CONF-8710249--2

DE88 012928

THE THERMAL CONDUCTIVITY OF BEDS OF SPHERES

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ABSTRACT

The thermal conductivities (k) of beds of solid and hollow microspheres were measured using two radial heat flow techniques. One technique provided k-data at 300 K for beds with the void spaces between particles filled with argon, nitrogen, or helium from 5 kPa to 30 MPa. The other technique provided k-data with air at atmospheric pressure from 300 to 1000 K. The 300 K technique was used to study bed systems with high k-values that can be varied by changing the gas type and gas pressure. Such systems can be used to control the operating temperature of an irradiation capsule. The systems studied included beds of 500 µm dia solid Al_2O_3, the same Al_2O_3 spheres mixed with spheres of silica-alumina or with SiC shards, carbon spheres, and nickel spheres.* Both techniques were used to determine the k-value of beds of hollow spheres with solid shells of Al_2O_3, $Al_2O_3 \cdot 7$ w/o Cr_2O_3, and partially stabilized ZrO_2.** The hollow microspheres had diameters from 2100 to 3500 µm and wall thicknesses from 80 to 160 µm.

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*Research sponsored by the Office of Fusion Energy, and the Energy Utilization Research, Energy Conversion and Utilization Technologies (ECUT) Program**, U.S. Department of Energy, under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

INTRODUCTION

Controlled mixtures of two phase systems, such as solids and gases, are the basis of many thermal insulation systems. The performance of an insulation depends on the individual components to heat transfer by convection, conduction by solid and gas, and radiation (1). Several such systems composed of beds of spheres of various materials were investigated in the present study. Poured beds of monosize solid spheres with gas-filled voids typically achieve about 60% of the theoretical density of the solid. The fraction of theoretical density of the poured bed can be increased to about 85% by filling the voids with smaller spheres (2). As such, these assemblies have the potential to yield thermal properties intermediate to those of the gas and the solid.

A specific use of these intermediate properties is to produce a relatively-high bed thermal conductance that can be varied by changing the gas type and the gas pressure. One primary goal of the present study is to determine if such a variable conductance bed can be applied to the temperature control of irradiation capsules (3). This part of the study was sponsored by the Office of Fusion Energy. The behavior at various exposure temperatures of metals and alloys subjected to neutron irradiation is important to the design of fission and fusion reactors. These irradiation studies have typically been conducted by exposing specimens imbedded in solid cylinders inside capsules that contained a small annular gap for a filling gas. From the estimated heat produced in the solid cylinder due to neutronic heating, and the known thermal conductivity of various gases, the gap could be sized to control the specimen exposure temperature. This can be accomplished through several reactor cycles needed to obtain the total neutron exposure, sometimes exceeding 10^{22} n/cm^2. Such designs have worked well where the gas gap is not too small to obtain accurately.

Table 1 shows the required gap sizes and gap conductances for various specimen irradiation temperatures in a typical irradiation capsule with heat generation rate of 27 kW/m inside the temperature control gap.

Table 1. Control gas gap conductance, C, (k/thickness) required at operating temperature in a typical irradiation capsule with a heat generation rate of 27 kW/m inside the gap.

Specimen Irradiation Temperature (°C)	Control Gas Gap Thickness (mm)	Control Gap Conductance (W/m^2·K)
400	0.117	950
330	0.085	1250
200	0.029	3300

The 200°C design requires a nominal gap of 0.029 mm at a power generation rate of 27 kW/m inside the gap for a reactor coolant near 55°C. One primary problem is machining and assembly of a concentric cylindrical system to maintain the needed gap dimensions and hence the needed gap thermal conductance. Obtaining a bed with higher thermal conductivity offers the possibility of increasing the gap dimension to one which is more easily achievable. The required metallic microsphere bed k for a 0.73 mm gap to match the helium gap conductance of 3300 W/m^2·K is 2.7 W/m·K.

A second goal of this study is to model and measure the thermal conductivity of beds of thin-wall hollow spheres produced from ceramic powder slurries using a coaxial nozzle process (4). In this process, slips of ceramics are blown into individual spheres at rates of 50 to 60 per second; these spheres are subsequently dried and fired to provide impervious walls. The modeling effort is to produce working computer programs based on existing, open-literature models for both hollow spheres and solid spheres. The measurement effort provides a base for comparison of thermal properties to commercially available insulation products being used for energy conservation products. This part of the study was sponsored by the ECUT program.

RADIAL HEAT FLOW APPARATUSES

Figure 1 depicts the unguarded radial heat flow apparatus used to measure k of beds of spheres near 300 K with the void spaces filled with argon, nitrogen, or helium at absolute pressures ranging from 5.1 kPa to 30 MPa. This apparatus was designated ORNL-7; its predecessors were described in references 6 and 7. Steady state k determinations are based on the temperature difference across the specimen in the radial direction, ΔT; the electrical power dissipated in the stainless steel core heater, $E \cdot I/L$; and the radial dimensions of the annular space, r_o and r_i. Accordingly, k is calculated from:

$$k = \frac{EI}{2\pi L} \cdot \frac{\ln r_o/r_i}{\Delta T} \qquad (1)$$

If significant axial heat flow occurs in the measurment section of ORNL-7, then a correction to k calculated from Eq. (1) is necessary. The length-to-diameter ratio of ORNL-7 is large, 22.5, which minimizes the amount of correction to k. The effect of axial heat flow for an unguarded radial measurement apparatus becomes increasingly important as the specimen k decreases. Two-dimensional thermal modeling with the HEATING5 computer

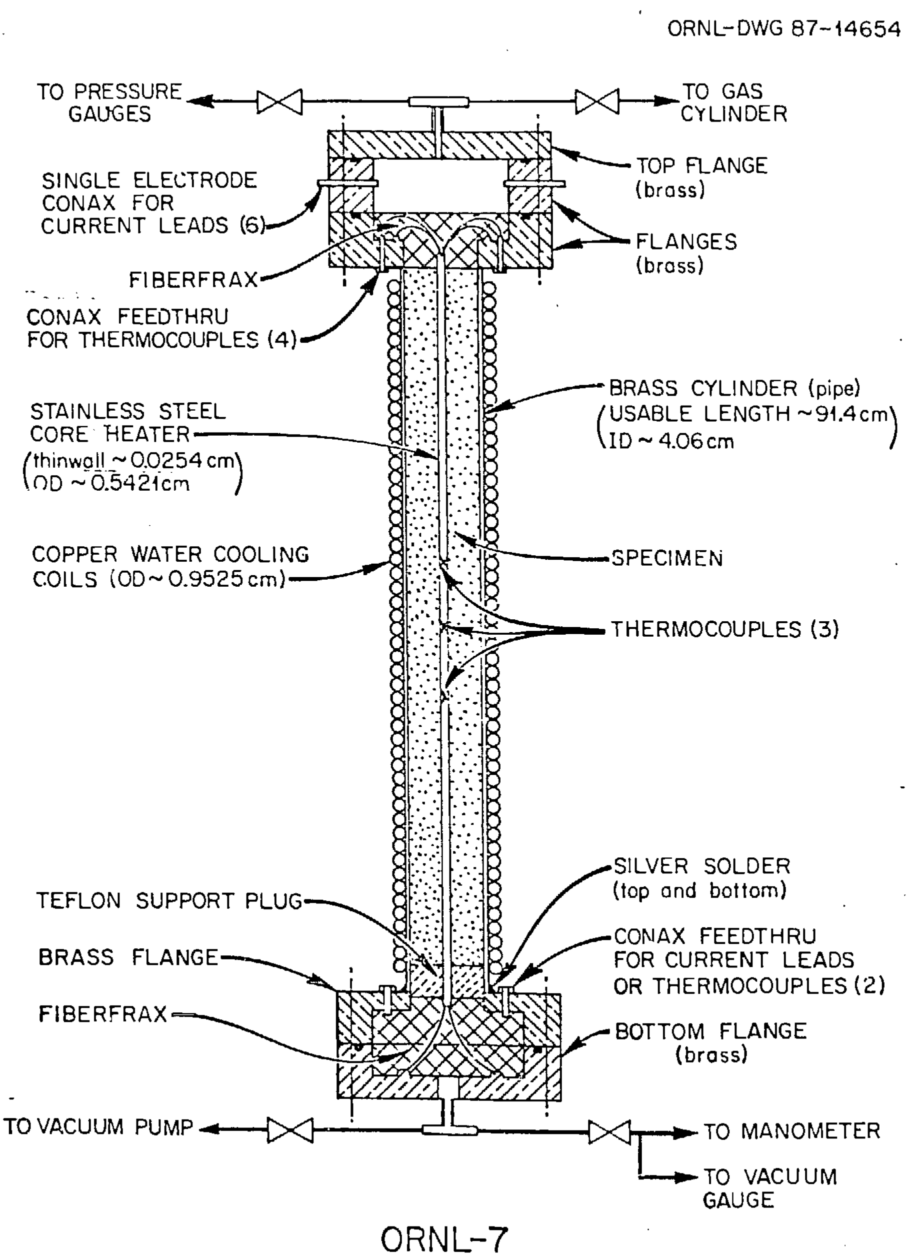

Figure 1. THE 300 K UNGUARDED RADIAL HEAT FLOW APPARATUS, ORNL-7

program (8) was performed to obtain the corrections required for a range of specimen k-values (see Table 2). A determinate error analysis (9) of ORNL-7 indicated a total error in k of ±3%.

Figure 2 depicts the unguarded radial heat flow apparatus (ORNL-8) used to measure k of beds of spheres from 300 to 1000 K in air. ORNL-8 is similar to ORNL-7 except the specimen annulus is formed by an instrumented stainless steel core heater tube and an instrumented outer cylindrical nichrome screen wire heater. Both are heated by dc power supplies. The core heater is instrumented with two Pt vs Pt-10% Rh thermocouples to measure the voltage drop in the central 10 cm of the heater and the surface temperature of the heater. Five Pt vs Pt-10% Rh thermocouples are welded to Pt studs welded near the central plane of the nichrome screen heater. The length-to-diameter ratio of ORNL-8 is about 17, which is sufficient to minimize the corrections to k calculated from Eq. 1. A determinate error analysis (9) of ORNL-7 indicated a total error in k of ±2.4%.

Table 2. Results of Thermal Modeling of ORNL-7 Using the HEATING5 Computer Program

Specimen Thermal Conductivity (W/m-K)	Percent Error in k Due to Axial Heat Flow %
5.0	0.20
2.0	0.20
1.0	0.20
0.5	0.20
0.2	0.22
0.1	0.20
0.05	0.20
0.007	0.70

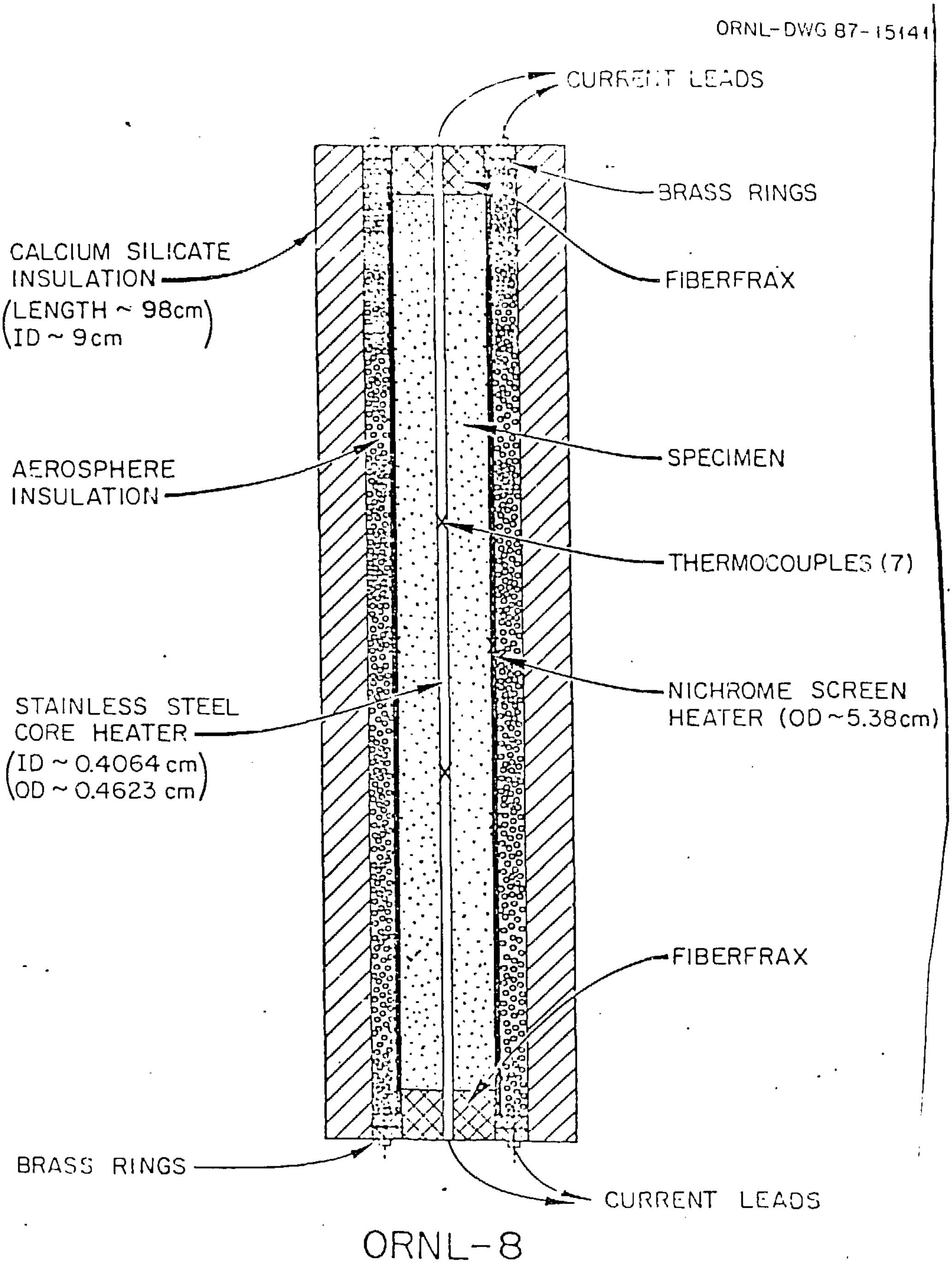

Figure 2. THE HIGH TEMPERATURE UNGUARDED RADIAL HEAT FLOW APPARATUS, ORNL-8

EXPERIMENTAL RESULTS

The characteristics of the beds of solid and hollow spheres tested in ORNL-7 and ORNL-8 are given in Table 3.

Table 3A provides the solid sphere materials, nominal diameters, and the density of the beds that were tested using ORNL-7. Table 4 lists the corresponding k-results obtained with N_2 and He gases at absolute pressures of 0.1, 1.0, and 30 MPa at 300 K.

The k-values of each bed increase with gas pressure; the percentage increase is much greater in helium than in nitrogen. The k-values with helium are 2.7 to 5 times larger than those with nitrogen. The k-values of the binary (coarse/fine) beds with Al_2O_3 (500 µm) are greater than the k-values of the beds of Al_2O_3 (500 µm) only, and of glassy carbon (355-425 µm) only. The bed of Al_2O_3 and SiC yielded the highest k-values for the ceramic materials. The binary bed of nickel spheres yielded k-values slightly higher than those of the bed of Al_2O_3 and SiC. Price (11) provides data for SiC that shows neutron irradiation decreases its k-value. Since similar decreases are expected for Al_2O_3 and glassy carbon, but not for metals, future work for application to irradiation capsule temperature control systems will focus on beds of metallic spheres.

Table 3B provides the sphere diameter and wall thickness for the ceramic hollow spheres forming the beds tested in ORNL-7 and ORNL-8. Figure 3 shows that the k-values increase with nitrogen gas pressure and depend on the material forming the hollow sphere and the sphere diameter. The bed

Table 3. Characteristics of Beds of Spheres Tested

A. Solid Sphere Beds

Bed Designation	Material	Diameter (μm)	Bed Density (kg/m³)	Apparatus (ORNL-7 or 8)
A	Al_2O_3	500	2375	7
AZ6	Al_2O_3 plus 600 Zeeospheres(a)	500 3-25	2410	7
AZ8	Al_2O_3 plus 850 Zeeospheres(a)	500 18-107	2440	7
ASC	Al_2O_3 plus 320 grit SiC	500 48	2590	7
C	Glassy Carbon	355-425	848	7
NN	Nickel (b) Nickel	700 200	5260	7

B. Hollow Sphere Beds (9)

Bed Designation	Material	Diameter (μm)	Wall (μm)	Bed Density (kg/m³)	Apparatus (ORNL-7 or 8)
11	Al_2O_3	3448	78	260	7
12	Al_2O_3	2809	104	480	7, 8
13	Al_2O_3	2229	92	590	7
14	Al_2O_3	2289	132	790	7
15	Al_2O_3	2106	130	900	7
21	$Al_2O_3 \cdot 7$ w/o Cr_2O_3	2852	91	410	7, 8
22	$Al_2O_3 \cdot 7$ w/o Cr_2O_3	3498	70	240	7, 8
31	Partially Stabilized ZrO_2	2250	55	560	7, 8

(a) Zeeospheres is an alloy of $Al_2O_3 \cdot SiO_2$ hollow spheres, Zeelon Industries, Inc., St. Paul, MN

(b) Federal Mogul Metal Powder

Table 4. Thermal Conductivity Results for Various Beds of Solid Spheres in Nitrogen and Helium as a Function of Gas Pressure at 300 K.

Bed Designation	Gas	Thermal Conductivity, W/m•K 0.1 MPa	1.0 MPa	30 MPa
A	N_2	0.274	0.293	0.30
	He	1.19	1.40	1.44
AZ6	N_2	0.45	0.47	0.50
	He	1.23	1.74	1.91
AZ8	N_2	0.40	0.45	0.47
	He	1.30	1.61	1.66
ASC	N_2	0.43	0.47	0.57
	He	1.42	2.36	2.46
C	N_2	0.39	0.40	-
	He	1.08	1.34	-
NN	N_2	0.49	0.51	0.53
	He	1.83	2.38	2.52

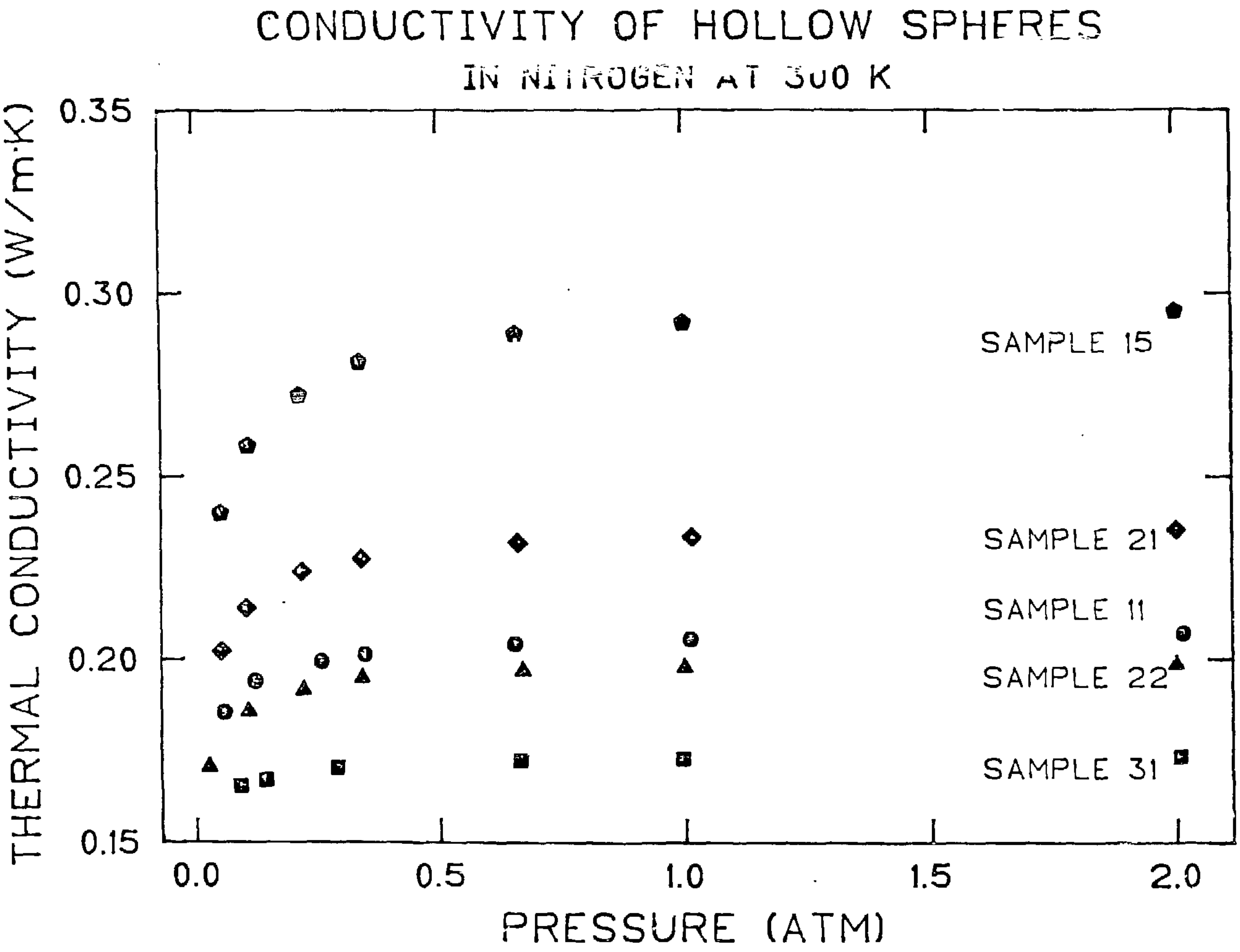

Figure 3. THE THERMAL CONDUCTIVITY AT 300 K OF VARIOUS HOLLOW CERAMIC SPHERE BEDS AS A FUNCTION OF NITROGEN GAS PRESENT

k-values decrease when the Al_2O_3 is alloyed with Cr_2O_3 and are lowest for spheres of partially stabilized ZrO_2.

Figure 4 shows that the bed k-values in helium for Al_2O_3 shells increase as bed density increases.

Figure 5 shows that the k-values measured for ceramic hollow spheres in air, using ORNL-8, increase with increasing temperature in the range 300 to 1000 K. These results were empirically fitted to equations of the form $a + bT + CT^3$. If the radiation contribution is associated with the cubic term, then extinction coefficients in the range 800 to 1200 m^{-1} are obtained using the Rosseland approximation:

$$k_r \simeq \frac{16}{3} \frac{n^2 \sigma T^3}{E}.$$

where n is the index of refraction, σ is the Stefan-Boltzman constant, E is the extinction coefficient, and T the absolute temperature.

MODELS

A calculational procedure developed by Moore, et al (2), has been used to calculate k_a, the apparent thermal conductivity, for solid sphere beds like those tested. The calculation has been implemented by a FORTRAN program that uses sphere size, solid properties, and gas properties as input data. Apparent thermal conductivities are then calculated for a range of accommodation coefficients. This model correctly predicts the pressure dependence of the apparent thermal conductivity and can produce results that agree with experiment by using the accommodation coefficient as an adjustable parameter.

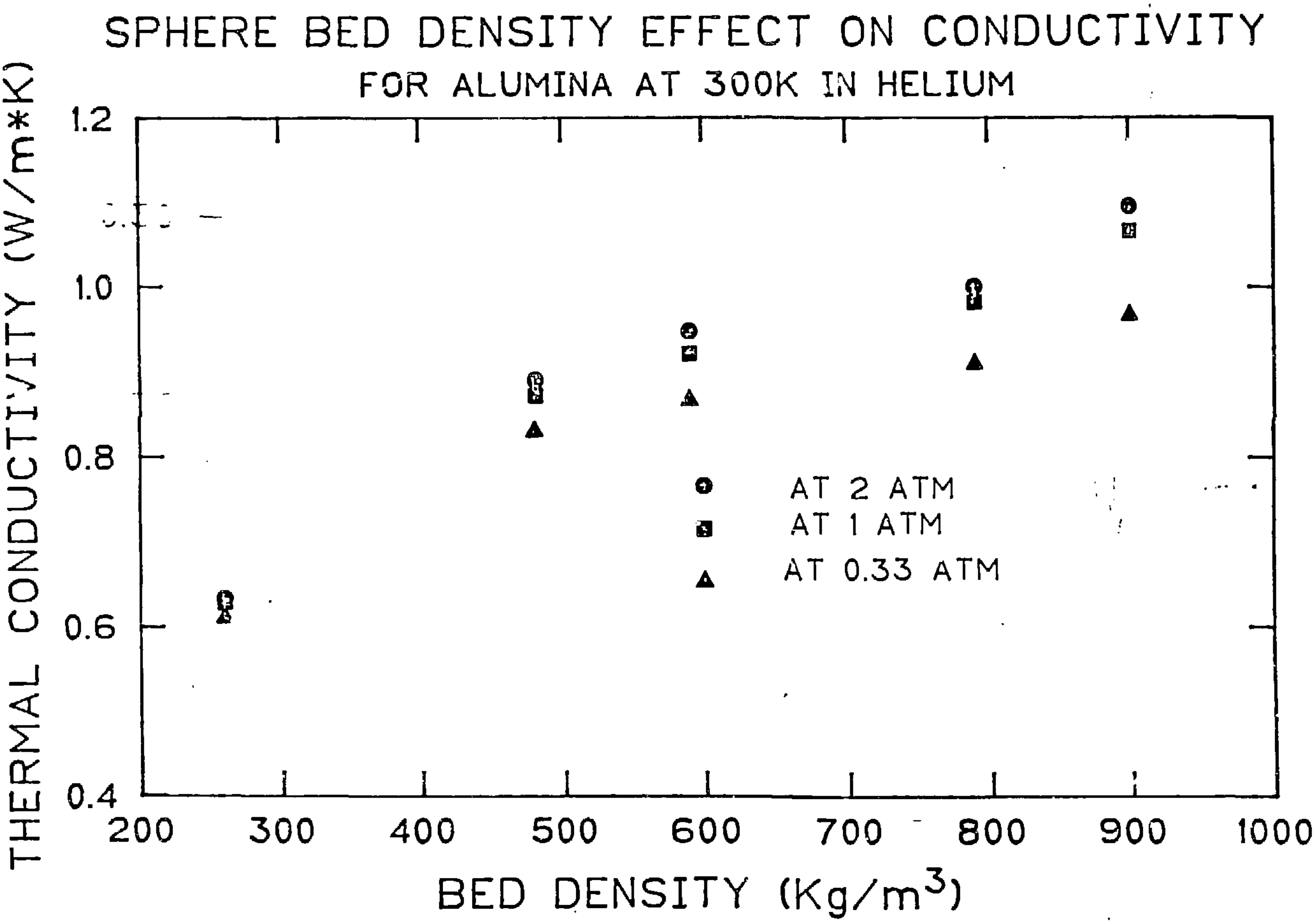

Figure 4. THE THERMAL CONDUCTIVITY AT 300 K OF VARIOUS HOLLOW Al_2O_3 SPHERE BEDS IN HELIUM AS A FUNCTION OF BED DENSITY

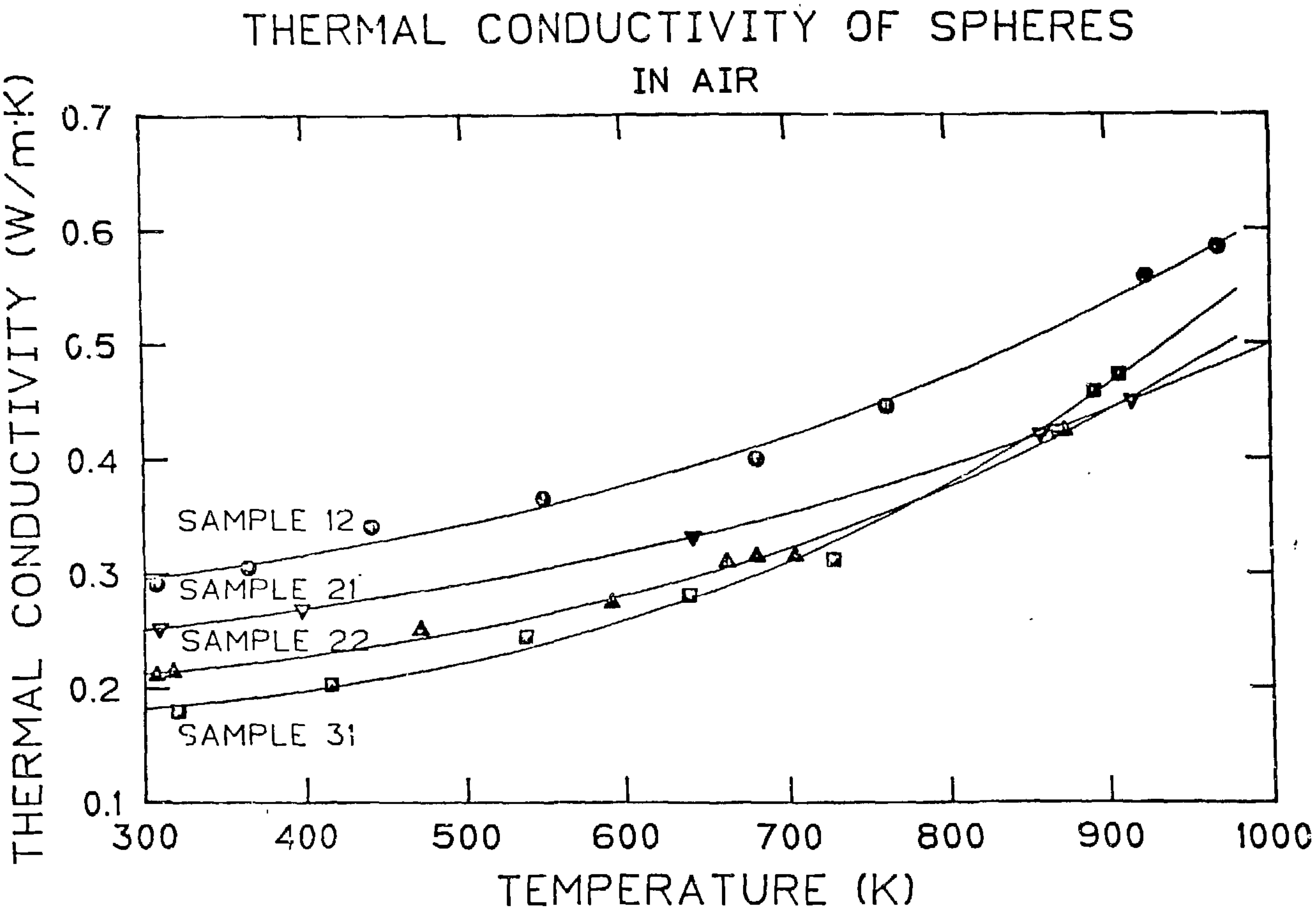

Figure 5. THE TEMPERATURE DEPENDENCY OF THE THERMAL CONDUCTIVITY IN AIR FOR VARIOUS HOLLOW CERAMIC SPHERE BEDS

The experimental k_a for hollow spheres have been compared with a modified version of a model developed by Parmley and Cunnington (12) and discussed by Cunnington and Tien (5). The model treats k_a as a sum of solid conduction, gas conduction, and radiation.

$$k_a = k_{sc} + k_{gc} + k_r \tag{2}$$

The solid conduction term, k_{sc}, is the product of a contact coefficient, a pressure term, and a bulk-solid thermal conductivity. The absolute pressure of the gas phase has been used in the k_{sc} calculation rather than the external compressive load used by Parmley and Cunnington. Agreement between calculated and measured k_a required k_{sc} proportional to P^n with $n = 1/2$ or less.

The gas conduction term, k_{gc}, includes the pressure dependent bulk gas thermal conductivity and the effective thermal conductivity of the hollow sphere. The material properties of the gas and the accommodation coefficient are included in the calculation of k_{gc} (12).

The radiative component, k_r, requires boundary emittances and an extinction coefficient for the sphere bed. An extinction coefficient of 1200 m^{-1} was obtained from the T^3 coefficient obtained for the Al_2O_3 spheres on the interval 300 to 1000 K.

The model can be viewed as having at least two adjustable parameters; the contact coefficient and the pressure exponent. The sphere bed void fraction was taken to be the ideal value $m = 0.4$, but subsequent analysis shows that the calculation of k_a is sensitive to this parameter.

Figures 6 and 7 show calculated k_a-values compared with measured k_a-values. Figure 6 is typical of the results obtained for hollow spheres in N_2 using $n = ½$ for the pressure exponent while Figure 7 shows similar results for hollow spheres in He. The agreement between calculated and experimental values can be improved to better than ±10% shown in the figures by adjusting the pressure exponent or the void fraction or both.

CONCLUSIONS

The thermal conductivity at 300 K of beds of solid spheres are dependent on sphere material and increase with gas pressure and bed density. Beds of metallic spheres are promising gap media for irradiation capsules.

The thermal conductivity of beds of hollow ceramic spheres are dependent on the sphere material and increase with temperature, gas pressure, and bed density.

The pressure dependence of bed k-values at 300 K can be predicted to ±10% by models with adjustable values for contact coefficients and pressure exponent.

Bed k-values in air at 300 K are between 0.2 and 0.3 W/m·K and increase to 0.5 to 0.6 W/m·K near 1000 K. These k-values are about twice those of lower-density, commercially-available, fibrous thermal insulations.

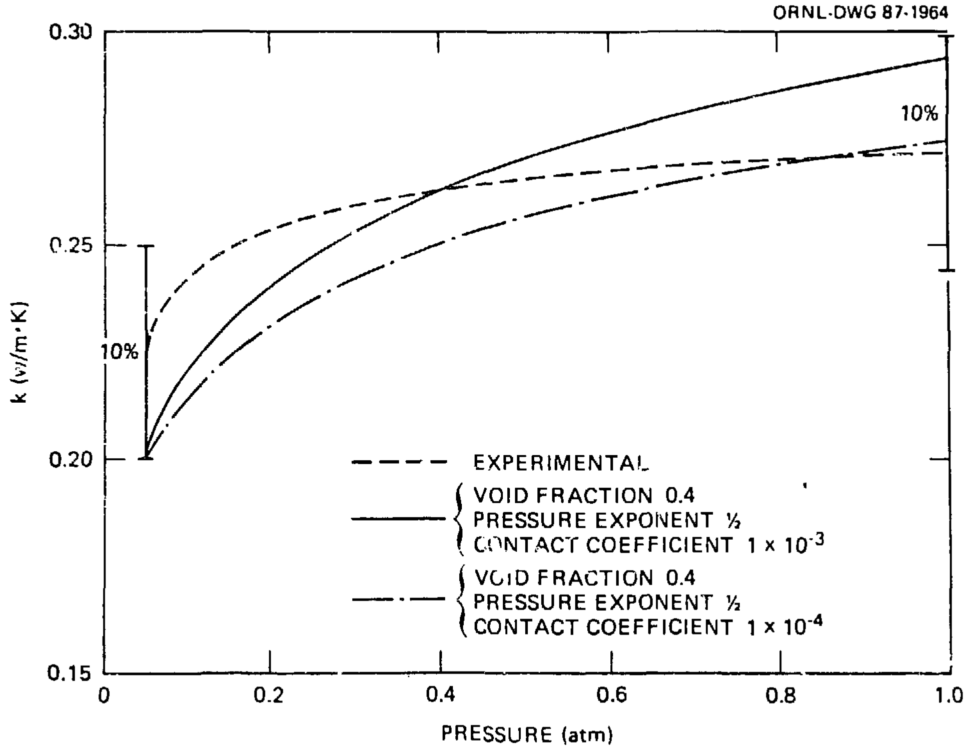

Figure 6. MEASURED k_a FOR 2229 μm-diam. HOLLOW SPHERES WITH 126 μm WALL THICKNESS IN NITROGEN COMPARED WITH CALCULATED VALUES. (a) EXPERIMENTAL DATA (b) CALCULATIONS WITH VOID FRACTION 0.4, PRESSURE EXPONENT 0.5, AND CONTACT COEFFICIENT 0.001 (c) SAME AS (b) EXCEPT CONTACT COEFFICIENT 0.0001.

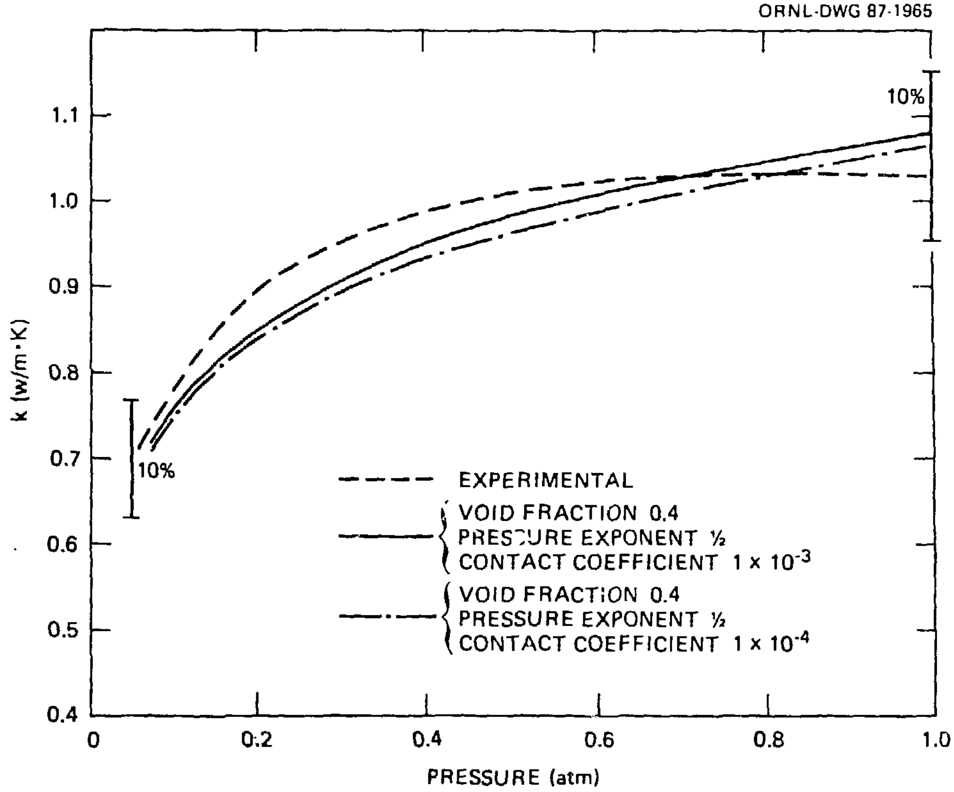

Figure 7. MEASURED k_a FOR 2106 μm-diam. HOLLOW SPHERES WITH 157 μm WALL THICKNESS IN HELIUM COMPARED WITH CALCULATED VALUES. (a) EXPERIMENTAL DATA (b) CALCULATIONS WITH VOID FRACTION 0.4, PRESSURE EXPONENT 0.5, AND CONTACT COEFFICIENT 0.001 (c) SAME AS (b) EXCEPT CONTACT COEFFICIENT 0.0001.

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