

Models for the Prediction of the Thermal Conductivities of Powders

Samuel S. Xue and Joel W. Barlow
Chemical Engineering Department
University of Texas at Austin
August 12, 1991

Abstract

Five models and equations for the prediction of the thermal conductivities of powders in the literature are compared with the data obtained in the experiments of the authors. A new modified model for the correlation of the experimental data is presented.

Key words: differential scanning calorimetry, porosity, solid content, specific heat, thermal conductivity.

Introduction

Knowledge of the thermal properties, especially thermal conductivity, k , of powders is essential to the study of many technical processes, including Selective Laser Sintering (SLS). This paper reviews models for predicting k found in the literature and compares the predictions of these models with experimental measurements on powder beds comprising a variety of materials, including various polymers, glass, and tin.

Literature Review

Five models for predicting the thermal conductivities of powders are reviewed here. All of the models attempt to predict the variation in k with bed porosity from the properties of the solid and of air. The models differ somewhat from one another by the "mixing rules" employed.

Maxwell-Eucken Model. This model was first derived by James Clerk Maxwell for the prediction of the electrical conductivity of heterogeneous media. A. Eucken (1940) suggested its use for predicting the thermal conductivity of powders and loose materials. It reads as follows:

$$k = k_s \frac{1 - (1 - ak_a/k_s)\epsilon}{1 + (a - 1)\epsilon} \quad (1)$$

when the solid content of the powder is greater than 50% by volume, where $a = 3k_s/(2k_s + k_a)$, k is the thermal conductivity of the powder bed, k_s and k_a are the thermal conductivities of the solid and the air at the same temperature respectively, and ϵ is the porosity of the volume fraction of air.

When the solid content of the powder is less than 50%:

$$k = k_a \frac{1 - (1 - ak_s/k_a)(1 - \epsilon)}{1 + (a - 1)(1 - \epsilon)} \quad (2)$$

where $a = 3k_a/(2k_a + k_s)$.

These equations are derived for small values of ϵ and $(1 - \epsilon)$, and their extension to ϵ or $(1 - \epsilon)$ values near 0.5 may be questionable.

Yagi-Kunii Model Yagi and Kunii (1957) proposed a model for porous beds of metals and other materials with high thermal conductivities. They considered interparticle radiation transfer as well as conduction to be important. Their model, at low temperatures where radiation transfer between particles is not important, takes the form,

$$k = \beta k_s(1 - \epsilon)/(1 + \phi k_s/k_a) \quad (3)$$

where

$$\phi = 0.02 \cdot 10^{(2 \cdot (\epsilon - 0.3))}, \quad \beta \approx 1.0, \quad \text{and the other symbols are as defined above.}$$

Yagi and Kunii considered k_s , k_a , and porosity in their treatment. But the present writers noticed that when powders of low porosities (e.g. porosity = 0.605, or 0.687) were used, their method gave too low values, and for some powders (e.g. glass powders), their method gave too high (two or three times higher) values.

Saxena, Chohan and Gustafsson Model. These authors (1986) provide two set of equations for the prediction of thermal conductivities of granular materials. The first set of equations is for small cubic dispersions in the continuous medium, while the volume fraction of these dispersions are not very small.

$$k = k_a(1 + 3.844(1-\epsilon)^{2/3}) \quad 0 < (1-\epsilon) < 0.5 \quad (4a)$$

$$k = k_s(1 - 1.545\epsilon^{2/3}) \quad 0 < \epsilon < 0.5 \quad (4b)$$

These authors thought the natural loose and granular materials are mixtures of solid and gas phases where each of the phases occupies a large volume fraction (0.2 to 0.8) of the sample. In this situation neither of the phases (solid or gas) provides the continuous medium. The authors suggested the following set of equations to consider for the case of a loose, granular, two-phase system which consists of small volume fractions of solid or gas phase in the effective continuous medium (so as to produce porosities ranging between 0.2 and 0.8):

$$k = k_{ec}(1 + 3.844(0.5-\epsilon)^{2/3}) \quad \epsilon < 0.5 \quad (5a)$$

$$k = k_{ec}(1 - 1.545(\epsilon-0.5)^{2/3}) \quad \epsilon > 0.5 \quad (5b)$$

In the above equations,

k_{ec} is the thermal conductivity of the effective continuous medium,

$$k_{ec} = 0.6132(k_a k_s)^{1/2}$$

It has been found by the present writers that generally Eqns. (4a) and (4b) gave higher values than the actual data, and Eqns. (5a) and (5b) gave very low values.

Heiji Enomoto, et al. Model. In their experiments, the effective thermal conductivities of coal powder and itaconic acid were measured as a function of the porosity and the particle size, as well as the temperature. For a sample of coal powder, they got the correlating equation:

$$k = (0.006T + (5 - 0.006T)(1 - \epsilon) + 20D_p - 1.5) \cdot 0.0001 \quad (6)$$

where k is the bed conductivity, cal/cm-sec-K, T the temperature, K, and D_p the particle size, cm. The correlation is limited to $0.35 \leq \epsilon \leq 0.6$, D_p between 0.004 and 0.015 cm, and temperature between 300 and 370 K.

The thermal conductivity of the solid is not clearly shown in Equation (6) although it may be included in the constants of the correlation. Despite this, Equation (6) is found to predict k for plastic powder beds about as well as the other models. This may suggest that the effective conductivity of the powder is a strong function of the conductivity of the gas and not of the solid.

Deissler-Boegli Model. The authors of this article gave a summary of results of their investigation of effective thermal conductivities of powders:

1) The effective conductivity of the powder is a strong function of the conductivity of the gas. For instance, the effective conductivity of magnesium oxide powder in helium was about five times that for the same powder in argon.

2) The effective conductivity of a powder becomes nearly independent of pressure at some ratio of mean free path of gas molecules to a characteristic dimension of the powder particle. For all of the powders investigated the breakaway pressure could be predicted by an equation based on the kinetic theory of gases.

3) A reasonably good correlation of the data was obtained by plotting the ratio of effective conductivity to gas conductivity against the ratio of solid conductivity to gas conductivity for various porosities. A relaxation solution for heat flow through spheres in cubical array (porosity = 0.475) was in reasonable agreement with the group of data for porosities from 0.42 to 0.50.

The Fig 8 of that article is appended below.

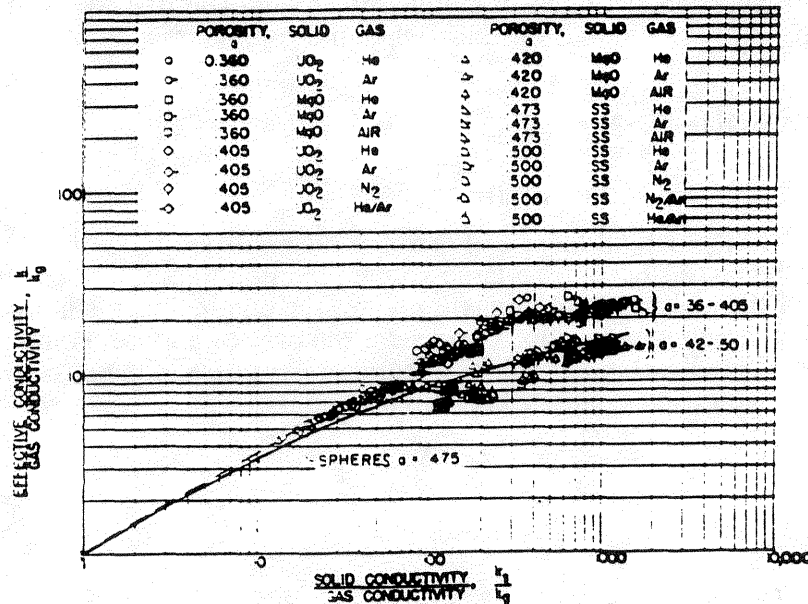


Fig. 1. The graph of Deissler et al. for the correlation of their experimental effective thermal conductivities of powders in various gases and comparison with solution for spheres.

The present writers found that the thermal conductivities found from the curve of this graph were a little higher than the actual data we collected.

Temperature Dependent Properties. In all the calculations for the k of air with the change of temperature for the various models, the present writers used the following relationship from the handbook:

$$k_a = 5.86 \cdot 10^{-5} + 1.7639 \cdot 10^{-7} T \quad \text{cal}/(\text{cm-sec-}^\circ\text{C}) \quad (7)$$

where T, °C is between 0 and 400°C.

The temperature dependency of k's of the solid was either measured directly by the authors or taken from the literature. In using these varying values for the solid thermal conductivities, a better approximation to the experimental data of the authors by the models in literature is often seen, than just using the solid thermal conductivity data from literature. For some of the calculations, a relationship found by tests done on polystyrene: $k_s = 3.203 \cdot 10^{-4} + 1.9193 \cdot 10^{-6} T$ cal/(cm-sec-°C). (T [=] °C) was used for approximation of the k of the solid polymer (E.C. Bernhardt, "Processing of Thermoplastic Materials"(1959),pp.630-631). For polycarbonate solid, the following relationship between k and temperature was found by the experiments of the authors.

following relationship between k and temperature was found by the experiments of the authors. The authors used a solid polycarbonate cylinder with a hole drilled at the center for the placement of a thermal couple, and proceeded with the same procedure mentioned above for the measurement of the k of powder beds to measure the k of the solid. The equation found by the authors for the polycarbonate solid cylinder was : $k_s = 0.0985 + 0.0036696T - 0.0000501T^2 + 1.7639 \cdot 10^{-7}T^3$ where k is in W/m-K, and T in °C. Specific heats were measured directly and found also to increase with increasing temperature.

For solid tin, the data of the change of k according to temperature was taken from Perry's Chemical Engineers' Handbook, 6th Edition, edited by Perry and Green.

Experimental Work

Differential Scanning Calorimetry (DSC) was used to obtain the specific heats. An unsteady heat transfer technique was developed for measuring bed thermal conductivity, Xue (1990). A synopsis of the methods of measurement is given here.

Differential Scanning Calorimetry

A sapphire sample was used each time as a standard after the baseline had been run. The tested standard and samples were held at two limiting temperatures (between these two limits, the samples were tested for the specific heats) for several minutes during every run. Each time, a line is drawn at the bases of the held temperature readings. The heights of the curves above this line at various temperatures were proportioned to the specific heats of the sapphire and the samples at the temperatures.

Unsteady State Conduction Measurements

An unsteady state method for the measuring of the thermal conductivities of powders has been used. A cylindrically symmetric aluminium tube, open at one end was first packed with the sample powder. A thermal couple was placed at the center of the tube from the opening above. This filled-up tube was brought to a constant temperature in a thermostat. Subsequently, the measuring tube was placed in a bath which was held constantly at a higher temperature, and the temperature development at the middle of the measuring tube was recorded by a computer. Small changes in bath temperatures, typically 10°C, were used and the thermal properties were assumed constant within that change to permit simplified analysis. The thermal diffusivity of the sample powder was calculated out from the temperature changing profile according to time at the center of the tube, and subsequently thermal conductivity was calculated from the specific heat and the bulk density of the sample powder.

Results and Discussion

Figures 2-7 summarize comparisons between observed bed thermal conductivities and those predicted by the models above for polycarbonate (PC), poly(vinyl chloride) (PVC), rubber modified poly(styrene-co-acrylonitrile) (ABS), nylon, glass, and tin powders. With the exception of tin powder beds, Figure 7, all thermal conductivities are observed to increase with increasing temperature as a result of k_a , Equation (7), and k_s both increasing with increasing temperature.

Generally for the non-metal powders, the Maxwell-Eucken, Enomoto, and Yagi-Kunii models most consistently predict k values near those which are experimentally observed. The Deissler and Saxena models, generally yield the most inaccurate predictions. Even within these generalities, however, there are exceptions. For example, the Deissler model does a better job of predicting k for PC beds than does either the Maxwell-Eucken or Enomoto models. The k of the tin powder bed is fairly well predicted by all except the Maxwell-Eucken and Saxena models. But it is to be noticed that all the models predict the increase of k according to increasing temperature, which is not the case for tin powder according to our experiments. (k decreases from 0.994 W/m-K at 41°C to 0.412 W/m-K at 83.5°C, according to our experiments.)

k of FC Compared with Various Eqns.

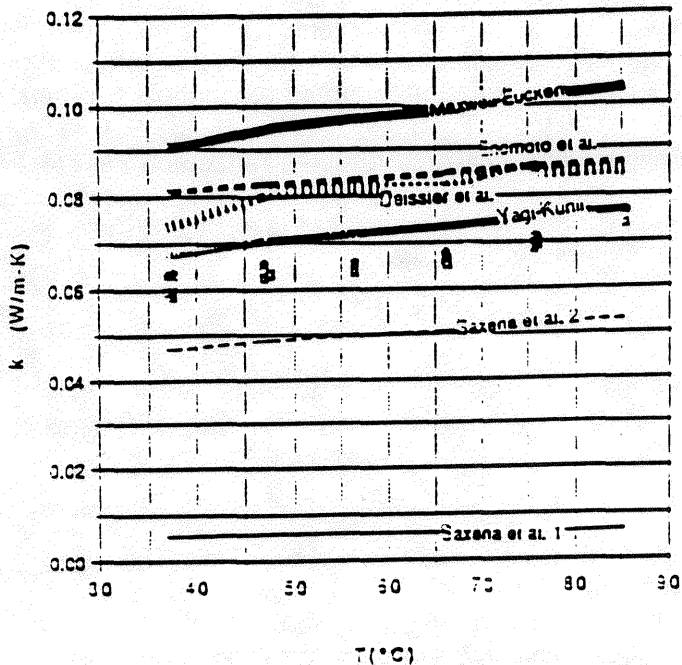


Figure 2. Comparisons of experimental and calculated values of k for polycarbonate powder ($\epsilon = 0.4976$) vs. temperature.

k of PVC Powder Compared with Eqns.

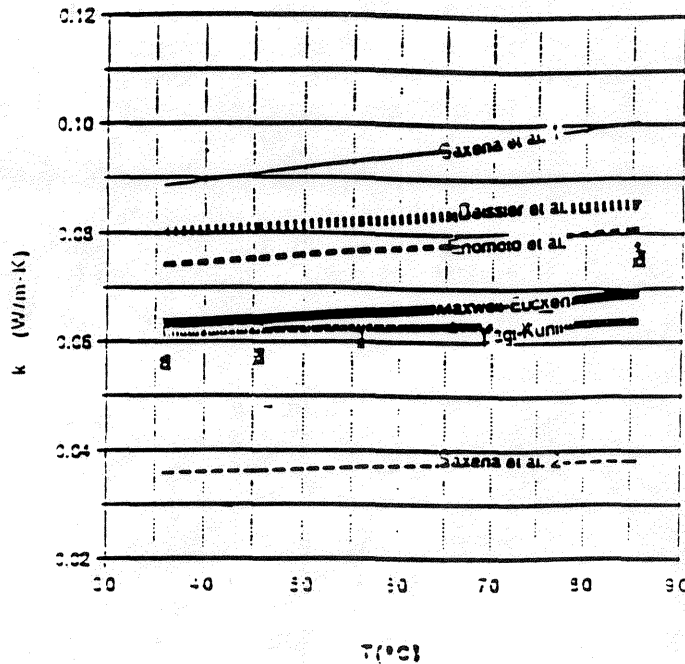


Figure 3. Comparisons of experimental and calculated values of k for PVC powder ($\epsilon = 0.550$) vs. temperature.

k of ABS Powder Compared with Various Eqns.

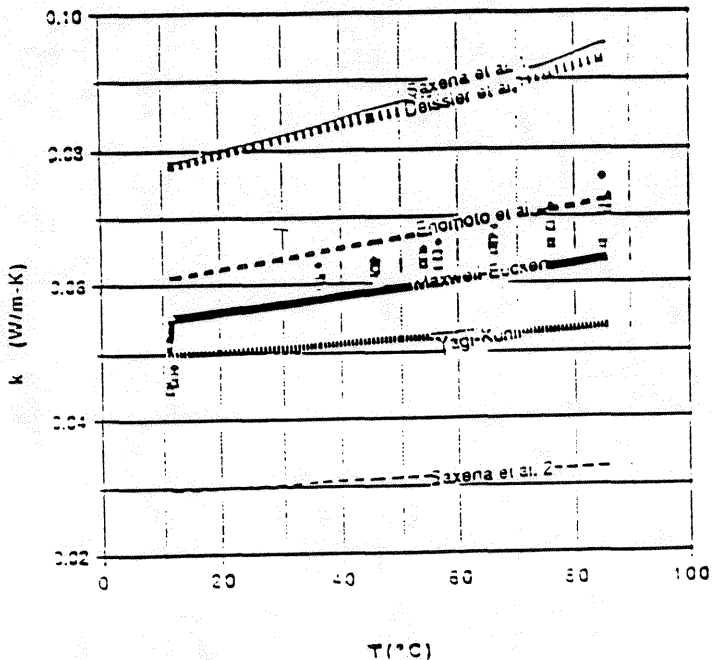


Figure 4. Comparisons of experimental and calculated values of k for ABS powder ($\epsilon = 0.605$) vs. temperature.

k of Nylon Powder Compared with the Eqns.

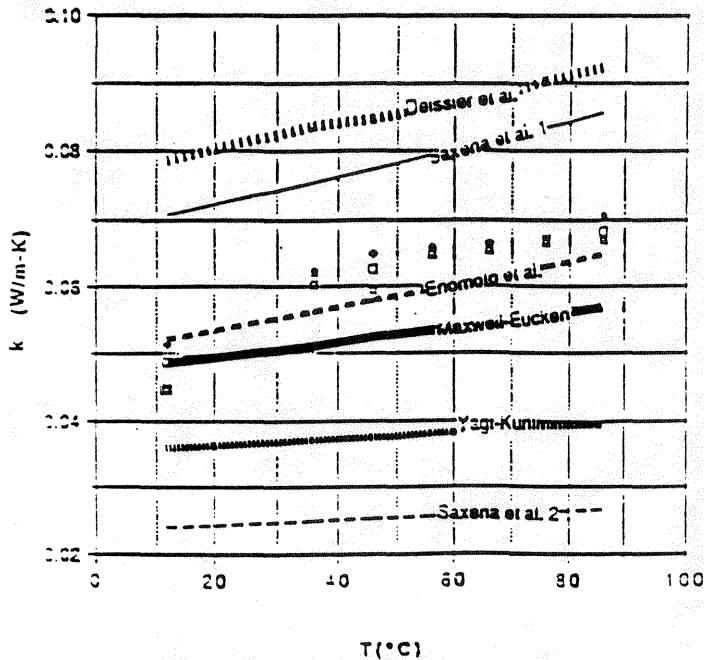


Figure 5. Comparisons of experimental and calculated values of k for nylon powder ($\epsilon = 0.687$) vs. temperature.

k of Glass Powder Compared with Various Eqns. k of Tin Powder Compared with Various Eqn.

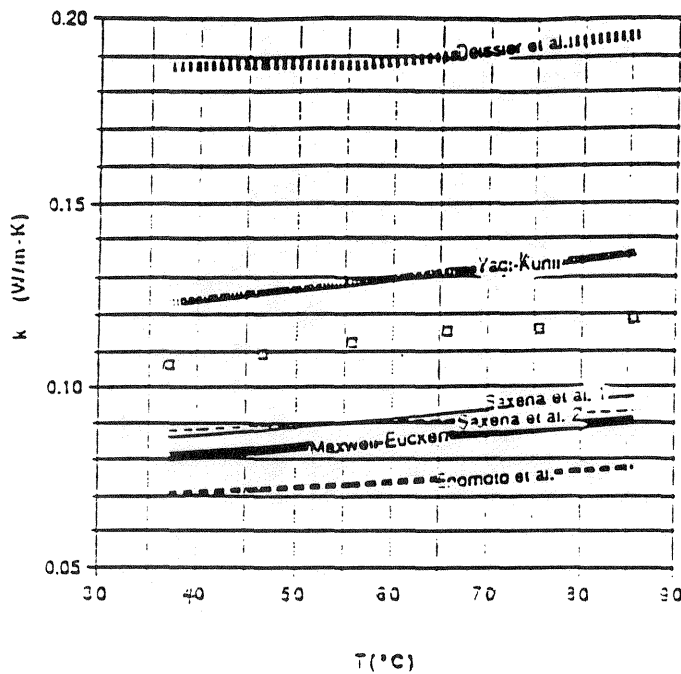


Fig.6. Comparisons of experimental and calculated values of k for glass powder ($\epsilon = 0.584$) vs. temperature

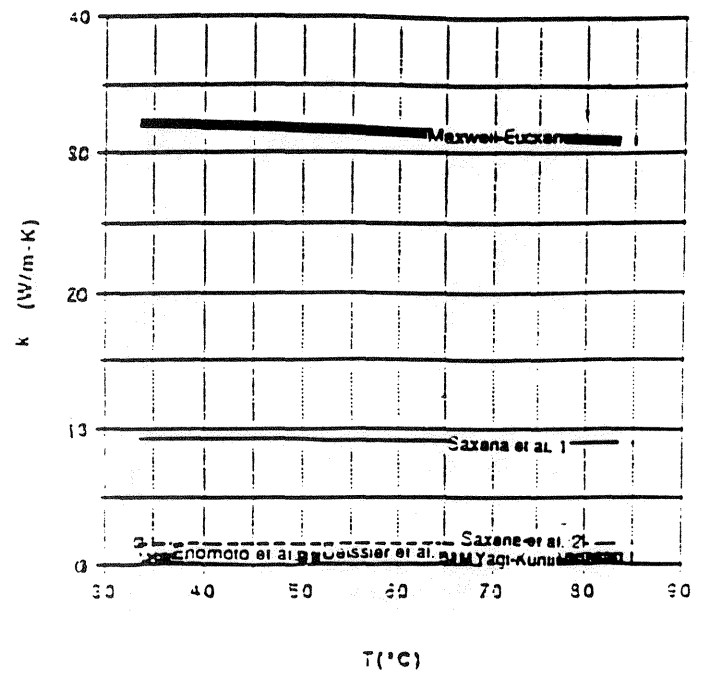


Figure 7. Comparisons of experimental and calculated values of k for tin powder ($\epsilon = 0.417$) vs. temperature

In an attempt to improve the prediction of bed thermal conductivities, we have selected and empirically revised the Yagi-Kunii model to increase the influence of temperature through the effect of temperature on k_s and k_a , and to decrease slightly the influence of porosity to yield,

$$k = (6.3 + 22\sqrt{0.09k_s - 0.016}) \times \frac{k_s(1-\epsilon)}{\frac{k_s}{k_a} [10^{(0.523-0.594\epsilon)}] - 1} \quad (8)$$

where k is the effective thermal conductivity of the powder,
 k_s is the thermal conductivity of the solid,
 k_a is the thermal conductivity of the gas (air) within the powder at the same temperature,
 ϵ is the porosity of the powder, and
 $1 - \epsilon$ is the solid content of the powder.

A comparison of the predictions from equation (8) with experiments are shown in Figures 8-12 for beds of non-conductors. Generally equation (8) fits these data quite well. The porosities used in our experiments ranged from about 0.4 to 0.7. The prediction for tin powder beds, Figure 13, just gave an approximate average value for the whole range of variations, but these were the same cases for the Yagi-Kunii, Saxena et al., Enomoto, and Deissler models.

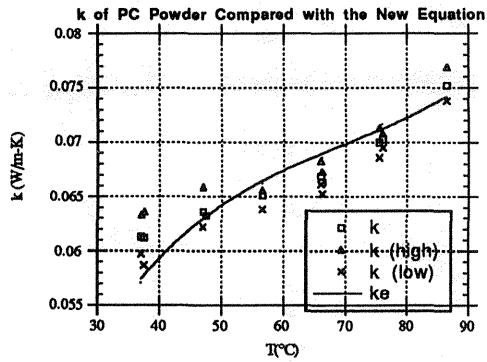


Figure 8. k of PC powder compared with the curve obtained from Equation (8)

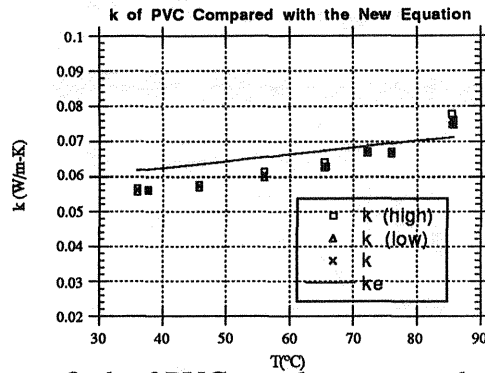


Figure 9. k of PVC powder compared with the curve obtained from Equation (8).

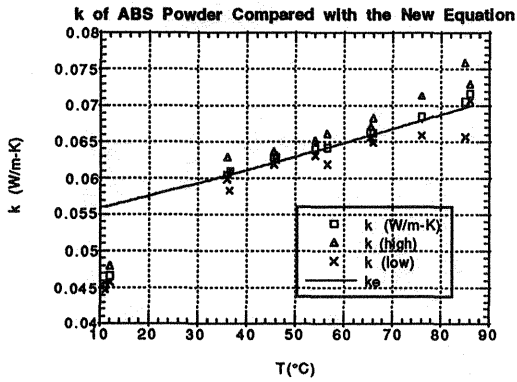


Figure 10. k of ABS powder compared with the curve obtained from Equation (8).

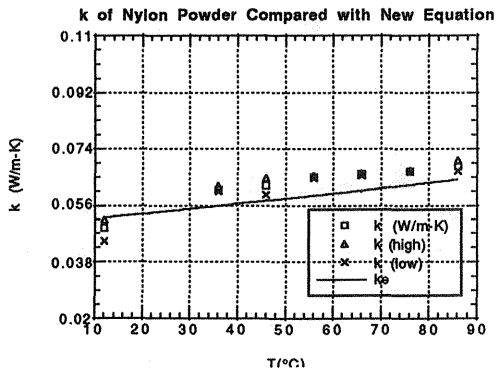


Figure 11. k of Nylon Powder compared with the curve obtained from Equation (8).

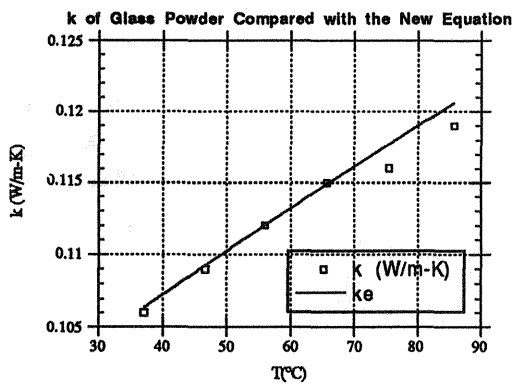


Figure 12. k of Glass Powder compared with the curve obtained from Equation (8).

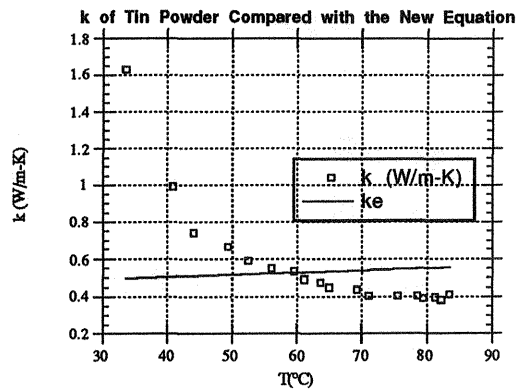


Figure 13. k of tin powder compared with the curve obtained from Equation (8)

Conclusion

The thermal conductivities of powders are smaller than those of their corresponding solids. The thermal conductivities of polymer powders and glass powder show an increase as the temperature is raised. The thermal conductivity of tin powder decreases as the temperature is raised. The equation introduced by the authors shows a better approximation to all the experimental data than can be obtained from the literature. Due to the large thermal excursions normally seen in the SLS process, the influence of temperature is possibly more important to the thermal conductivity of powder beds than the influence of porosity. Further studies in this realm are still to be done.

References

1. Samuel S. Xue and Joel W. Barlow: "Thermal Properties of Powders" in the Solid Freeform Fabrication Symposium Proceedings, the University of Texas at Austin, Austin, Texas (1990), pp.179-185.
2. Max Jakob: Heat Transfer, Vol. I, Wiley, New York (1949), pp. 83-85.
3. Maxwell, James C.: A Treatise on Electricity and Magnetism, vol. I, Oxford (1873), p365.
4. A. Eucken: "Allgemeine Gesetzmäßigkeiten für das Wärmeleitvermögen verschiedener Stoffarten und Aggregatzustände," Forschung auf dem Gebiete des Ingenieurwesens. vol.11, No.1 (1940), pp.6-20.
5. Yagi and Kunii: "Studies on Effective Thermal Conductivities in Packed Beds," J. AIChE, vol. 3, No. 3 (1957), pp. 373-381.
6. Saxena, Chohan and Gustafsson: "Effective Thermal Conductivity of Loose Granular Materials," J. Phys. D: Appl. Phys. **19** (1986), pp.1625-1630.
7. Heiji Enomoto, Tomoyuki Fukuchi, Hiroshi Kiyohashi, Munesuke Kyo, and Shozo Tanaka: "Measurement of Thermal Conductivity of Powdery Materials and the Influence of the Temperature Dependency of Thermal Conductivity," in Safety Technology (in Japanese), Vol. 26, No. 2 (1987), pp.85-90.
8. R. G. Deissler and J. S. Boegli: "An Investigation of Effective Thermal Conductivities of Powders in Various Gases", "Transactions of the ASME, October(1958), pp. 1417-1425.
9. E.C. Bernhardt, "Processing of Thermoplastic Materials", Van Nostrand Reinhold, New York (1959), pp.630-631.
10. David L. Swift: "The Thermal Conductivity of Spherical Metal Powders Including the Effect of an Oxide Coating", International Journal of Heat and Mass Transfer, Vol.9 (1966), pp.1061-1074.