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# A Transient Hot-Wire Instrument for the Measurement of the Thermal Conductivity of Solids up to 590 K[1]

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A novel application of the transient hot-wire technique for measurements of the thermal conductivity of solids up to 590 K is described. The method makes use of a soft silicone material between the hot wires and the solid of interest. Measurement of the transient temperature rise of the wires in response to electrical step heating over a period of 20 μs to 20 s allows an absolute determination of the thermal conductivity of the solid, as well as of the silicone paste. The method is based on a full theoretical model with equations solved by a finite-element method applied to the exact geometry. Two sets of thermal-conductivity measurements up to 590 K, employing different silicone pastes and samples of Pyroceram 9606, are reported. At the 95% confidence level, the standard deviation of the thermal conductivity measurements is less than 1.5%.

**KEY WORDS:** ceramic; pyroceram 9606; solid; thermal conductivity; transient hot-wire.

## 1. INTRODUCTION

In a previous paper [1], a novel application of the transient hot-wire technique for the measurement of the thermal conductivity of solids was described. The advantages of the proposed application are:

(1) Contact resistance between wire sensor and solid was minimized, as the wire sensor was placed in a soft silicone paste between two

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blocks of the solid. At very short times (20 μs to 0.2 s) the generated heat wave is confined to the paste, hence, the properties of the paste can be obtained. At larger times the properties of the solid can be obtained.

(2) The method is based on a full theoretical model with equations describing the heat generated in the wire, and the heat transferred in the intermediate layer and the solid. The equations are solved by a finite-element method applied to the exact geometry.

(3) The technique can be validated by employing the sensor for measurements of the thermal conductivity of liquids.

(4) Very low temperature rises are necessary, typically 3 to 4 K over a period of 20 s.

The measurements were shown to be absolute and characterized by a low uncertainty.

In this paper, the technique will be applied to the measurement of the thermal conductivity of Pyroceram 9606, a proposed thermal conductivity reference material, up to 590 K.

## 2. EXPERIMENTAL

The description of the technique, as employed for the measurement of the thermal conductivity of solids at 298.15 K is described elsewhere [1] and will only be briefly presented here. The instrument employs two tantalum wires as hot wires, differing only in length, placed in a silicone layer between two blocks of the solid of interest. A Wheatstone-type electronic bridge is employed to measure the evolution of the resistance change of a finite segment of infinite wire (axial heat conduction from the wire ends is also automatically compensated), and to ensure that a known constant heat flux is generated in the hot wires.

The instrument employed here for the measurements up to 590 K is shown in Fig. 1. The two wires, made out of 25-μm-diameter tantalum wire of 2.1 and 5.1 cm lengths, are placed one after the other, and spot-welded to flattened 0.5-mm-diameter tantalum wires. These in turn are spot-welded to 1-mm-diameter Chromel metal-sheathed wires, which act both as electrical contacts and supports. This support with the two wires is also employed as is, for measurements of the thermal conductivity of liquids. For measurements of the thermal conductivity of solids, the wires' support is placed in a stainless steel (310S) half cylinder, which holds one of the two solid blocks. Each solid block has dimensions of $10 \times 5 \times 2$ cm$^3$. The other solid block is held in the other half cylinder (Fig. 1). As already

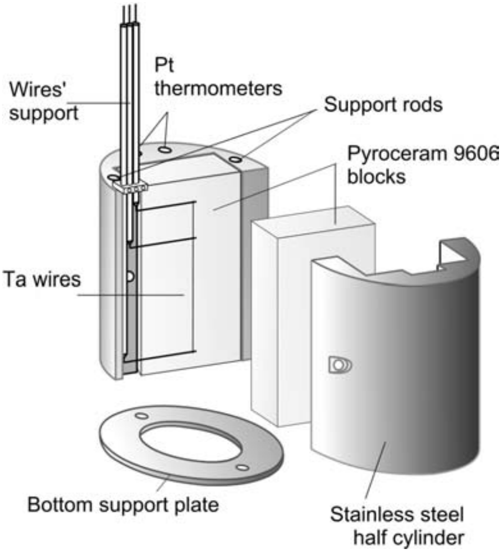

**Fig. 1.** Schematic diagram of the wires and the two Pyroceram 9606 blocks.

described [1], in order to reduce contact resistance between the hot wires and the solid, a soft silicone paste is used.

The whole arrangement is placed in the center of an accurate, vertical three-zone tubular furnace (Carbolite, Model TVS 12). The temperature was determined using two platinum-resistance thermometers embedded on the top and bottom of the half cylinder. The thermometers were calibrated versus a Class 1 NPL-certified, Tinsley platinum resistance thermometer, to a maximum uncertainty of less than ±60 mK. All temperatures refer to ITS-90. Preliminary measurements with platinum resistance thermometers placed in the position of the wires, showed a 0.7 K maximum difference between the top and the bottom of the half cylinder and a 0.2 K maximum difference radially. These differences have no significant effect in the final quoted uncertainty of the thermal conductivity measurement.

To heat the wires and measure their resistance at the same time, a computer-controlled automatic bridge was employed [1]. During one run a thousand measurements of the temperature rise are accumulated. Temperatures rises employed are between 3 to 4 K over a maximum period of 20 s, and the thermal conductivity, $\lambda$, and the product (density × specific heat), $(\rho C_p)$, of the solid and the intermediate layer, as well as the thickness of the intermediate layer are uniquely determined.

## 3. MEASUREMENTS

### 3.1. Validation of Technique

An advantage of the proposed configuration is that it can also be employed to measure the thermal conductivity of fluids. Liquid toluene has been proposed by the Subcommittee on Transport Properties of the International Union of Pure and Applied Chemistry as a standard with an uncertainty of 0.5% [2].

In this case, the wires with their support, were placed in toluene at 295.15 K, and the equations were solved with the intermediate and solid layers substituted by liquid toluene. The thermal conductivity and the product $(\rho C_{\mathrm{p}})$ measured were in excellent agreement (standard deviations of 0.5% and 1% respectively, at the 95% confidence level) with the above referenced literature values.

### 3.2. Pyroceram 9606

Following the toluene measurements, the wires were subsequently placed between two pieces of Pyroceram 9606 (Corning U.S.A.), as shown in Fig. 1. Pyroceram 9606 is a glassy ceramic, originally developed by NASA, and since it is particularly well defined and thermally stable, it has been used as a reference material for thermal conductivity industry-wide for over three decades, and is currently considered a candidate Reference Material by the National Physical Laboratory, United Kingdom. As mentioned earlier, to reduce contact resistance, the gap between the wire and the ceramic blocks was filled with silicone paste.

Two series of tests were carried out:

- Series 1 employed a high-temperature red silicone paste (BORO 650, VersaChem, U.S.A.), while
- Series 2 employed a white silicone paste (heat transfer compound, HTCO2S, Electrolube, U.K.).

The results for the thermal conductivity and the product $(\rho C_{\mathrm{p}})$ are shown in Table I. Figure 2 shows excellent agreement of the experimental data with values predicted by FEM curves, indicating accurate modeling of the surfaces involved and the absence of any other layer, such as an air gap. The results of Table I are plotted in Figs. 3 and 4. As expected, independent of the different properties of the two pastes, the thermal conductivity and the product $(\rho C_{\mathrm{p}})$ of the test solid, are in very good agreement compared to each other.

**Table I.** Measured Properties of Pyroceram 9606 and of the Two Silicone Pastes as a Function of Temperature

| $T$ (K) | $\lambda$ ($\mathrm{W \cdot m^{-1} \cdot K^{-1}}$) | $100(\lambda_{\mathrm{exp}} - \lambda_{\mathrm{fit}})/\lambda_{\mathrm{fit}}$ (%) | $(\rho C_{\mathrm{p}})$ ($\mathrm{kJ \cdot m^{-3} \cdot K^{-1}}$) | $\lambda^{a}$ ($\mathrm{W \cdot m^{-1} \cdot K^{-1}}$) | $(\rho C_{\mathrm{p}})^{a}$ ($\mathrm{kJ \cdot m^{-3} \cdot K^{-1}}$) |
|---|---|---|---|---|---|
| Series 1 | Pyroceram 9606 | | | Red Silicone Paste | |
| 298.652 | 3.88 | 0.56 | 1909 | 0.1742 | 7905 |
| 318.181 | 3.70 | −1.60 | 2028 | 0.1743 | 8568 |
| 351.926 | 3.63 | −0.27 | 2246 | 0.1767 | 9690 |
| 391.065 | 3.55 | 0.54 | 2395 | 0.1769 | 10098 |
| 439.397 | 3.44 | 0.15 | 2584 | 0.1771 | 11220 |
| 484.475 | 3.36 | −0.48 | 2621 | 0.1778 | 13311 |
| 524.350 | 3.29 | −0.80 | 2647 | 0.1791 | 14841 |
| 569.238 | 3.21 | −0.94 | 2674 | | |
| Series 2 | Pyroceram 9606 | | | White Silicone Paste | |
| 296.546 | 3.90 | 0.61 | 1827 | 0.4600 | 2701 |
| 322.930 | 3.71 | −1.08 | 2007 | 0.4475 | 3915 |
| 361.400 | 3.63 | 0.68 | 2240 | 0.4375 | 4725 |
| 405.263 | 3.55 | 1.15 | 2518 | 0.4270 | 5940 |
| 449.227 | 3.43 | 0.35 | 2604 | 0.4205 | 8370 |
| 484.063 | 3.33 | −1.19 | 2708 | | |
| 513.825 | 3.32 | −0.41 | 2781 | | |

[a] Values at the highest temperatures are not quoted due to the higher uncertainty associated with the solidification of the silicone pastes.

The thermal conductivity, $\lambda$ ($\mathrm{W \cdot m^{-1} \cdot K^{-1}}$), values shown in Table I, were fitted as a function of the absolute temperature $T$ (K) to an equation,

$$\lambda = 8.22384 - 2.7241 \times 10^{-2} T + 5.312 \times 10^{-5} T^{2} - 3.626 \times 10^{-8} T^{3}. \tag{1}$$

The maximum deviation of the experimental points presented in Table I, from the above equation is 1.6%, and the standard deviation at the 95% confidence level is 0.8%. Both sets, i.e., Series 1 and Series 2, as well as the above equation are shown in Fig. 3. At the 95% confidence level, the standard deviation of the thermal conductivity measurements is less than 1.5%, and of the product (density × heat capacity) less than 5%. The present values are slightly lower than our previously reported values [1] at 298.15 K. This is probably attributed to the lack of good temperature control unit for the previous set.

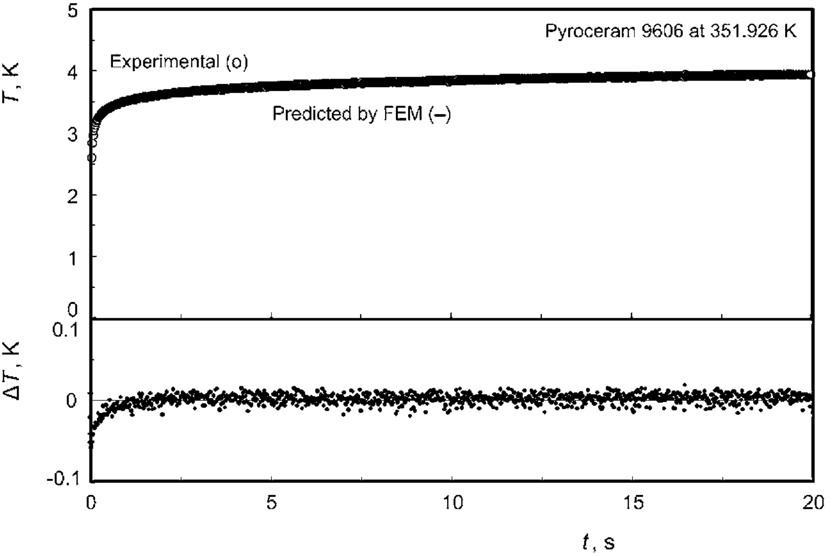

**Fig. 2.** Experimental and predicted temperature rise in Pyroceram 9606.

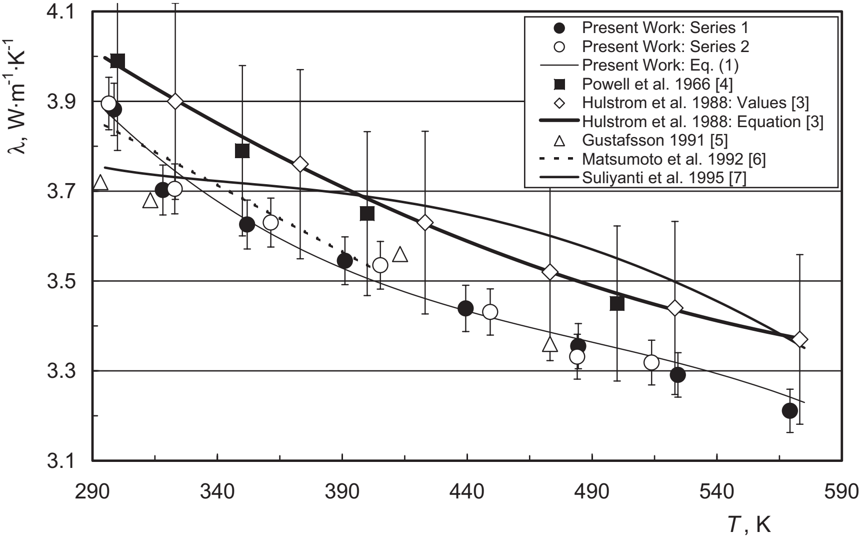

**Fig. 3.** Thermal conductivity of Pyroceram 9606 as a function of temperature.

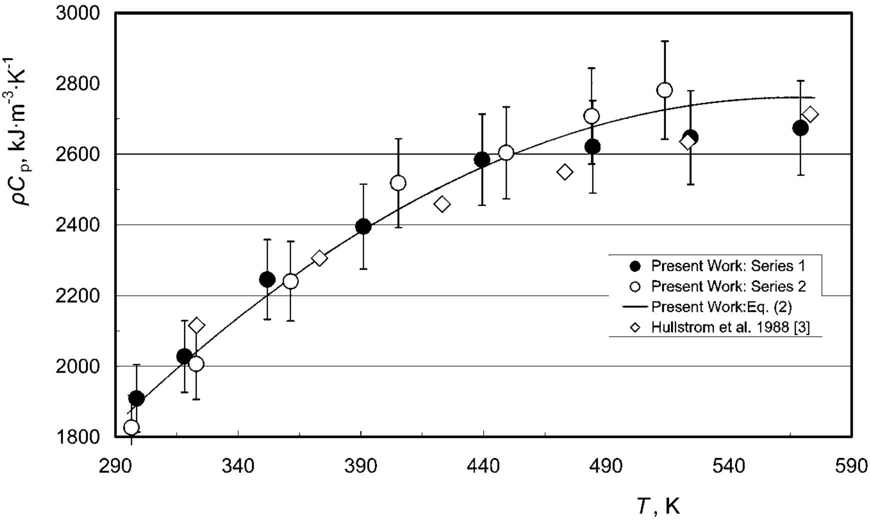

**Fig. 4.** Product ($\rho C_p$) of Pyroceram 9606 as a function of temperature.

## 4. DISCUSSION

The National Physical Laboratory, United Kingdom, is currently considering Pyroceram 9606 as a thermal conductivity reference material, although no recommendation has yet been published. In 1988, the results for round-robin tests for the same material were published by Hulstrom et al. [3]. Their recommended values and equation, characterized by a 5.7% standard deviation at the 95% confidence level, are shown in Fig. 3, together with the previously recommended values of Powell et al. [4], of 5% maximum uncertainty. The agreement with both these sets is excellent. In the same figure three other, more recent sets of measurements are also included:

- the thermal conductivity measurements of Gustafsson in 1991 [5], performed with a spiral wire in a hot disc arrangement, and a claimed uncertainty of 3% (no confidence level was specified),
- the measurements of Matsumoto and Ono in 1992 [6], performed in a radiative heat exchange instrument, with a claimed uncertainty of 2.5% (no confidence level was specified), and
- the derived values from thermal diffusivity measurements of Suliyanti et al. [7], performed with the laser flash method, with a claimed uncertainty of 3% (no confidence level was specified).

In all cases, the deviations are within the mutual uncertainties of the instruments.

In Fig. 4, the values obtained for the product $(\rho C_{\rm p})$ for Pyroceram 9606 are shown. These were fitted as a function of the absolute temperature $T$ (K),

$$(\rho C_{\rm p}) = -1109.9 + 13.63T - 0.012T^2. \tag{2}$$

The agreement of the two sets, as well as with those previously reported by Hullstrom et al., [1988] is very good.

## 5. CONCLUSIONS

A novel application of the transient hot-wire technique for measurements of the thermal conductivity of solids up to 590 K has been described. The wires were placed in a soft silicone paste between the ceramic material allowing the calculation of the properties of the soft paste and subsequently the calculation of the thermal conductivity, $\lambda$, and the product $(\rho C_{\rm p})$ of the solid material. The method is based on a full theoretical model with equations solved by finite elements for the exact geometry. At the 95% confidence level, the standard deviation of the thermal conductivity measurements is less than 1.5%, and of the product $(\rho C_{\rm p})$ is less than 5%.

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