

Short communication

Estimating correlations for the effective thermal conductivity of granular materials

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Abstract

Based on the well-known solution of Maxwell [1] for the thermal conductivity of dilute dispersions of spheres and the improved form of Maxwell's equation developed by Chiew and Glandt [2], two general correlation for k_{eff} estimation are proposed. One of them apply to medium dense dispersions ($0.15 \leq \phi \leq 0.85$) and the other one for dense porous materials ($\phi > 0.9$). Both correlating equations encompass a wide range of phase conductivity ratio values. The comparison with experimental measurements shows very good agreement. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Heat transport through complex and often dynamic porous materials is an essential requirement for modern technology such as high performance cryogenic insulation, heterogeneous catalysts and catalytic reactors, construction materials and powder metallurgy, between others.

This subject has been an active research field, both theoretically and experimentally, for over a century.

In general, there are three parameters that are used in this type of studies. One is the relation between the thermal conductivities of the two phases present in the porous material:

$$\alpha = \frac{k_2}{k_1} \quad (1)$$

The reduced thermal polarizability, defined as follows:

$$\beta = \frac{k_2 - k_1}{k_2 + 2k_1} = \frac{\alpha - 1}{\alpha + 2} \quad (2)$$

and the fractional volume ϕ of the dispersed phase (spheres).

In this paper, we are concerned with the effective conductivity k_{eff} estimation of materials consisting in a random assembly of microspheres. In this two-phase dispersion, one of the phases is a continuous matrix of conductivity k_1 , while the other (spheres, dispersed phase) have a conductivity k_2 and a volume fraction ϕ .

The specific objective is to obtain simple but useful correlating equations for the effective thermal conductivity of these materials. It is also considered porous materials obtained by compression and sintering of spherical aggregates or granular materials.

Series [3,4] and asymptotic [5] solutions for several regular arrays of spheres have been derived. However, as was shown by Churchill [6] the k_{eff} of dispersion is independent of the size distribution and arrangement of the spheres. For practical purposes, these parametric dependences are generally negligible when theoretical solutions and experimental data are compared.

Models that are a combination of the series and parallel phase-phase distribution were also suggested [7].

Krupiczka [8], derived a numerical solution for k_{eff} based on a model made up of spheres in cubic lattice.

Churchill [6], in a deep review on this subject, analyzed most the theoretical solutions published. He also made suggestions to produce correlations with minimal parametric dependence.

However, the greater the range of α value considered, the greater the divergence between the predictions of different models.

2. Analysis

Three main groups of materials may be recognized. Type (A): low-dense porous materials (volume fraction of spheres ϕ up to 10%), Type (B): medium-dense materials with ϕ in

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the range 0.15–0.85, and Type (C): high-dense materials (ϕ higher than 90%).

According to the Maxwell [1] model, which assumes randomly distributed and non-interacting spherical particles in a homogeneous continuous medium, the effective conductivity is given by the expression:

$$\frac{k_{\text{eff}}}{k_1} = \frac{1 + 2\beta\phi}{1 - \beta\phi} \quad (3)$$

This model predicts very well effective thermal conductivity of Type (A) porous materials with ϕ up to 10%.

The Type (B) porous materials address a specific class of sol–gel derived porous material used as catalysts support [9]. It is also found in catalyst particles obtained by compression and sintering of alumina, silica or glass powder. In the early 1980s, Chiew and Glandt [2] proposed an improved form of Maxwell's equation (3), which is correct to order ϕ^2 :

$$\frac{k_{\text{eff}}}{k_1} = \frac{1 + 2\beta\phi + (K_2 - 3\beta^2)\phi^2}{1 - \beta\phi} \quad (4)$$

where K_2 takes into account higher order contribution. For computing terms to the order of ϕ^3 in Eq. (4), a linear relation between K_2 and ϕ is assumed, e.g. ($K_2 = a + b\phi$). Parameters a and b , being the functions of β . According to K_2 values presented by Chiew and Glandt and considering that α , for materials and fluid commonly used in catalysis, varies from 10^{-3} to as high as 10^4 ($\beta = -0.499$ to ≈ 1), the best a and b fitting parameters values gives the following expression for estimating k_{eff} :

$$\frac{k_{\text{eff}}}{k_1} = \frac{1 + 2\beta\phi + (2\beta^3 - 0.1\beta)\phi^2 + \phi^3 0.05 \exp(4.5\beta)}{(1 - \beta\phi)} \quad (5)$$

In Table 1, experimental results of (k_{eff}/k_1) obtained with different porous materials are compared with Eq. (5) estimations. The comparison shows a remarkably good agreement. A real challenge was predict the effective thermal conductivity of silver catalyst made by pelleting microporous powder

Table 1
Comparison between predicted Eq. (5) and experimental values of (k_{eff}/k_1) (Type (B) materials)

ϕ	k_1 (W/m K)	k_2 (W/m K)	α	(k_{eff}/k_1) Estimated	(k_{eff}/k_1) Experimental	System	Reference
0.563	0.0261	0.418	16.0	5.1	5.23	Air–coal	[8]
	0.1927		2.17	1.60	1.53	H ₂ –coal	[8]
0.575	0.1724	18	104.4	7.8	5.7	H ₂ –silicium carbide	[8]
	0.0242		743.8	8.4	10.8	Air–silicium carbide	[8]
	0.0146		1232.9	8.5	9.3	CO ₂ –silicium carbide	[8]
	0.1387		129.8	7.9	5.1	He–silicium carbide	[8]
0.63	0.0332	1.858	56.0	9.3	9.8	Air–sand	[8]
0.612	0.3460	133.5	385.8	9.95	10.7	Copper–ethanol solution	[8]
	0.527		253.3	9.8	12.8	Copper–glycerol solution	[8]
0.595	0.166	11.61	69.9	8.2	8.3	Uranium oxide–He $d = 81 \mu\text{m}$	100 °C [8]
	0.1811		64.1	8.1	7.82		160 °C
	0.1927		60.3	8.0	7.30		200 °C
0.58	0.0306	18.58	607.2	8.62	10.2	Air–MgO $d = 0.2 \text{ mm}$	100 °C [8]
	0.0332		559.6	8.6	9.8		140 °C
	0.0344		540.1	8.59	10.1		160 °C
	0.0369		503.5	8.58	9.13		200 °C
0.575				8.1	8.21		
0.6	0.0257	4.148	161.4	9.0	8.86	Air–sand	[7]
0.62				9.9	9.29		
0.645				11.2	9.94		
0.55				6.5	6.6		
0.55	0.0257	1.424	55.4	6.5	6.6	SiO ₂ gel	[15]
0.62	0.0257	1.259	49.0	8.6	7.1	Glass beads 29–80 μm	[15]
0.14	20	0.026	1.3×10^{-3}	0.80	0.77	Air in alumina	[6]
0.55				3.6	4.0	Alumina	
0.725	0.0260	0.21	8.1	5.7	6.6	Behmite	[16]
0.866				8.8	8.3	Powder	
0.31	0.461	34.6	75	2.71	2.6	Lead in	[6]
0.43				4.1	3.8	Acrylic res.	
0.77	0.0311	0.45	14.5	10.1	9.8	Air/Ni/W catalyst	[8]
0.655	0.0311	0.293	9.4	5.2	5.7	Air/Cr/Al catalyst	[8]
0.71	0.0311	0.243	7.8	5.3	4.45	Air/Co/Mo catalyst	[8]
0.655	0.0311	0.36	11.6	5.8	5.8	Air/SiO ₂ –Al ₂ O ₃ /catalyst	[8]
0.70	0.0311	0.21	6.8	4.6	4.2	Air/Pt/Al ₂ O ₃ /catalyst	[8]
0.59	0.026	427	1.64×10^4	9.2	9.3	Silver	[10]
0.71				15.1	17.0	Powder	

Table 2
Comparison between predicted [Eq. (11)] and experimental values of (k_{eff}/k_1) (Type (C) materials)

ϕ	k_1 (W/m K)	k_2 (W/m K)	α	β'	Estimated (k_{eff}/k_1)	Experimental (k_{eff}/k_1)	System	Reference
0.9	0.0257	30	1167.3	-0.5	1000.9	992	304L s.s. powder	[17]
0.925	0.0257	48.3	1879.4	-0.499	1675	1606	Ni alloy powder	[12]
0.959	0.1125	0.0257	0.228	0.5296	0.245	0.26	Polystyrene Foam	[18] [19]
0.945	3.632	0.0257	7.1×10^{-3}	0.9791	8.3×10^{-3}	9.5×10^{-3}	Glass foam	[18]
0.929	0.8648	0.0261	3.02×10^{-2}	0.9146	0.0365	0.0379	Epoxy Resin foam	[18] [19]

particles. The experimental results showed very low conductivities in comparison with the high value of k_2 for solid silver. Eq. (5) was able to predict (k_{eff}/k_1) values that differ in no more than 10% with experimental data [10] (last two rows in Table 1).

For porous materials of the Type (C), Bauer [11] derived the following relation for metal powder packing of arbitrary pores shapes and concentration:

$$\frac{k_{\text{eff}}}{k_1} = \alpha \phi^{3/2} \quad (6)$$

This equation is almost identical, above $\phi = 0.85$, to that obtained by Maxwell:

$$\frac{k_{\text{eff}}}{k_1} = \alpha \frac{2\phi}{3 - \phi} \quad (7)$$

Few years' ago, Argento and Bouvard [12] have been empirically chose the following relation for predicting k_{eff} :

$$\frac{k_{\text{eff}}}{k_1} = \alpha \left(\frac{\phi - \phi_0}{1 - \phi_0} \right)^{(3/2)(1 - \phi_0)} \quad (8)$$

where ϕ_0 is the initial density of the packing before densification (compression).

In Eqs. (6)–(8), k_{eff} depend only on ϕ and k_2 , but not on k_1 . This is the reason these equations predict very well the effective conductivity of dense porous materials with high values of α ($\alpha \geq 10^3$). The contribution of the continuous phase is insignificant in this case.

Taken into account the equation derived by Maxwell for a dilute dispersion of spheres, it is reasonable to think that a similar relation should be a solution for the diametrically opposed situation, $(1 - \phi) < 0.1$. Under these conditions, the notion of touching particles need to be replaced by a model of dense matrix (compressed packed of spheres) with distributed pores. Then the same model proposed by Maxwell can be assumed but with changing roles. Instead of ϕ , β' and k_{eff}/k_1 now $(1 - \phi)$, β' and k_{eff}/k_2 should be used in Eq. (3), given:

$$\frac{k_{\text{eff}}}{k_2} = \frac{1 + 2\beta'(1 - \phi)}{1 - \beta'(1 - \phi)} \quad (9)$$

with

$$\beta' = \frac{k_1 - k_2}{k_1 + 2k_2} = \frac{1 - \alpha}{1 + 2\alpha} \quad (10)$$

Nevertheless, k_2 continues being the conductivity of the spheres that can be of the “fluid” or “solid” type.

Considering the relation generally used, that is the ratio of effective conductivity to that of the phase between spheres (k_1):

$$\frac{k_{\text{eff}}}{k_1} = \alpha \frac{[1 + 2\beta'(1 - \phi)]}{[1 - \beta'(1 - \phi)]} \quad (11)$$

Surprisingly, the experimental data of high dense porous materials are very well represented by expression (11) for a large range of α values. Table 2 shows a comparison between experimental and calculated (k_{eff}/k_1) values of high dense materials with α ranging from 0.0071 to 1879.4. This model encloses porous materials of “air-cell” (foam) and “granular-solid” type both with $\phi > 0.9$.

For aggregates of touching impenetrable spheres the maximum values of ϕ are 0.534, 0.680 and 0.740, for simple cubic, centered-cubic and face-centered cubic arrays [6], respectively. Nevertheless, when the porous material is prepared by compression and sintering of spherical or granular aggregates substantial overlapping take place. Then, ϕ values higher than the maximum one can be achieved. Eq. (11) encompass this type of porous materials.

The influence of convection can be neglected for the cases here studied where granular materials and spheres of diameters smaller than 0.1 cm was considered [8,13]. The radiative heat transfer also contributes to the effective thermal conductivity. Although this effect can be important at temperatures above 200 °C, it has been shown [13,14] that radiation contribution was negligible compared with the solid phase conduction of metal powders used in powder metallurgy. However, we must point out that the correlating Eqs. (5) and (11) do not take into account the effects of convection and radiation.

3. Conclusions

1. The Maxwell's solution for the thermal conductivity of low dense porous materials besides to predict very well (k_{eff}/k_1) values provides an insight for developing correlations for Types (B) and (C) materials.
2. The correlating equation constructed from the Maxwell solution and the extended solution of Chiew and Glandt

provides excellent estimations of (k_{eff}/k_1) for porous materials with α ranging from 10^{-3} to 10^4 and ϕ from 0.15 to 0.85.

3. The improved solution for high dense porous materials, Eq. (11), predicts (k_{eff}/k_1) values in close agreement with experimental data for a large range of α values. This model encompasses the “air-cell” (foam) and “granular-solid” type dispersions.

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References

- [1] J.C. Maxwell, A Treatise on Electricity and Magnetism, Vol. 1, Clarendon Press, Oxford, 1873, p. 365.
- [2] Y.C. Chiew, E. Glandt, The effect of structure on the conductivity of a dispersion, *J. Coll. Interf. Sci.* 94 (1983) 90–104.
- [3] D.A. de Vries, The thermal conductivity of granular materials, *Bull. Inst. Intern. Froid Annexe* 1952-1 32 (1952) 115–131.
- [4] C. Fernandes, N. Mendes, P. Philippi, Determinação de correlações matemáticas para condutividade térmica de materiais porosos, in: Proceedings of the Eight Latin American Congress on Heat and Mass Transfer, Veracruz, Mexico, 2001, pp. 620–625.
- [5] G.K. Batchelor, R.W. O’Brien, Thermal or electrical conductivity through a granular material, *Proc. Roy. Soc., London*, A355 (1977) 313–333.
- [6] S.W. Churchill, The thermal conductivity of dispersions and packed beds: an illustration of the unexploited potential of limiting solution correlation, in: A. Mujumdar, R. Mashelkar (Eds.), *Advances in Transport Processes*, Vol. IV, Wiley, New Delhi, 1986, pp. 394–418.
- [7] I.H. Tavman, Effective thermal conductivity of granular porous materials, *Int. Commun. Heat Mass Transfer* 23 (1996) 169–176.
- [8] R. Krupiczka, Analysis of thermal conductivity in granular materials, *Int. Chem. Eng.* 7 (1967) 122–144.
- [9] S.C. Reyes, E. Iglesia, Y.C. Chiew, Monte Carlo simulation of effective diffusivities in three-dimensional pore structure, *Proc. Mater. Res. Soc.* 195 (1990) 553–561.
- [10] S. Masamune, J.M. Smith, Thermal conductivity of porous catalyst pellets, *J. Chem. Eng. Data* 8 (1963) 54–58.
- [11] T.H. Bauer, A general analytical approach toward the thermal conductivity of porous media, *Int. J. Heat Mass Transfer* 36 (1993) 4181–4191.
- [12] C. Argento, D. Bouvard, Modeling the effective thermal conductivity of random packing of spheres through densification, *Int. J. Heat Mass Transfer* 39 (1996) 1343–1350.
- [13] C.L. Tien, K. Vafai, Convective and radiative heat transfer in porous media, *Adv. Appl. Mech.* 27 (1990) 225–281.
- [14] C. Argento, D. Bouvard, A ray tracing method for evaluating the radiative heat transfer in porous media, *Int. J. Heat Mass Transfer* 39 (1996) 1079.
- [15] C.N. Satterfield, *Heterogeneous Catalysis in Industrial Practice*, McGraw-Hill, New York, 1991, pp. 510–515.
- [16] R.A. Mischke, J.M. Smith, Thermal conductivity of alumina catalyst pellets, *Ind. Eng. Chem. Fundam.* 1 (1962) 288–292.
- [17] J.S. Agapiou, M.F. de Vries, An experimental determination of the thermal conductivity of a 304L stainless steel powder metallurgy material, *ASME J. Heat Transfer* 111 (1989) 281–286.
- [18] R.H. Perry, C.H. Chilton, *Chemical Engineers’ Handbook*, 6th Edition, McGraw-Hill, New York, 1984.
- [19] D.R. Lide (Ed.), *CRC Handbook of Chemistry and Physics*, 73rd Edition, CRC Press, Boca Raton, FL, 1993.