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Evaluation of a Pulse-Heating Reflectometric Technique[1]

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The classical millisecond-resolution pulse-heating method makes use of tubular specimens with a rectangular blackbody hole for accurate measurements of several thermophysical properties at high temperatures. An alternative measurement technique that uses simple strip specimens has been developed at the IMGC (Italy). The normal spectral emissivity of the pulse-heated strip is measured by integrating sphere reflectometry, using a modulated laser beam to discriminate between the reflected radiation and that self-emitted by the specimen. After a brief description of the measurement system and a review of the main experimental results, attention is devoted to current developments and possible improvements of the method, with particular attention to experiments performed at the melting point. The measurement technique is analyzed and evaluated, with a brief description of potential future measurement possibilities.

KEY WORDS: high temperatures; normal spectral emissivity; pulse heating; reflectivity; subsecond thermophysics; transient techniques.

1. INTRODUCTION

Several measurement techniques based on subsecond pulse-heating methods were developed in the last thirty years. Details of the original measurement method (indicated here as the "classical" technique) are available in the literature [1, 2]. The classical technique, as developed at the National Bureau of Standards (NBS, U.S.A., presently the National Institute of Standards and

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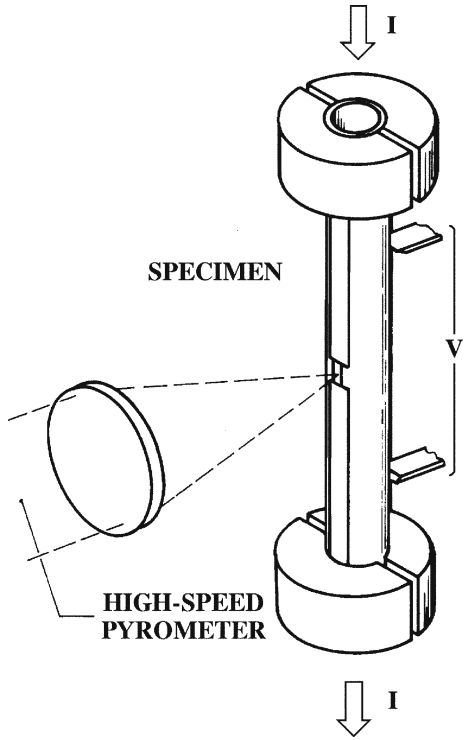

Fig. 1. Schematic representation of the classical technique (from Ref. 3). I, current through the tubular specimen; V, voltage drop in the central portion of the specimen.

Technology, NIST) made use of tubular specimens with a rectangular blackbody hole (Fig. 1). Using this type of specimens, it was possible to perform accurate determinations of several thermophysical properties (heat capacity, electrical resistivity, hemispherical total emissivity) of electrical conductors at high temperatures, from above 1500 K up to the melting point. Further developments through the years extended the subsecond pulse-heating measurement techniques to normal spectral emissivity, thermal expansion, thermal conductivity and to measurements at the melting point. A recent review [3] presents an introduction to the pulse-heating method and to the different techniques developed over several decades, while another review [4] discusses the application of high-speed pyrometry to these measurement methods.

The classical technique provides the most accurate experimental results, but the machining of tubular specimens with a blackbody hole

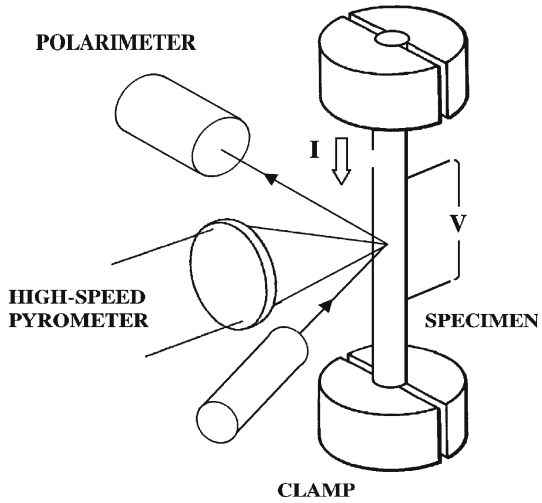

Fig. 2. Schematic representation of the laser polarimetric technique. I, current through the rod specimen; V, voltage drop in the central portion of the specimen.

is costly and time consuming. In the last decade alternative techniques were developed to perform similar measurements on specimens with simpler geometries. NIST, in collaboration with Containerless Research Inc., developed laser polarimetry to measure the normal spectral emissivity of rod-shaped specimens during the pulse-heating experiment (Fig. 2). The National Research Laboratory of Metrology (NRLM, Japan, presently the National Metrology Institute of Japan, NMIJ) also used laser polarimetry combining the pulse heating of a strip with a brief steady-state experiment [5]. An alternative approach was adopted at the CNR Istituto Metrologia G. Colonnetti (IMGC, Italy) that realized a high-speed version of an integrating sphere reflectometer to measure the normal spectral emissivity of a strip specimen (Fig. 3) during a pulse-heating experiment.

All these recently developed techniques that use specimens without a blackbody hole share some common important features: (i) the need of direct determination of the normal spectral emissivity of the specimen surface during the pulse-heating experiment, (ii) the fact that normal spectral emissivity is measured at one point, and (iii) the necessity that the specific location where emissivity is measured be truly representative of the entire specimen surface. Measurements inside a blackbody are highly forgiving of possible emissivity variations inside the cavity because the blackbody integrates emitted radiation. Using specimens of simple geometry without a blackbody hole, a poor selection of the surface point where emissivity is

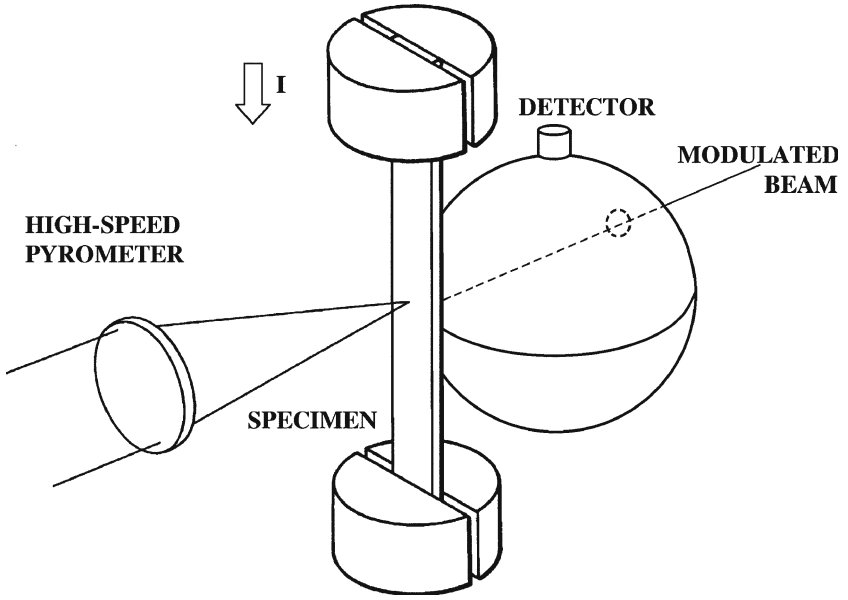

Fig. 3. Schematic representation of the reflectometric technique (from Ref. 3). I, current through the strip specimen.

measured may lead to completely erroneous results. Consequently, the uniformity of the specimen emissivity becomes a crucial factor.

This paper analyzes in detail the fast reflectometric technique developed at the IMGC in the last decade, presenting a summary of experimental results and evaluating the research work necessary for further development.

2. DEVELOPED SYSTEM AND EXPERIMENTAL RESULTS

A short description of the high-speed reflectometric technique and a summary of experimental results and related literature references are presented in this section. The pulse technique based on integrating sphere reflectometry for the determination of the normal spectral emissivity of electrical conductors was developed at the IMGC in the mid-1990s [6]. An improved version of the apparatus was described later, in a publication that also presented experimental results of the normal spectral emissivity at 900 nm of niobium up to temperatures around 2700 K [7]. Subsecond pulse-heating experiments were performed on strip specimens, computing true temperatures from radiance temperatures and normal spectral emissivity data. The experimental results of thermophysical

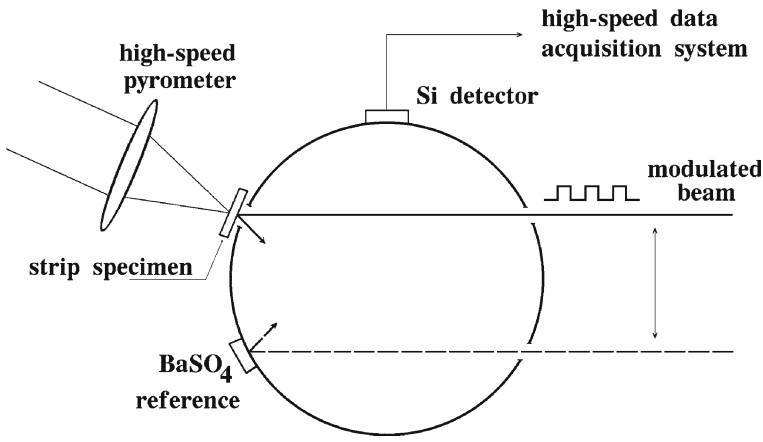

Fig. 4. Cross-sectional view of the reflectometric technique (from Ref. 9).

properties of niobium [8], as computed from measurements performed on strips, were found to be compatible with similar measurements performed fifteen years earlier on specimens with a blackbody hole, confirming the accuracy of the new technique. Measurements were then extended to the melting-point region [9], observing rapid emissivity variations due to the change of the surface structure during melting. A detailed study of normal spectral emissivity changes in niobium strip specimens under pulse-heating conditions was also performed [10], revealing that the emissivity is unstable during the initial heating–cooling cycles because surface changes are governed by a diffusion type phenomenon that evidently is not completed until the specimen has been brought several times to its melting point. Such an observation has important implications for the accuracy of measurements performed by pulse-heating strips.

A schematic cross-sectional diagram of the technique developed at the IMGC is presented in Fig. 4. It is a high-speed version of an integrating sphere reflectometer of the comparison type, in which the reflectivity of the specimen, that undergoes pulse heating, is always measured relative to the known reflectivity of a barium sulfate ($BaSO_4$) reference standard. The measured quantity is the spectral directional-hemispherical reflectivity of one side of the specimen at the wavelength of the interference filter placed in front of the silicon detector of the sphere. According to Kirchoff's law for opaque materials, this quantity is the complement to one of the normal spectral emissivity of the specimen.

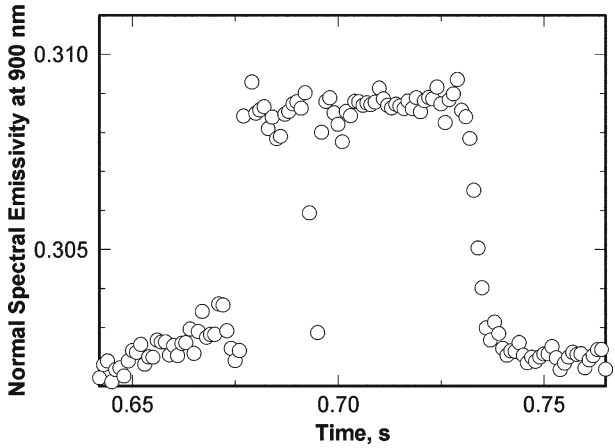

Fig. 5. Typical emissivity measurements at the melting point of niobium for specimens with a well characterized surface.

The radiance temperature on one side of the strip is measured with a high-speed pyrometer [11]. The other side of the strip is placed outside the porthole of a small integrating sphere located inside the environmental chamber. A modulated beam generated by a laser diode is reflected by the side of the strip facing the sphere. The reflected beam is collected by the integrating sphere and measured by a silicon detector in the sphere ceiling. The detector is operated at the same wavelength of the high-speed pyrometer, presently at 900 nm. A numerical lock-in technique is used to discriminate between the reflected modulated beam and the radiation self-emitted by the specimen at high temperatures. Until now the apparatus has been operated in a relative mode, using the reference value of the normal spectral emissivity at 900 nm of niobium at its melting point as a calibration point [12]. Additional details on the measurement technique and on the pulse-heating system can be found in earlier publications [6, 7, 13]. A multi-wavelength version of this type of apparatus has been developed recently at the Harbin Institute of Technology (HIT, P. R. China) [14].

3. CURRENT DEVELOPMENTS AND POSSIBLE IMPROVEMENTS

The fast reflectometric technique has been operational for some years producing significant experimental results. A typical emissivity measurement at the melting point of niobium is presented in Fig. 5. This type of behavior is typical for a strip specimen that has been taken to its melting point several times. On melting of the surface layer, the normal

spectral emissivity of the niobium strip exhibits a sudden jump and then remains constant during the melting process. An electrical disturbance is noticed when the current is interrupted, but the emissivity does not change until the surface starts to re-solidify. The liquid–solid transition on cooling takes a longer time, but the emissivity value of the fully solidified surface returns to the previous solid surface value.

The rest of this section describes recent developments, possible further advances, and some experimental limitations of the present apparatus. The experimental system is not a static one: continuous modifications and improvements are implemented when appropriate electronic and optical components become available. A recent advance was the substitution of electro-mechanical switches in the pulse-heating system with mosfet switches. These new electronic components, capable of interrupting large currents in sub-microsecond times, have simplified the operation of the apparatus making it possible to reach melting in a single experiment, while programming via software the duration of the melting plateau. The study of normal spectral emissivity changes in niobium strips taken to their melting point [10] was made possible by the availability of these components and by their implementation into the apparatus. Other possible developments, like the feedback control of the mosfet switches to regulate current flow, might make possible programmable heating and cooling rates, improving the control of the experiment duration that is presently defined only by the time base generator controlling the current pulse in combination with imparted power and specimen geometry.

One of the interesting features of such an experimental system is the possibility to perform accurate studies on the specimen during melting. The present experimental apparatus evolved from an earlier version, where measurements were made on tubular specimens with a blackbody hole [13]. A small integrating sphere was inserted into an existing experimental chamber (as shown in Fig. 3), accepting the inevitable limitations due to the adaptations. One can see from the cross section of the apparatus in Fig. 4 that the specimen is not symmetrical with respect to current flow. During the experiment, large currents flow into the specimen and in its support columns, creating electro-dynamic forces acting on the various parts of the circuit. As long as the specimen is in the solid phase, its stiffness prevents any movement. When the beginning of melting occurs, the specimen loses much of its mechanical strength and, in the present geometry, there are some force components that tend to bend it away from the sphere. If this happens and the strip moves, the experiment is lost and the specimen no longer usable. Care must be exercised, both in the clamping action and in the correct duration of the melting plateau, to be able to perform repeated experiments at the melting point. The construction

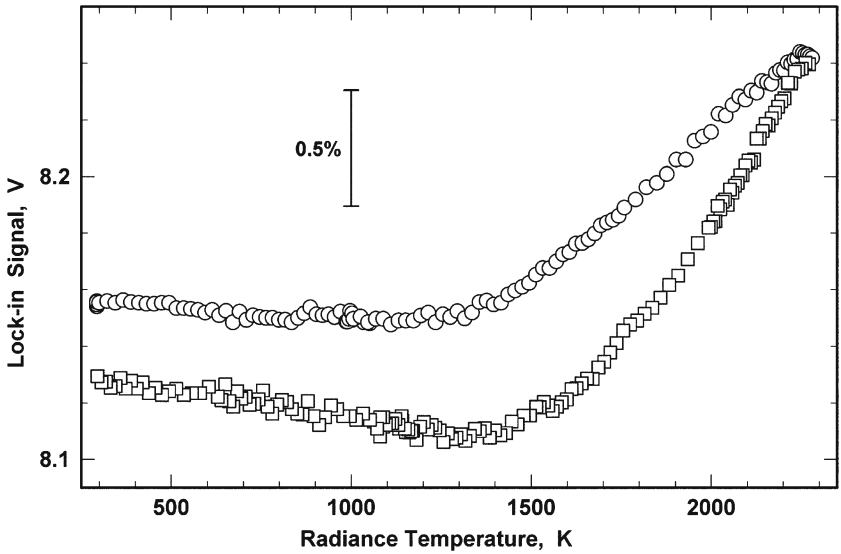

Fig. 6. Change in the lock-in signal during a measurement on $BaSO_4$ (circles, heating; squares, cooling). Data from Ref. 7.

of a new experimental chamber, where the specimen is exactly symmetrical between the support columns, would improve considerably the possibility to obtain longer melting plateaus and limit the danger of specimen bending.

Another limitation of the present experimental system is related to the size of the integrating sphere, currently 100 mm in internal diameter. The three holes for the laser diode beam are 9 mm in diameter, and similar size holes are used for the silicon detector and for the $BaSO_4$ reflectivity standard. A good integrating sphere is generally large, with little openings with respect to the total surface. The present small sphere with fairly large openings is not the ideal solution, and geometrical correction formulas to take into account the openings fail to provide adequate results. Therefore, in all experiments performed so far, it has been necessary to perform a relative measurement, requiring a knowledge of the correct normal spectral emissivity value at the melting point. This is a severe limitation in performing experiments on materials for which such a value at the melting point is unknown. Work is continuing regarding this topic to evaluate if some experimental correction factors may be usable when the emissivity reference value is unknown.

Two other experimental problems were consistently noticed but not yet properly explained. Measurements are performed with respect to a reflectivity standard, a small $BaSO_4$ specimen placed on the sphere inner surface. When a pulse-heating experiment on the niobium specimen is performed, but the laser diode beam is directed to the reflectivity standard, one would expect a constant signal value from the $BaSO_4$ specimen. A typical experimental result for this type of experiment is shown in Fig. 6, clearly indicating that some stray radiation due to the heating of the niobium specimen somehow reaches the reflectivity standard modifying its constant value. The fact that the lock-in signal during cooling is lower is also the indication of a possible coating of the sphere inner surface with vapors emitted by the melting specimen. This problem requires a correction to be applied to the results of experiments performed on the niobium strips, assuming that the phenomenon is repeatable when the experimental configuration is modified.

Another experimental problem is related to the poor applicability of the measurement technique to very thin specimens. Experiments with strips with several thicknesses over 0.25 mm are accurate and reproducible. Experiments with thinner specimen are sometime unpredictable, showing preferential melting paths on the specimen surface for unknown reasons, as shown in Fig. 7. The uneven surface emissivity distribution makes results from such an experiment unusable for accurate thermophysical property measurements. This uneven emissivity distribution is observed during and after the first experiment only, with the second experiment to the melting point providing a uniform emissivity distribution over the entire specimen surface.

4. FUTURE POSSIBILITIES

This final section discusses some possible developments of the pulse-heating reflectometric technique. One of the most interesting scientific possibilities is the extension of the method to the measurement of other thermophysical properties. At the IMGC this has already been done using high-speed scanning pyrometry [15] and developing measurement techniques for both thermal expansion [16] and thermal conductivity [17] using tubular specimens. In the future similar experiments might be performed using strip specimens and the fast reflectometric technique.

Another interesting possibility is the extension of this measurement method to lower temperatures, performing experiments starting from room temperature. Such an extension would require the development of high-speed pyrometry at longer wavelengths: the feasibility of this approach has been demonstrated by the Japanese metrological laboratory, that

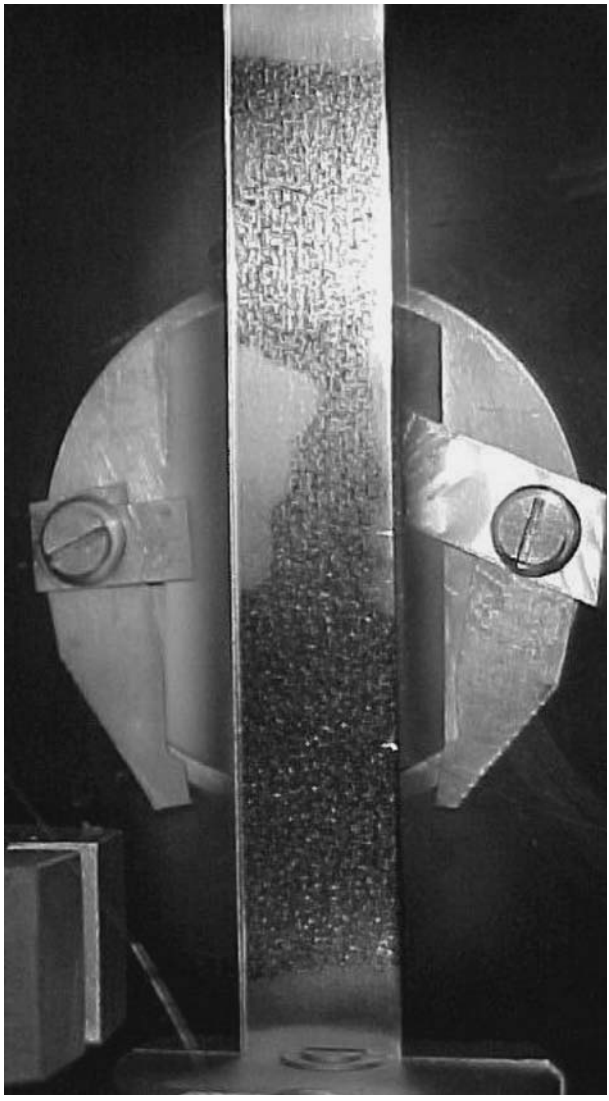

Fig. 7. Thin specimen after pulse-heating with uneven surface melting distribution.

has developed a microsecond resolution pyrometer operating with an InSb detector [18]. Another possibility could involve the development of InGaAs pyrometers, operating around 1.6 μm. Such a pyrometer might not be able to perform measurements at room temperature, but would

extend considerably the range to lower temperatures, possibly down to 500 K. Another alternative possibility could be the use of the specimen itself as a resistance thermometer. The electrical resistance experimental data, performed by the four-wire method, are already available from room temperature upward: it would be necessary to perform appropriate studies for each measured material about the relation between electrical resistivity and temperature, combining experimental results with studies of the reproducibility of the strip thermometer in various experimental conditions.

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