

Measuring normal spectral emissivities of niobium by a pulse-heating technique: 1000 K to the melting point

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Received June 8, 2006

The normal spectral emissivity of niobium strip specimen is measured using a new pulse-heating reflectometric technique. The hemispherical spectral reflectivity of the surface of a strip tangent to an integrating sphere is determined by a high-speed lock-in technique. At the same time, the radiance temperature of the strip is measured by the multi-wavelength high-speed pyrometer from approximately 1000 K to the melting point. Details of the measurement method and of the related calibration techniques are reported. Results of the normal spectral emissivity of niobium at 633, 753, 827, and 905 nm from room temperature to its melting point are presented. The accuracy of spectral emissivities is estimated better than 5%.

OCIS codes: 120.0120, 120.4570, 000.6850, 120.6780, 300.2140.

Pulse-heating techniques have gained wide acceptance for the determination of several thermophysical properties at high temperatures. The development of pulse-heating apparatus is aimed at the measurement of thermophysical properties for martial and space materials. The most accurate measurements of the mentioned thermophysical properties using the pulse-heating technique are performed on tubular specimens with a blackbody hole, which are difficult to produce and require very careful machining. In practical applications measurements must often be performed in short times on simple specimens. Two techniques have been proposed to perform direct measurements of thermophysical properties on simple specimens^[1-4]. The method developed at the National Institute of Standards and Technology (NIST, USA) used cylindrical specimens and performs direct measurements of the normal spectral emissivity using laser polarimetry^[1]. The technique developed at the CNR Istituto di Metrologia "G. Colonnetti" (IMGC, Italy) made use of specimens in the form of strips, and the normal spectral emissivity was measured by integrating sphere reflectometry in pulse-heating conditions^[2-4]. A millisecond pulse-heating apparatus was developed at the Harbin Institute of Technology (HIT, China)^[5]. The design was based on systems previously developed both at NIST and IMGC for the measurement of several thermophysical properties with millisecond time resolution. This paper describes experimental measurements of the normal spectral emissivity of niobium strip specimens at different wavelengths over a wide temperature range (from room temperature to the melting point) obtained recently at HIT.

The measurement technique is schematically shown in Fig. 1. It combines multi-wavelength pyrometry with a high-speed version of integrating sphere reflectometric technique. Measurements are performed on a niobium strip specimen placed in high vacuum (better than 10^{-3} Pa) and pulse heated by an electric power circuit with the maximum current up to 1700 A. The radiance temperatures are measured on one side of the strip at several wavelengths by a four-wavelength high-speed pyrometer with microsecond time resolution. Four operating

channels of the pyrometer have been selected to measure the radiance temperature at 633, 753, 827, and 905 nm. The other side of the strip faces the porthole of a high-speed version of integrating sphere reflectometer of the comparison type, in which the reflectivity of the pulse-heated specimen is measured in relation to the known reflectivity of a barium sulfate (BaSO_4) reference specimen. The measured quantity is the spectral directional-hemispherical reflectivity ρ of the specimen at the wavelength of the interference filter placed in front of the silicon detector. Considering the small changes of directional properties measured at small angles ($< 10^\circ$) from the normal, this property can be considered the complement to the normal spectral emissivity ε of the specimen (Kirchhoff's law, $\varepsilon = 1 - \rho$). Using this apparatus, the normal spectral emissivity of the specimen at one wavelength can be measured in one experiment over a wide temperature range. The HIT method has been described previously^[5]; a brief summary is reported here, including some significant modifications of the apparatus.

A modulated beam generated by a laser diode strikes the side of the strip facing the integrating sphere. The reflected beam is collected by the integrating sphere, taken out with an optical fiber, and measured by a silicon detector placed outside the experimental chamber. The detector operates at the wavelength of the laser diode (at

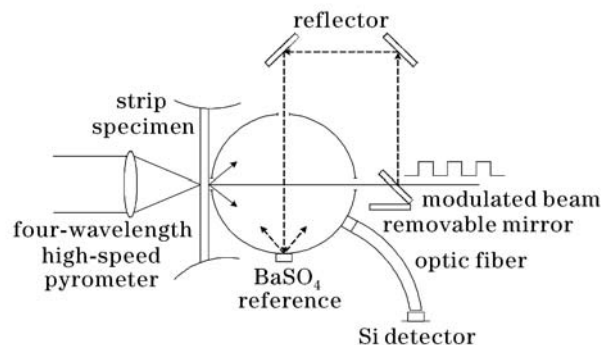


Fig. 1. Schematic diagram of the multi-wavelength pulse-heating reflectometric technique.

present, near 633 nm) that is the same as one of the four-wavelength high-speed pyrometer. A numerical lock-in technique is used to discriminate between the reflected modulated beam and the radiation emitted by the specimen itself at high temperature. The specimen is pulse heated, and can be brought to high temperatures (up to its melting point) in times ranging from 1.1 s to several seconds, depending on the signal-to-noise ratio (SNR) acceptable in the lock-in measurements.

The normal spectral emissivity $\varepsilon_\lambda(T)$ is measured by an integrating sphere reflectometer. At the same time, the spectral temperature T_λ at the same wavelength is measured by a high-speed multi-wavelength pyrometer. A simpler and in many cases very useful expression can be derived using the Wien approximation:

$$T = \frac{c_2 T_\lambda}{\lambda T_\lambda \ln \varepsilon_\lambda(T) + c_2}. \quad (1)$$

So the normal spectral emissivity data $\varepsilon_\lambda(T)$ measured simultaneously is used to obtain the true temperature T from the spectral temperature T_λ measured in experiment. According to the direct spectral temperature measurements of the other three wavelengths at the same time and the true temperature of the specimen, we can figure out the normal spectral emissivities of the specimen at the other several wavelengths. So the multi-wavelength emissivities are obtained by dynamic measurement in the experiment.

Measurements are performed on three niobium strip specimens with the nominal dimensions of 69 mm in length, 10 mm in width, and 1 mm in thickness. The strip specimens are made in Goodfellow Cambridge Limited (LS1234165SAC NB000380/6) from niobium foil of mass purity 99.9%. All experiments are performed in high vacuum (better than 10^{-3} Pa). Typical operational conditions to cover the range from room temperature to the melting point are current pulse duration 2.34 s, heating rate 1000 K/s. The lock-in reference frequency during all experiments is 800 Hz^[6,7]. The normal spectral emissivity is strongly dependent on surface conditions, and all specimens exhibit large changes in normal spectral emissivity during the initial experiments. A stable surface condition is reached only after the specimens have been taken to their melting point a few times, interrupting the current pulse before the destruction of the specimen. After several experiments to the melting point, the specimen surface becomes shiny and exhibits very large grains.

Integrating sphere reflectometry is not an absolute method and therefore requires an appropriate reflectivity reference standard to take into account different factors, including the geometry of the integrating sphere and possible imperfections in its internal coating. This is done in a separate calibration experiment using mirrors placed in an appropriate holder to direct the collimated beam to a BaSO₄ specimen (Fig. 1) and measuring the silicon detector output for all the temperatures of interest. In the calibration experiment, the removable mirror is set at an appropriate angle, so that the collimated beam is directed toward the reference specimen. This calibration can be repeated as many times as necessary by simply moving the reflectors to the appropriate position. A typical measured signal when the collimated beam is directed toward the BaSO₄ specimen is shown in Fig. 2. Theoretically, a

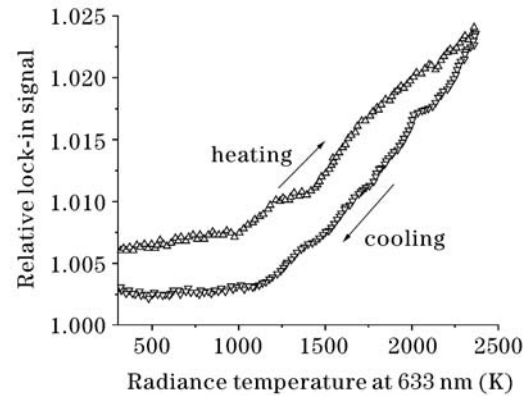


Fig. 2. Typical digital lock-in signal as a function of radiance temperature during an experiment on a BaSO₄ specimen.

constant value should be observed because the BaSO₄ specimen remains at room temperature during the entire experiment, but a variation of about 1% is observed, with the signal during the heating and cooling phases following slightly different paths. On account of this phenomenon, all experimental measurements on niobium strips are related to BaSO₄ reference measurements performed exactly in the same operational conditions.

In the direct experiment, the mirror is removed to send the collimated beam toward the niobium specimen. Additional corrections are necessary as a result of the following causes. 1) For safety reasons, the hot strip specimen must be placed at an appropriate distance from the sphere (generally about 2 mm), with partial loss of the reflected beam. 2) Specimens taken to their melting point exhibit large grain growth and become strongly specular. This condition increases the radiation lost from the beam entrance hole after a single reflection. 3) The BaSO₄ calibration experiment requires a slightly different optical path with three additional mirrors. 4) The BaSO₄ and niobium experiments are performed at different times.

These potential problems are taken care of by using the melting point of niobium as a calibration point. All the measurements presented in this paper have been obtained by adopting the values 0.3482, 0.3284, 0.3172, and 0.3083 for the normal spectral emissivity (at 633, 753, 827, and 905 nm) and 2742 K for the true temperature of niobium at its melting point, as measured earlier at IMGIC (0.3087 for the normal spectral emissivity at 900 nm and 2749 K for the true temperature of niobium at its melting point)^[3].

During these experiments, reflectivity is measured for the entire temperature range, starting before the application of the current pulse (when the specimen is at room temperature) and performing reflectivity measurements during both heating and cooling until the specimen is cooled freely back to room temperature (the total time was in the range of 40 to 60 s). The radiance temperature is measured by high-speed pyrometry from the moment when the specimen reaches approximately 1000 K during heating to the time it reached the same radiance temperature during cooling. Below these temperatures the SNR of the pyrometer does not permit accurate temperature measurements in the wavelength region of the instrument. Temperatures between 300 and 1000 K are

computed by interpolation during heating and by extrapolation during cooling. The normal spectral emissivity is dependent on the surface conditions, and the speed of experiments has a certain influence on the modifications occurring on the surface of the strip.

Experiments are performed on three niobium strips, but final results are presented for a typical specimen that was pulse-heated to its melting point six times. The typical curves of the normal spectral emissivity of niobium at 633 nm versus temperature during both heating and cooling are presented in Fig. 3. The experimental data below 1000 K are measurements of the normal spectral emissivity and temperatures computed by interpolation during heating and by extrapolation during cooling. It may be clearly seen that the normal spectral emissivity of the strip specimen has a different temperature behavior between the heating and cooling curves, and this trend is clearly visible in all experiments. The general trend indicates a decrease of the normal spectral emissivity of niobium (at 633 nm) with temperature from approximately 1100 K to the range of 1800–2000 K, followed by a moderate increase with temperature up to the melting point. The data are presented up to 2700 K; the convergence toward a common value at the melting point (2742 K) is a consequence of the calibration procedure described previously. This is probably due to the evaporation of some surface oxide layer during heating and is consistent with similar results obtained at IMG C at 900 nm^[4]. After reaching a minimum, the normal spectral emissivity increases with temperature up to the melting point.

The most likely explanation of this phenomenon is that probably a very thin layer of oxide is presenting at the surface of the specimen kept in the experimental chamber. When the specimen is pulse heated, the oxide layer evaporates. The trend above 1800 K for the experiment represents the normal spectral emissivity versus temperature for a clean niobium surface. During cooling the trend is reversed and emissivity follows part of the heating curve. The formation of the thin oxide layer is not instantaneous, but it takes some time, and therefore the normal spectral emissivity will follow a different temperature trend with respect to the heating period. It is also clearly seen that the emissivity when the specimen cools freely back to room temperature is the same as that when the experiment starts. This result demonstrates that a stable specimen surface has been reached.

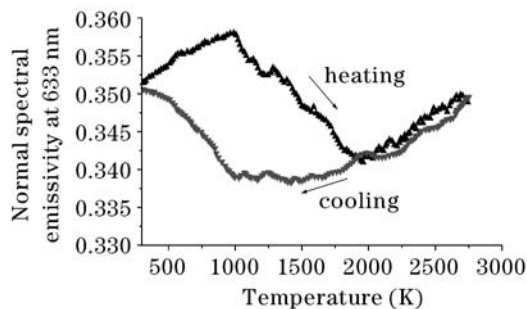


Fig. 3. Experimental results for the normal spectral emissivity at 633 nm as a function of temperature for a niobium strip specimen.

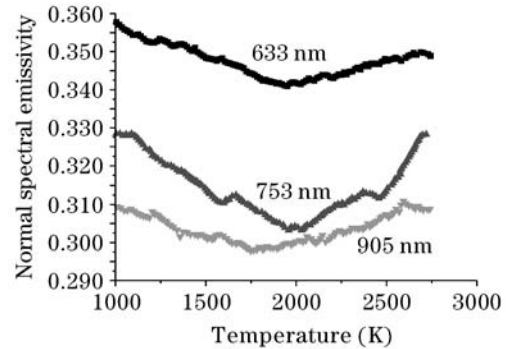


Fig. 4. Experimental results for the normal spectral emissivity of niobium at 633, 753, and 905 nm as a function of temperature during the heating period.

The specimen is fully stabilized and no further changes of emissivity take place. In relation to the experimental results described previously, other different wavelength normal spectral emissivity curves are computed and used directly in the related experiments. The result of the normal spectral emissivity at 753, 827, and 905 nm can be obtained using the true temperature of the specimen and the spectral temperature at 753, 827, and 905 nm of niobium strip in the experiment. The typical shape of the normal spectral emissivity at 633, 753, and 905 nm versus temperature during the heating period is presented in Fig. 4. Thus the multi-wavelength emissivities are obtained by dynamic measurement.

The estimate of uncertainties (two-standard deviation level) is based on uncertainties in measured quantities, operational conditions, and calibrations.

In reflectivity techniques, uncertainties in temperature measurements cause only errors in the temperature associated with the reflectivity (emissivity) value. Considering the very weak dependence of emissivity on temperature, the uncertainty in temperature measurements has negligible effects on the uncertainty in emissivity. Long-standing primary calibrations of the effective wavelength and of the linearity of the pyrometer detector are used in these experiments. The radiance temperature at the melting point of niobium^[3] is used as a reference temperature; therefore, the melting plateau in each experiment provides the exact value for obtaining correct radiance temperature measurements, including the transmission of all-optical components in the pyrometer target path. All temperatures reported in this paper are based on the International Temperature Scale of 1990 (ITS-90)^[4].

Uncertainties in reflectivity (emissivity) measurements depend on uncertainties in measured signals, the stability of operational conditions, and the uncertainty of calibration point. Lock-in signals are measured with a 16-bit data acquisition system that is calibrated periodically. Electronic noise, remaining uncertainties after the calibration, and possible drifts between calibrations contribute a 0.1% uncertainty to signal. The total uncertainty in the niobium and BaSO₄ signals is therefore 0.2%. Measurements of the stability of the laser diode in the time necessary for completion contribute an uncertainty of 0.5%. Possible variations among different BaSO₄ specimens and remaining uncertainties after the BaSO₄ reference measurements contribute another 1% uncertainty to the emissivity. All the different effects re-

lated to emissivity calibration at the reflected signal are taken into account by the emissivity calibration at the melting point of niobium. The uncertainties of this emissivity value depend on uncertainties associated with the melting point of niobium (10 K), the radiance temperature at the melting point (8 K), and the wavelength (2 nm). The wavelength uncertainty depends on a possible mismatch among the laser diode wavelength (center, 633 nm; bandwidth, 1.2 nm), the sphere detector wavelength (center, 633 nm; bandwidth, 10 nm), and the pyrometer wavelengths (centers, 633, 753, 827, and 905 nm; bandwidths, 78, 56, 53, 41 nm). The uncertainty in the emissivity value at the melting point of niobium is 4.3% and represents the largest contribution to the total uncertainty. It may be concluded that the total uncertainty in these emissivity measurements is not more than 5%.

In conclusion, experimental results of the normal spectral emissivity at three wavelengths of the niobium strip specimen in the temperature range from room temperature to the melting point have been presented. The measurements were performed with a new pulse-heating technique developed at HIT, which is a multi-wavelength pyrometry with a high-speed version of integrating sphere reflectometer. The thermophysical property results validate these normal spectral emissivity measurements because thermophysical properties of strip specimens closely repeat values obtained on tubular specimens with a blackbody. The dependence of normal spectral emissivity on surface conditions has been confirmed, indicating the presence of a thin oxide layer that evaporates during the heating phase of the experiment but is formed again during cooling at lower temperatures. Consequently the

normal spectral emissivity of the niobium strip does not follow the same trend during heating and cooling. Additional experiments on other materials are necessary to verify whether this behavior is specific to niobium or whether it reflects a general pattern in-pulse heating experiments.

The work was supported by the National Natural Science Foundation of China (No. 60377037) and the National Fund of Author of Most Excellent Doctoral Dissertation (No. 199929). The authors gratefully acknowledge Dr. F. Righini, the Istituto di Metrologia "G. Colonnetti" (IMGC), Italy, for the useful suggestions. P. Xiao is the author to whom the correspondence should be addressed, his e-mail address is rocshore@hit.edu.cn.

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