A MODEL FOR PREDICTION OF THE EFFECTIVE THERMAL CONDUCTIVITY OF GRANULAR MATERIALS WITH LIQUID BINDER

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A model is developed to predict the effective thermal conductivity of a three-component system containing sand particles bonded with liquid binders. The effective thermal conductivities of the bonded and unbonded sands are measured by the line-heat-source method at room temperature. The parameters of a two-component model are determined from the measurements for unbonded sands. Finally the model for the three-component systems such as bonded sand is developed using the assumption of the coalescence of the liquid binder at the particle contact points. Comparison between the experimental results and the model predictions shows that the model developed in this study predicts well the effective thermal conductivities of fluid-saturated sand or sand bonded with liquid binders. Also, this model can be used to determine the effects of the parameters such as the thermal conductivities and the volume fractions of components and the geometrical characteristics of solid particles on the effective thermal conductivity of granular materials with or without the liquid binder.

Key Words: Effective Thermal Conductivity, Granular Material, Bonded and Unbonded Sands, Liquid Binders, Line-Heat-Source Method

NOMENCLATURE

a : Constant given in(Eq. 2)
a_b, a_f : Constants given in(Eq. 9)
h : Constant given in(Eq. 5)
K : Thermal conductance
k : Thermal conductivity
R : Characteristic radius in (Fig. 4)
r : Radius in(Fig. 4)
x, y : Length coordinates
ϕ : Volume fraction
ϕ_{ef} : Volume fraction given in(Eq. 6)

Subscripts
b : Binder
e : Effective
f : Fluid
s : Solid particle

1. INTRODUCTION

Numerous papers on the prediction of the effective thermal conductivity of granular materials have been published in the last thirty years. Many of these models require only the porosity and the thermal conductivities of two components to calculate the effective thermal conductivity of a two-component system. But the effective thermal conductivity of unbonded sands, which is a two-component system, is known to be dependent also on the characteristics of the sand particles. The characteristics of sand particles incude average particle size, particle shape and particle size distribution. Also, binders are usually added to sands to improve mechanical properties and to improve the contact between sand particles and to increase the effective thermal conductivity of the bonded sands.

There are numerous experimental data on the thermal conductivity of sands bonded with various types of binders. For example, Jackson(1982) measured the effective thermal conductivity of Ottawa silica sand when mixed with liquid binders such as Gulf wax and Dow Therm-A to develop a method to increase the effective thermal conductivity of bonded sands. Even so, the available data are most often insufficient because the variety of bonded sands is innumerable. Furthermore, no reliable mathematical model is available to predict the effective thermal conductivity of bonded sands from a knowlege of their constituents.

Thus, in this study, the effects of these constituents and their properties on the effective thermal conductivity of bonded and unbonded sands are examined and the development of a predictive model is attempted. Finally, experimental verification of this model is provided by comparing model predictions with experimental results.

2. MEASUREMENTS USING LINE-HEAT-SOURCE METHOD

For the measurements of the effective thermal conductivity of sands, transient methods utilizing a thermal conductivity probe that approximates a line heat source are adopted because these methods require a relatively simple apparatus and measurements can be completed very rapidly. The principles of this method, also called the hot-wire method, were proposed by Schleiermacher(1938).

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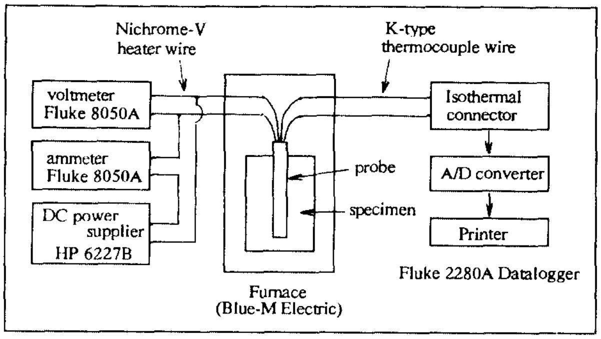

Fig. 1 Schematic diagram of measurement system

The thermal conductivity probe designed for this study is similar to the laboratory probe constructed by Steinmanis (1982) to measure the thermal conductivity of soil. The probe consists of a 30 AWG Chromel-Alumel thermocouple wire, a Nichrome-V heater wire, a ceramic insulator and a stainless steel tube. The outer diameter of the probe is 2.41 mm and the length-to-diameter ratio is 58. The basic measurement system for the probe method is composed of a thermal conductivity probe, a regulated DC power supply, a furnace, and a probe temperature and electric power measurement system as shown in Fig. 1.

3. MODEL DEVELOPMENT FOR TWO-COMPONENT SYSTEMS

Among the models that predict the effective thermal conductivity of granular materials, the models developed by Kunii and Smith(1960), Woodside and Messmer(1961), de Vries(1952), and Masamune and Smith(1963) are selected for comparison with the measured results. These models require only the porosity and the thermal conductivities of the two components to calculate the effective thermal conductivity of a two-component system.

For comparison between prediction and experiment, the effective thermal conductivity of Ottawa silica sand saturated with air and saturated with water are measured by the thermal conductivity probe method. The measured values are compared with the calculated values from the two-component models and the results are shown in Figs. 2 and 3.

These figures show that neither model is entirely adequate for predicting the thermal conductivity of these two-component systems. Thus, development of an improved model for two-component systems and an extension of this model to three-component systems were initiated.

The effective thermal conductivity of granular materials depends on the thermal conductivities of the solid material and of the saturating fluid, the apparent density, the surface properties, and the geometrical characteristics of the solid particles. When a sand type is selected, the thermal conductivity, the surface property, and the geometrical characteristics of the sand particles are completely determined. Thus the effective thermal conductivity of a particular and will be a function of the volume fraction of sand particles and the thermal conductivity of the saturating fluid only. One of the desirable features of a theoretical model for two-component systems would be that the geometrical parameters be entirely independent of the thermal conductivities of the existing components. This would facilitate the extension of the model

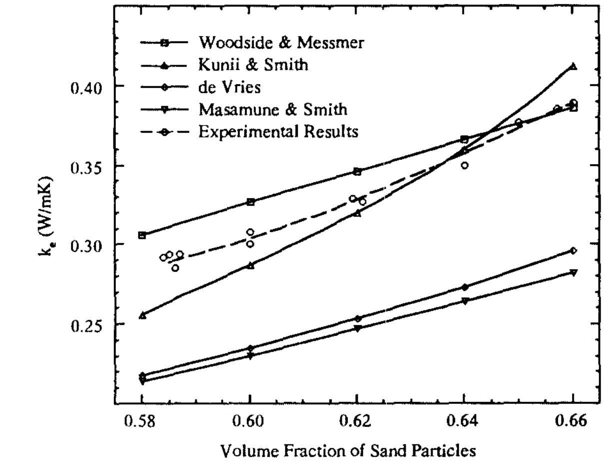

Fig. 2 Comparison of two-component models with experimental results for dry, unbonded ottawa sand

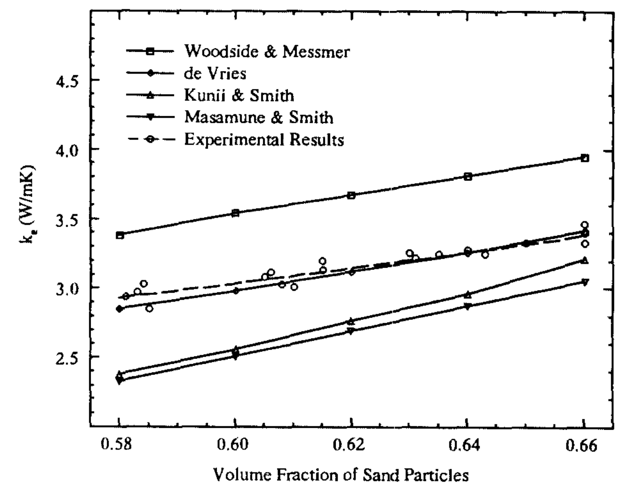

Fig. 3 Comparison of two-component models with experimental results for sand saturated with water

to a three-component system.

Heat transfer can be assumed to occur in the granular materials by the following mechanisms.

(1) conduction through the voids

(2) conduction through a series path consisting of an effective solid-path length and a fluid-path length

(3) conduction through the direct-contact area from one solid particle to neighbouring particles

(4) transgranular radiation and intergranular radiation

(5) convection in the voids

The thermal conductivity of the solid particles is usually higher than that of the saturating fluid. For silica sands, the ratio of thermal conductivity of silica sand particles to that of air is about 340 at room temperature. Therefore, the heat transfer through the pore spaces is relatively small. Also, the convective effect is negligibly small because the pore size in typical sand aggregates is very small. In addition, the radiation contribution to the total heat transfer is negligible near room temperature.

Heat transfer will be occur mainly by conduction through and near the contact points between solid partcles. Thus, the

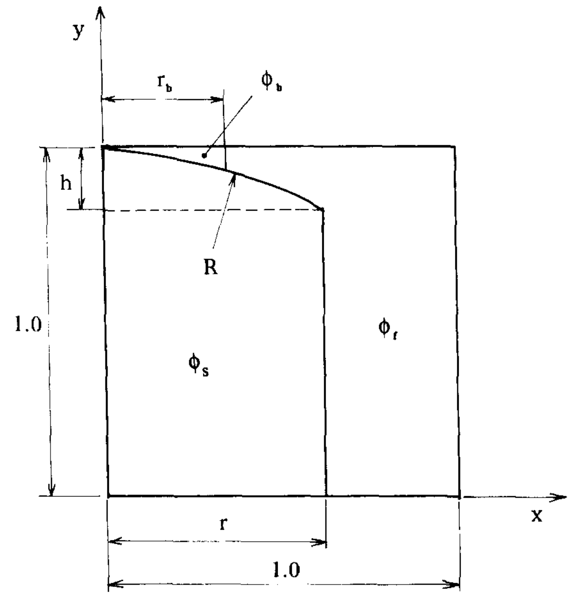

Fig. 4 Geometry of two-or three-component model

geometry near the contact area should be the most important parameter controlling the heat transfer through sand particle aggregates. To develop a model for two-component unbonded sands, the sand particle surface near the contact points is assumed to be a spherical surface with a characteristic radius. The solid-to-solid contact area can be related to approximately to the measured effective thermal conductivity under vacuum condition (i.e., no saturating fluid present). However, the measured effective thermal conductivity of quartz particles under vacuum conditions is relatively small according to Woodside and Messmer(1961), and the heat transfer through the direct contact area should decrease as the thermal conductivity of the saturating fluid is increased because the resistance to heat flow through the fluid decreases. Thus, the heat transfer through the direct contact area is assumed to be negligible.

In this way, a model is developed to include the assumptions of a spherical surface with a characteristic radius and negligible direct-contact heat transfer as shown in Fig. 4. The cylindrical core has radius r, and a spherical surface of radius R.

When the spherical surface of the particle is employed, the mathematical formulation for the geometry of the model is too complex. Being able to describe each heat transfer mechanism separately using a simpler model is desirable. Thus, the assumption of one dimensional heat flow is kept in the model in this study. The separation distance between solid particles is assumed zero. A solid particle may have many contact points with its neighbouring particles which can be assumed to have a spherical shape near contact points. The number of contact points of a particle increases with the volume fraction of solid particles because of changes in the arrangement of the solid particle. The effect of increasing the number of contact points may be represented by an increase in the characteristic radius of the solid core in the model as the volume fraction of solid particles is increased.

In this study, the characterisic radius of the model is assumed to increase linearly with the volume fraction of solid particles for simplification and this assumption is verified by comparing the calculated values from the model with the measured ones.

4. EFFECTIVE THERMAL CONDUCTIVITY OF TWO-COMPONENT MODEL

With the assumption of one-dimensional heat flow, a closed form solution can be obtained for the effective thermal conductivity from the model. The total heat flow is calculated by summing all contributions by conduction through infinitesimal cylindrical tubes of unit length. Thus, the conductance of a small cylindrical volume element can be expressed as

$$dK=\frac{2\pi x dx}{y/k_s+(1-y)/k_f} \tag{1}$$

By integrating Eq. 1, the total conductance through the solid particles and interstitial fluid is found to be

$$K=\frac{2\pi}{1/k_s-1/k_f}\left[\mathrm{a}\ \ln\left(\frac{\sqrt{R^2-r^2}+a}{R+a}-\sqrt{R^2-r^2}+R\right)\right] \tag{2}$$

where

$$a=\frac{(1-R)/k_s+R/k_f}{1/k_s-1/k_f}$$

The relationship between the conductance and the effective thermal conductivity of the two component model is

$$K=\pi k_e \tag{3}$$

Thus, the effective thermal conductivity of the two-component model is determined by

$$k_e=\frac{2}{1/k_s-1/k_f}\left[\mathrm{a}\ \ln\left(\frac{\sqrt{R^2-r^2}+a}{R+a}-\sqrt{R^2-r^2}+R\right)\right] \tag{4}$$

The volume fraction of solid particles and the effective fluid volume are determined from the geometrical pamaters of the model as follows.

$$\phi_s=h(6R-3Rh-3h+2h^2)/3 \text{ where } h=R-\sqrt{R^2-r^2} \tag{5}$$

and

$$\phi_{ef}=h^2(3R-2h) \tag{6}$$

The dimensions to be determined in the two-component model are the parameters R, and r. The procedure to determine these geometrical parameters from the measured thermal conductivities of a typical sand is outlined as follows :

(1) Measure the effective thermal conductivities of a dry sand which is saturated with air at two different volume fractions of solid particles.

(2) Using Eq. (4) with two different values of k_e from (1) and Eq. (5) with ϕ_s known, give two equations to solve for R and r for each value of ϕ_s.

(3) Determine R as a linear function of ϕ_s.

The dimensions R and r can be determined with ϕ_s, and then the effective thermal conductivity of the two-component model is calculated by Eq. (4) with the known values of R and r. Thus two experimental measurements are required for each sand before effective thermal conductivity predictions can be made.

5. EXPERIMENTAL VERIFICATION OF TWO-COMPONENT MODEL

From the experimental measurement of the effective thermal conductivity of dry Ottawa silica sand saturated with air, the geometrical parameters of the two-component model are

Table 1 Comparison between model prediction and measurement for unbonded dry sands

Sand Type	Volume fraction of sand particles	Effective thermal conductivity, W/mK	
		measured	calculated
Ottawa	0.58	0.2839	0.2791(1.7%)
Ottawa	0.62	0.3281	0.3325(1.3%)
Ottawa	0.66	0.3918	0.3867(1.3%)
Masonry	0.50	0.2528	0.2511(0.7%)
Masonry	0.54	0.2957	0.2973(0.6%)
Masonry	0.58	0.3429	0.3441(0.4%)
Masonry	0.62	0.3945	0.3907(1.0%)

*The values in parentheses represent the relative error

calculated. As a result, R for Ottawa sand is determined to be

$$R=7.62\phi_s-3.29 \text{ when } 0.58<\phi_s<0.66 \qquad (7)$$

While the particle shape of Ottawa sand is relatively round, the Masonry sand has a rather angular particle shape. To see the effect of particle shape, the characteristic radius of the two component model for Masonry silica sand is determined to be

$$R=6.50\phi_s-2.22 \text{ when } 0.5<\phi_s<0.62 \qquad (8)$$

With given values of R, the effective thermal conductivities of Ottawa sand and Masonry sand saturated with air are calculated. These values are compared with the experimental values at various volume fractions of sand particles in Table 1. The results show agreement to within two percent for both sands. Thus, the assumptions of a linear dependence of R on ϕ_s and zero separation distance between solid particles seem justified.

6. EFFECT OF SATURATING FLUID

The geometrical parametes (R and r) are independent of the thermal conductivity of the saturating fluid. The other parameter that affects the effective thermal conductivity of the model is the thermal conductivity of the saturating fluid. Thus, the model for two-component systems is examined to determine if the model could predict the effect of the saturating fluid. For this purpose, transformer oil (982-68), supplied by Gulf Oil Company, Prestone Ⅱ which is a form of ethylene glycol, and water are selected as test masterials. The thermal conductivity of each of these fluids is higher than that of air and less than that of water. The values are listed in Table 2.

The effective thermal conductivities of Ottawa sand and Masonry sand saturated with these fluids are measured and are compared with values predicted from the two-component model in Table 3. The percent deviation between measured and calculated values is shown in parentheses in the table. The results indicate that the model accurately predicts the effective thermal conductivity of the unbonded sands when the thermal conductivity of the saturating fluid varies between the value for air and the value for water.

Table 2 Thermal conductivites of saturating fluids

Saturating fluids	Thermal conductivity at 25°C(W/mK)
air	0.0254
transformer oil	0.11
Prestone Ⅱ	0.263
water	0.609

7. MODEL EXTENSION FOR THREE-COMPONENT SYSTEMS

The geometry of the two-component model is independent of the thermal conductivity of the saturating fluid for a specific sand. The model provides rather accurate predictions of the effective thermal conductivity of fluid-saturated sands. This indicates that the model rather accurately describes the representative configuration near the contact points through which most heat transfer occurs. Therefore, the model in this study should be expected to predict the heat transfer through sand bonded with liquid binders even though the sand is not saturated with the liquid.

Liquid binder will coalesce at the contact points between sand particles and form a meniscus structure. Since one-dimensional heat flow is assumed in developing the model for the thermal conductivity of unbonded sands, all of the liquid binder is assumed to be placed at the contact area in the two-component model as shown Fig. 4. Again parallel conduction paths are assumed present in the model. The total conductance of the model for three-component systems can be determined by adding all conductances of conduction heat transfer paths in the model. Thus, the effective thermal conductivity of the three-component model is expressed as

$$k_e=\frac{2}{1/k_s-1/k_b}\left[\mathrm{a}_b\ln\left(\frac{\sqrt{R^2-r_b^2}+a_b}{R+a_b}\right)-\sqrt{R^2-r_b^2}+R\right]$$
$$+\frac{2}{1/k_s-1/k_f}\left[a_f\ln\left(\frac{\sqrt{R^2-r^2}+a_f}{\sqrt{R^2-r_b^2}+a_f}\right)-\sqrt{R^2-r^2}\right.$$
$$\left.+\sqrt{R^2-r_b^2}\right]$$

where

$$a_b=\frac{(1-R)/k_s+R/k_b}{1/k_s-/k_b} \qquad (9)$$

Table 3 Effect of thermal conductivity of saturating fluid

Sand Type	ϕ	Effective thermal conductivity, W/mK					
		Transformer oil		Prestone Ⅱ		Water	
		measured	calculated	measured	calculated	meausred	calculated
Ottawa	0.60	—	—	1.851	1.722(7.5%)	3.036	3.007(1.0%)
Ottawa	0.62	1.094	1.003(9.1%)	1.939	1.828(6.1%)	3.153	3.135(0.6%)
Ottawa	0.64	1.147	1.074(6.8%)	2.002	1.938(3.3%)	3.276	3.277(0.0%)
Masonry	0.50	—	—	—	—	2.652	2.572(3.1%)
Masonry	0.534	0.956	0.874(9.4%)	—	—	2.769	2.761(0.3%)
Masonry	0.56	—	—	1.797	1.726(4.1%)	2.914	2.922(0.3%)
Masonry	0.62	—	—	—	—	3.226	3.319(2.9%)

Table 4 Thermal conductivity of liquid binders

Type of binder	Thermal conductivity (W/m K)
kerosene	0.145
Dow Therm-A	0.142
Epoxy	0.188
Ethylene Glycol	0.260
Gulf Wax	0.270

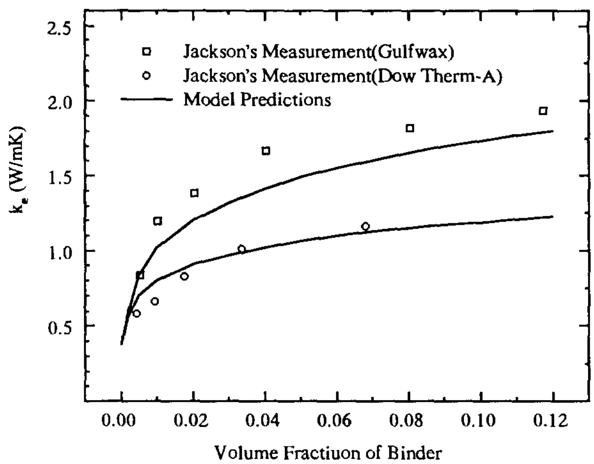

Fig. 5 Comparison of model predictions with experimental results for bonded Ottawa sand(ϕ_s=0.66)

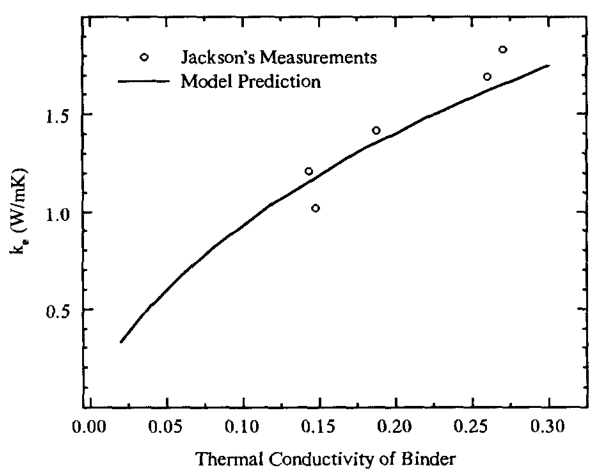

Fig. 6 Effect of thermal conductivity of binder for bonded Ottawa sand(ϕ_s=0.66)

$$a_f = \frac{(1-R)/k_s + R/k_f}{1/k_s - 1/k_f}$$

The assumptions made to extend the model are examined indirectly by comparing model predictions to experimental measurements. The effective thermal conductivities of Ottawa sand bonded with liquid binders are calculated using the three-component model in order to compare them with the experimental results of Jackson(1980). Gulfwax, ethylene glycol, epoxy, Dow therm-A and kerosene were used as liquid binders. The thermal conductivities of these binders are given in Table 4.

Jackson measured the effective thermal conductivity of Ottawa sand when mixed with Gulf wax and with Dow Therm-A while maintaining the volume fraction of solid particles equal to 0.66. In these tests, the volume fraction of binder was varied up to 0.12 for Gulf wax and up to 0.07 for Dow Therm-A. The values predicted from the model are compared with Jackson's results in Fig. 5. This figure show that the calculated values from the three-component model agree well with the measured values and have an average error of 13 percent.

The effective thermal conductivities of Ottawa sand mixed with the binders listed in Table 4 were also measured by Jackson. The volumeteric binder content was 0.08, and the volume fraction of sand particles was 0.66. Fig. 6, which compares the measured values and the values predicted by the three-component model, shows that the model for three-component systems predicts the effect of thermal conductivity of the liquid binder and the volumetric binder content very well. With the exception of the lowest experimental values shown in the figure, the model predictions agree to within five percent of the experimental values of Jackson. Thus, the model can be used to predict the effective thermal conductivity of sands bonded with liquid binders.

8. THEORETICAL RESULTS OF MODEL PREDICTION

8.1 Two-Component Systems

For a given sand type, the effective thermal conductivity of unbonded sand depends only on the thermal conductivity of the saturating fluid and the volume fraction of sand particles. The effects of these parameters are considered with the theoretical model developed in this study. The geometry of the two-component model for unbonded Ottawa sand can be used for unbonded sands having the same geometrical characteristics as Ottawa sand even though the particle thermal conductivity may be different. Using the model geometries for Ottawa and Masonry sands, the effective thermal conductivity is calculated to investigate the effects of volume fraction of solid particles ranging from 0.4 to 0.7 and the thermal conductivity of the saturating fluid. The calculated effective thermal conductivity is normalized by the thermal conductivity of the solid particles and is shown in Fig. 7 when the normalized thermal conductivity of the saturating fluid varies from 0.0 to 1.0.

This figure shows that the calculated effective thermal conductivity and its rate of increase are greater for greater volume fraction of solid particles, and the rate of increase of the effective thermal conductivity decreases as the thermal conductivity of the saturating fluid is increased. The effect of the volume fraction of solid particles on the effective thermal conductivity becomes small as the thermal conductivity of the saturating fluid increases. For a constant volume fraction of solid particles, the thermal conductance of the liquid film between solid particles depends only on the normalized thermal conductivity of the saturating fluid. As the difference between the thermal conductivities of the solid particles and the saturating fluid become smaller, the contribution of the thermal conductance between the solid particles to the effective thermal conductivity of unbonded solid particles become less significant. Thus, the increase in the effective thermal conductivity of unbonded particles in Fig. 7. represents the effect of an increase in the thermal conductance between solid particles owing to the increase in the thermal conduc-

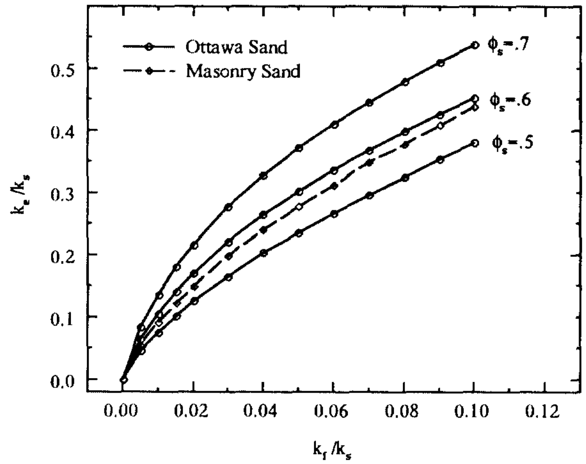

Fig. 7 Effecitive thermal conductivity of unbonded sand as a function of thermal conductivity of saturating fluid

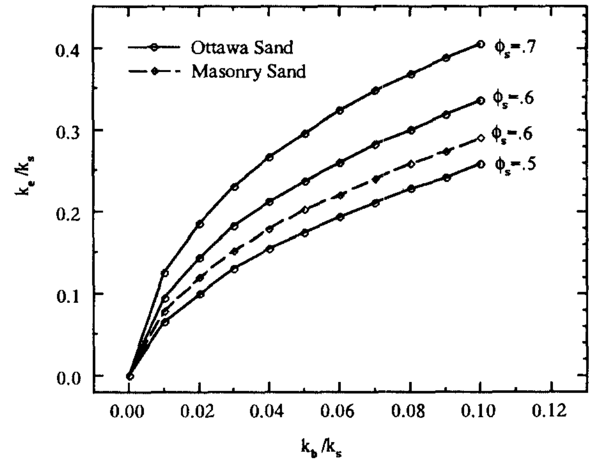

Fig. 8 Effective thermal conductivity of bonded sands as a function of binder thermal conductivity ($\phi_b = .08$, $k_f/k_s = .003$)

tivity of the saturating fluid.

The effective thermal conductivity of unbonded solid particles having angular shape is slightly greater than that of unbonded particles having round shape, and the influence of particle shape becomes small when the thermal conductivity of the saturating fluid is large. Fig. 7 also shows that the model predicts that the effective thermal conductivity approaches zero when the normalized thermal conductivity of the saturating fluid becomes zero. This is because solid-to-solid contact has been neglected in the model. Also, when the thermal conductivity of the fluid is zero, the resistance of the fluid film assumed to be present between solid particles is infinite, and the effective thermal conductivity of unbonded solid particles is zero regardless of the volume fraction of solid particles.

8.2 Three-Component System

The only assumption used in the development of the three-component model for bonded solid particles with liquid binders is that all of the liquid binder is located at the particle contact points. Thus, for the solid particles bonded with liquid binders, the effective thermal conductivity depends upon the thermal conductivities of the components, the volume fraction of solid particles and the volume fraction of binder.

The normalized effective thermal conductivity of solid particles bonded with homogeneous liquid binders, calculated as a function of the normalized binder thermal conductivity using the three-component model, is shown in Fig. 8 for various values of the volume fraction of solid particles. The binder volume fraction and the normalized thermal conductivity of saturated fluid in the pore is 0.08 and 0.003, respectively.

This figure shows that the effective thermal conductivity increases as the normalized binder thermal conductivity increases for a constant volume fraction of solid particles. Thus, the variation of the effective thermal conductivity of bonded particles with the binder thermal conductivity is similar to that for unbonded particles. The argument used to explain the effect of the volume fraction of solid particles on the effective thermal conductivity of unbonded particles is also applicable to solid particles bonded with liquid binders.

When compared with the results in Fig. 7, the effect of the geometrical characteristics of the sand particles is more significant and the difference between the effective thermal conductivities of Ottawa sand and Masonry sand increases with the normalized binder thermal conductivity. This is because the characteristic radius in the model is larger and thus the heat transfer area through the binder is greater at the same volume fraction of solid particles for Masonry sand.

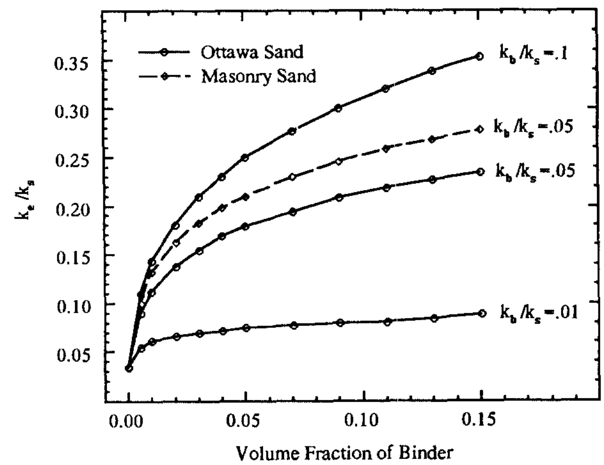

Fig. 9 Variation of normalized effective thermal conductivity of bonded sands with volumetric binder content ($\phi_s = .6$, $k_f/k_s = .003$)

The effective thermal conductivity of bonded particles is also influenced by the volume fraction of the binder and the binder thermal conductivity as shown in Fig. 9.

The normalized effective thermal conductivity increases rapidly as the binder content increases and the rate of increase is greatest at low volumetric binder contents. This also can be explained with reference to the geometry of the three-component model. As the volumetric binder content increases, the area available for heat transfer through the binder between solid particles is increased rapidly at low binder contents. Increasing the binder content at greater binder contents causes an increase in the thickness of the binder film between solid particles and only a small increase in the contact area. Thus, the increase in the conductance of the bond between solid particles will be small also. The thermal conductivity of the bidner is also an important parameter, and its effect on the normalized effective thermal conductivity of bonded particles can be seen in Fig. 9.

9. CONCLUSIONS

(1) The geometry of the two-component model for the fluid-saturated solid particles can be determined by two measurements of the effective thermal conductivity. The influence of the solid particle characteristics on the effective thermal conductivity of unbonded solid particles can be determined by the geometrical parameters in the two-component model. Comparison of the experimental results with the predicted effective thermal conductivity of fluid-saturated silica sands indicates that the accuracy of the predicted values is better than 10 percent. The two-component model is useful to estimate the effects of the thermal conductivity of saturating fluid, the volume fraction of solid particles and the geometrical characteristics of solid particles when the thermal conductivity of the saturating fluid is higher than that of air and lower than that of water.

(2) The three-component model can be used to predict the effective thermal conductivity of solid particles bonded with liquid binders. The liquid thermal binders can be assumed to coalesce at the particle contact points. Model predictions using Eq. (9) agree will with the experimental results and provide a method for determining the effects of the thermal conductivity and the volume fraction of solid particles and thermal binders on the effective thermal conductivity of bonded solid particles.

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