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An Improved Comparative Thermal Conductivity Apparatus for Measurements at High Temperatures

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An apparatus developed for the measurement of thermal conductivity of solids at temperatures from 350 to 1250 K in air, vacuum, or any other controlled atmosphere is described. It is based on the steady-state axial heat flow comparative method and can be used for measurements of conductivities in the range 1 to 100 $W \cdot m^{-1} \cdot K^{-1}$. New heat source layout gives uniform heat flux across the specimen column, improving the accuracy of the measurements. The specimen stack is fixed in a rigid frame. It incorporates convection current breakers, eliminating thermal insulation of the stack and thereby considerably increasing the ease of specimen mounting. The accuracy of measurements was assessed by measuring the thermal conductivity of approved reference materials and is found to be within $\pm 3\%$. The results of measurements on nickel of known purity are also presented. Error analysis of the system shows that the determinate error leaving the uncertainty in the thermal conductivity of the reference materials, is less than $\pm 2\%$.

KEY WORDS: comparative method; inconel; nickel; pyrex; pyroceram; thermal conductivity.

1. INTRODUCTION

Different types of apparatus based on the comparative method for the measurement of thermal conductivity of solids have been reported in the literature. In this method, the specimen to be measured, shaped into the form of a cylinder or a squre plate with smooth top and bottom surfaces, is sandwiched between two similar-shaped reference materials of known thermal conductivity. An axial heat flow is established through this three-element stack by sandwiching them between a heat source and a sink. The reference materials serve as gauges to monitor the heat flux along the stack,

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and the conductivity of the specimen is determind from the heat flux and the thermal gradient across the specimen.

The development of the method is described by Francl and Kingery [1], Morris and Hust [2], and Mirkovich [3]. Though the accuracy reported varied from 3 to 7%, Flynn [4] and Laubits [5, 6] have questioned the claim of this level of accuracy by the comparative method.

Recently, Moss et al. [7] and Sweet et al. [8] at the Sandia National Laboratories (USA) conducted a systematic study to ascertain the accuracy limit of the measurements by the comparative method and confirmed that accuracy of $\pm 5\%$ can be achieved by this method. In the present paper, an equipment developed in our laboratory which provides a measurement accuracy of $\pm 3\%$ is reported.

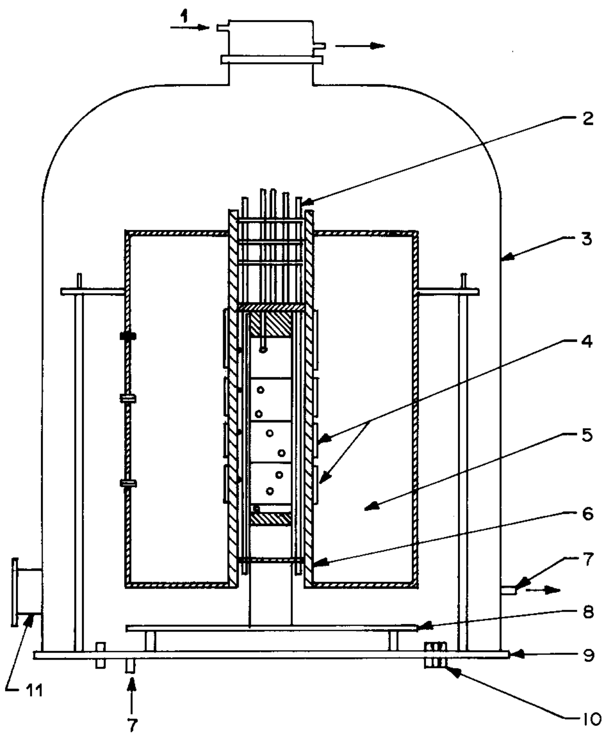

Fig. 1. Schematic diagram of the thermal conductivity apparatus. (1) Inlet and outlet for cooling water, (2) thermocouples, (3) stainless-steel vacuum chamber, (4) guard heaters, (5) thermal insulation, (6) alumina furnace tube, (7) inlet and outlet for cooling water, (8) supporting stand, (9) stainless-steel base plate, (10) feed-throughs, and (11) viewing port.

2. DETAILS OF APPARATUS

The apparatus is shown schematically in Fig. 1. A cylindrical specimen, 2.5 cm in diameter and 2 to 3 cm in height, is inserted between two identical reference materials having the same diameter as the specimen. To one end of this, a heat source is coupled through a thermal stabilizer and to the other end a heat sink with an auxiliary heater is provided. This entire stack is clamped in a rigid frame (Fig. 2) with spring loading (with

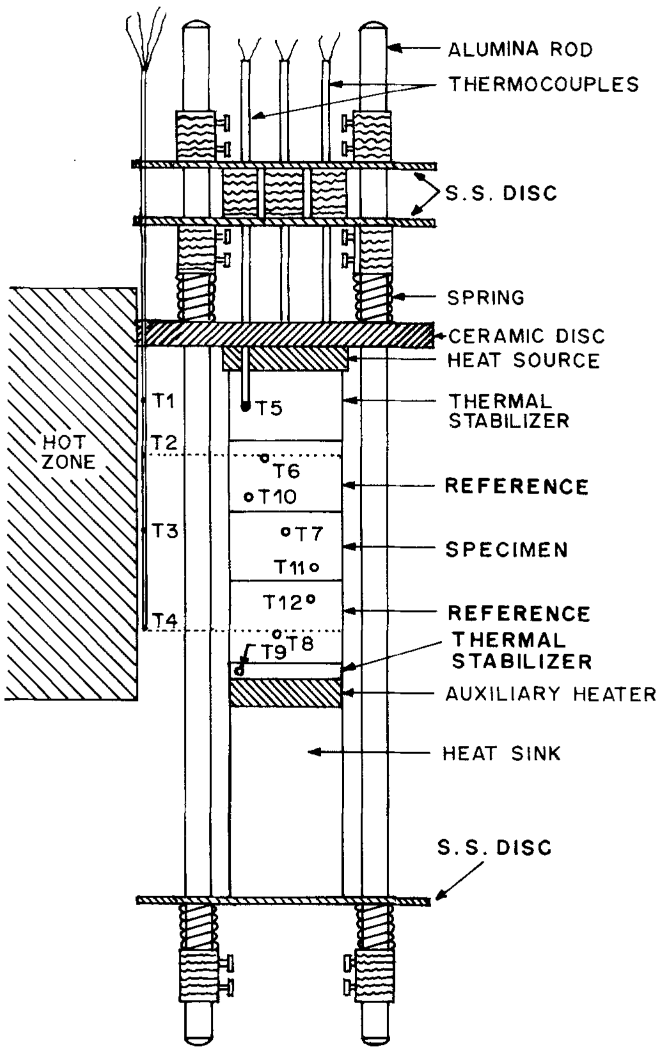

Fig. 2. Specimen stack (not to scale). T_1, T_2, etc., are thermocouple positions.

loads of 500 to 5000 N). In order to achieve the desired temperature profile without lateral heat loss, the stack is placed axially within a cylindrical guard furnace.

Both the source and the auxiliary heaters are flat, having the same diameter as the specimen and reference. They were fabricated by embedding coiled kanthal-resistive heating wire in alumina cement. Their configuration helped to achieve a radially uniform heat input to the stack, unlike in the earlier designs. The diameters of the specimen column and the heaters being the same, guarding of the stack temperature is easily achieved.

Stainless-steel cylinders, 2.5 cm in diameter and 2 cm in thickness, placed between the heaters and the top and bottom references serve as thermal stabilizers. These stabilizers help to prevent sharp temperature drops between the heaters and the references and also avoid heat channeling through the references.

The heat sink removes heat from the stack and the auxiliary heater regulates the flow of heat to the sink without distorting the temperature gradient. The sink is also a stainless-steel cylinder, 2.5 cm in diameter, and is coupled to the water-cooled stainless-steel base plate of the vacuum system.

Calibrated thermocouples, used for temperature measurements and monitoring, were located on the specimen and the references as shown in Fig. 3. The thermocouples were insulated using twinbore recrystallized alumina sleeves. Six calibrated 28-SWG chromel–alumel thermocouples (T_6 to T_8 and T_{10} to T_{12} in Fig. 2) were used for the measurement of temperature in the specimen and the references. Six 24-SWG platinum and platinum–13% rhodium thermocouples (T_1 to T_5 and T_9) were employed to control the temperatures of the guard furnace and auxiliary heater to establish the linear steady-state condition. The thermocouple leads, taken out from the guard furnace through alumina sleeves, were fixed rigidly to the specimen stack frame. Such an arrangement greatly helped in fixing the positions of the thermocouples precisely. When the measurements are performed on electrically conducting materials, a thin alumina cement coating is given to the thermocouple beads for electrical insulation.

The guard furnace consists of four separate heaters of 22-SWG kanthal wire wound on a 45-cm-long recrystallized alumina tube, 5 cm in inside diameter. The furnace is thermally insulated using zirconia felt. The position of the first heater is in level with the heat source and the thermal stabilizer. The other three heaters are in level with the specimen and the references, respectively. The guard furnace with the specimen stack is housed inside a hydraulically operated stainless-steel chamber (37.5 cm in diameter and 75 cm in height) of the vacuum unit.

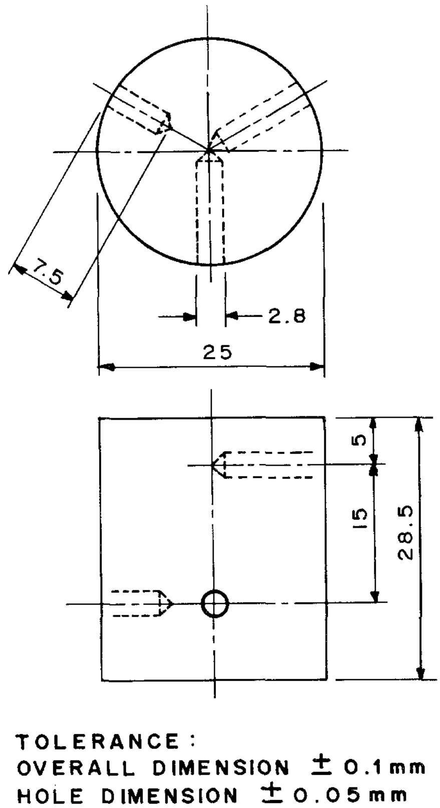

Fig. 3. Configuration and dimensions of specimen, references, and holes for fixing the thermocouples.

The thermal resistances at the interfaces of the elements in the stack are minimized by introducing thin platinum foil, 0.01 mm in thickness, between the finely polished interfaces of each cylinder.

The stainless-steel and alumina disks in the specimen stack (Fig. 2), besides holding the stack elements, also act as convection current brakers, minimizing air currents inside the furnace.

Power to the guard heaters and to the auxiliary heater is supplied through separate temperature controllers having a control accuracy of better than 1 K. A stabilized dc power supply having a stability of ± 1 mV is used for energizing the source heater. The continuous monitoring of the temperatures at different points is achieved by feeding the thermocouple output to a six-pen potentiometric recorder having a measurement accuracy of 5 μV.

The reference materials used for the measurements are pyrex-7740, pyroceram-9606, and inconel-718, obtained in the required configurations from Dynatech Corporation (USA). Their thermal conductivity versus temperature charts with the NBS references were supplied by the firm.

3. MEASUREMENT PROCEDURE

The specimen stack, with the specimen and thermocouples held rigidly in position, is inserted into the guard furnace and the electrical and thermocouple leads are connected to the respective feed-throughs fixed in the base plate. The power inputs to the source heater, the guard heater adjacent to it, and the auxiliary heater are regulated in such a manner as to stabilize the stack at or near the desired temperature level with a temperature gradient of approximately 50 to 100 K over the column of specimen and references. Subsequently, the other guard heaters are energized separately to achieve the same temperature gradient in the guard furnace as that in the specimen column. The guard heaters are adjusted until the thermocouples in the guard furnace, T_1 to T_4, match the temperatures at corresponding points in the stack within 1 K. Generally 1 to 4 h is required to reach the steady-state condition. Once the desired temperature gradient in the steady-state condition is obtained as mentioned, the isothermal profile condition along the radial direction of the specimen stack is ensured by noting the temperatures of two radially displaced thermocouples fixed at the same horizontal plane on one of the references (Fig. 3). The uniformity of the heat flux through the stack was ensured by computing the fluxes of the top and bottom references. Though ideally the two fluxes are supposed to become equal, under actual experimental conditions it is rarely achieved. When the two fluxes are equal to within $\pm 4\%$ of the average, it is considered that the system is ready for the measurement.

The annular space between the specimen stack and the guard furnace was thermally insulated by filling it with bubble alumina ($\lambda \sim 0.01 \ W \cdot m^{-1} \cdot K^{-1}$) and its effect on the accuracy of the measurements was evaluated. No noticeable improvent was observed by the insulation and hence in subsequent measurements no thermal insulation was employed.

When the measurements are carried out in vacuum or inert gases, the required atmosphere is created after mounting the stack in the furnace and before energizing the heaters.

4. ERROR ANALYSIS

The errors associated with thermal conductivity measurement methods have been discussed by several authors [9] and those for comparative method have been analyzed particularly by Sweet et al. [8]. Proceeding in a way similar to that of Sweet et al., the determinate errors of the apparatus were evaluated and are given below.

In the comparative method the thermal conductivity λ_s of the specimen is determined by equating the average heat flux of the references to the specimen flux as

$$\lambda_s(\bar{T}_2) = \phi/(\Delta T/\Delta L)_s \tag{1}$$

where the flux ϕ is given by the relation

$$\phi = 1/2[\lambda_r(\bar{T}_1)(\Delta T/\Delta L)_{tr} + \lambda_r(\bar{T}_3)(\Delta T/\Delta L)_{br}] \tag{2}$$

where $\lambda_r(\bar{T})$ is the thermal conductivity of the reference; at temperature $\bar{T}$, the subscripts s, tr, and br denote, respectively, the specimen and the top and bottom references, and $(\Delta T/\Delta L)$ is the temperature gradient. $\bar{T}_2$, $\bar{T}_1$, and $\bar{T}_3$ are the average temperatures of the specimen and the top and bottom references, respectively.

Considering the measurement errors to be random, the most probable value of the error E is [10]

$$E = \left(\sum_i \varepsilon_i^2\right)^{1/2} \tag{3}$$

where ε_i is the fractional uncertainty for each term obtained by differentiation of Eq. (1). If the heat flux in either reference denoted by $\phi_r = \lambda_r\, \Delta T_r/\Delta L_r$ and assuming that there is no correlation between ε_i and ε_j, then

$$(\delta\lambda_s/\lambda_s)^2 = (\delta\lambda_r/\lambda_r)^2 + 1/2 \cdot \{\delta(\Delta T_r)/\Delta T_r\}^2 + 1/2 \cdot \{\delta(\Delta L_r)/\Delta L_r\}^2 + \{\delta(\Delta T_s)/\Delta T_s\}^2 + \{\delta(\Delta L_s)/\Delta L_s\}^2 \tag{4}$$

The factor 1/2 in the second and third terms of RHS of Eq. (4) is due to the presence of two references in the measurements.

Generally, a temperature gradient of 15 K across each element in the stack was maintained. Since the potentiometer used has an accuracy of 5 μV, corresponding to 0.12 K for the chromel–alumel thermocouples, and since the temperature gradients are measured by differential thermocouples and the respective thermocouple wires were taken from the same spools, the above value can be taken to be the error limit for the temperature

Table I. Determinate Error from Various Parameters Other than That of the Reference Thermal Conductivity Term[a]

Parameter	$10^{+2}\varepsilon_i$	$10^{+4}A\varepsilon_i^2$
ΔT	0.80	0.64
ΔT	0.80	0.32
ΔL	0.67	0.45
ΔL	0.67	0.22
Nonuniform heat flux	0.50	0.25
Sum		1.88

[a] A = 1 or 1/2 corresponding to the terms of Eq. (4). Percentage error $(E \times 100) = \pm 1.37$.

measurements. The fractional uncertainty involved in $\delta(\Delta T)/\Delta T$ terms of Eq. (4) was accordingly calculated and is given in Table I.

The $\delta(\Delta L)/\Delta L$ terms were calculated employing the dimensional tolerances shown in Fig. 3 and are also given in Table I.

It is also important to evaluate the effect of a mismatch in the temperature gradients between the guard furnace and the specimen stack.

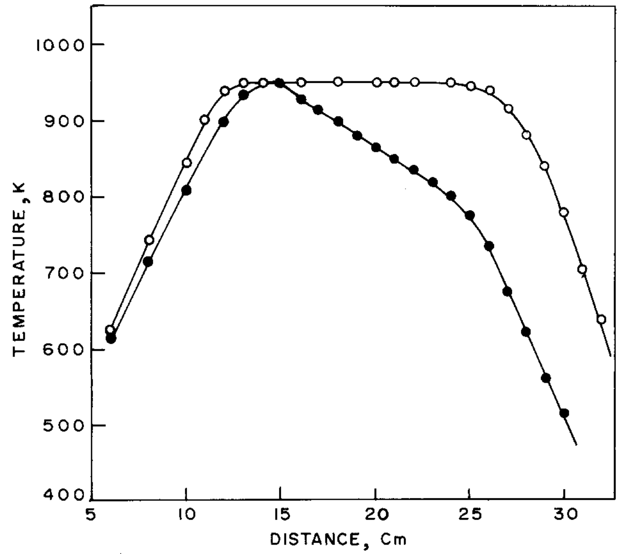

Fig. 4. Typical axial temperature profile when the guard heaters are energized simultaneously; (○) all the heaters are adjusted to the same temperature level and (●) different temperature levels.

The axial temperature profiles of the guard furnace, when guard heaters are energized for two different slopes, are shown in Fig. 4. Any temperature profile of varying slope is easily obtained by proper adjustments of the power inputs to the guard heaters. This type of a guard profile demonstrates that a nearly ideal temperature gradient match can be obtained. And it is observed experimentally that even for a specimen-to-reference thermal conductivity ratio of ~5, the deviation introduced in the results due to the mismatch is insignificant [11]. In the light of these observations, it was assumed that contribution from factors such as uncertainty due to displacement of guard to specimen, that due to mismatch of thermal conductivities, etc., proposed by Laubitz [5] is not substantial and, as noted by Sweet et al. [8], is only around ±0.5%, which is also included as the nonuniform heat flux in Table I. The total determinate error exclusive of the reference conductivity term $\delta\lambda_r/\lambda_r$

Table II. Determination of Experimental Error Using Pyrex-7740 as Specimen and Pyroceram-9606 as References; Measurements Carried Out in Air[a]

Avg. temp. (K) of specimen & references			Temp. gradient in specimen & references			$\lambda_{meas.}$	$\lambda_{recom.}$	Deviation
$\bar{T}_1$	$\bar{T}_2$	$\bar{T}_3$	ΔT_1	ΔT_2	ΔT_3	$(W \cdot m^{-1} \cdot K^{-1})$	$(W \cdot m^{-1} \cdot K^{-1})$	(%)
424	386	358	8.0	23.6	7.4	1.20	1.22	−1.64
449	418	391	7.6	20.9	6.9	1.25	1.26	−0.79
469	431	405	8.4	22.7	7.8	1.27	1.28	−0.78
523	485	453	10.5	25.7	9.7	1.36	1.34	+1.49
572	533	500	12.1	27.8	11.3	1.42	1.40	+1.43
603	565	530	11.5	25.0	10.6	1.48	1.44	+2.78
652	605	573	12.3	25.7	11.5	1.52	1.50	+1.33
691	645	613	12.5	24.5	11.6	1.59	1.55	+2.58
733	696	663	13.1	24.5	12.4	1.66	1.64	+1.22
781	738	707	12.8	21.4	11.5	1.73	1.73	0.00
403	365	335	8.3	25.4	7.5	1.16	1.19	−2.52
433	394	365	9.2	26.5	8.4	1.22	1.23	−0.81
493	453	420	10.7	28.0	10.0	1.30	1.31	−0.38
547	504	473	11.2	27.0	10.5	1.38	1.36	+1.47
593	544	513	11.8	26.8	11.2	1.44	1.41	+2.13
632	584	558	12.4	26.8	11.9	1.50	1.47	+2.04
681	635	613	12.8	25.9	12.1	1.56	1.54	+1.30
722	681	652	11.9	22.8	11.4	1.64	1.62	+1.23
767	726	697	12.7	22.9	12.0	1.70	1.70	0.00

[a] RMS deviation, ±0.016.

estimated according to Eq. (4) is thus only $\pm 1.37\%$. This value is found to be very much comparable with the value obtained for the radial heat flow apparatus reported by Godfrey et al. [10].

5. MEASUREMENT ACCURACY

Since indeterminate errors cannot be quantified, the practical approach to assess the accuracy and reproducibility of any measuring equipment is to carry out measurements on standard materials recommended for the particular property and to identify the deviations from the specified values. Hence, evaluation of the accuracy and reproducibility of the apparatus was carried out in air and in vacuum ($>10^{-5}$ Torr) in the temperature range 350 to 1250 K by measuring the thermal conductivities of pyrex-7740, inconel-718, and pyroceram-9606 as specimen and references. The results of the measurements with pyroceram as references are tabulated in Tables II and III and are shown graphically in Figs. 5 and 6.

In Tables II and III, $\bar{T}_2$, $\bar{T}_1$, and $\bar{T}_3$ are the arithmetical averages of

Table III. Determination of Experimental Error Using Inconel-718 as Specimen and Pyroceram-9606 as Reference Materials; Measurements Carried Out in Vacuum[a]

Avg. temp. (K) of specimen & references			Temp. gradient in specimen & references					
$\bar{T}_1$	$\bar{T}_2$	$\bar{T}_3$	ΔT_1	ΔT_2	ΔT_3	$\lambda_{meas.}$ ($W \cdot m^{-1} \cdot K^{-1}$)	$\lambda_{recom.}$ ($W \cdot m^{-1} \cdot K^{-1}$)	Deviation (%)
368	340	315	20.1	6.4	18.5	11.60	11.90	−2.52
415	387	363	22.2	6.2	21.3	13.07	12.60	+3.73
461	430	405	23.8	6.2	21.9	13.14	13.40	−1.94
548	515	488	24.2	5.6	22.8	14.40	14.80	−2.70
583	550	523	24.2	5.3	23.0	15.03	15.40	−2.40
675	641	613	26.3	5.0	24.9	16.66	17.00	−2.12
734	698	675	28.2	5.1	26.7	17.58	18.00	−2.33
767	731	704	28.3	4.8	26.8	18.02	18.50	−2.59
811	773	745	30.0	4.5	28.6	20.09	19.30	+4.09
820	823	798	31.0	4.6	29.3	20.12	20.20	−0.40
943	900	869	34.5	4.8	32.8	21.20	21.40	−0.90
964	923	888	35.0	4.7	32.9	21.85	21.80	+0.23
1045	999	966	37.5	4.9	36.6	22.42	23.00	−2.52
1093	1048	1013	38.0	4.6	36.1	23.53	23.80	−1.13
1124	1074	1038	40.7	4.8	39.1	24.26	24.10	+0.66
1196	1141	1102	45.6	5.0	43.0	25.66	25.00	+2.23
1248	1192	1148	47.8	5.1	46.7	26.70	26.00	+2.69

[a] RMS deviation, ± 0.023.

measured temperatures in the specimen and the top and bottom references, respectively, and ΔT_2, ΔT_1, and ΔT_3 are the temperature gradients. Thermal conductivity of the specimen, $\lambda_{meas.}$, represents the average value of the conductivities obtained from each references. The percentage and RMS deviations of the measured values from those of Dynatech Corporation, $\lambda_{recom.}$, are also included in Tables II and III. It is found that the overall deviations of the measured values from those recommended are within $\pm 3\%$.

In order to observe the performance of the setup with materials having a high thermal conductivity, measurements were performed on nickel (Ni—99.4%; Co—0.43%; Mn—0.13%; Fe—0.013%; Zn, Ti, Pb, Mg, etc.—0.015%) in air and vacuum using inconel-718 as reference. For these measurements fresh specimens were fabricated from the same nickel rod. The results are shown in Fig. 7. The plot clearly highlights the accuracy and reliability of the data vis-à-vis those reported in the literature [12, 13].

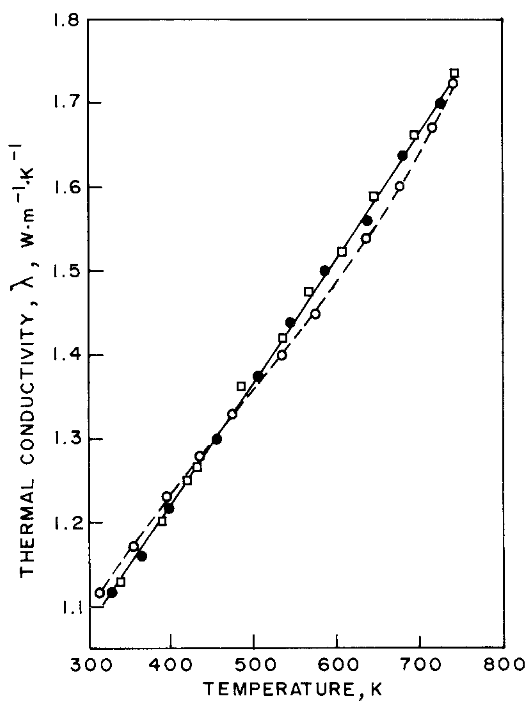

Fig. 5. Thermal conductivity of pyrex-7740 measured in air, using pyroceram-9606 as reference. (○) Recommended values; (●, □) measured values.

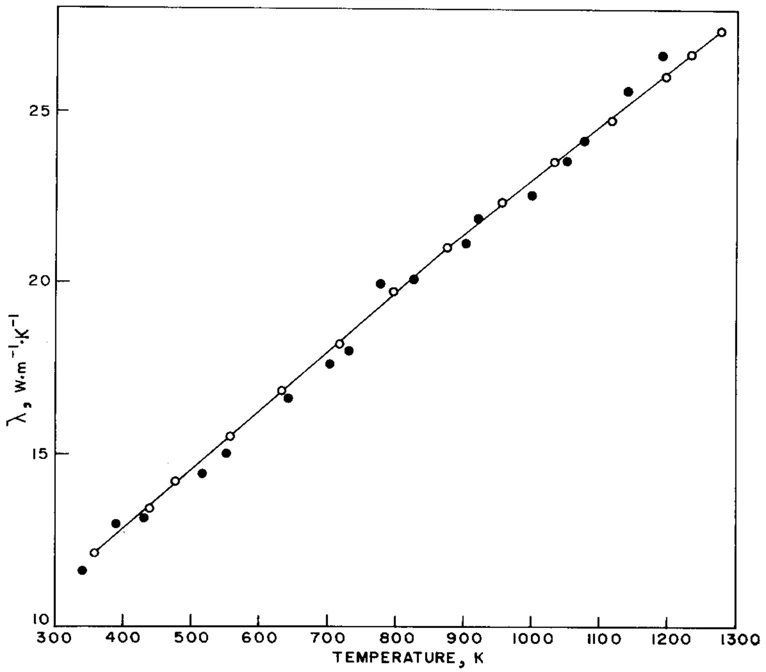

Fig. 6. Thermal conductivity of inconel-718 measured in vacuum ($>10^{-5}$ Torr), using pyroceram-9606 as reference. (●) Measured value; (○) recommended values.

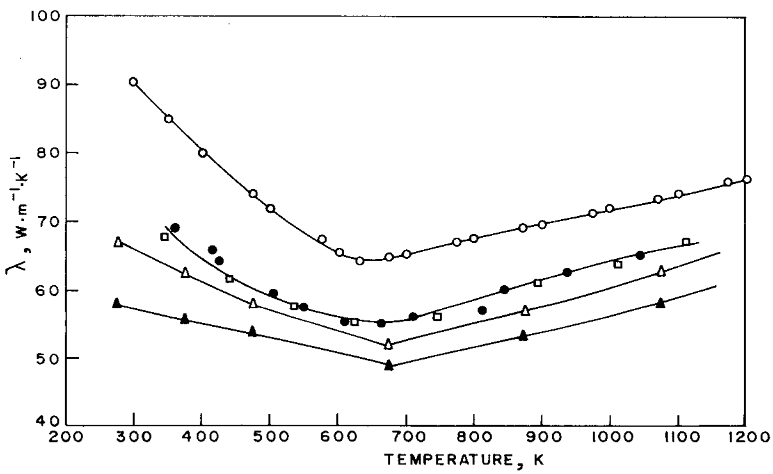

Fig. 7. Thermal conductivity of nickel of purity 99.4% measured in air (●) and vacuum (□), using inconel-718 as reference with literature data: (○) very high purity [12]; (△) 99.2% purity; (▲) ~97% purity [13].

The effect of purity and the well-known magnetic transition on thermal conductivity is clearly observed.

The reproducibility of the data was verified with repeated measurements on the same specimen by successive measurements using different references. The spread in data during such measurements was found to be well within the accuracy limit. It was also observed experimentally, as mentioned earlier, that for a specimen-to-reference thermal conductivity ratio of ~ 5 ($\lambda_{\text{inconel}}/\lambda_{\text{pyroceram}}$), the error introduced in the results due to the mismatch of the conductivities between specimen and reference was insignificant.

6. CONCLUSION

Evaluation of the thermal conductivity data obtained shows that the apparatus can yield measurements with an accuracy of $\pm 3\%$. This accuracy has been achieved primarily by establishing a uniform linear axial temperature gradient in the measuring stack through improved heat source layout and precise heat guarding of the stack. It is found that determinate error, leaving the uncertainty in the thermal conductivity of reference material, is $< \pm 2\%$. Hence the accuracy of the comparative thermal conductivity method can be considerably improved, comparable to that of the absolute methods, if reference materials with less uncertainty in their thermal conductivity can be identified.

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