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Report on the Workshop on Transient Methods for Measuring Thermophysical Properties

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A Workshop on Transient Methods for Measuring Thermophysical Properties was organised during the 15th European Conference on Thermophysical Properties, Würzburg, Germany, 4 – 7 September 1999. The aim of the Workshop was to bring together researchers using transient methods to exchange knowledge regarding theory, instrumentation, and applications. The Workshop was a working meeting with a goal to create a common attitude towards measurement accuracy, data reliability, and the choice of suitable reference materials (SRM).

In his introduction, the chairman **L'udovít Kubičár** (Institute of Physics, SAS, Slovakia) gave an overview on transient methods and listed potential topics for discussion:

1. Various methods operating with dynamic temperature fields are used for measuring specific heat, thermal diffusivity, and thermal conductivity. The aim of the workshop was to focus on advances in theory, instrumentation, and applications of transient methods. These methods involve measuring probes (usually the heat source and the thermometer) embedded into materials. Depending on the size of the probe and of the specimen, measurements can be performed on materials with considerable inhomogeneities. These methods can be characterised by:

- form of the heat source (line, plane, or disk);
- way of generation of the temperature field (pulse of heat or heat flux in the form of a stepwise function);
- number of probes (one probe when the heat source is also the thermometer or two probes when they are separated).

2. The measurement process is characterised by the application of a small disturbance to a specimen held at a stable and uniform temperature. The disturbance is applied in the form of a heat pulse or a heat flux characterised by a stepwise function. The temperature response is recorded. A model of the method and the experimental temperature response are used to calculate the thermophysical parameters. Depending on the method one or two parameters can be measured directly. Theoretical and experimental analysis of the discrepancies between the model and the experiment is of major interest. A set of real models corresponding to various boundary and initial conditions needs to be used to understand the measuring process by any transient method. Intercomparison is a key strategy of the verification of the measuring technique, and introduction of the ideal and the real model is here of major importance.
3. The availability of SRMs certified simultaneously for thermal conductivity, thermal diffusivity, and specific heat is necessary to help evaluate differences in any intercomparison.

Following the introduction, **Ron Tye** (Sinku-Riko, Japan) made a brief presentation indicating several basic issues that should be addressed during the Workshop. From a historical view he appreciated the fact that the developments and introduction of these methods of measurement were in direct response to the escalation of the growth of new and modified materials often in special forms and for a widely varying set of applications. The requirements for performance data were such that the classical methods were too complex and time-consuming and often required test specimens of larger size and of different configurations than could be fabricated. The introduction of relatively simple and fast techniques makes them very attractive and popular.

However, the response has been a proliferation of such methods and their modifications each claiming a high precision, often $\pm 3\%$ to 5% or better, for one or more properties being measured or derived. Now that more results are being published for different material types, some of which have been measured by classical techniques, we find that such claims cannot be substantiated. Furthermore, in some cases the results for individual properties are often internally inconsistent, particularly with respect to measured and derived thermal conductivity.

Essentially the techniques are often based upon somewhat similar simplistic models developed as solutions of the basic heat transfer equation but involving a number of assumptions and then trying to make the experimental setup parameters represent the solution. The issues here are that the models require refinement, as may the experimental parameters representing them. For example, account should be taken of radiative heat transmission, contact resistances, optimum temperature limits for technique, and also under what circumstances the normally accepted thermal conductivity/specific heat capacity/thermal diffusivity relationship does not apply.

Using the thermal probe version of the line source method as an example, one finds that there are dimensional limitations such that one requires different probes for material types, different applications, and range of thermal properties. In addition, there are both internal and external contact resistances to be considered as does their consistency, and/or behavioural reproducibility under heating and cooling.

Obviously, to verify the precision and efficacy of a method, the availability of one or more reference materials is a major, but not the sole, contribution. It is clear that we need a variety of such reference materials for the range of material types, thermal properties, and temperatures that are claimed acceptable for these methods. However, these methods involve two or three properties and thus reference values should, ideally, be available for all three properties. Furthermore, for the range of applications for which the methods are said to be suitable, one needs to determine from the users the acceptable levels to which the performance values are required. These may vary for different applications and it may not be necessary always to determine them to a high precision, say 3% or better. Finally, it is clear that there is or will be a need to standardise one or more of these techniques. It is unlikely that one basic standard could be developed to cover all versions but as a first exercise an essential feature is for intercomparison(s) of different techniques to be undertaken. This should be a prime activity for near future and one which should involve a great deal of international cooperation both in the modelling and experimental areas.

Aaron Nabi (RAFAEL, Israel) commented on the consequences of the difference between the model used and the real experimental setup. Thermophysical properties measured by the various transient techniques and by different laboratories on the same kind of materials exhibit, in some cases, quite large deviations from each other (10% to 20% or even more). These deviations are much larger than the measurement errors reported by the individual laboratories (typically 2% to 5%). Part of the scatter in reported properties can be attributed to real differences in material properties due to small differences in composition (for different batches), material inhomogeneities (in the same batch), anisotropy, different suppliers, etc. It seems that most of the remaining differences are due to systematic measurement errors not accounted for in the model simulating the experiment, or, as in many cases, even unknown to the experimenter. Systematic errors include all the errors that can be attributed to differences between the actual experimental setup and the model being used to derive the property. Many models relate to an ideal situation and do not represent the precise physics of the experiment. To name only a few of the systematic errors sources: thermal resistance and capacitance of the heater in the hot wire, hot plate, and hot disk methods; two- and three-dimensional effects; semi-transparency effect; heat losses from the sample by convection and radiation and by conduction through the sample holder; non-uniform heating; and many more, some we are even unaware of.

Transient thermophysical measurement techniques have in common the measurement of time response of a variable (mostly temperature) to heat input. In order to enable the experimenter and other reviewers to detect the presence of systematic errors in the experiment, it is suggested to include with the reported thermal properties a plot of the measured temperature together with the temperature predicted by the model and residuals. Most of the systematic errors (including those we are not aware of) will result in residuals not being scattered evenly throughout the measurement time interval, but rather, have a shape (horizontal 'S' shape or 'U' shape). Unfortunately, the residuals plot is not a quantitative tool and cannot tell the source or the exact nature of the systematic error; this still remains a major task of the experimenter. Despite the fact that it is not a quantitative tool, this approach can help a great deal in tracing non-ideal phenomena and in assigning a more realistic error to the reported properties.

A comment of **Libor Vozar** (Constantine the Philosopher University, Slovakia) was related to the use of more complicated models. Improvement of the analytical model of a transient method that takes into account more realistic (non-ideal) initial and boundary conditions results in general in an increase of the accuracy of the measurement. The disadvantage this brings is that increasing the number of parameters in a data reduction process based on a least-squares fitting reduces the sensitivity of the experimental method to the estimated parameter(s) and prolongs the optimal duration of an experiment. The other approach—performing complementary measurements (calibrations)—may easily incorporate significant errors. The approach he recommended is to use simple analytical models while trying to maintain the assumptions of the ideal experimental conditions, where possible.

Ulf Hammerschmidt (PTB, Germany) dealt with the importance of an appropriate time window for data evaluation. The transient hot-strip method for determining thermal conductivity and thermal diffusivity is characterised by its experimental simplicity. In fact, the measuring process consists of monitoring of the resistance change of the strip by the four-wire method and using a constant electrical current source. In the case of dielectrics, the measuring process takes several minutes. However, the evaluation of thermal conductivity and thermal diffusivity from the data set is a sophisticated process and easily subject to errors because of the complex mathematical model. A crucial problem is to find a time window for data evaluation that gives 'closest agreement' when one intercompares data. Therefore, the complex model has been analysed to find a time window in which thermal conductivity can be evaluated in a similar way as with the transient hot-wire method. The mathematical model consists of three different terms that correspond to different symmetries of the temperature field. The symmetry of the temperature field changes in time from plane to concentric cylinders. The last one agrees with the symmetry of the temperature field generated by the transient hot-wire method. Thus, for long times, t, the temperature of a strip is a linear function of $\ln t$. Plot of the strip temperature against $\ln t$ gives a linear part used for data evaluation.

Anne Kibble (NPL, United Kingdom) highlighted some of the difficulties in assessing the reliability of thermal property data produced with transient techniques, a problem which is further compounded when thermal conductivity values obtained by a steady-state technique are compared with those calculated from measurements of thermal diffusivity, specific heat capacity, and density of a material. Data discrepancies that arise could be due to a number of causes including:

- the uncertainty of the individual measurement methods (whereas thermal conductivity can be measured directly to within ±3%, the combined uncertainty in thermal conductivity arising from measurements of thermal diffusivity and heat capacity can amount to ±7% or more);
- high-thermal-expansion materials (typically metals) can have significant changes in density with temperature which may need to be included;
- literature data often provide insufficient materials characterisation, eg material purity and thermal history.

The above and other issues related to data reliability and consistency merit further attention from the measurement and standards community.

Mattias Gustavsson (Chalmers University, Sweden) concentrated his contribution on the hot disk technique—referred to by some as the Gustafsson-probe technique. A major advantage of this technique, and similar techniques, for the common user is the flexibility in sample preparation—virtually any sample geometry may be studied—including a large flexibility in sample size and range in thermal conductivity. The only requirement is that the sample surface facing the sensor should be relatively flat. Also, it is possible to perform tests in short times. One fact of importance for industry is that testing may be performed on real samples taken directly from the manufacturing process or when the material is in use. In this way, thermal transport properties are obtained for samples in which the effects stemming from the manufacturing process are incorporated in material structure.

The real model used in curve fitting is quite different from the ideal model. Among the differences between the real and the ideal model, the dominating one—and which is fully accounted for—is the thermal contact resistance between the sensor and the sample. The results thus represent 'bulk' properties of the structure, and not method-specific 'apparent' or 'effective' thermal conductivity values. Several other and less significant differences accounted for in the real model need not be fitted in the data reduction process, for instance the variation in output of power during the stepwise pulse ($<1\%$), and the amount of heat consumed by the sensor itself during the transient. Accounting for these effects in the real model thus improves the general accuracy without reducing sensitivity in the fitting procedures. Fully reproducible thermal conductivity results are possible even when reassembling a sample set at different mounting pressures, or if the surface roughness has changed. This also holds true if a deviating surface layer is present, or even if the sensor insulation has changed.

Two topics need some more attention. The first concerns consistency between conductivity, diffusivity, and specific heat, obtained by one method compared with the results obtained by another method. Discrepancies in these properties were noted, also for the hot disk, to some extent in the thermal diffusivity and specific heat. An approach was suggested how to avoid these discrepancies so as to achieve full consistency.

The other topic of interest concerns accuracy and intercomparison testing. Apart from deviations in intercomparison results caused by anisotropy and material stability, a general cause of error is discrepancies between models used for a particular method compared to the real experimental situation. This is not only the case with transient methods. The accuracy issue needs to be further studied, and this apparently coincides with the issue of SRM and the need for reliable reference data.

David Salmon (NPL, United Kingdom) presented an overview of the EU project to establish Pyroceram 9606 as a Certified Reference Material for thermal conductivity and diffusivity over the temperature range 100 to 1000 °C. Measurements are being made of thermal expansion, specific heat capacity, density, thermal diffusivity, and thermal conductivity by several European laboratories to characterise a large batch of Pyroceram 9606 material obtained from Corning Glass Co. in the United States.

Thermal expansion measurements, ultrasound techniques, and studies of the microstructure are being carried out to quantify the material anisotropy. Measurements of its thermal properties are also being carried out on several blocks of the material to determine the degree of uniformity within the batch.

After the certification stage, six laboratories will measure the thermal diffusivity of Pyroceram 9606 by two techniques. Thermal conductivity will be measured with the transient hot-wire, steady-state axial heat flow, and guarded hot-plate methods. The thermal conductivity will also be determined by calculation from the diffusivity (and heat capacity) measurements, and thus thermal conductivity determined by various transient and steady-state measurements can be compared. It is expected that the proposed Pyroceram 9606 Certified Reference Material will be available from IRMM in about three years time.

As a result of this significant amount of interest generated in transient methods and the apparent will of the attendees to approach the subject by means of a concerted cooperative effort, NPL staff suggested that one approach that could be investigated is via an EU funded Network as a concerted action. This would allow interested partners to meet once or twice a year to discuss and better focus research activities on transient thermal property measurement techniques. At some stage it should be possible to hold a workshop to discuss the standardisation of transient techniques. The Workshop participants were asked to add their names to a list if they were interested in participating, and 18 names were collected. David Salmon agreed to enquire about details of possible EU funding and to coordinate any action on behalf of the group.

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