Prediction of the Thermal Conductivity of Beds Which Contain Polymer Coated Metal Particles

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Abstract

Structural parts of ceramics or metals can, in principle, be made by laser sintering polymer coated ceramic or metal powders, followed by conventional methods for removing the binder and sintering in ovens. Understanding the laser sintering of coated materials requires knowledge of the behavior of beds containing composite particles. Many correlations for predicting the effective thermal conductivity of a bed of solid particles exist in literature, but little work has been done on beds of coated particles. We coated lead shots (high conductivity) with a styrene acrylic acid copolymer (low conductivity) to study the effect of coating thickness on the thermal conductivity. The thermal conductivity of the coated particle bed was found to drop rapidly in the beginning and then level off with increasing coating thickness. We also developed an equation that yields the equivalent conductivity of a coated spherical particle subjected to axial heat flow. The predicted results agree with the experimental measurements of bed conductivity obtained by an unsteady state method.

Introduction

The measurement of the effective thermal conductivity of coated particle beds has assumed increasing importance following the development of the Selective Laser Sintering (SLS) process that allows for production of parts from powders. The application of this process for production of metal and ceramic parts requires the respective metal and ceramic powders to be coated with low melting binders or polymers with low glass transition temperatures. These powders are then sintered by the action of a moving laser beam. One of the important parameters in this process is the effective thermal conductivity of the powder bed, which determines the heat penetration into the bed and controls the sintering rate.

Effective thermal conductivity of particle beds has been studied in the past and correlations for predicting the same are available in the literature. However only one paper has extended the approach for predicting the effective thermal conductivity of coated particle beds [D.L. Swift, 1966]. This method assumes a fixed bed voidage (0.4) due to orthorhombic packing and this limits the applicability of the predictive equation. In the present work we obtained an expression for the equivalent conductivity of a coated sphere and used this in conjunction with available correlations for predicting the effective bed conductivity. The resistor model of Woodside and Messmer (1961), the orthorhombic model of Swift (1966), the correlation of Yagii and Kunii (1957) as well as the correlation provided by Saxena et al. (1986) were used to predict the bed conductivity and the results are compared with the experimental data.
Experimental Procedure

Lead spheres 1.33 mm diameter were coated with a styrene acrylic copolymer in a pill coater. The coating thickness was varied and the bed conductivity was then measured. The thermal conductivity measurement was carried out by imposing a thermal gradient on a bed of particles packed in a thin walled cylindrical aluminium tube (dia = 1.8 mm). The cylinder is immersed in a isothermal bath and after it has attained steady state it is transferred to another isothermal bath at a different temperature. The measurement of temperature with time was carried out using a thermocouple in conjunction with a Data Acquisition System. The heat conduction equation in the cylindrical coordinate system yields a solution with Bessel functions [D. Naumann, 1983]. If we limit on the temperature development at the tube axis the solution can be expressed as an infinite series of exponential functions. A first order approximation allows one to determine thermal diffusivity from the following equation:

\[ \alpha = (R^2/(5.8t)) \ln[1.6* (T_b-T_0)/(T_b-T(t))] \]

where \( \alpha \) = thermal diffusivity (m²/s)
\( R \) = tube radius (m)
\( t \) = time (sec)
\( T_b \) = bath temperature (deg K)
\( T_0 \) = initial temperature of the particle bed (deg K)
\( T(t) \) = bed temperature at time t (deg K)

From the thermal diffusivity, the thermal conductivity is obtained by using the relation:

\[ K_{\text{bed}} = \alpha \cdot \rho \cdot C \]

where \( K_{\text{bed}} \) = Effective thermal conductivity of the bed
\( \rho \) = bulk density of the bed
\( C \) = specific heat of the solid.

Model for equivalent conductivity of a coated spherical particle

We wish to obtain an expression for the equivalent conductivity of a coated spherical particle subjected to an axial heat flow. This can be used in conjunction with the correlations available in literature to predict the bed conductivity. We can thus treat a coated particle as a single solid. The expression for the equivalent conductivity is derived from heat transport equations and is shown below.
Heat Balance Equation:

\[
q_x y^2 = -\frac{2q_0}{\pi} (R-x)^2 + c
\]

\[
q_x y^2 = \frac{q_0}{\pi} (R-x)^3 + c
\]

COATING:

Heat Balance Equation:

\[
q_x \pi y^2 |_x - q_x \pi y^2 |_{x+dx} + q_2 2\pi ydy = 0
\]

\[
y^2 = R^2 - (R-x)^2 \quad \rightarrow \text{Eqn. of outer Circle} = 2R_x x + x^2
\]

\[
(q_x y^2) = 2q_0(R-x)
\]

\[
q_x = -k_1 (dT/dx)
\]

Boundary Conditions:

\[
\text{@ } x=0, \quad T=T_0 \quad \text{T-Tm}
\]

We obtain:

\[
k_1(T_0-T) = q_1 x
\]

\[
\frac{d}{dx}(q_x y^2) = 2q_0 R - 2q_0 x
\]

\[
q_x y^2 = 2q_0 R x - q_0 x^2 + c = x(2R_x - x) q_x = \ldots
\]

\[
\frac{d}{dx} - \frac{1}{\pi} \frac{dT}{dT} = \frac{2q_0 R x}{x(2R_x - x)} - \frac{q_0 x^2}{x(2R_x - x)} + \frac{c}{x(2R_x - x)}
\]

Figure 1: Heat Balance for Equivalent Conductivity
**SOLID + COATING:**

Heat Balance Equation gives:

\[(q_x \pi y_1^2)' + (q_x \pi (y_2^2 - y_1^2))' = 2q_l \pi (R - x)\]

where:

\[y_1^2 = r^2 - (R - x)^2\]

\[y_2^2 = R^2 - (R - x)^2\]

Solid: \[q_x = -k_2 (dT/dx)\]

Coating: \[q_x = -k_1 (dT/dx)\]

\[
\frac{dT}{dx} = \frac{-q_l (2R-x)}{2(R-r)k_1 + k_2(2r-x)}
\]

\[T = \int \left[ - \frac{q_l (2R-x)}{2(R-r)k_1 + k_2(2r-x)} \right] dx + c_1\]

where \(c_1\) : constant of integration

Boundary Conditions:

\[\text{at } x = R - r \quad T = T_m\]

\[\text{at } x = R \quad T = T_c\]

From the above, we obtain:

\[T_c - T_1 = q_l \left[ \frac{r}{a} - (2R/a) \ln \left( \frac{aR+b}{a(R-r)+b} \right) - (b/a^2) \ln \left( \frac{aR+b}{a(R-r)+b} \right) \right] \quad (2)\]

where \(a = -k_2\)

\[b = 2(R-r)k_1 + 2r(k_2)\]
From (1) & (2):

\[ T_c - T_0 = q [ \frac{r}{a} (2R/a) \ln \left( \frac{aR+b}{a(R-r)+b} \right) \]
\[- \left( \frac{b}{a^2} \right) \ln \left( \frac{aR+b}{a(R-r)+b} \right) + \left( R-r \right) \left( \frac{1}{k_1} \right) \]

For the Equivalent spherical particle we have from eqn (1):

\[ T_c - T_0 = - \left( \frac{qR}{k_{eq}} \right) \]

From (3) & (4):

\[ k_{eq} = R \left[ \frac{2R/a}{\ln \left( \frac{aR+b}{a(R-r)+b} \right)} + \left( b/a^2 \right) \ln \left( \frac{aR+b}{a(R-r)+b} \right) + \left( R-r/k_1 \right) - \left( r/a \right) \right] \]

Results and Discussion

The equivalent conductivity model was used to calculate the values of the coated solid conductivities and these were used in conjunction with the correlations available in literature for predicting effective bed conductivities. Of the many correlations in literature the orthorhombic model relation, the Yagii and Kunii equation, the empirical correlation of Saxena et al., and the resistor model for effective thermal conductivity were used to compare with the experimental data. Table 1 lists the various correlations. Figure 3 shows the variation of effective bed conductivity with coating thickness. The predictions of the various models and the experimental data are shown. We see that the effective bed conductivity drops rapidly in the beginning and then levels off with increasing coating thickness. This can be explained by observing the behavior of the equivalent conductivity of the coated solid with the ratio of the coating volume to the metal volume which is shown in Figure 4. We also find that the orthorhombic model, by Smith, gives the best prediction for bed conductivity. The other three models also give a good prediction of bed conductivity. The Saxena model gives good prediction for low values of equivalent solid conductivity. One of the main reasons for the good predictions of the orthorhombic model is the fact that the void fractions in the bed were very close to 0.4, which is the assumption of the orthorhombic packing. However, if the void fraction of the bed is much different from 0.4 it would be better to use the resistor model equation which has shown a good agreement with literature data. The equivalent conductivity model can be applied in a similar manner for a coated cylinder as well as a coated slab to account for non-spherical geometries.

These results suggest that the thermal conductivities of beds containing polymer coated metal particles will be quite similar to those of beds which contain polymer particles alone. Consequently, we expect to be able to form parts from polymer coated particles with only minor variations in machine settings relative to those used to process polymer powders.
<table>
<thead>
<tr>
<th>Authors</th>
<th>Correlations</th>
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<tbody>
<tr>
<td>1. Yagii and Kunii</td>
<td>( K_{\text{bed}} = \beta K_{\text{eq}} \frac{(1-\varepsilon)}{D} )</td>
</tr>
<tr>
<td></td>
<td>where ( D = 1 + k_s \varphi )</td>
</tr>
<tr>
<td></td>
<td>( \beta = 1 )</td>
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<td></td>
<td>( \varphi = 0.2086 \exp(1.8845) )</td>
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<tr>
<td>2. Woodside and Messmer</td>
<td>( K_{\text{bed}} = \left[ a k_{\text{eq}} k_f / (k_{\text{eq}} (1-d) + dk_f) \right] + b k_{\text{eq}} + c k_f )</td>
</tr>
<tr>
<td></td>
<td>where ( b = 0, c = \varepsilon - 0.03, a = 1 - c, )</td>
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<tr>
<td></td>
<td>and ( d = (1-\varepsilon)/a )</td>
</tr>
<tr>
<td>3. Swift</td>
<td>( K_{\text{bed}} = 0.5777 \pi k_f [\left( \lambda_s - 1 \right)^{-1} - (\lambda_s - 1)^{-2} \ln \lambda_s] + 0.093 k_f )</td>
</tr>
<tr>
<td></td>
<td>where ( \lambda_s = k_f / k_{\text{eq}} )</td>
</tr>
<tr>
<td>4. Saxena, Chohan and Gustafsson</td>
<td>( K_{\text{bed}} = k_{\text{ec}} (1 + 3.844 e^{0.667}) ) for ( \varepsilon &gt; 0.5 )</td>
</tr>
<tr>
<td></td>
<td>( K_{\text{bed}} = k_{\text{ec}} (1 - 1.545 e^{0.667}) ) for ( \varepsilon &lt; 0.5 )</td>
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</table>
Figure 3. Experimental and predicted values for effective bed conductivity with coating thickness.

Figure 4. Variation of equivalent conductivity with ratio of coating volume to volume of the metal. (k1=0.1, k2=33.85, t=123.5 microns)
Nomenclature

\( \alpha \) : thermal diffusivity \([\text{m}^2/\text{sec}]\)
\( c \) : heat capacity \([\text{J/kg deg K}]\)
\( \varepsilon \) : void fraction
\( k_1 \) : thermal conductivity of coating \([\text{W/m deg K}]\)
\( k_2 \) : thermal conductivity of the solid \([\text{W/m deg K}]\)
\( k_{eq} \) : equivalent thermal conductivity of the coated solid \([\text{W/m deg K}]\)
\( k_{bed} \) : effective thermal conductivity of the bed \([\text{W/m deg K}]\)
\( q \) : heat flux \([\text{W/m}^2]\)
\( R \) : tube radius \([\text{m}]\)
\( r \) : radial distance \([\text{m}]\)
\( t \) : time \([\text{sec}]\)
\( T \) : temperature \([\text{deg K}]\)
\( V \) : volume \([\text{m}^3]\)
\( x, y \) : coordinate axes

Greek symbols
\( \rho \) : bulk density of bed \([\text{kg/m}^3]\)
\( \varepsilon \) : void fraction of the bed
\( \varphi \) : parameter in Yagii and Kunii equation

Subscripts
\( b \) : bath
\( c \) : coating
\( l \) : laser
\( m \) : metal
\( o \) : initial
\( x \) : x-direction

REFERENCES