Journal of Thermal Analysis, Vol. 34 (1988) 249–257

HEAT TRANSFER IN MICROSPHERE INSULATION

R. Wawryk and *J. Rafałowicz*

INSTITUTE FOR LOW TEMPERATURE AND STRUCTURE RESEARCH, POLISH ACADEMY OF SCIENCES, 95 PRÓCHNIK STR., 53–529 WROCŁAW, POLAND

(Received April 14, 1987)

The results of an investigation of heat transfer in a new type of insulation (microsphere insulation) are presented. The effects of the microsphere diameter, the concentration of metallized microspheres and the residual gas pressure on the thermal conductivity of the insulation were investigated. Measurements were made of the thermal conductivity at 77 to 300 K of microspheres with differing diameters (e.g. 95, 130 and 270 μm) and of samples with silver metallized microsphere concentrations of 7 and 32%. Measurements of average thermal conductivity (77–296 K) were made at residual gas pressures $k(p)$ in the range from 10^{-3} Pa to 10^5 Pa for pure nitrogen. The component of heat transfer by gas, $k_{gc}(p)$, was estimated.

In 1973, the idea of using hollow glass microspheres was developed [2]. The radius of the spheres varied from 20 to 200 μm and their thickness from 0.5 to 2 μm. This kind of material was called high-performance thermal insulation. As compared with multilayer insulation microsphere insulation has thermal conductivity about half an order higher, but is characterized by certain advantages which are not present in multilayer insulation. As advantages, we may list ease of use in complicated geometrical shapes, no thermal conductivity anisotropy, and a high degree of resistance to destruction in use under pressures of the order of 10^6 to 10^7 Pa. In addition, the microsphere insulation is characterized by stable thermal parameters.

Theory

The transfer of heat in microsphere insulation is due principally to three fundamental mechanisms of heat exchange:

— thermal conductivity by a solid-state material,

The experimental results presented here were obtained in the framework of a Ph.D. thesis [1], and have in part been published previously.

John Wiley & Sons, Limited, Chichester
Akadémiai Kiadó, Budapest

— heat transfer by radiation,

— heat conductivity by a residual gas.

The effective coefficient of thermal conductivity of a layered medium made from tightly packed spheres may be presented as the sum of the following components:

$$k_{\text{eff}}(T,p) = k_c(T,p)+k_r(T) = k_{ss}(T)+k_{gc}(T,p)+k_r(T) \tag{1}$$

where: k_c = conduction component,
k_r = radiation component,
k_{ss} = solid-state heat conduction component, and
k_{gc} = component of heat conduction by residual gas.

Chan and Tien [3] have considered a situation in which a pressure P acts on a layered medium of regularly packed solid or thick-walled spheres. In this case, the relations for thermal conductivity through a solid body such as this medium have the following equations:

$$k_{ss}(T) = S_p\left(\frac{1-\mu^2}{E}P\right)^{1/3}\cdot k_s(T) \qquad \text{for } P>0 \tag{2}$$

and

$$k_{ss}(T) = S_N\left[\frac{(1-\mu^2)\varrho_s V_s L}{Er_0^3}\right]^{1/3}\cdot k_s(T) \qquad \text{for } P=0. \tag{3}$$

From these formulae, two important results are evident. First, in both cases of loading the thermal conductivity of microsphere layers does not depend on the microsphere diameter d, but depends on the ratio of the wall thickness t to the microsphere radius r_0; second, microsphere insulation is directly proportional to the thermal conductivity of the microsphere material $k_s(T)$.

Klein [4] has presented an equation for a component of heat transfer by radiation in a medium of regularly packed spheres of the same diameter:

$$k_r(T) = \frac{4\sigma d}{\delta_s}\left(\frac{\varepsilon}{2-\varepsilon}\right)T^3. \tag{4}$$

The component of heat transfer by gas, $k_{gc}(T,p)$, in microsphere insulation is a function of the gas thermal conductivity k_g, the thermal conductivity of the spheres k_{gr}, the average local distance between the surfaces of contacting microspheres δ_a and the mean free path of the gas particles $\overline{L}$. The equation for the thermal conductivity of a gas in a porous granular material is as follows [5]:

$$k_{gc}(T,p) = k_g\left[\frac{5.8\delta_s^2}{K}\left(\frac{1}{K}\ln\frac{k_{gr}}{k_g}-1-\frac{K}{2}\right)+1\right] \tag{5}$$

where:

$$K = 1 - \frac{k_g}{k_{gr}}; \quad k_g = \frac{k_{g0}}{1 + \frac{19}{6} \frac{2-\alpha}{\alpha} Kn}.$$

Results of measurements

With the aim of estimating the influence of the diameter of the microspheres on the thermal conductivity of insulation, three samples were prepared, using microspheres of differing diameters, and measurements were carried out within the limits of the effective coefficient of thermal conductivity relative to temperature in the range from 77 to 300 K [6]. The average diameters of the microspheres d_m in samples 1 to 3 were 270 μm, 130 μm and 95 μm, and their density ranged from 350 kg/m³ to 390 kg/m³.

The material composition of the microspheres included:

SiO_2 56%, Al 15%, Fe 2%, K 2.9% and Na 0.6%.

Metals are involved as oxides. The microspheres are filled with the gases N_2, CO_2 and CO.

Measurements of thermal conductivity were carried out in a spherical calorimeter with an insulation layer 2.95 cm thick, using a differential method with a temperature difference $\Delta T < 20$ deg.

The results of the measurements are presented in Fig. 1. Points marked on the graph are experimental, and the continuous lines are represented by $k(T) = AT + BT^3$. The result of the measurements, an increase in the component for heat transfer by radiation $k_r(T)$ with increase of the diameter conforms with Klein's equation (4). The coefficient B in the analytical equation increases with increase of the microsphere diameter. It may also be seen that, with increase of the diameter of the microsphere, the component of thermal conductivity through the solid state also increases (coefficient A is larger). This is not in keeping with the theoretical results of Chan and Tien [2, 3], who explain that such a relation does not hold.

This discrepancy is related to the different densities of packing in the three samples, resulting in different numbers of thermal contacts. The coefficients δ_s for samples 1 to 3 are 0.64, 0.57 and 0.43, and the average numbers of thermal contacts $\bar{N}$ are 8.8, 7.1 and 4.8. After cleaning of the insulated chamber containing sample 3 with pure nitrogen, a lowering of the thermal conductivity was found (curve 4). This shows that particles of water vapour, carbon dioxide and other gases were evacuated. Subsequently, solidification of the gases created thermal bridges between the contacting microspheres in sample 3 as well as in samples 1 and 2.

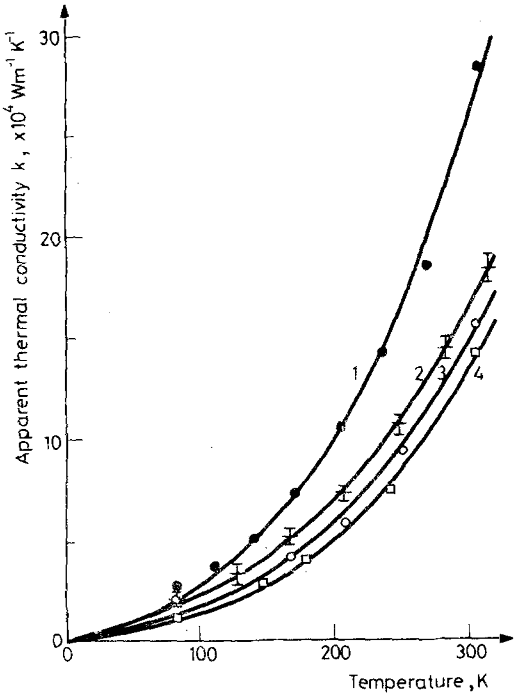

Fig. 1 Apparent thermal conductivity versus temperature for uncoated microspheres. Curve: 1 – d_m = 270 μm, $k = 2.0275 \times 10^{-6} T + 0.7373 \times 10^{-10} T^3$; 2 – d_m = 130 μm, $k = 1.9242 \times 10^{-6} T + 0.3958 \times 10^{-10} T^3$; 3 – d_m = 95 μm, $k = 1.3646 \times 10^{-6} T + 0.3912 \times 10^{-10} T^3$; 4 – d_m = 95 μm, $k = 0.9885 \times 10^{-6} T + 0.3876 \times 10^{-10} T^3$, $\mathrm{Wm^{-1} K^{-1}}$

The analysis of heat transfer in a medium of tightly packed microspheres shows that in the whole process of heat exchange an important role is played by radiation. The intensity of radiation passing through a layer of spheres is diminished by spheres having characteristics of absorption and scattering, while the intensity is increased by the emission of radiation from the surface of the spheres. For purposes of diminishing the radiation intensity, the spheres are coated with a thin layer of metal with a low coefficient of emission ε, i.e. metals such as aluminium, nickel, rhodium, cobalt, tantalum and silver.

For measurements of thermal conductivity, three samples were prepared, with different concentrations of metallized microspheres [7]. The first sample was composed of non-metallized microspheres. In samples 2 and 3 the concentration of silver metallized microspheres was 7% and 32%, respectively. The thickness of the silver was 0.8 μm. The resulting measurements are presented in Fig. 2. A slight diminishing of the effective thermal conductivity may be observed in the range of room temperature for the samples involving metallized microspheres. The

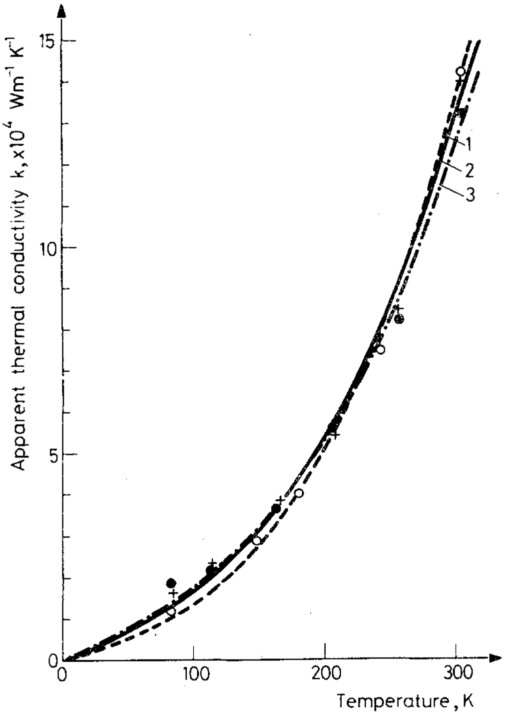

Fig. 2 Apparent thermal conductivity versus temperature for coated and uncoated microspheres. Curve 1 - - - (○) – $k = 0.9862 \times 10^{-6} T + 0.3879 \times 10^{-10} T^3$ (uncoated); 2 —— (+) – $k = 1.3584 \times 10^{-6} T + 0.3319 \times 10^{-10} T^3$ (7% Ag coated); 3 -·-·- (●) – $k = 1.4767 \times 10^{-6} T + 0.2954 \times 10^{-10} T^3$ (32% Ag coated); $Wm^{-1} K^{-1}$

diminishing of the thermal conductivity is higher for sample 3, with a 32% concentration of metallized microspheres. However, in the range of liquid nitrogen temperature, the samples of metallized microspheres have a higher thermal conductivity. This is caused by the diminishing of the thermal resistivity of the contacting surfaces of the metallized spheres, and by the increasing conductivity of the metallized coating of the spheres.

In order to estimate the influence of gas on the quality of insulation, measurements of the average thermal conductivity of the microspheres (from 77 to 296 K) relative to residual gas pressure were made. As a non-condensing gas, nitrogen was used to fill the spaces between the spheres. The measurements were made in a calorimeter with spherical symmetry, with its outer wall immersed in liquid nitrogen. Before the measurements, the microsphere sample was evacuated at a higher temperature (about 400 K) and cleared with pure nitrogen gas. The resulting measurements of the average value of the thermal conductivity coefficient of microsphere insulation (d_m = 95 μm) in the pressure range of the residual gas

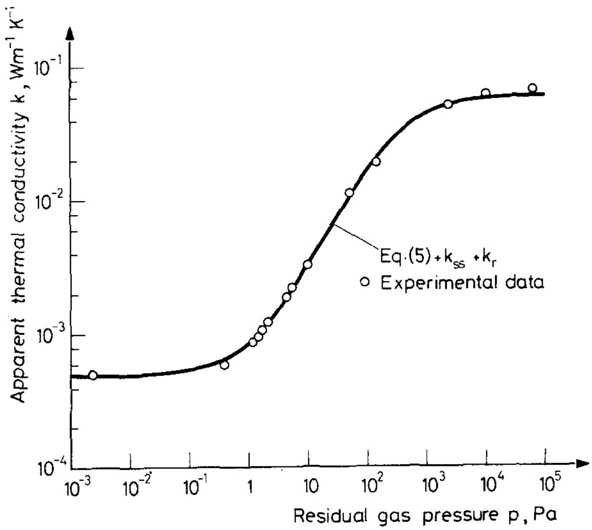

Fig. 3 Average thermal conductivity versus residual gas pressure for uncoated microspheres; $d_m = 95$ μm, N_2 atmosphere, $\bar{T} = 187$ K

(N_2) are presented in Fig. 3. The two axes are plotted in logarithmic units. The points on the graph are experimental and the curve is plotted on the basis of Eq. (5), to which are added the values of the components of solid-state and radiation heat transfer for every point, measured under conditions of high vacuum ($k_{ss}+k_r = 5\cdot 10^{-4}$ Wm^{-1} deg^{-1}).

At pressures below 0.5 Pa, the influence of the residual gas (between the microspheres) on the thermal conductivity of the medium was small. In this range of pressure, the total thermal conductivity is due only to the solid-state and radiation heat transfer components. Above 0.5 Pa, the contribution of the gas to the thermal conductivity starts to increase. In the pressure range from 1 Pa to 10^3 Pa, the $k(p)$ curve increases steeply. The effective thermal conductivity at 10^3 Pa is two orders higher than that at $p=1$ Pa. Above 10^3 Pa up to atmospheric pressure, the increase in the $k(p)$ curve is flat and approaches saturation asymptotically. The increased contributions of the gas heat transfer components become evident for those through which the mean free path of the gas molecules tends towards the values of the microsphere diameters. At $p=1$ Pa, the mean free path of nitrogen molecules is 5 mm, whereas at 10^2 Pa it is 5 μm.

Conclusions

Our experimental investigations revealed an increase in the radiation component of thermal transfer (k_r) with increase of the average microsphere diameter (d_m), in keeping with Klein's equation. An increase in the solid-state component of thermal transfer on microsphere diameter increase does not agree with formula (3). This fact is caused by the different densities of sphere packing in the different samples. The heat flux flowing through one sphere is proportional to the number of thermal contacts with its neighbours [8]; $Q_s = \frac{1}{6}(Q_{1c} \cdot N)$. The dependence of effective thermal conductivity on temperature for the metallized and non-metallized microspheres produced here is well described by the analytical formula $k(T) = AT + BT^3$. Qualitative agreement has been found between the experimental points of the thermal conductivity of microsphere insulation in relation to the pressure of residual gas and the curve obtained from Eq. (5).

* * *

The authors express their thanks to Dr. J. Hycnar for the samples of microspheres, to the team of the Chemical Institute of the Jagiellonian University in Cracow for the chemical analyses of the microsphere material, and to Mr. T. Tyc for the computer calculations.

List of symbols

A, B = constants
d = microsphere diameter
d_m = average microsphere diameter
E = Young's modulus for sphere material
k or k_{eff} = effective (apparent coefficient of thermal conductivity
k_c = conduction component
k_g = thermal conductivity of gas
k_{gc} = component of heat conduction by residual gas
k_{g0} = thermal conductivity of gas under atmospheric pressure
k_{gr} = thermal conductivity of microspheres
k_r = raduation component
k_s = thermal conductivity of sphere material
k_{ss} = solid-state heat conduction component
$Kn = \frac{\bar{L}}{\delta_a}$ = Knudsen's number
L = thickness of layer of spheres

$\bar{L}$ = mean free path
N = number of thermal contacts
P = externally applied pressure
p = residual gas pressure
Q_{1c} = heat flux flowing through one thermal contact
Q_s = heat flux flowing through one sphere
r_0 = sphere radius
S_p and S_N = parameters depending on the mode of sphere packing
T = temperature
t = wall thickness
ΔT = temperature difference
V_s = volume of sphere
α = accommodation coefficient
δ_a = average local distance between surfaces of contacting microspheres
δ_s = solid fraction
ε = effective emissivity coefficient of the packing
μ = Poisson's ratio
ϱ_s = density of sphere
σ = Stefan–Boltzmann constant

References

1 R. Wawryk, Heat transfer in selected kinds of microsphere insulation in the temperature range 80–300 K. PhD thesis, Wrocław, 1984.
2 N. H. Deschamps and G. L. Bernier, Thermal super insulation, Patent USA, No. 3., 769, 1973, 770·
3 C. K. Chan and C. L. Tien, Conductance of packed spheres in vacuum. J. Heat Transfer 95 (1973) 302.
4 J. D. Klein, Symposium on thermal radiation of solids. NASA SP-55 (1965) 73.
5 M. G. Kaganer, Thermal insulation in low temperature technique, Moscow 1966 (in Russian).
6 R. Wawryk, J. Rafałowicz and K. Balcerek, The influence of microsphere diameter on the coefficient of thermal conductivity of microsphere insulation. Cryogenics, 23 (1983) 441.
7 R. Wawryk and J. Rafałowicz, Thermal conductivity of coated microspheres, Cryogenics, 25 (1985) 33.
8 M. G. Kaganer, Heat and mass transfer in low temperature constructions, Moscow 1979 (in Russian).

Zusammenfassung — Es werden die Ergebnisse einer Wärmetransmissionsuntersuchung einer neuartigen Isolation (Mikrokugelisolation) dargestellt. Der Einfluß von Mikokugeldurchmesser, von Konzentration an metallisierten Mikrokugeln und Restgasdruck auf die Wärmeleitfähigkeit der Isolation wurden untersucht. Die Messungen der Wärmeleitfähigkeit wurden zwischen 77 und 300 K an Mikrokugeln verschiedenen Durchmessers (95, 130, 270 μm) mit einem Gehalt an versilberten

Mikrokugeln von 7 und 32% durchgeführt. Die Messungen der durchschnittlichen Wärmeleitfähigkeit (77–296 K) wurden bei einem Restgasdruck $k(p)$ im Bereich von 10^{-3} Pa zu 10^{+5} Pa reinem Stickstoffes getätigt. Die den gasförmigen Stoffen zuzuschreibende Wärmetransmissionskomponente $k_{gc}(p)$ wurde abgeschätzt.

Резюме — Представлены результаты исследования переноса тепла в новом типе изоляции (микросферной изоляции). Изучено влияние диаметра микросфер, концентрации металлизированных микросфер и остаточно давления газа на теплопроводмость.. Измерения термической проводимости микросфер с различным диаметром (95, 130 и 270 мкм) и образцов с посеребрянными микросферами с концентрацией 7–32% были проведены в интервале температур 77–300 К. Измерения средней термической проводимости в интервале температур 77–296 К были проведены при давлении чистого азота 10^{-5}–10^{5} Па. Установлена компонента теплопереноса, обусловленная газом.