \\ &= \pi R \Delta t \lambda_f \left(\frac{2\kappa_1}{2\kappa_1 - 1 - c} \right)^2 \\ &\quad \times \left[\ln \left\{ \frac{2\kappa_1 - (2\kappa_1 - 1 - c) \cos \theta_0}{1+c} \right\} \right. \\ &\quad \left. - \left(\frac{2\kappa_1 - 1 - c}{2\kappa_1} \right) (1 - \cos \theta_0) \right] \quad (\text{A-3}) \end{aligned}$$

As the heat flux per sphere is nq , the effective thermal conductivity of solid part, λ_{es3} , is

$$\begin{aligned} \frac{\lambda_{es3}}{\lambda_f} &= \frac{(nq/\pi R^2)/(\Delta t/2R)}{\lambda_f} = 2n \left(\frac{2\kappa_1}{2\kappa_1 - 1 - c} \right)^2 \\ &\quad \times \left[\ln \left\{ \frac{2\kappa_1 - (2\kappa_1 - 1 - c) \cos \theta_0}{1+c} \right\} \right. \\ &\quad \left. - \left(\frac{2\kappa_1 - 1 - c}{2\kappa_1} \right) (1 - \cos \theta_0) \right] \quad (\text{A-4}) \end{aligned}$$

2. Derivation of Eq. (23)

The heat flux dq across the small area $2\pi l dl$ is presented by the next equation.

$$\begin{aligned} dq &= 2\pi l dl \Delta t / [\sqrt{R_1^2 - l^2}/\lambda_{s1} + \sqrt{R_2^2 - h^2}/h^2 \lambda_{s2} \\ &\quad + (R_1 + R_2 - \sqrt{R_1^2 - l^2} - \sqrt{R_2^2 - h^2})/h \lambda_f] \quad (\text{A-5}) \end{aligned}$$

In this equation, the first term of the denominator corresponds to the heat transfer within the sphere with R_1 , the second term to that within the sphere with R_2 and the third term to that in the region of fluid between two spheres. And in these three

regions the heat flux is constant, so that the second term and the third term have h^2 and h of the denominator respectively.

The total heat flux q is given by Eq. (A-6).

$$q = \int_0^{R_1'} dq \quad (\text{A-6})$$

The apparent effective thermal conductivity for one contact point is as follows.

$$\begin{aligned} \lambda_{e12}/\lambda_f &= (q/\pi R_1^2)/[\Delta t/(R_1 + R_2)]/\lambda_f \\ &= \int_0^{\sqrt{2r_1/(1+r_1)} \sin \theta_0} 2(1+r_1)dl/[\sqrt{1-l^2}/\kappa_1 \\ &\quad + \sqrt{r_1^2 - h^2 l^2}/h^2 \kappa_2 + (1+r_1 - \sqrt{1-l^2})/\kappa_1 \\ &\quad - \sqrt{r_1^2 - h^2 l^2}/h] \end{aligned} \quad (\text{A-7})$$

3. Determination of $\sin \theta_0$

We can calculate the effective contact numbers, n_{eff} , equivalent to that of homogeneous beds for occupying area.

$$n_{eff} = n_{11} + n_{12}(2r_1)/(1+r_1) \quad (\text{A-8})$$

Substituting Eq. (22), n_{eff} becomes n , that is, the effective contacts number of homogeneous granular beds. Therefore, we can calculate $\sin \theta_0$ by Eq. (7).

Nomenclature

A_s	= fractional area of solid part of bed	[—]
A_v	= fractional area of macro void part of bed	[—]
c	= ratio of solid thermal conductivities	[—]
D	= diameter of particle	[m]
E	= deviation between observed and predicted values	[—]
m	= ratio of particle diameters	[—]
N	= number of contact points per particle	[—]
n	= effective number of contact points per hemisphere	[—]

q	= heat flux	[W]
R	= radius of sphere	[m]
X	= volume fraction of component	[—]
Y	= probability of case	[—]

ε	= void fraction of bed	[—]
θ	= angle	[—]
λ	= thermal conductivity	[W·m ⁻¹ ·K ⁻¹]
λ_e	= effective thermal conductivity of bed	[W·m ⁻¹ ·K ⁻¹]
λ_{es}	= apparent thermal conductivity of solid part	[W·m ⁻¹ ·K ⁻¹]

<Subscripts>

f	= fluid
obs	= observed
pred	= predicted
s	= solid

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