

## **Thermal Conductivity by a Pulse-Heating Method: Theory and Experimental Apparatus<sup>1</sup>**

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A new dynamic technique for the measurement of thermal conductivity at high temperatures has been developed at the IMGC. The specimen is brought to high temperatures with a current pulse; during cooling the heat content is dissipated by radiation and by conduction. The differential equation describing this process contains terms related to the heat capacity, the hemispherical total emittance, and the thermal conductivity of the material. If the first two properties are determined using the same specimen during subsecond pulse heating experiments, thermal conductivity may be evaluated by accurate measurements of the round-shaped temperature profiles established on the specimen during cooling. High-speed scanning pyrometry makes possible accurate measurements of temperatures and of temperature derivatives (with respect to space and time), which enables the differential equation describing the power balance at each point of the specimen to be transformed into a linear equation of the unknown thermal conductivity. A large overdetermined system of linear equations is solved by least-squares techniques to obtain thermal conductivity as a function of temperature. The theory underlying the technique is outlined, the experimental apparatus is described, and details of the measurement technique are given.

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**KEY WORDS:** dynamic measurements; high temperature; scanning pyrometry; thermal conductivity.

### **1. INTRODUCTION**

In the last two decades, pulse techniques were proved to be valid experimental tools for the measurement of several thermophysical properties. The advantages of short exposures of the specimen to high

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temperatures (subsecond experiment) and the high precision and accuracy possible with modern electronic instrumentation were combined in experimental apparatus capable of simultaneous multiproperty determinations. The development of high-speed pyrometry with submillisecond time resolution was the key to the improved accuracy of the pulse method.

The first apparatus was developed at the National Bureau of Standards (NBS) in the United States [1]. In Europe, a similar system was set up at the Istituto di Metrologia "G. Colonnetti" (IMGC) in Italy [2, 3]. Both systems were initially aimed to the simultaneous measurement of several thermophysical properties (heat capacity, electrical resistivity, hemispherical total emittance, normal spectral emittance) in pulse experiments of subsecond duration. The basic idea of the pulse technique involves performing the experiment in a time scale such that the central portion of the specimen is self-heated to high temperatures with minimal loss of energy by thermal conduction toward the clamps; in this way a high-temperature specimen (central zone) is obtained without the need of cumbersome furnaces and auxiliary equipment. The thermal properties are obtained by simple power balance equations during heating and the initial part of cooling; experimental quantities (input power, temperature, heating and cooling rates) are measured with submillisecond resolution.

The pulse method was later extended in both laboratories to thermal expansion using different techniques: at the NBS a polarized beam interferometer was used to measure the expansion across the central region of the specimen [4]; at the IMGC the longitudinal expansion was measured interferometrically with a simultaneous measurement of the temperature profile with a high-speed scanning pyrometer [5].

It is clearly desirable to extend further the measurement capabilities of the apparatus developed for the pulse technique: measurements performed with the same equipment on the same specimens provide the added bonus of the possible determination of correlation properties and the minimization of certain measurement errors.

This paper describes the theory and the experimental apparatus developed at the IMGC to extend the measurements to thermal conductivity. A fundamental modification of the pulse technique is necessary: any experiment that attempts to measure thermal conductivity must take place in a time scale compatible with the thermal conduction process. This is possible either by slowing down the experiment during the heating phase (lasting 20–30 s) or by following the specimen during cooling. In either case round-shaped temperature profiles are established on the specimen (due to heat conduction from the central part of the specimen toward the clamps) and these evolve dynamically during the experiment: their accurate measurement by high-speed scanning pyrometry provides information on

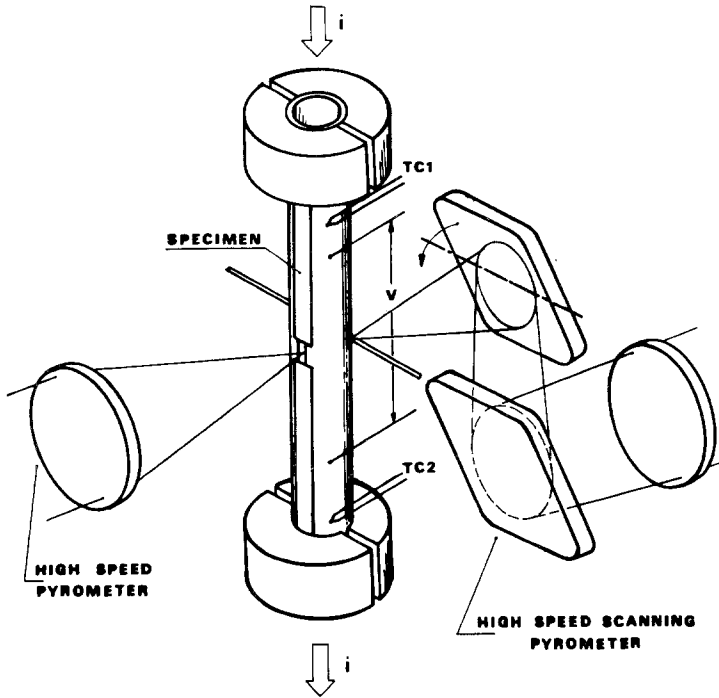


Fig. 1. Schematic representation of the new dynamic technique for the measurement of thermal conductivity. I, current passing through the specimen; V, voltage drop across the central part of the specimen; TC1 and TC2, Chromel-Alumel thermocouples.

the thermal conductivity of the material. Figure 1 presents a schematic diagram of the experimental apparatus developed at the IMGC for the measurement of thermal conductivity.

## 2. MEASUREMENT THEORY

Direct heating methods for the measurement of thermal conductivity using steady state techniques are well known. A comprehensive review of the work until 1968 is given by Flynn [6]; a later survey by Taylor [7] covers in detail direct heating methods at high temperatures (above 1500 K) and the development of a steady-state multiproperty apparatus. Following Ref. 7, the "long thin rod approximation" of the general equation describing energy transport in a directly heated homogeneous solid specimen under dynamic conditions is

$$\frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right) + \frac{\rho I^2}{S^2} - \frac{\varepsilon_{ht} \sigma p (T^4 - T_a^4)}{S} - \frac{\mu I \partial T}{S \partial x} = \delta C_p \frac{\partial T}{\partial t} \quad (1)$$

Equation (1) contains different thermal properties ( $\lambda$ , thermal conductivity;  $\rho$ , electrical resistivity;  $\epsilon_{ht}$ , hemispherical total emittance;  $\mu$ , Thomson coefficient;  $\delta$ , density;  $C_p$ , heat capacity), geometrical quantities ( $S$ , cross-sectional area;  $p$ , perimeter), temperature  $T$  with its space and time derivatives, current  $I$ , ambient temperature  $T_a$ , and the Stefan-Boltzmann constant  $\sigma$ . Equation (1) is valid for a long thin rod or tube with radial symmetry and bound by an electrical insulator. The steady state form of Eq. (1) (time derivative equals zero) is the basis of the stationary direct heating technique developed at the Thermophysical Property Research Laboratory (TPRL) of Purdue University (see Ref. 8 for a complete review). The experiment at the IMGC attempts to realize similar measurements under dynamic conditions, with the specimen being kept at high temperature for a few seconds only.

At the IMGC the various thermophysical properties necessary for Eq. (1) (heat capacity, electrical resistivity, hemispherical total emittance, normal spectral emittance) may be measured in a pulse experiment (subsecond duration) using the same experimental apparatus and the same specimens. This procedure ensures additional advantages regarding the minimization of measurement errors, as discussed in Ref. 7.

The subsecond pulse experiments for the measurement of heat capacity, electrical resistivity, and hemispherical total emittance may be considered a variant of Eq. (1) under special conditions. If one considers the flat central region of the specimen heated by a subsecond pulse (where  $\partial T/\partial x = 0$ ) and applies what remains of Eq. (1) to the region of uniform high temperature (between the voltage probes), the result is the power balance equation used for the pulse experiment.

The development at the IMGC of high-speed scanning pyrometry [9] provides the additional information necessary to work with Eq. (1). The accurate real-time measurement of temperature profiles gives the temperature values and the temperature vs time and space derivatives necessary to transform the differential equation into a linear equation of the unknown thermal properties.

The determination of thermal conductivity in these dynamic experiments is possible during both heating and cooling. The control of input power during heating makes possible many different experimental situations, but unfortunately to maintain adequate dynamic conditions the thermal conduction term at high temperatures is always the difference between two rather large terms (related to Joule heat and to the sum of absorbed and radiated heat). One has to work with the complete equation and needs also to determine (or use an estimate for) the small Thomson heat term.

Dynamic experiments for thermal conductivity performed during

cooling provide much simpler experimental conditions. The specimen may be brought to high temperatures with a current pulse lasting from less than 1 s to several seconds, according to the desired initial conditions. With no current flowing ( $I=0$ ) Eq. (1) becomes

$$\frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right) - \frac{\varepsilon_{\text{ht}} \sigma p (T^4 - T_a^4)}{S} = \delta C_p \frac{\partial T}{\partial t} \quad (2)$$

where each term is a power per volume. Equation (2) expresses the power balance of each small portion of the specimen, with the heat content being dissipated by radiation and by thermal conduction. The disadvantage of a cooling experiment is that cooling rates cannot be controlled and the dynamic evolution of the temperature profiles and their shape depend on the thermal properties of the material and on geometrical factors (length, cross-sectional area, radiating area) defined for each specimen at manufacturing time.

The determination of thermal conductivity over a wide temperature range from a single experiment is a potential advantage of the pulse technique. In principle, one may follow the power balance of a small part of the specimen from very high temperatures down to the lowest temperature measurable by pyrometry (with present experimental conditions from approximately 3000 K to 1000 K). Such an approach requires a careful choice of the reference frame in which properties are computed, because a pyrometer focused on one point in absolute space will actually "see" different parts of the specimen during a pulse experiment, on account of the specimen's thermal expansion. Additional problems arise with the method of measurement of properties needed for Eqs. (1) and (2) that may or may not be corrected for thermal expansion effects, as pointed out in Ref. 10. The problem was solved at the IMGCC by using Eq. (2) in an idealized reference frame where no expansion takes place. During computations all quantities are referred to an ideal tube-space (the one existing when the specimen was at room temperature); temperature profiles measured in real space are made to shrink to the shape they would have had if the specimen did not expand and space derivatives are calculated accordingly. Careful consideration is made of the way the other properties were determined and adequate thermal expansion corrections are applied to work in the ideal tube-space. Once this is done, the temperature vs time derivative is simply obtained by observing the temperature evolution of the same point in tube-space. The final thermal conductivity results need again to be partially corrected to take fully into account expansion effects.

Equation (2) can provide conductivity values at various temperatures,

but since the number of data soon become very large, an alternative computation is followed. Equation (2) is rewritten as

$$\lambda \frac{\partial^2 T}{\partial x^2} + \frac{d\lambda}{dT} \left( \frac{\partial T}{\partial x} \right)^2 = \delta C_p \frac{\partial T}{\partial t} + \frac{\varepsilon_{ht} \sigma p (T^4 - T_a^4)}{S} \quad (3)$$

Thermal conductivity over a wide temperature range is generally represented by a low-order polynomial in temperature:

$$\begin{aligned} \lambda(T) &= a_1 + a_2 T + \dots + a_n T^{n-1} \\ \frac{d\lambda}{dT} &= a_2 + \dots + (n-1) a_n T^{n-2} \end{aligned} \quad (4)$$

If the expressions in Eq. (4) are inserted in Eq. (3) and terms rearranged, one obtains

$$\begin{aligned} a_1 \frac{\partial^2 T}{\partial x^2} + a_2 \left[ T \frac{\partial^2 T}{\partial x^2} + \left( \frac{\partial T}{\partial x} \right)^2 \right] + \dots \\ + a_n \left[ T^{n-1} \frac{\partial^2 T}{\partial x^2} + (n-1) T^{n-2} \left( \frac{\partial T}{\partial x} \right)^2 \right] \\ = \delta C_p \frac{\partial T}{\partial t} + \frac{\varepsilon_{ht} \sigma p (T^4 - T_a^4)}{S} = N \end{aligned} \quad (5)$$

where all the quantities on the right-hand side are measured or are known. Equation (5) is a linear equation in the unknowns  $a_1, a_2, \dots, a_n$  and is applicable to any point of any profile. So using  $P$  different points (chosen arbitrarily in different profiles and/or in different experiments where conditions are favorable to thermal conductivity determinations), when  $P \gg n$ , one obtains an overdetermined system that may be solved by least-squares techniques. A set of values ( $a_1, a_2, \dots, a_n$ ) is thus obtained for the best thermal conductivity polynomial covering the entire temperature range.

The least-squares approach is advisable in these dynamic experiments because the number of data becomes immediately unmanageable as soon as a few experiments are performed. The raw numbers available are approximately 200 temperatures per profile and 80 profiles per experiment. Thermal conductivity computations are possible everywhere on the profiles, but for accurate measurements it is clearly advisable to use only those parts of the profiles where thermal conduction plays an important part. By selecting 10 points in tube-space, one experiment provides 800 equations; when data from three specimens (five experiments each) are available the system reaches 12000 equations. These are not difficult to solve, but they require a fair-sized computer to handle all the data.

### 3. EXPERIMENTAL APPARATUS

Measurements are performed with the apparatus developed for pulse experiments [2] with subsequent modifications for thermal expansion [5]. A block diagram of the thermal conductivity apparatus is shown in Fig. 2: only modifications to the earlier descriptions are presented. Heating pulses are provided by a set of series connected batteries: input power may be modified by manually changing the power supply output. The massive brass clamps developed for thermal expansion measurements were retained:

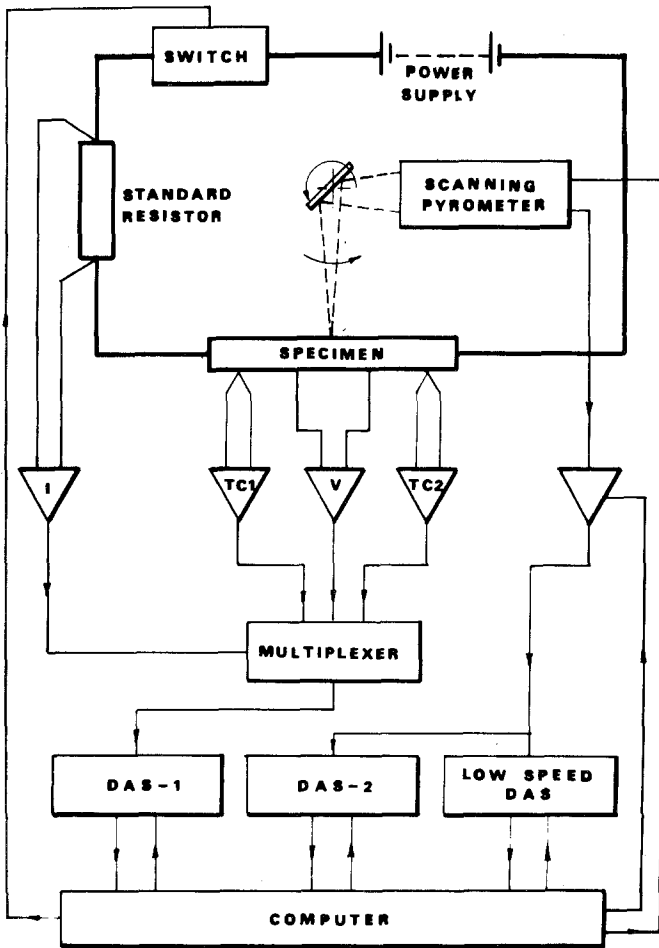


Fig. 2. Block diagram of the experimental apparatus. DAS, data acquisition system; rest of notation as in Fig. 1.

their large mass helps to maintain sharp temperature gradients by providing a sink for the heat conducted away from the center of the specimen. Measurements are possible both in vacuum or in an inert atmosphere: the use of a dynamic technique with the specimen at high temperatures only for 10–20 s makes convection losses either negligible or accountable with a small correction.

Several quantities are measured before and after the profile measurement with a multiplexed 14-bit data acquisition system (DAS-1 in Fig. 2; 20 kHz; range, 0–10 V; lsb, 0.61 mV). Current is measured by the voltage drop across the standard resistor (1 m $\Omega$ ); the partial voltage drop (central part) is defined by two spot-welded wire probes (0.05-mm diameter) of the same material being measured. Two Chromel–Alumel thermocouples spot welded at the ends of the specimen provide temperature values in regions where pyrometric measurements are not possible. For thermal conductivity measurements only TC1 is needed to reconstruct the temperature profile from the upper clamp (fixed in space) and to compute profiles in tube-space. Both thermocouples were retained as experiments were performed on the same specimens previously used for thermal expansion. Each data acquisition system channel and its associated differential amplifier were calibrated each measurement day with computer controlled procedures.

A scanning pyrometer with microsecond resolution was developed to measure the longitudinal temperature profiles of the specimen. The instrument was designed and built at the IMGC specifically for these experiments and only its main features are reported here: detailed technical descriptions and performance data may be found in an earlier publication [9]. The scanning pyrometer consists of (see Figs. 1 and 2)

- (a) a fast monochromatic pyrometer with microsecond resolution (wavelength, 900 nm; bandwidth, 82 nm);
- (b) an optical scanning system with related electronics for synchronization and triggering purposes; and
- (c) a fast data acquisition system (DAS-2; 250 kHz; 12 bits; range, 0–10 V; lsb, 2.5 mV).

Pyrometer autoranging during both heating and cooling is provided by a programmable differential amplifier inserted between the pyrometer and the DAS-2. Other technical features for the measurement of temperature profiles (zero position defined by a wire across the specimen, procedures for *in situ* measurement of spatial positions, etc...) are identical to those described for thermal expansion measurements [5]. No significant changes in the specimen temperature must take place during a profile measurement by the scanning pyrometer. Since the time spent by the rotating mirror in



front of the specimen is always less than 2 ms, the profile will change by less than 1 K during the active scan ( $500 \text{ K} \cdot \text{s}^{-1}$  cooling rate), and this may be corrected by assuming linear temperature changes during the profile measurement.

At present experiments are performed with a small minicomputer available in the laboratory. Its memory size can accommodate a maximum of 80 profiles (224 temperatures each); provisions to bypass this limitation are gradually being implemented. After the experiment, the raw data are processed on the same machine to obtain temperatures and spatial positions. These profiles are sent via an IEEE-488 line to a larger minicomputer (HP Model 320; 2-Mbyte RAM) for computations in tube-space and for thermophysical property determinations.

#### 4. MEASUREMENT TECHNIQUE

Thermal conductivity experiments with this dynamic technique should be performed on cylindrical or tubular specimens compatible with the long thin rod approximation. At present, extensive experience is available only with tubular specimens with a blackbody hole, because most of the measurements were performed with the specimens previously used for the determination of heat capacity, electrical resistivity, hemispherical total emittance, and normal spectral emittance of niobium [11] and for the measurement of its thermal expansion [12]. The presence of a small blackbody hole ( $0.7 \times 1 \text{ mm}$ ) and of the cross-section compensation flats on the side opposite from the temperature profile measurements clearly does not satisfy completely the radial symmetry required by the long thin rod approximation; however, the largest disturbance (due to the presence of the rectangular blackbody hole) is in the central region of the specimen, where thermal conductivity measurements are practically impossible because the temperature profile is flat and very little thermal conduction takes place. On the other hand, the presence of a blackbody hole with high emissivity ( $>0.99$ ) creates direct ties between the temperatures of the profiles and a blackbody temperature scale, with immediate potential advantages.

The dynamic technique for thermal conductivity is entirely based on accurate temperature measurements via scanning pyrometry. Radiance temperature measurements by the pyrometer must be transformed into true temperatures and therefore the accuracy of this transformation is crucial to the accuracy of the technique. A similar problem was solved in the development of the pulse thermal expansion technique [5]: in that case, measurements were performed during heating and two possible different methods to obtain true temperatures from radiance temperatures (via normal spectral emittance and via resistivity) were described. For the

thermal conductivity experiment with cooling profiles, the only method available is via normal spectral emittance. A direct measurement of the normal spectral emittance of the specimen was tried with acceptable results, but it did suffer from a main problem: either normal spectral emittance measurements were repeated very often (with the need to stop the rotating mirror, position both pyrometers and use a different hardware setup) or there was always uncertainty whether some minimal change (specimen surface change, dust collected on optical surfaces, window coating,...) had occurred in the conditions which existed during the emittance determinations.

A valid alternative was found in performing "apparent emittance" measurements during heating and in using the results for the analysis of data taken during cooling. These measurements require a fixed pyrometer focused inside the blackbody and the scanning pyrometer (see Fig. 1). Fast heating is necessary so that temperature profiles are flat in the central region between the probes (this is verifiable with the scanning pyrometer). Under these conditions the true temperature of the central portion is given by the blackbody measurement and the average radiance temperature (between the probes) is obtained by the scanning pyrometer measurements. From these data over the entire heating an "apparent emittance" as a function of true temperature is computed and it is used during the following cooling to compute the temperature profiles from the scanning pyrometer signals. The apparent emittance might differ from the true normal spectral emittance of the specimen, but it takes into account all possible changing conditions (transmission changes, coatings,...) in the optics and in the specimen right at the time of profile measurements. It also represents an average emittance over a considerable portion of the specimen surface and does not depend on a single point.

A possible variation of this scheme was successfully tried for the occasions when only one high-speed pyrometer might be available. In this situation additional measurements are needed to determine the electrical resistance of an individual specimen as a function of true temperature using the fixed pyrometer focused on the blackbody hole. This is an accurate and reproducible function and the major uncertainties affecting electrical resistivity determinations (knowledge of probe separation and cross-sectional area) are bypassed. During the heating phase of the experiments to measure the temperature profiles, the true temperature is obtained using the specimen itself as a resistance thermometer and the apparent spectral emittance is computed as before.

A typical set of experimental temperature profiles is shown in Fig. 3. The subsequent processing involves three different computer programs. A first program transforms the temperature profiles from the absolute space

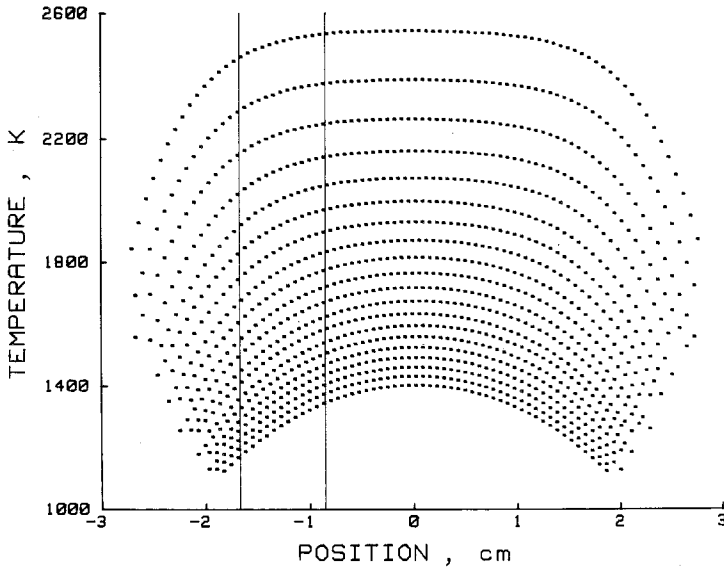


Fig. 3. Experimental temperature profiles and typical subset (between  $-0.87$  and  $-1.67$  cm) used for computations. For viewing purposes, a reduced data set is presented: one profile of every three is plotted, and on each profile shown, only one temperature of every two is plotted.

of measurements to tube-space. At the same time the first derivative with respect to tube-space is computed for all pyrometric measurements on each profile. A second interactive computer program prepares the experimentally measured data necessary for the solution of Eq. (5) at each selected spatial position; a symmetrical portion of tube-space is used for all computations (see typical subset in Fig. 3). On each portion of tube-space (generally consisting of 25 positions) the temperatures of each profile are fitted with respect to tube-space using third-degree polynomials: the fitted temperature and the first derivative in the center position of each profile are retained in a file for further processing. The space derivatives (computed in the first program) are similarly fitted with respect to tube-space: the first derivative of the fit (second derivative with respect to space) is added to the file. The entire process is repeated for all profiles. Then the center temperatures are fitted with respect to time using various polynomials (from third to seventh order). The degree of the chosen fit is determined on the basis of the standard deviation and the shape of the cooling rate function ("wobbling" rates have no physical meaning and are the result of mathematical approximations). The cooling rate of the chosen fit is added to the file: the calculation is then repeated for another portion of tube-space or for other experiments.

A certain amount of subjective judgment is necessary to select the most useful tube-space positions for computations. The following criteria have proved useful in extracting information from the profiles without excessive calculations.

- (a) Select the regions with sharp temperature gradients. As a pragmatic rule, points on profiles where the conduction term is less than 25% of the heat capacity term are not considered.
- (b) Avoid regions where humps and bumps in the  $T(x)$  function are shown (graphically visible on the screen during the fit). These regions either exhibit a small cross-section difference with respect to the rest of the specimen or have a normal spectral emittance problem on the surface. In both cases that region is not usable for calculations.
- (c) Calculations in four regions (two per each side) away from the center generally enable the extraction of all possible information from each experiment. This rule sets a limit of 320 equations to be solved for each experiment.

Finally, a third computer program collects all the data from the different subset files and builds an overdetermined system of equations as given by Eq. (5). The data may come from different portions of the same experiment, from different experiments, and/or from different specimens. A polynomial function of thermal conductivity vs temperature is computed for various orders (generally up to a third degree polynomial); selection of the best function depends both on practical considerations and on how the various polynomials fit the data.

## 5. CONCLUSIONS

This new dynamic technique has the potential of extending the pulse method to measurements of thermal conductivity. Its main advantages are

- measurements in a large temperature range with the specimen remaining at high temperatures for 10–20 s only;
- operation in vacuum or in inert atmosphere;
- experiments performed on specimens which can also be used for the determination of other thermophysical properties; and
- possible direct ties to a blackbody temperature scale.

The work is continuing with a full assessment of dynamic heating experiments. Measurements of the thermal conductivity of niobium in a large temperature range are being performed, along with a complete study of the accuracy of the method.

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