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AN APPARATUS TO MEASURE THE THERMAL CONDUCTIVITY OF GRAPHITE NEAR ROOM TEMPERATURE

by

J. VAN de VELDE, H. OCKFEN and T. NOELS

1968

Report prepared at the CEN
Centre d'Etude de l'Energie Nucléaire, Mol - Belgium

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The heat transfer principle is used for measuring the coefficient K near room temperature.

The graphite specimen placed in a vacuum is held coaxially between the end faces of the two electrolytically pure copper probes which have the same cross-section as the sample, and of which, one is heated and the other is cooled.

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SUMMARY

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KEYWORDS

THERMAL CONDUCTIVITY
MEASUREMENT
GRAPHITE
INSTRUMENTS
HEAT TRANSFER
VACUUM
COPPER
MECHANICAL STRUCTURES
HEATING
COOLING

CONTENTS

Introduction

The thermal conductivity of solid material is of great importance in all applications involving the exchange and transport of thermal energy.

Nuclear graphites are used as moderator and some specially fabricated graphites are used as fuel cans. In any case the determination of the thermal exchanges between the graphite and the cooling medium implies the knowledge of the method of how the heat propagation takes place.

In this report we indicate only one method for determining the thermal conductivity and which is based on the heat transfer principle and permits thermal conductivity to be determined near room temperature.

An apparatus for heat flow measurements adaptable to various graphite parallelepipided blocks has been developed in the LMA's graphite laboratory and has been used for a large number of specimens examined for the Deutsche Babcock-Wilcox Dampfkessel Werke program.

1. Laws of Heat Flow.

The thermal conductivity determines the heat flow through a specimen for a given temperature gradient, or determines the temperature gradient obtained when a fixed heat flow is employed.

Usually three methods of heat transfer are considered : conduction, convection and radiation. It is possible, however, to develop conditions where the amount of heat transferred by one of these methods is much greater than that transferred by the others, and then to study this case as an example of transfer by that method alone.

Convection is not considered in this investigation because it is only applicable in the case of fluids.

1.1. Conduction

The fundamental equation of thermal conductivity and which serves as a definition of that term is : [1, 2]

$$Q = K.A.\frac{\Delta\theta}{\Delta x}. \qquad \text{steady-state regime} \qquad (1)$$

Q = amount of heat flowing per unit time through an area A.

$\frac{\Delta\theta}{\Delta x}$ = temperature gradient in a direction perpendicular to A.

K, the thermal conductivity coefficient is defined by the relation of a thermal flux in unit time and a temperature gradient.

If Q is given in calories, θ in degrees centigrade, x in cm and A in cm^2; then K has the following dimensions : $cal.\ cm^{-2}.\ cm.s^{-1}.\ {}^\circ C^{-1}$ or $cal.cm^{-1}.\ s^{-1}.{}^\circ C^{-1}$.

The determination of K will result in measuring the temperature gradient and the heat flux.

1.2. Radiation

When heat is transferred by radiation between two surfaces, one at temperature T_1 and the other at temperature T_2, the amount of heat transferred per unit area of surface in a unit time is [1.]

$$Q = C\ (T_1^{\ 4} - T_2^{\ 4}) \qquad (2)$$

C = a constant, dependent of effective emissivity and geometrical shape of surfaces. Temperatures are on the absolute scale.

More general studies on thermal conductivity and heat flow can be found in references [3, 4, 5 .]

2. Static measurement of thermal conductivity.

In static methods, the sample is allowed to come to a steady state and the temperature distribution measured to determine the thermal conductivity, K, by equation (1).

Absolute methods of measurement require a precise measurement of both heat input to the system and the path of heat flow. In general, elaborate methods of heating and guarding and quite large specimens are required.

In contrast, a comparative method employs a material of known thermal conductivity to measure the heat flow, and can employ simple shapes of relatively small size.

The principle of the comparative method is simple. A uniform heat flow is established through two or more samples. The temperature drop, heat flow, and thermal conductivity are related by the equations:

$$Q = K_1 . A_1 . \frac{\Delta\theta_1}{\Delta x_1} = K_2 . A_2 . \frac{\Delta\theta_2}{\Delta x_2} = K_3 . A_3 . \frac{\Delta\theta_3}{\Delta x_3} = \cdots \quad (3)$$

As in our apparatus the graphite specimen is positionned between the end faces of two samples of known thermal conductivity and of the same cross-section as the graphite sample, equation (3) becomes

$$Q = K.A. \frac{\Delta\theta_1}{\Delta x_1} = K_s.A. \frac{\Delta\theta_2}{\Delta x_2} = K.A. \frac{\Delta\theta_3}{\Delta x_3} \quad (4)$$

$$\text{and } K_s = K \frac{\Delta\theta_1}{\Delta\theta_2} . \frac{\Delta x_2}{\Delta x_1} = K \frac{\Delta\theta_3}{\Delta\theta_2} . \frac{\Delta x_2}{\Delta x_3} \quad (5)$$

K_s : thermal conductivity coefficient of the sample

Q : heat flow

A : mean area

$\Delta\theta$: temperature decrease

Δx : thermocouples separation

The determination of K_s will result in measuring the temperature gradients, which are determined by thermocouple emf determinations.

The graphite sample is held coaxially between the end faces of two electrolytically pure copper probes each of them having two thermocouples a certain distance apart.

In the specimenitself, the thermocouples are positionned in small holes drilled in the material. This of course can cause perturbations in the temperature field.

One copper probe is heated by an electrical resistance in which a current is passing. The other copper probe is cooled by water circulation.

The entire system is placed in vacuum chamber.

At thermal equilibrium, the gradients are compared to provide thermal-conductivity values for the sample relative to that for copper.

The thermal conductivity of this material was taken as $0{,}91\ cal.cm^{-1}s^{-1}\ °C^{-1}$ (or $3{,}81\ W.\ cm^{-1}.°C^{-1}$)

For comparative reasons, the heat flux can be determined directly in deducing the calorific heat by measurement of the electrical power dissipated in the resistance of the hot source. The current I gives a tension drop V over the heating resistance R, and the dissipated quantity of calories is given by

$$W = 0{,}24\ R\ I^2 = 0{,}24.\ V.I.\quad \text{calories.}$$

This quantity of heat is not only necessary for creating a heat flux in the probes and specimen, but counts also for compensation of all thermal losses, such as thermal losses along the supporting elements, losses along the thermocouple wires, losses by conduction in the air andby radiation.

Working in a vacuum, the losses by conduction in air are negligible. Interfacial resistance between samples may cause a nonuniformity of temperature distribution and adds thermal resistance which has generally been included with that of the sample, giving erroneous results. This resistance is reduced by forming smooth surfaces and applying mechanical pressure to ensure good contact.

In any case, between the heat flux calculated by the electrical power dissipated in the hot source, and the flux measured by the thermocouples in the copper probes,there should be always a good relationship if everything is settled up in the right way.

3. Apparatus and Experimental Procedure.

3.1. Apparatus

The equipment evolved after consideration and testing of several modifications is shown in fig. 1 and 2. The apparatus has been adopted for the following specimen cross-sections: 16 x 16 mm / 5 x 7 mm / 2,5 x 6 mm /.
It consists of a hot-and a cold source positionned in a vacuum chamber.

The hot source is formed by an electrical resistance positionned around a cylindrical specimen in copper, isolated and contained in a steel envelope. Thermocouple-and current conductors are brought out through an insulated hole. The resistance stands in a regulating device having a mA meter and voltage meters. In our measurements the current I changed from about 150 mA to 250 mA. As shown in fig. 1, mechanical pressure ensuring good contact, is applied through a champing device mounted on supporting rods. Attached to this hot source is the copper probe containing two thermocouples positionned in 1 mm diameter holes in order to measure the heat flow entering the graphite sample.

The seoond copper probe containing two (or three) thermocouples is related directly to the cold source. This source consists of a chamber cooled by circulating water. The circulation system is of the closed circuit type and uses a thermostat and pump.

Control of the heat flow entering the sample, and leaving it can be made in this way.

The graphit sample, see fig. 3, is clamped between the end faces of the two copper probes. Small holes are drilled to mount the thermocouples in the specimen.

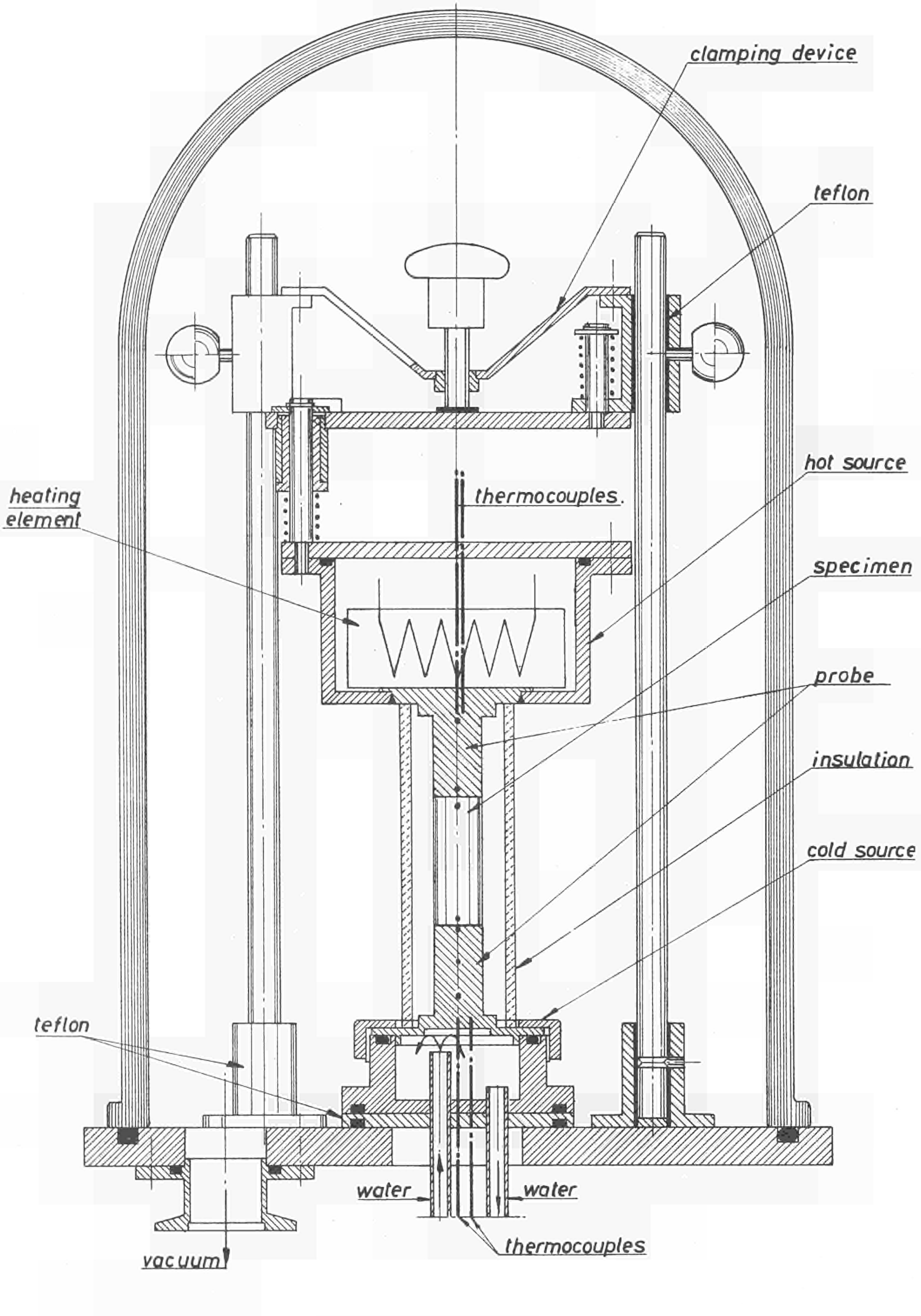

Fig. 1. PRINCIPLE.

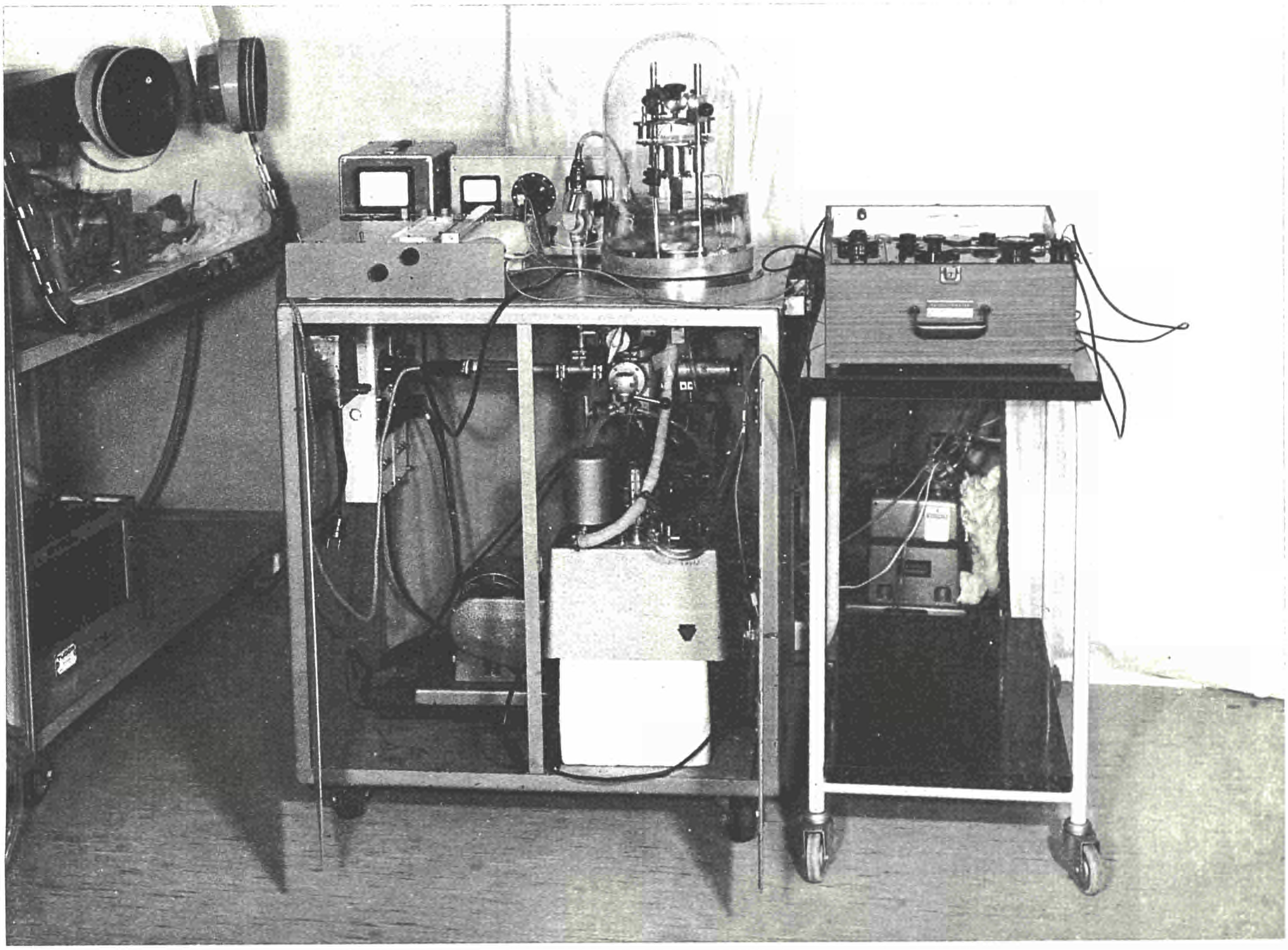

fig. 2

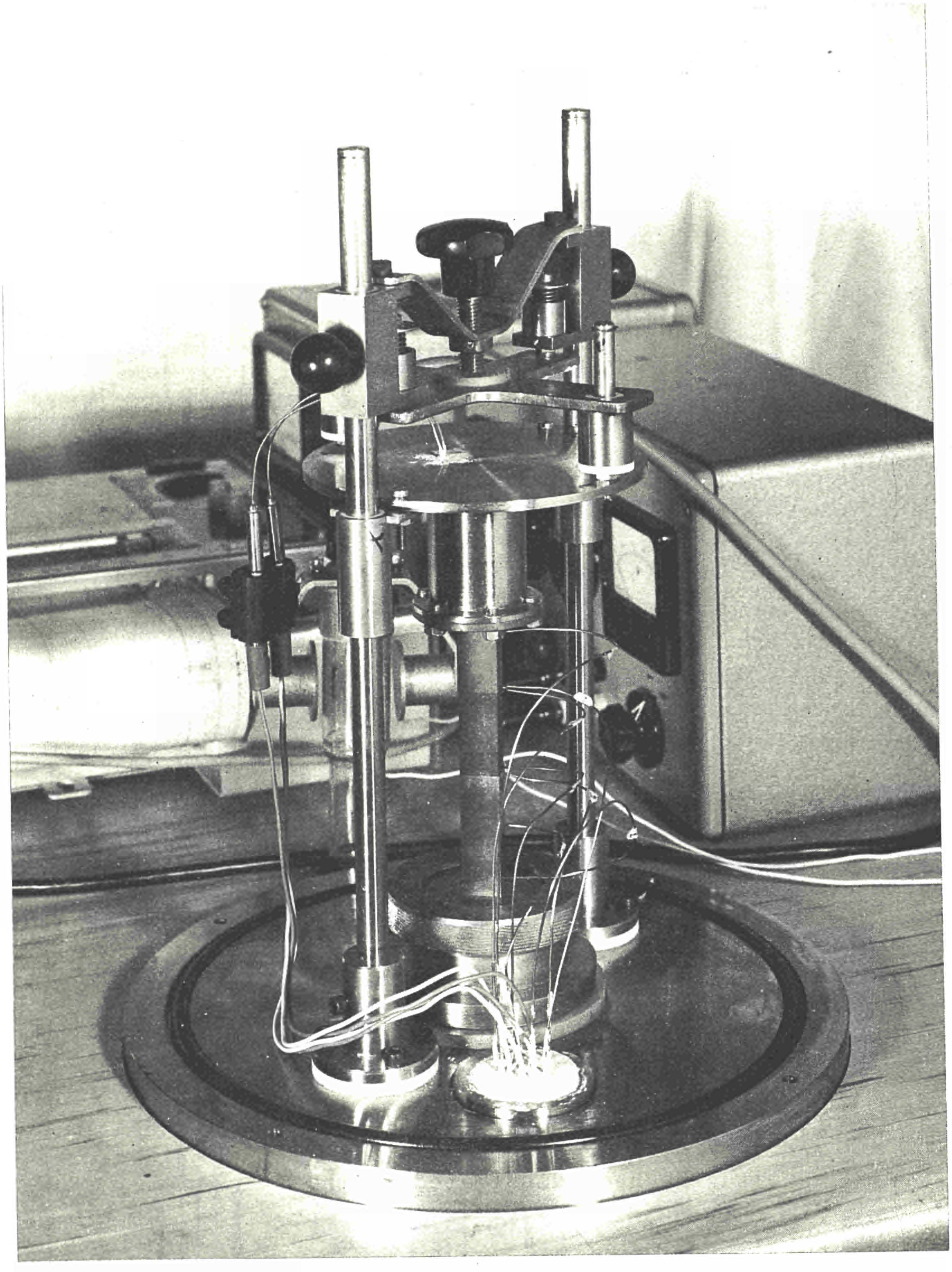

fig. 2a

The entire installation is connected to a vacuum circuit having a mechanical fore pump and a diffusion pump with control instrumentation.
All thermocouples current-and tension wires are connected to the control panels, passing through the mounting plate by means of a special vacuum tight plug.

Thermal insulators can be placed around the sample and probes.
Other apparatus technology can be found in references [6 and 7].

3.2. Experimental Procedure

The determination of thermal conductivity requires the assembly of standard probes and sample and the adjustment of the heater to give satisfactory heat flow and temperature distribution.

Once the sample is clamped between the copper probes, care being taken to assure good contacts, and the thermocouples positionned; the electrical current is adjusted for heating up the hot source.

When steady state conditions are attained in a sufficient vacuum, the emf. of all thermocouples are determined by a portable precision potentiometer.

The heat flow, passing the two probes, is calculated and compared with the heat dissipated in the electrical resistance.

In order to calculate the coefficient of thermal conductivity of the sample, we consider the mean value of the two calculated probe heat flows as the heat flow passing through the graphite sample applying equation (1).

4. Application to Nuclear graphites

Thermal conductivity measurements have been done on several pile graphite types using specimens of different shapes. See fig.3.

4.1. Graphite types

A. A French nuclear graphite. One bitumen impregnation. Bulk density 1,68 g/cc.

B. Nuclear graphite of German fabricate. Bulk density 1,67 g/cc.

C. British pile graphite A. PGA : Bulk density 1,69 g/cc.

D. British pile graphite - isotropic type. Bulk density 1,77 g/cc.

4.2. Results on virgin graphite.

A series of specimens taken in parallel and perpendicular direction has been measured, giving the following mean values of $\bar{K}_0$

Graphite	$\bar{K}_0$ // (.)	$\bar{K}_0 \perp$ (.)
A	0,408	0,368
B	0,437	0,372
C	0,478	0,257
D	0,336	0,324

(.) cal. cm^{-1}. $°C^{-1}$. s^{-1}.

Interesting studies on thermal conductivity of virgin and irradiated pile graphite have been done by CEA and others. See references [6, 8, 9, 10] .

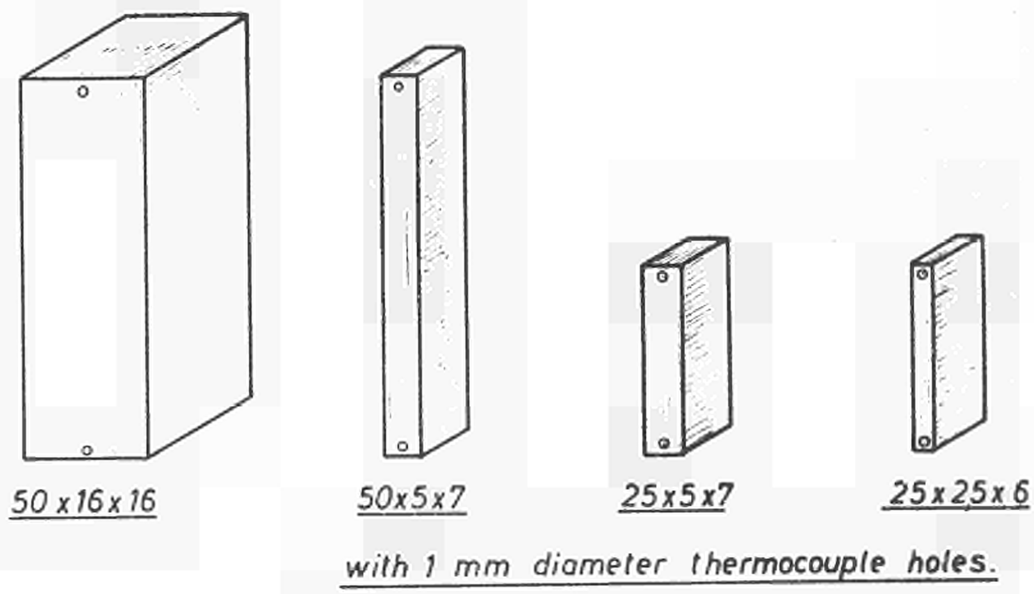

Fig. 3. GRAPHITE SPECIMENS.

4.3. Results on irradiated specimens

Some results on graphite specimens, which have been irradiated in a CO_2 atmosphere are given here.

graphite type	irradiation conditions flux th.	<temperature> °C	oxidation < % >	$\overline{K}_{i//}$ (.)	$\overline{K}_0 / \overline{K}_{i//}$
A	$\sim 1,12 \times 10^{20}$ n/cm^2	309	1,75	0,119	3,4
B		307	1,52	0,097	4,5
C		319	1,58	0,115	4,2
D		249	0,20	0,099	3,4

(.) cal. cm^{-1}. $°C^{-1}$. s^{-1}.

5. Appendix - Experimental Errors.

In general, the greatest difficulty in these comparative methods is obtaining heat flow which coincides exactly with that assumed in deriving the mathematical equations (3) and following. This heat flow through the copper probes and graphite sample must be maintained constant and equal for satisfactory results.

The design of the present apparatus is such that experimental errors have been eliminated as far as possible. Improvements consist mainly in specimens sizes, thermocouples location and guarding the heat flow path.

Errors occur mainly by a. lateral heat flow
and b. interfacial ressistance between probes and sample.

a. Lateral heat flow results in an unequal heat flow in the copper probes and graphite specimen i.e. some of the heat entering the heated probe is lost for the sample and following cooled probe. A reduction of this error will be the employment of larger samples, suitable insulation and guarding methods. Increasing the size of the probes and sample gives a more suitable heat distribution, makes assembly more precise, gives a better temperature uniformity and decreases also the effect of small errors in measurements of sample and probes dimensions.

b. Interfacial resistance cause a nonuniformity of the temperature distribution, this complicates eventually the guarding. If thermocouples are placed only in the probes, the temperature drop in the graphite sample must be determined by extrapolation. Interfacial resistance will be unknown and unmeasured and is included with the resistance of the sample giving erroneous results. An improvement has been the position of the thermocouples in the standard probes and graphite sample itself eliminating any extrapolation. Mechanical pressure ensuring good contact is applied through the clamping device and helps to decrease the influence of interfacial resistance.

Heatlosses by conduction in air are rendered negligible by operating in a vacuum of about 10^{-5} Torr.

Losses by radiation are calculated by eq. (2)

$Q = C\,(T_1^{\,4} - T_2^{\,4})$ or

$Q = 0{,}24 . \alpha . \sigma . \; (T_1^{\,4} - T_2^{\,4})$ calories

with α = emissivity factor

σ = radiation coefficient

T_1 = absolute temperature of the specimen

T_2 = absolute temperature of the apparatus chamber

For "black bodies" $\alpha = 1$ and $\sigma \sim 5{,}7 \times 10^{-12}$ Watts.cm^2 [6]

Calculations in our experiments show that Q_{rad} is only a very small part of the heat flux, and thus negligible.

Other factors affecting the experimental errors are :

- accurate positioning of the thermocouples in the probes and sample ;
- thermocouple accuracy
  - temperature measurements are made with chromel-alumel TC (sample) and iron-constantan TC (probes) in connection with a portable precision emf.bridge.
  - in normal conditions $\Delta\theta$ sample was about 2° to 5°C.
- dimensional tolerances of sample and probes ;
- fluctuations in heater block and water flow.

Considering all these points we believe to have thermal conductivity coefficient determinations within an accuracy of ± 6%. This precision has been verificated by calculations from standard samples like electrolytically pure copper samples having a thermal conductivity coefficient of 3,81 W.cm^{-1}. °C^{-1} (0,91 cal.cm^{-1}. s^{-1} °C^{-1}) and with zircaloy-2 specimens having a coefficient of 0,14 W. cm^{-1}.°C^{-1} (0,0334 cal.cm^{-1}. s^{-1} °C^{-1}). In both cases the precision and reproducibility was within the limits mentioned. All graphite types examinated show a coefficient lying between the two values.

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M. MASON and KRIBBS

To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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