Indian Journal of Pure & Applied Physics
Vol. 55, December 2017, pp. 857-863

Investigation of fixed point of copper in the metal-in-graphite blackbody cavity using standard photoelectric linear pyrometer

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Received 28 December 2016; accepted 4 July 2017

The phase transition temperature of high purity copper has been used as fixed point temperature to calibrate standard thermometers of both contact as well as non-contact type. In the present work the freezing point of copper has been measured by a spectral radiation pyrometer using copper-in-graphite black body crucible fabricated in the form of a cavity with an aperture of 10 mm. A recently procured detector based photoelectric pyrometer, LP4 has been used as transfer standard in the measurement process. The output signal of photocurrent due to radiation from the source of copper blackbody cavity at wavelength of 650 nm in vacuum has been measured during the period when metal changes its phase from solid to liquid and liquid to solid. The stability of fixed point has been determined from the melting and freezing curves plotted between the photocurrent or corresponding temperature and the time period of the whole cycle. The uncertainty in the measurement of copper point has been evaluated to be ± 0.530 ºC and thus the copper freezing point has been determined to be 1083.41 ºC (± 0.53 ºC, at *k* =2) differing from the assigned value in the international temperature scale of 1990 (ITS-90) by 1.21 ºC lower while the melting point of copper has been determined to be 1084.23 °C differing by 0.39 °C lower respectively than the defined value of ITS-90.

Keywords: Fixed point, Photoelectric radiation pyrometer, Blackbody source, ITS-90, Photocurrent, Melting and freezing plateaus, Heat pipe, Uncertainty

1 Introduction

Above the freezing point of silver, the temperature on the international temperature scale of 1990 (ITS-90) is defined in terms of the radiance ratio of source of interest to that of any one of the three fixed points, i.e., silver, gold or copper freezing points[1-4]. ITS-90 assigns agreed temperatures to the phase transition (such as melting or freezing) of pure materials, which can then be used to calibrate standard thermometer or pyrometer. The freezing point is depressed slightly by presence of impurities; therefore the use of materials with sub-ppm impurity levels is essential. Fixed point set-ups are used in calibration laboratories for realizing the melting or freezing temperature of fixed point materials. These set-ups consist of a furnace and standard fixed point blackbody cavity with a several cubic centimeters of highly pure metal. A well-known phase transition temperature arising inside the cavity during melting and freezing is used to calibrate standard tungsten lamps with variable intensity of radiation by estimating the true value of temperature. In practice the fixed point blackbody radiator suffers from practical limitations. Realization of fixed points for radiation thermometry is a complex time-consuming process and the graphite parts are to be replaced often, even though the cavities are heated in an inert gas environment using argon gas over pressure. In addition, the cavity apertures are relatively small in order to realize a high effective emissivity. Consequently, an excellent size-of-source characteristics and relatively small measurement spot is required for a primary radiation pyrometer. In order to overcome these practical limitations, the authors used a sodium heat-pipe blackbody source to realize the ITS-90 above 900 ºC and to realize copper point. For most defined metal fixed points in the ITS-90, the freezing plateaus are used because of the best stability and reproducibility compared to their melting plateaus[5-12]. Although the temperature uncertainty of melting plateau is higher than that of freezing plateau but operation and realization of melting are easier than freezing, primarily because of super-cooling and induction of nucleation during realization of the freezing point. Realization of melting plateaus avoids these problems and the duration is also longer than that of the freezing plateau. In the freezing process,

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the performance of plateau is influenced by the purity of metal and current operation procedure but not prior freezing and melting history while in contrast, the performance of melting plateau is influenced by the prior freezing history. As a consequence the melting plateaus are not used in the ITS-90 for most metal fixed points. In this paper comparison of melting and freezing plateaus of copper point were carried out and discussed. The fixed point of copper was measured and uncertainty was evaluated considering the most affecting components which are contributing towards the limit of accuracy in determining the fixed point.

2 Mathematical Equations

The thermodynamic temperature of copper using blackbody is found from the spectral responsivity and Planck's radiation law by use of:

$$i_c = \int S_L . L(\lambda, T) d\lambda \quad \text{... (1)}$$

where i_c is the measured photocurrent, S_L is the absolute spectral responsivity and $L(\lambda,T)$ is the spectral radiance.

$$L(\lambda,T) = (c_1/n^2\lambda^5)[\exp(c_2/n\lambda T)- 1]^{-1} \quad \text{... (2)}$$

where c_1 and c_2 are first and second radiation constants, T is thermodynamic temperature of Cu, λ is the wavelength of radiation in air and n is the refractive index. For these calculations, the CODATA values[13] for the radiation constants and the refractive index of air n=1.00029 were utilized. Any unknown temperature can be evaluated if photocurrents are determined using the Eq. (2) above. Thus taking the ratio:

$$L(\lambda,T)/L(\lambda,T_{Cu})=[\exp(c_2/\lambda T_{Cu})-1]/[\exp(c_2/\lambda T)-1] = I_{ph}(T)/I_{ph}(T_{Cu}) \quad \text{... (3)}$$

3 Equipment Section

3.1 Fixed-point blackbody cavity

Figure 1 shows the physical structure diagram of the copper in blackbody cavity. The graphite crucible is held in a stainless steel tube with an argon purge at the front. The stainless steel tube is placed in a sodium heat pipe line constructed from Inconel 601 for temperature uniformity and is sealed except for front opening. The cell having 120 mm long and 45 mm in outer diameter, having centralized cavity aperture of 10 mm and a length of 50 mm immersed in the metal. In the present work we have used copper-in-graphite blackbody cavity commercially procured from Isothermal Technology, UK. The blackbody comprises of high purity copper filled in the graphite cell of significant purity. The cell inserts in the stainless steel cylindrical enclosure provided with a tube for purging of argon gas. The assembly is inserted inside the high temperature heat pipe blackbody heating source exactly at the middle position.

3.2 High temperature blackbody source

A horizontal blackbody heating furnace having a cylindrical sodium heat pipe lining assembly, automatically controlled with eurotherm make built-in-controller was used to provide stable temperature covering a range from 450 ºC to 1200 ºC. The source stability was measured by a Type-S thermocouple at a temperature near at 1085 ºC to use it for measurement of copper point. The stabilization curve is as shown in Fig. 2. The stability of the source was evaluated to be of the order of ±60 mK in the above range which makes it suitable source for realization of these fixed points by radiation technique. Figure 3 shows the heat-pipe high temperature blackbody source of radiation temperature.

Fig. 1 — Structure of copper point blackbody cavity.

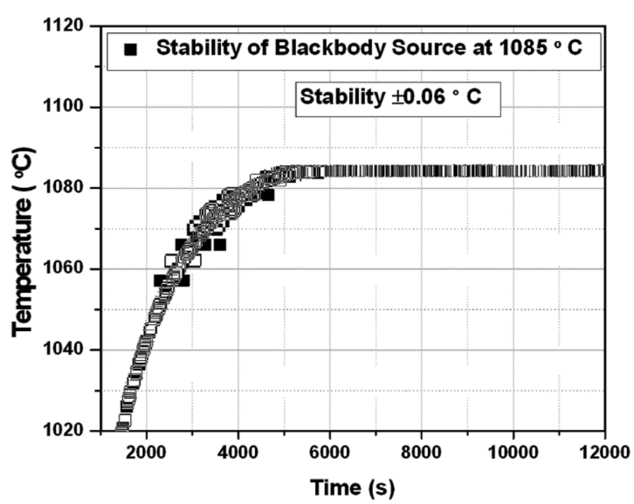

Fig. 2 — Stability of blackbody source at 1085 ºC measured by Type-S thermocouple.

3.3 Description of photoelectric spectral pyrometer

The internal structure of the photoelectric spectral linear pyrometer, LP4 is shown in Fig. 3. It has been procured from M/s K E Technologies, Germany and calibrated at Physikalisch-Technische Bundesanstalt (PTB), Germany. The pyrometer is a unique instrument used as transfer standard for measurement of radiation temperature of a black body or non-blackbody sources like metal-in-graphite cavity or a tungsten strip filament lamp. The photoelectric pyrometer has specific feature that the photocurrent generated by the photo-detector varies proportionally linear with the temperature of the source. It is used with a high precision silicon photocell detector for measuring photocurrent. It is used with a high precision silicon photocell detector for measurement of photocurrent. It has an especial vacuum photocell with a guard ring and a homogenous cathode as receiver in connection with high standard electronics and is equipped with interference filters of suitable wavelength 650 nm and 970 nm, respectively. The instrument has a nominal spot size of 0.25 mm (called measurement field stop) at a focal distance 600 mm. The image of spot aperture is reflected over the plane mirror placed at an angle of 45º in parallel position to view it by an eyepiece using an optical alignment. The image of aperture (as black spot) of the pyrometer is focused over the image of the exact centre of the cavity aperture of a metal fixed point black body cavity. This confirms the radiation exactly falling over the detector surface of the pyrometer after making all required alignments between the radiating source and the pyrometer[14]. The optical system along with detector is a vacuum sealed chamber whose temperature is controlled and stabilized during measurement of photocurrent. The pyrometer has its capability of measuring photo-current in the range from 1×10^{-12} A to 1×10^{-8} A and temperature in the range from 500 K to 3500 K. The pyrometer has facility for displaying the chamber temperature, photocurrent and the temperature in the scales of ºC and K. The pyrometer, LP4 can be directly hooked to computer through RS232 cable so that the temperature of the source, the photocurrent and the time variation curve of temperature can be displayed and recorded simultaneously.

3.4 Argon gas flow system

The fixed point cell of copper is a high purity metal-in-graphite cavity. The cavity was fabricated from high purity graphite rod. When graphite is exposed to high temperature, it starts oxidizing in the presence of air. It is therefore, required to make arrangement of pure dry argon gas to be purged all around the fixed point cell in order to avoid oxidation of the graphite material. The pressure from the gas cylinder is so adjusted by using a flow meter to maintain the flow rate of argon gas to in-circulate it all around the cavity and at the same time does not affect the melting or freezing plateaus of the copper. The outer cylinder of stainless steel holding the graphite cell has a cover fixed with a tube that is connected to an argon gas cylinder through a calibrated flow meter. Figure 4 shows the experimental setup for measurement of copper point using blackbody source and spectral radiation pyrometer LP4.

4 Measurement Techniques

The fixed point of copper has been realized by measuring melting and freezing plateaus by heating the metal-in-graphite blackbody cavity in the high temperature heat pipe blackbody source. The heating source comprised of an automatically controlled

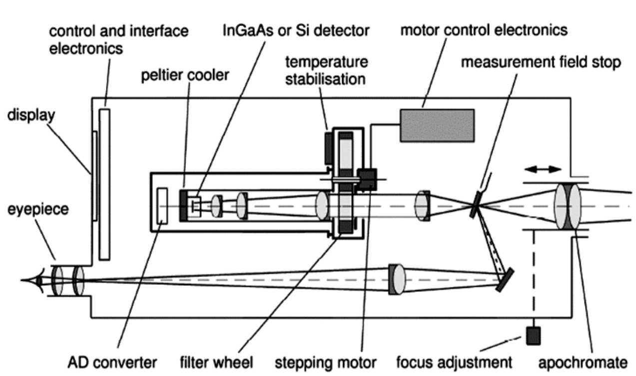

Fig. 3 — Internal structure of photoelectric spectral linear pyrometer, LP4.

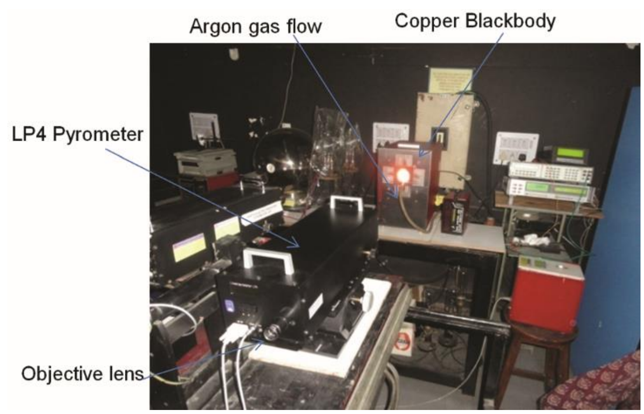

Fig. 4 — Experimental set-up for realization of Cu-point using blackbody.

sodium heat pipe cylindrical tube. The cell is inserted in the heating source and placed at the central location for uniform heating. The cell is set to heat initially at 600 ºC and the pyrometer, LP4 is set to focus the middle point of the cell and centrally align it by using laser lamp. The source temperature is then increased at every 100 ºC range till the temperature reaches to 10 ºC higher than the melting point of copper, i.e., at 1094 ºC. The melting of sample starts and the temperature is stabilized. The pyrometer is hooked to computer and the display of temperature, corresponding photocurrent generated due to radiation from the cell and the plot between time and temperature can be shown in the computer screen. The data on time variation of temperature in both scales (ºC, K), and with photocurrent can be stored in a file in order to analyze later. The radiation from the fixed point cell is measured at different locations axially in order to study the profiles for uniformity and stability of temperature.

5 Measurement of Melting and Freezing Plateaus

Copper point temperature is one of the reference temperatures for realizing international temperature scale 1990 (ITS-90). The work has been carried out on realization of point by using metal-in graphite crucible in the form of a blackbody cell. The complete equipment set-up comprises of a radiation pyrometer, LP4, high stability heat pipe black body source, a 8-1/2 digit digital voltmeter (DVM), a type-S thermocouple to monitor the cell temperature, a high current rating (0-40 A) DC power-supply and the argon flow arrangement to inflow a required purging of argon gas inside the metal cavity in order to avoid oxidation of copper metal. Although melting and freezing points of copper have been realized but freezing is more preferred because of natural control of temperature for better precision achieved during the measurement. In the work it was carried out to measure the points using commercially made crucible as well as by self-designed and fabricated blackbody cavities and the results were compared. The temperature profile of the furnace used was measured in order to know the influence of temperature on the performance of melting and freezing plateau. Figure 8 shows the temperature profile of the source before realizing the copper point. The temperature difference between melting and freezing was investigated and the stability of copper point was measured with a precision of ±0.03 ºC using the source having stability within ±0.06 ºC. The high precision in copper point was possible because of uniform temperature obtained around the metal-in-graphite cavity. The controlled flow of argon across the blackbody cavity above 400 °C significantly reduced the oxidation of graphite.

6 Results and Discussion

Copper point is the reference temperature for realizing ITS-90 using contact and non-contact measurement techniques. We have made effort to measure the melting and freezing states of the metal in graphite blackbody cavity using high precision spectral radiation pyrometer in terms of stabilized photocurrent under the condition of monochromatic radiation of wavelength 650 nm in vacuum. Figures 5 to 7 show the melting and freezing plateaus of copper, both in time variation of temperature and photocurrent. Figure 5 shows the melting state in terms of time versus photocurrent signals while Fig. 6(a,b) shows the melting of Cu along with precision of best curve measured. Figure 7(a,b) shows the freezing state along with precision of best curve measured for the freezing point. A precision of ±0.03 ºC has been achieved for the best of repeated measurement plots at the freezing state of copper for a period of about 33 min while for freezing the state of stabilization has been studied for a period of about 80 min. In order to ascertain the best stability measurement, data on temperature variation due to change in distance of the pyrometer detector from the bottom of the cavity has been collected. Figure 8 shows the signal variation with location of pyrometer with respect to the radiation source during the freezing of copper metal. A stability of ±0.01 ºC has been achieved along a distance of 120 mm in the profile which is a significant stability under the

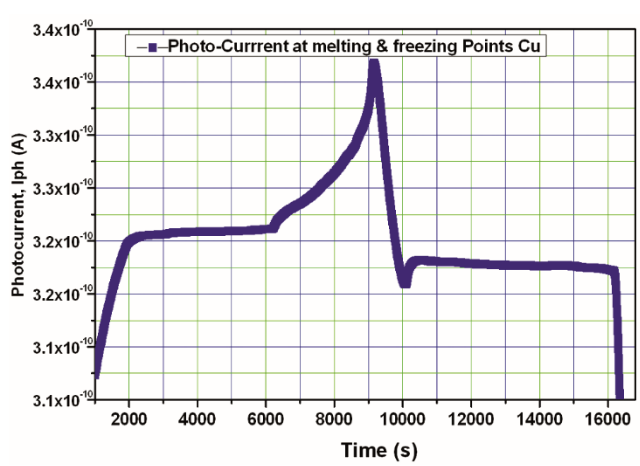

Fig. 5 — Melting and freezing plateaus of Cu in terms of time variation of photocurrent.

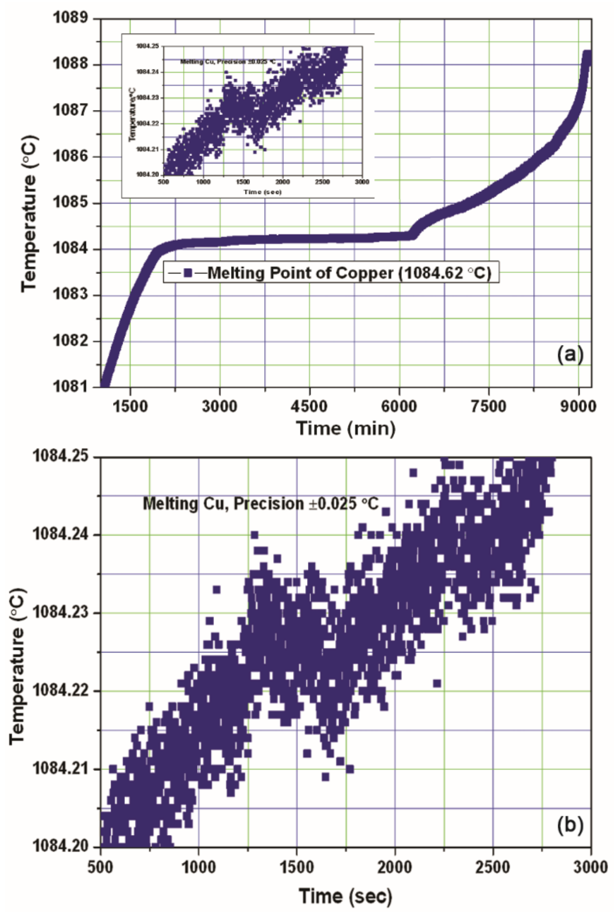

Fig. 6 — (a) Melting plateau of copper and (b) melting plateau of Cu-point with its precision of realization.

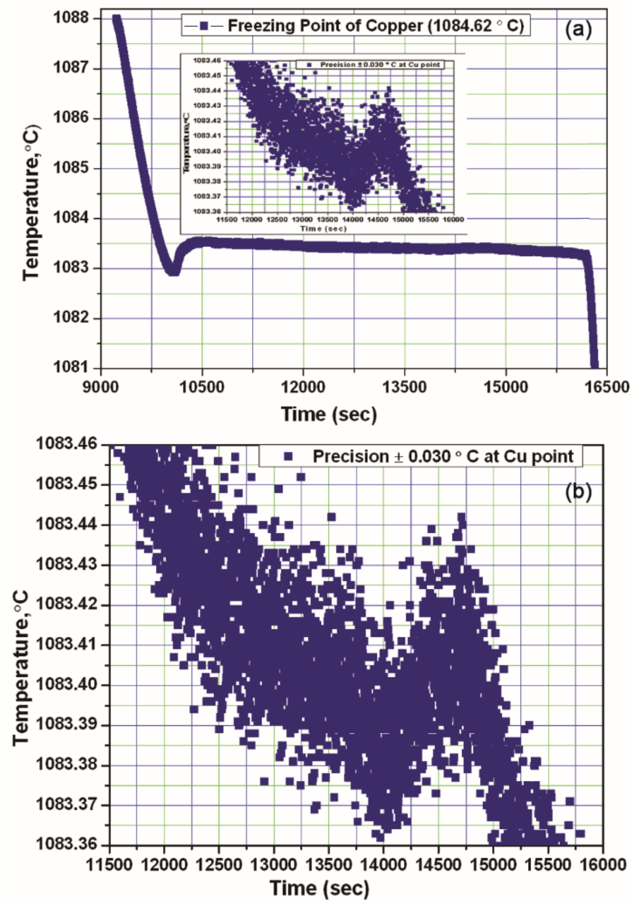

Fig. 7 — (a) Freezing plateau of copper and (b) freezing plateau of Cu with its precision of realization.

experimental limitation. The mean value of photocurrent measured at the copper point realization for melting state is determined to be 3.250×10^{-10} A while the same as determined at freezing state realization is to be 3.225×10^{-10} A. The I_{ph} at freezing is measured to be slightly lower than at melting state because of impurity contents in the metal which lowers the state under practical condition of realization. The melting and freezing states are affected by the impurity contents present in the metal, though these may be present at the ppm level. Due to the impurities or deviation caused due to several runs of measurements performed earlier with the metal, a significant difference is observed between the melting and freezing points.

The uncertainty of measurement of copper freezing point has been evaluated by taking a significant number components contributing towards the uncertainty associated with the black body crucible, spectral responsivity, size of source effect and the output signal of the pyrometer detector. The standard uncertainty of copper point measurement calculated from different sources is evaluated to be ± 0.530 K (at k=2) and thus the copper freezing point is determined to be 1083.41 ºC (± 0.53 ºC, at k =2) differing from the assigned value in the international temperature scale of 1990 (ITS-90) by 1.21 ºC lower while the melting point of copper was determined to be 1084.23 °C differing by 0.39 °C lower, respectively, than the defined value of ITS-90. The uncertainty budget is shown in Table 1.

7 Evaluation of Uncertainty in the Measurement

The uncertainty in the measurement of each radiance temperature determined at freezing of silver source has been estimated by using two different methods namely the Type-A, where the uncertainty component has been evaluated using statistical analysis of a series of repeated observations taken at stable freezing temperature and the other by Type-B estimation, where the uncertainty components have been determined arising from different

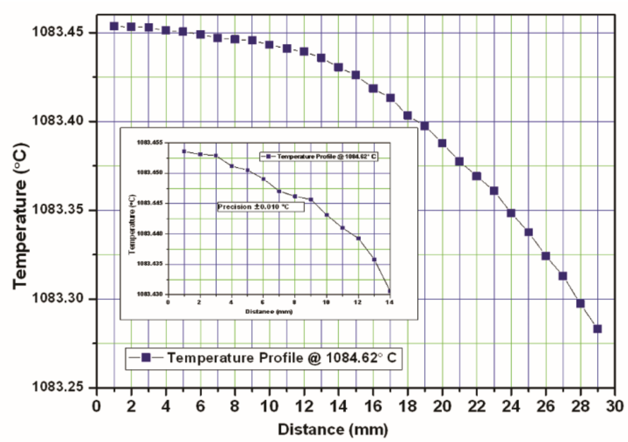

Fig. 8 — Temperature profiles of the blackbody source at copper point.

Table 1 — Uncertainty components for ITS-90 realization based on copper point blackbody realization using a heat pipe blackbody radiator.

S. No.	Source of uncertainty	Type	Standard uncertainty At Cu freezing point (K)
1.	Impurities	B	0.008
2.	Emissivity	B	0.005
3.	Plateau identification	B	0.020
4.	Immersion effect	B	0.060
5.	Repeatability	A	0.030
6.	Uncertainty due to pyrometer stability	B	0.250
7.	Uncertainty due to spectral responsivity due to cell	B	0.015
8.	Size of Source Effect	B	0.020
9.	Drift of signal in the measurement	B	0.026
10.	Nonlinearity	B	0.028
	Combined uncertainty at k=1		0.2637
	Expanded uncertainty of measurement at k=2		0.53

sources contributing to the measurement[15-20]. The mathematical model for uncertainty evaluation is defined as following:

$T_m = T_F \pm U(\Delta T)$

$U(\Delta T) = k.\ u_c$

$$u_c = \sqrt{\left[\sum_{i=1}^{9} u^2 i\left(\delta Ti\right)\right]} \quad \ldots (4)$$

The uncertainty components designated from u_1 to u_9 are evaluated by Type-A or Type-B methods depending upon the source of uncertainty. Here T_m is measured temperature, T_F is fixed point temperature as defined in ITS-90 scale, $U(\Delta T)$ *is* expanded uncertainty of measurement, k is coverage factor, u_c is combined uncertainty and $u_i(\delta T_i)$ is uncertainty components arising from various sources as described below (u_1 to u_{10}).

u_1 = uncertainty due to impurity content of the copper point cell

u_2 = uncertainty due to emissivity at the copper point blackbody

u_3 = uncertainty due to identification of the best plateau measured

u_4 = uncertainty due to immersion effect over the cell

u_5 = uncertainty due to repeatability of data measured

u_6 = uncertainty due to stability of spectral pyrometer

u_7 = uncertainty due to spectral responsivity due to cell

u_8 = uncertainty due to size of source effect

u_9 = uncertainty due to drift of signal in measurement

u_{10} = uncertainty due to non-linearity of output signal

The uncertainty budget is as shown in Table 1, describing the significant contributions of uncertainty in the measurement of copper point. The combined standard uncertainty and finally the expanded uncertainty of measurement at a coverage factor, k=2, for a level of confidence approximately 95% was evaluated.

8 Conclusions

The facility at CSIR-NPL, India has been created to establish primary standards of temperature by radiation pyrometry as per definition of ITS-90. The copper point is one of the three fixed points on ITS-90 to measure it by a non-contact method (by spectral pyrometer) to ascertain the linearity, uniformity and continuity of the temperature scale. The practical use of sodium heat-pipe radiation source is far more convenient to bring the fixed point to its freeze. Experimental results show that the sodium heat pipe radiation source could provide melting and freezing plateaus of longer duration because of the minimized gradients caused along the fixed point cavity. The uncertainty could be reduced due to long stable plateau at copper point. The results in this study leads to the conclusion that in the context of realizing the fixed point, it provides a reference temperature for calibrating the spectral pyrometers and high stability tungsten strip lamps using the Planck's equation. A calibrated lamp works as a temperature radiation source and could be employed as primary standards of temperature for calibrating optical pyrometers.

Acknowledgement

The authors express their gratitude to the Director of the CSIR-National Physical Laboratory, New Delhi for his continuous encouragement and support. The authors also express their thanks to Dr D D Shivagan for fruitful discussions and Mr R B Sibal, M K Gandhi and Hans Raj for the technical assistance while carrying out the experimental work. One of the authors AR is thankful to the University Grant Commission for grant of RGNF-Fellowship to support her research work at the CSIR-NPL, New Delhi.

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