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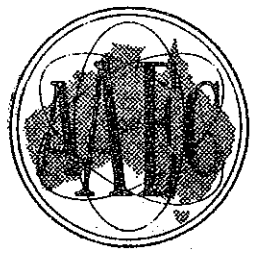

AUSTRALIAN ATOMIC ENERGY COMMISSION

RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

HIGH TEMPERATURE THERMAL CONDUCTIVITY OF BERYLLIUM OXIDE

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D.L. McDONALD
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ABSTRACT

The thermal conductivity of beryllium oxide samples produced by isopressing at 20 ton in^{-2} and sintering at 1505 °C was measured in the range 360 °K to 1200 °K. The BeO compacts measured had a density of 2.85 g cm^{-3} and an average grain size of 6μ. The thermal conductivity was found to be given by the relation:

$$k = 1.01 \times 10^{4}\ T^{-1.47}\ ,$$

where k is the thermal conductivity (watt (cm deg K)$^{-1}$) and T is the absolute temperature. This result is compared with other thermal conductivity measurements on similar materials.

CONTENTS

1. INTRODUCTION

Accurate values of the high temperature thermal conductivity of beryllium oxide are required for the assessment of its use in high temperature reactors since thermal stress resistance is an important limitation in its utilization. There is a wide variation in published values for the thermal conductivity of BeO (See Figure 1) over the range 0 °C to 1000 °C. Deviation from the mean curve drawn through the published values are as high as 50 per cent. at some temperatures. A critical evaluation of these results is made difficult by the lack, in many cases, of information concerning the purity and microstructure of the material under test, and estimates of the experimental accuracy of the measurements themselves.

It was therefore thought necessary to make accurate absolute measurements of the thermal conductivity of BeO compacts produced by the Australian Atomic Energy Commission since, in terms of the microstructure required for stability under neutron irradiation, these are not directly comparable with any of the other reported measurements.

This report gives the results of an absolute determination of the thermal conductivity of A.A.E.C. fabricated BeO below 1000 °C. The results form part of a general study to investigate the effect on thermal conductivity of high temperatures, neutron irradiation, grain size, porosity, impurities, and fuel additions.

2. DETAILS OF SPECIMENS

The specimens for the measurement of thermal conductivity were produced from Brush UOX grade BeO powder. The material as received had the following impurities (in p.p.m.).

Na	Mg	Ca	Al	Si	Fe	S	F	C
<35	60	<20	35	50	25	470	<5	310

The powder was homogenised by water grinding with BeO cylinders and isopressed at 20 ton in^{-2} . The compacts were then sintered for 1 hr 15 min at 1505 °C in dry nitrogen and the specimens machined to size (2.0 cm dia. by 5.1 cm long) using a diamond wheel.

The final density of the material was 2.85 g cm^{-3} , that is, 95 per cent. of the theoretical density of BeO. The microstructure was examined by means of electron micrographs of shadowed replicas of polished and etched samples cut from the specimens after measurements of thermal conductivity had been completed. The micrographs showed a considerable lack of uniformity in grain size. Most grains were in the range from 2 to 10μ but large grains up to 20μ in diameter were occasionally observed. The average grain size was about 6μ. A typical micrograph is shown in Figure 2.

3. EXPERIMENTAL DETAILS AND RESULTS

The method chosen for measuring the thermal conductivity was dictated by sample size and the requirement for a reasonable degree of accuracy. Large discs suitable for a radial heat flow approach could not be fabricated easily and would not have been suitable for high temperature irradiation in high flux positions in the research reactor HIFAR. The "Unmatched Guard Method" developed by Laubitz (1963) was used for the measurement since it combines the advantage of a reasonably small specimen diameter and good accuracy. The method has been described in detail by Laubitz (1963,1964).

The chief advantages of the method over other axial flow and comparator methods lie in the insensitivity of the results to guard temperature, lining-up of components of the apparatus, and thermocouple calibration errors.

The apparatus is shown schematically in Figure 3. It has cylindrical symmetry with an outside diameter of 6.5 cm and an overall length of 25 cm. The central specimen assembly consists of 5 pieces with an outside diameter 2.0 cm. A_2 and A_4 are the actual specimens under test and are 5.1 cm long. A_1 and A_5 are BeO spacers, each 6.8 cm long, and A_3 is a central BeO spacer 1.2 cm

long. The specimen assembly is surrounded by a cylindrical guard having three independently controlled heater windings which are used to help raise the specimen to a particular temperature and to provide a reasonably isothermal region for the thermocouples on the specimen assembly. The regions between the specimen assembly and guard and the guard and water jacket were filled with alumina powder insulation.

As shown in Figure 2, A_1, A_2, A_4, and A_5 have heaters mounted on their end faces. These were wound from 0.0075 -inch (0.019 cm) dia. Pt-40 per cent. Rh wire in the form of coils and cemented in grooves cut in the face of each cylinder. The spacer heaters were used to raise the specimens to the required temperature and the specimen heaters to establish small temperature gradients in the specimens from which the conductivity could be calculated. All power supplies to these heaters were regulated to 0.05 per cent. or better to minimize drift.

Each specimen had 15 Pt/Pt-13 per cent. Rh thermocouples located on its surface in 5 planes and 3 columns displaced 120° from each other. The thermocouples, which were made from 0.0075 inch (0.019 cm) dia. wire, were firmly imbedded in 0.017 inch (0.043 cm) dia. holes drilled in the specimen surface by means of an ultrasonic drill and a cylindrical drilling jig.

The central spacer A_3 had a ten junction thermopile located around its surface on two planes 1 cm apart. The whole specimen assembly including the thermocouple locations, was symmetrical about the centre plane of A_3.

To measure thermal conductivity, the specimens were raised to the required temperature by use of the guard and spacer heaters and the heating was adjusted so that the output of the thermopile on the central spacer was nominally zero (that is, less than 1μV) and the guard temperature was slightly higher than the specimen temperature. At equilibrium, the outputs of the 15 thermocouples on each specimen were measured by means of a Leeds and Northrup 6-dial potentiometer. The specimen heaters were then switched on and again balanced to give zero output from the thermopile and a suitable temperature gradient along the specimens. At equilibrium the thermocouple outputs were again measured and the power dissipated in the specimen heaters was determined. The specimen heaters were then turned off and the assembly returned to its original condition, the spacer and guard heaters being untouched during this sequence of operations. A further set of readings at equilibrium served to indicate any drift in the guard and spacer heaters.

Using this procedure Laubitz (1963) showed that for the symmetrical arrangement under consideration:

$$\frac{1}{2} \Sigma \Delta T \ (Z_i) = Z_i \ G + C_o - \sum_{n \text{ odd}} C_n \sin \frac{n \pi Z_i}{L} \quad , \qquad (1)$$

where $\Delta T \ (Z_i)$ is the difference in temperature of the thermocouple located on the surface of the specimen at $Z = Z_i$ taken with the specimen heaters on and off, $\frac{1}{2}\Sigma \ \Delta T \ (Z_i)$ is the average of this difference for the two corresponding thermocouples symmetrically located on the top and bottom specimens, and G is the linear temperature gradient related to the power outputs of the specimen heaters $\dot{Q}(2)$ and $\dot{Q}(3)$ by:

$$\dot{Q}(2) + \dot{Q}(3) = 2\pi a^2 \, k \, G \ ,$$

where a is the specimen radius, L the total length of the specimen assembly, and k the average thermal conductivity of the two specimens.

C_o is a constant and the C_n are a converging sequence of functions of the geometrical parameters of the system as a whole, and of the ratio of the conductivity of the specimen to that of the surrounding insulation. The form of these functions is given by Laubitz (1963). In the computation it was found necessary to carry the summation out to at least n = 40 to ensure convergence of the calculated value of conductivity.

G was computed from (1) by a least squares analysis of the experimental data on an IBM 7040 computer with G one of 3 disposable parameters. (The other two parameters (ϵ and κ) enter into the C_n and are defined by Laubitz (1963)). As mentioned above, the C_n are functions of the

ratios of the thermal conductivities, and the calculated values of k were re-used in an iterative procedure in which k converged rapidly to a terminal value which was independent of the initial choice of k .

As described by Laubitz, the relationship (1) is derived assuming that the thermal conductivity along the specimen at any particular temperature is the same, whereas, in practice, with temperature differences of the order of 30 °C along each specimen during a measurement, this is not the case and a correction must be applied to allow for this effect. This correction was calculated from the temperature variation of the thermal conductivity given by the initial uncorrected values of conductivity to obtain a new set of ΔT's and the cycle of computations was reiterated to obtain the final measured values. This correction for the temperature variation of the conductivity amounted to from 1 to 2 per cent.

Using the above procedure the conductivity of the BeO specimens was measured at a series of temperatures from 360 °K to 1200 °K and the results of two successive runs of increasing temperature are shown in Figure 4. The solid curve is the least squares fit to these measured values, and is given by the relation:

$$k = \frac{1.01 \times 10^4}{T^{1.47}} ,$$

where k = Thermal conductivity (watt/cm deg K)

T = Temperature (°K).

These values are the measured absolute values of the material under test, and have not been corrected to 100 per cent. density. Figure 4 also shows thermal conductivity values from other sources.

The estimated maximum error in the measured thermal conductivity values for BeO is ± 7 per cent. This arises from two main sources. The first, amounting to ±4 per cent, is from a consideration of the accuracy of the experimental parameters such as uncertainty in power measurement and dissipation, and uncertainty in thermocouple location and the measured temperature differentials due to long-term drifts.

The second source is uncertainties in the value assumed for the thermal conductivity of the powder insulation. Powder conductivity is extremely sensitive to such variables as grain size, grain size distribution, and packing fraction, and as mentioned above, the ratio of specimen conductivity to that of the surrounding powder insulation enters into the calculation of k .

Computations to test the dependence of k on this ratio showed that, for example, 20 per cent. error in powder conductivity results in a 2 per cent. error in the calculated value for the conductivity of the specimen.

To minimize any error arising from this source, the alumina powder was screened to give a grain size "d" such that:

$$200\mu < d < 250\mu ,$$

and tested to determine an average packing fraction. This gave the packing fraction as 59 per cent. ± 4 per cent. Values for powder conductivity were then taken from the values reported for a similar powder by Godbee and Ziegler (1966). In assigning a maximum error of ± 3 per cent. from this source, we have estimated that the assumed value from the powder conductivity could still be in error by up to about 30 per cent.

As a further test of the apparatus and to determine if any unsuspected systematic errors are present, measurements are in progress on samples of "Hylumina" * . Its thermal conductivity has been measured by the National Physical Laboratory in Britain and this will provide an independent check on the apparatus.

* Commercial alumina-based material prepared by S. Smith and Sons (Australia) Pty. Ltd.

4. DISCUSSION

In Figure 4, the present results are compared with the values obtained by Elston et al. (1963) and with a selected curve drawn through the high temperature values of Lillie (1961) and the room temperature measurements by Pryor et al. (1964). Above 900 °K the three curves are in agreement. The selected curve was derived in the absence of any experimental values for A.A.E.C. fabricated BeO above room temperature and is seriously in error below 800 °K. The differences could have large effects on any thermal stress calculations on A.A.E.C. fabricated BeO carried out using this selected curve at temperatures below 800 °K.

A comparison between the present results and those of Elston is of interest because French material consistently gives a higher value of conductivity than A.A.E.C. material. At room temperature the French material is reported to have a conductivity of 3.5 watt/cm deg K. An extrapolation of the present results to 300 °K gives a value of 2.3 watt/cm deg K which is comparable with the value of 2.4 watt/cm deg K obtained for this material at 273 °K (Dullow Unpublished) using a low temperature thermal conductivity apparatus (Pryor et al. 1964). At low temperatures the difference is even more pronounced, the French material having a peak in the thermal conductivity vs. temperature curve of 15 watt/cm deg K at 80 °K, compared with a peak of 5 watt/cm deg K at 120 °K for A.A.E.C. material (Pryor et al. 1964).

The cause of this difference is not known, since the two materials have similar purity and density. At 273 °K, large grain size (50μ) material produced at the A.A.E.C. was found by the authors (unpublished) to have a conductivity of 3.1 watt/cm deg K so that difference in grain size accounts for some of the difference between Elston's values and the measurements reported here for 6μ BeO since the French material was reported to have grain sizes between 6 and 60μ with an average of 25μ. Moreover, the shape of the two curves from 300 to 900 °K is consistent with a grain size effect, the difference decreasing, as it does with increasing temperature. It is surprising however, that differences due to this effect should occur at such high temperatures since the phonon mean free path at, say, 300 °K at 1100 °K is only 170Å and 15Å respectively (Taylor 1962).

The whole question of the inter-related effects of porosity, grain size, and impurities is difficult to resolve. It is hoped that the planned measurements to examine the effects of each of these variables independently of the others will bring about a solution to these problems.

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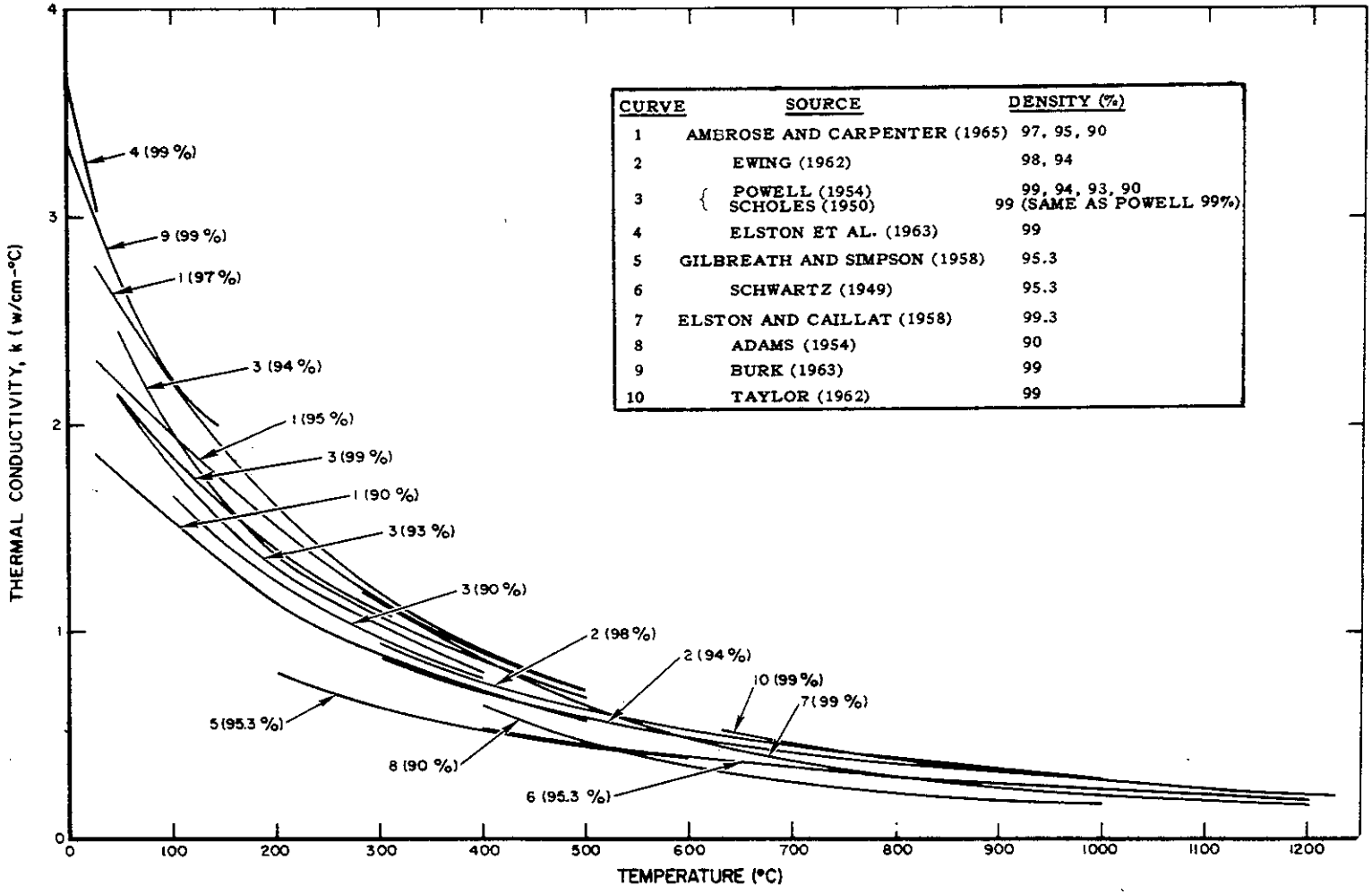

FIGURE 1. SOME PUBLISHED VALUES OF THE THERMAL CONDUCTIVITY OF BeO

(After Ambrose and Carpenter 1965)

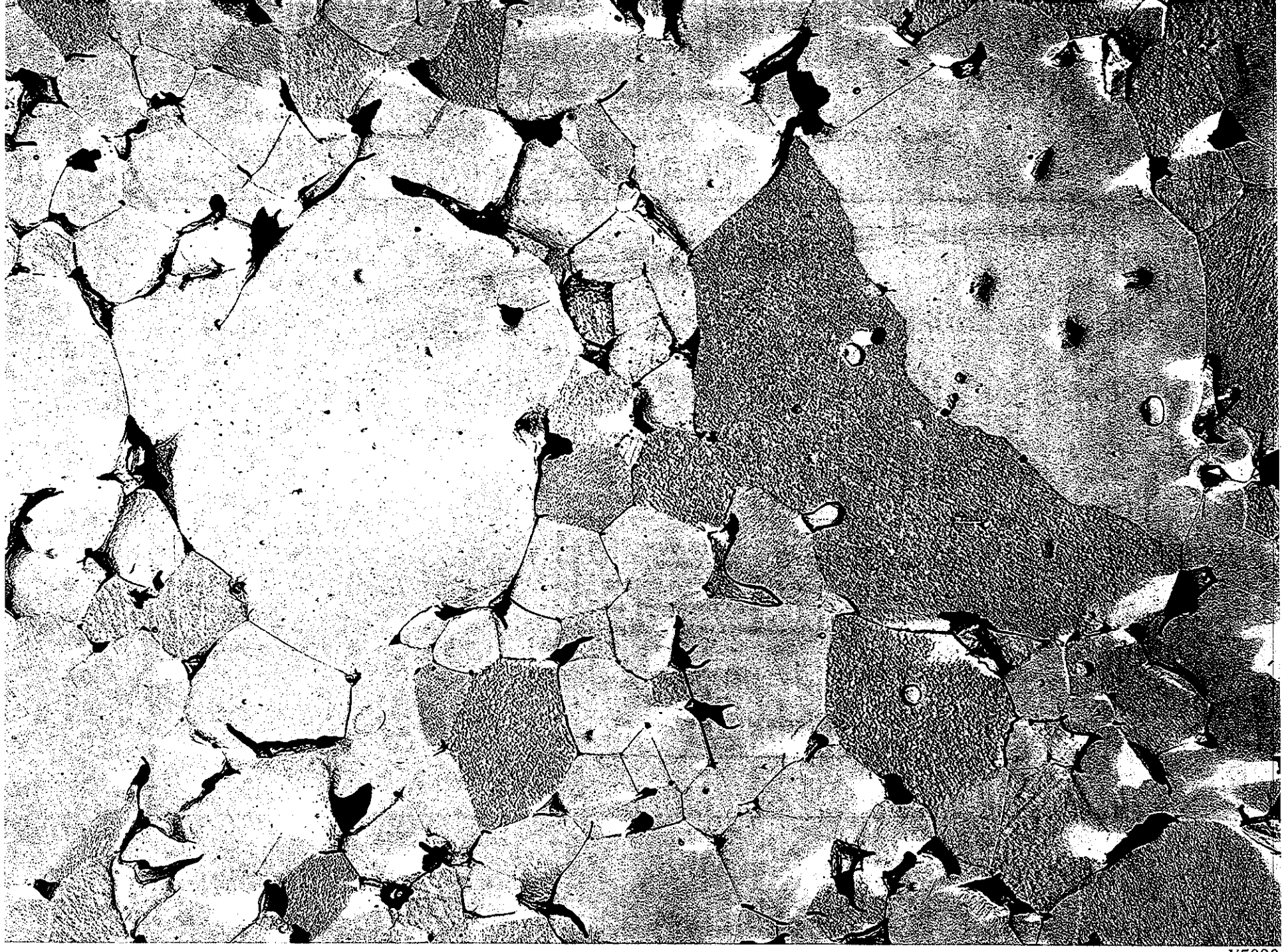

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FIGURE 2. TYPICAL ELECTRON MICROGRAPH OF SHADOWED REPLICA TAKEN FROM POLISHED AND ETCHED SURFACE OF BeO SPECIMEN

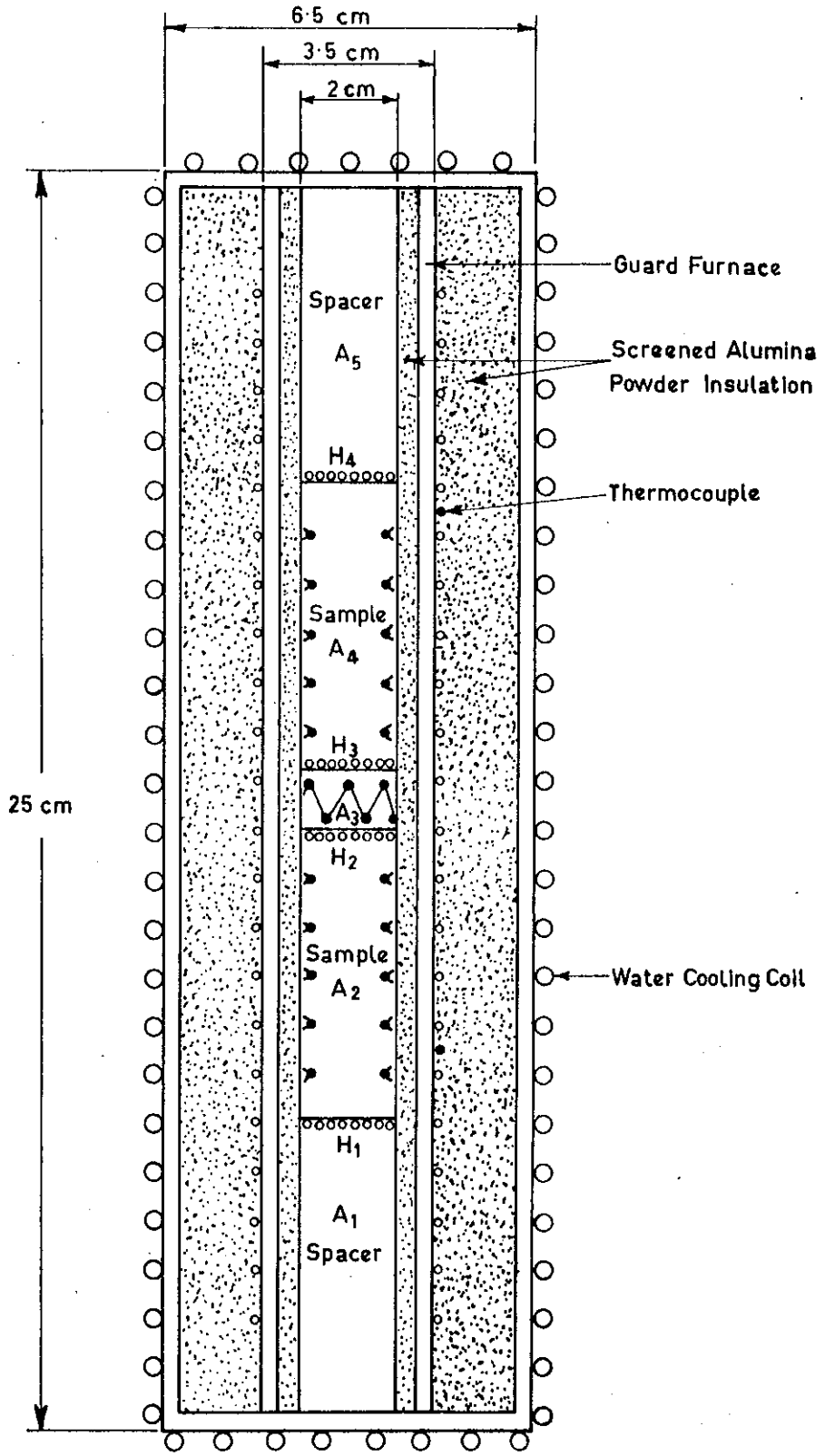

FIGURE 3. SCHEMATIC SECTION OF 'UNMATCHED GUARD' THERMAL CONDUCTIVITY APPARATUS

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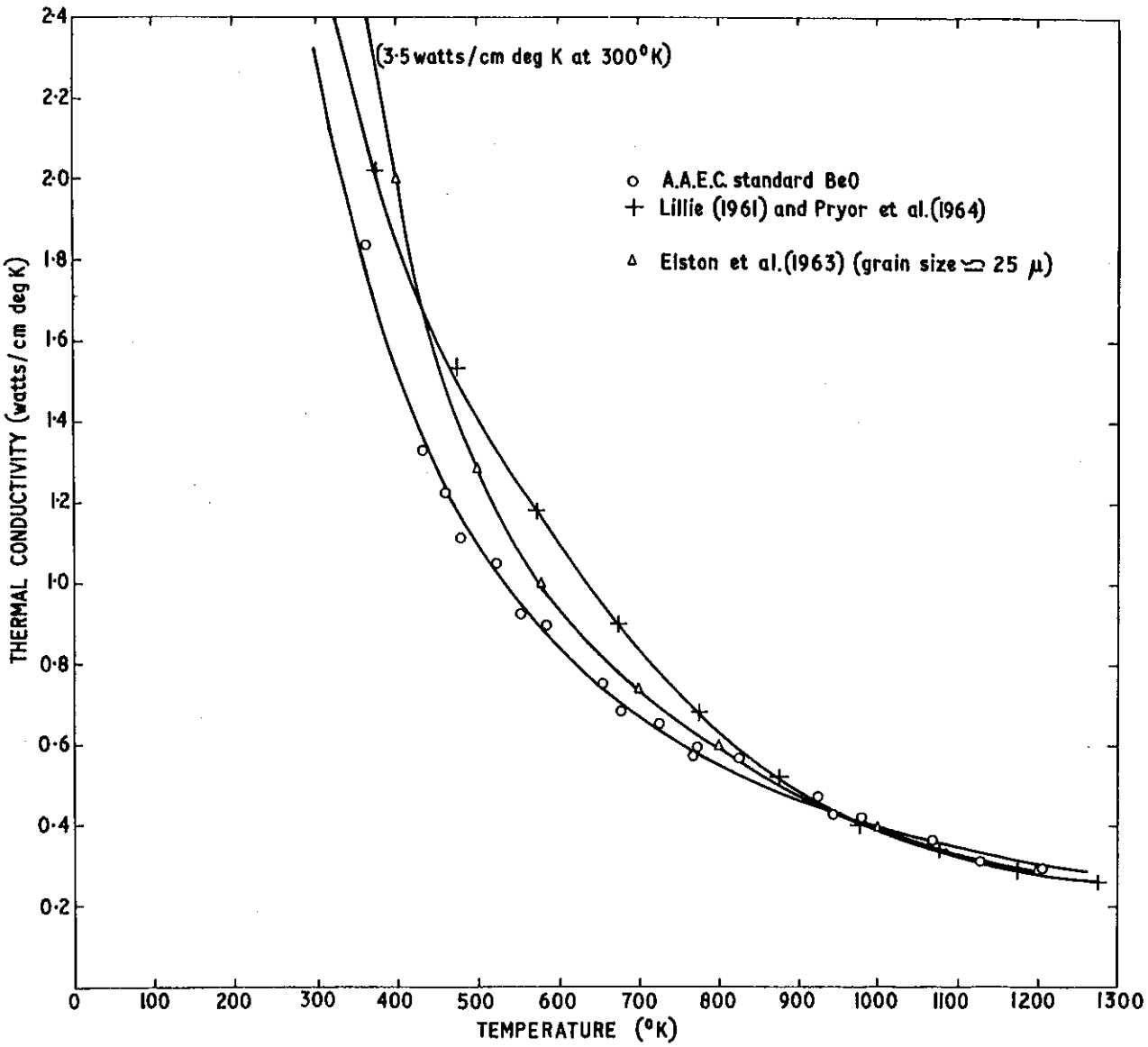

FIGURE 4. THERMAL CONDUCTIVITY OF BERYLLIUM OXIDE

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