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The Spectral Emissivity and Total Emissivity of Beryllium Oxide*

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A method has been devised for the measurement of spectral and total emissivities of non-metallic surfaces at high temperatures. The maximum temperature for spectral emissivity measurements is about 1700°C. With an extrapolation of spectral emissivity values it is possible to obtain experimental values for total emissivities at even higher temperatures. This method has been applied in the measurement of the emissivities of polished surfaces of three types of commercially prepared beryllia. Ceramic firing of hot-pressed beryllia was found to have very little, if any, effect on the spectral or total emissivity of a surface formed by polishing the final material. The impurity present in dark samples and/or the larger crystal size increases both the spectral and total emissivity by a factor of about two.

I. INTRODUCTION

VARIOUS methods[1] have been used to measure the total emissivity of non-metallic surfaces. These methods are limited either to a relatively low temperature range of measurement or to the use of surfaces prepared in a manner required by the experimental method. The latter limitation is very restricting since the emissivities of substances are influenced greatly by the nature of the surface. The method used in this investigation permits the use of surfaces prepared by any method desired. In this investigation measurements were made up to 1900°C.

Several investigators[2] have developed or suggested methods for the measurement of the spectral emissivity of non-metallic surfaces. Liebmann[3] discussed the various factors involved in obtaining reliable measurements and determined the spectral emissivity of a sintered deposit of powdered beryllia. The method reported in this paper was an adaptation of the methods of Skaupy[2] and of Burgess and Waltenberg.[2] Values thus obtained were checked at the melting point of gold by a procedure similar to that of Wiegand[2] and at the melting point of potassium sulfate by a method dependent upon the discontinuity in the transmissivity of potassium sulfate at its melting point.

II. EXPERIMENTAL METHOD

Beryllia Samples

Sample disks, 1.9 cm diameter×0.3 cm, were cut from blocks of fabricated beryllia. The faces were ground parallel and polished on No. 500 carborundum paper. Measurements were made with three different types of fabricated beryllium oxide. These types were prepared by the Norton Company.

Type 1 was fabricated by hot-pressing high fired beryllia powder in graphite forms. This type was uniformly colored black throughout the entire fabricated piece. The black coloration remained after the sample disks were heated in air. The density was 2.85 g cm^{-3}.

Type 2 was fabricated by hot-pressing Brush S.P. powdered beryllia. This type was white throughout the fabricated piece except for occasional small grey or black regions. There appeared to be a small hole at the center of many of the dark regions. This discoloration also remained after the sample disks were heated in air. The density was 2.844 g cm^{-3}.

Type 3 was fabricated the same as Type 2 except that the fabricated blocks were annealed by ceramic firing after being formed by the hot-pressing method. The specimen used in these measurements had a slight grey color but turned white when heated in air. The density was 2.778 g cm^{-3}.

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[1] C. P. Randolph and M. J. Overholzer, Phys. Rev. **2**, 144 (1913); G. Liebmann, Zeits. f. Physik **71**, 416 (1931); P. F. McDermott, Rev. Sci. Inst. **8**, 185 (1937).

[2] G. K. Burgess and R. G. Waltenberg, Bull. Bur. Stand. **11**, 591 (1915); F. Skaupy, Zeits. f. Physik **12**, 177 (1922); E. Wiegand, Zeits. f. Physik **30**, 40 (1924); W. E. Forsythe, Phys. Rev. **25**, 252 (1925); G. Herrman, Zeits. f. Physik. Chemie **35**, **Abt B4**, 298 (1937).

[3] G. Liebmann, Zeits. f. Physik **63**, 404 (1930).

Apparatus

The apparatus used to obtain radiation data for the calculation of total emissivities is shown in Fig. 1. A heated beryllia surface was obtained free of temperature gradient by use of the tungsten-tantalum heating block, *B* and *C*, and the beryllia shield and diaphragm, *I* and *J*. The metal block was heated by induction. The tungsten disk, *B*, had to be placed between the tantalum block, *C*, and beryllia sample, *A*, because of the chemical reaction between beryllia and tantalum, forming volatile tantalum oxide and beryllium. The radiation shield, *I*, and diaphragm, *J*, were fabricated from Type 1 beryllia.

In order to eliminate undesirable reflections the inner wall of the quartz tube was coated with Aquadag colloidal graphite from the edge of the optical quartz window, *O*, down to and including the upper surface of the constriction in the quartz wall. The shutter, *P*, protected the quartz window, *O*, from volatilized deposits at all times that measurements were not being made. The transmissivities of the prisms *M* and *T* and windows *N* and *O* remained constant throughout the investigation.

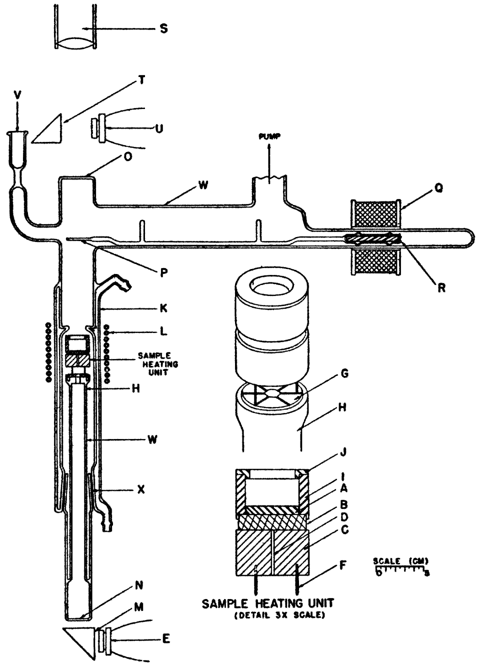

FIG. 1. Apparatus for heating of samples in vacuum. *A*, sample disk; *B*, tungsten heating disk; *C*, tantalum heating block; *D*, "black body" hole; *E* and *U*, calibrated Leeds and Northrup optical pyrometers; *F*, tungsten legs; *G*, grooved beryllia table; *H*, quartz support; *I*, radiation shield; *J*, diaphragm; *K*, water cooled quartz jacket; *L*, induction heating coil; *M* and *T*, silvered prisms; *N*, optical Pyrex window; *O*, optical quartz window; *P*, shutter; *Q*, solenoid; *R*, iron rod sealed in Pyrex; *S*, total radiation pyrometer; *V*, quartz supporting tube; *W*, quartz-Pyrex graded seal; *X*, 34/45 standard taper joint.

The Leeds and Northrup Rayotube total radiation pyrometer, *S*, was equipped with a quartz lens. When the pyrometer was focused on a surface the thermopile in it intercepted radiation from an area 0.6 cm in diameter. The Rayotube was securely mounted in a special holder which permitted adjustment of the vertical distance from specimen to Rayotube but eliminated all lateral motion. The silvered prism *T* was mounted in a special holder which permitted the prism to be moved into place for use with pyrometer *U* and moved out of the way when measurements were made with pyrometer *S*.

Joint *X* was sealed with Apiezon "*W*." To insure reproducibility in the assembly of the apparatus a reference mark was placed on the lower Pyrex tube and a sturdy metal pointer was mounted on the vacuum frame in coincidence with the reference mark.

Mercury vapor pumps with liquid nitrogen traps maintained a pressure of 10^{-6} to 10^{-5} mm of Hg while the heating unit and sample were at the maximum temperature obtainable.

The oscillator used with coil *L* was supplied by two parallel constant voltage transformers. This combination permitted maintaining a very constant temperature of the sample heating unit.

The apparatus used to measure the spectral emissivity of the beryllia surfaces is shown in section in Fig. 2. The beryllia sample disk was placed on a platinum ribbon $1\times6\times0.001$ inches. This ribbon was heated electrically by the output of a filament transformer supplied by a variable transformer. To maintain a constant ribbon temperature the variable transformer was supplied by a constant voltage transformer. A maximum current of 76 amp. was passed through the platinum ribbon.

The beryllia flats shown in Fig. 2 furnished a smooth supporting surface and prevented the platinum ribbon from buckling away from the supporting surface and producing hot spots.

Thus the center of the platinum ribbon could be heated to just slightly below the melting point of platinum without danger of the ribbon melting at any point. The top beryllia flats also served as a guard ring about the sample disk and helped eliminate temperature gradients in the surface of the disk.

Undesirable convection currents were eliminated by use of an insulating firebrick chimney which was shaped as shown in Fig. 2. The inner surface of the chimney was coated with Aquadag colloidal graphite. During a series of measurements the graphite was burned from no more than the lower half inch of the chimney.

Calibration of Rayotube

The Rayotube was calibrated *in situ* by use of the tungsten disk, B, as a known radiating surface. The top surface of the tungsten was polished with 4/0 emery paper. The tungsten was heated in vacuum to approximately 2200°C for several hours to outgas the tungsten and age its surface. The tungsten disk was then heated to constant temperature at about 900°C. The true temperature of the tungsten disk was measured with the calibrated pyrometer E (Fig. 1) and the brightness temperature of the top surface was measured with the calibrated pyrometer U. The voltage of the Rayotube was measured with a Leeds and Northrup precision potentiometer. This procedure was repeated at approximately 50-degree intervals in the range 900°C to 2000°C. The measurements were reproducible and the equilibrium potentials of the Rayotube remained constant within 0.03 percent over a period several times as long as required to make all the readings at each temperature. After proper corrections were made for the transmissivity of prisms and windows the readings of pyrometers E and U agreed very well with the spectral emissivity data of Worthing.[4]

The total radiant energy (in cal. cm^{-2} sec.$^{-1}$) was calculated at each temperature by the use of Worthing's[4] values for the total emissivity of tungsten. With this data a calibration curve was drawn for the Rayotube under the conditions of the experiment. This procedure did not completely correct for the absorption of radiant energy by the quartz window and lens since the spectral distribution of radiant energy is different for tungsten[5] and beryllia[3] surfaces. Nor does this method allow for possible variation between the two surfaces in the ratio of total hemispherical emissivity and total normal emissivity.

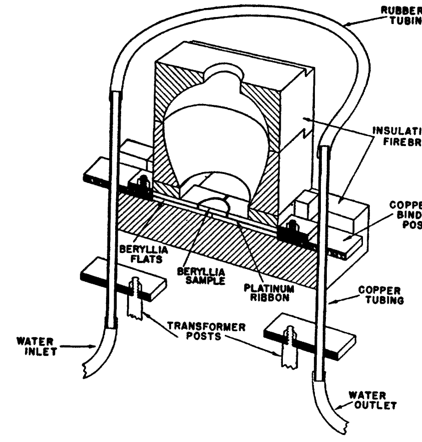

FIG. 2. Apparatus for resistance heating in air (section).

Determination of Total Emissivity

After an initial heating in air to about 1300°C each beryllia sample disk was placed in the apparatus shown in Fig. 1 for preliminary heat treatment in vacuum. It was heated to the maximum temperature obtainable for several hours in order to outgas the sample and volatilize any impurities remaining from the polishing of the upper surface. The Rayotube was adjusted to focus it on the center of the beryllia surface. After the heat treatment the heating unit was brought to a constant temperature; the radiant energy was determined by measurement of the equilibrium potential difference of the Rayotube; and the brightness temperature of the beryllia surface was determined with pyrometer U. These measurements were repeated at different temperatures and the energy radiated from the surface was plotted as a function of the brightness temperature of the beryllia surface. Measurements with the apparatus shown in Fig. 2 later gave the relation between brightness temperature and true temperature for the same beryllia surface. The total emissivity, ϵ_t, was

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[4] *International Critical Tables* (McGraw-Hill Book Company, Inc., New York, 1929), Vol. 5, pp. 243 and 245.

[5] L. S. Ornstein, Physica **3**, 561 (1936).

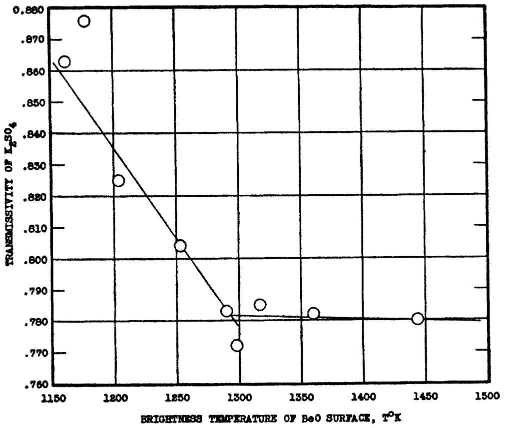

FIG. 3. Transmissivity curve for potassium sulfate layer on beryllia surface.

calculated at various temperatures by the relation

$$\epsilon_t = W/\sigma T^4 \qquad (1)$$

where W is the total radiant energy in cal. cm^{-2} sec.$^{-1}$ at the true surface temperature T°K, and σ is the Stefan-Boltzmann radiation constant (1.379×10^{-12} cal. cm^{-2} sec.$^{-1}$ deg.$^{-4}$).

Determination of Spectral Emissivity

Platinum was vaporized on one-half of the upper beryllia surface to a thickness of 0.001 to 0.002 cm and polished with 4/0 emery polishing paper. The beryllia disk was then placed on the platinum ribbon of the apparatus shown in Fig. 2. The ribbon was heated to approximately 1600°C for 30 min. During this heating the upper surface was approximately 1400°C. This preliminary heating was necessary in order to obtain reproducible measurements of the relative brightness temperatures of the platinum and beryllia surfaces. With the platinum ribbon at a constant temperature, the brightness temperatures of the beryllia surface and of the adhering platinum surface were measured with the same optical pyrometer. This procedure was repeated at different surface temperatures throughout the available temperature range. The entire temperature range was covered three or four times to check the reproducibility of the measurements. There was no indication of changing emissivities during the time of these measurements.

The brightness temperature of the platinum surface was plotted as a function of the corresponding brightness temperature of the beryllia surface. Stephens[6] determined the brightness temperature of a platinum surface at different absolute temperatures. By interpolation on the above smoothed curve the corresponding brightness temperature of the beryllia surface, T_B, was obtained at each absolute temperature, T. The spectral emissivity, ϵ_λ, at temperature T was then calculated from the relation

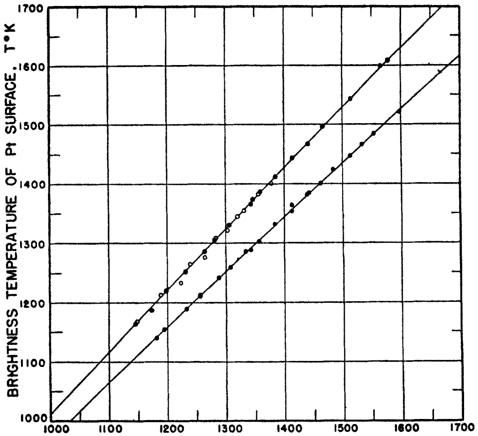

FIG. 4. Brightness temperatures of beryllia surface and adhering platinum surface. Lower line: Type 1 beryllia. Upper line: Type 2 beryllia (dark circles) and Type 3 beryllia (light circles).

$$\log_{10}\epsilon_\lambda = [c/2.303\lambda][(1/T)-(1/T_B)]. \qquad (2)$$

The effective wave-length of light, λ, transmitted by the optical pyrometers used in this investigation was taken as 0.665μ for all temperatures. Wien's radiation constant, c, is 1.432 cm °K. Therefore Eq. (2) reduces to

$$\log_{10}\epsilon_{0.665\mu} = 9352[(1/T)-(1/T_B)]. \qquad (3)$$

The spectral emissivity of each sample was checked at the melting point of gold by an independent method. The spectral emissivity of Type 1 beryllia was also checked at the melting point of potassium sulfate. (These measurements were actually made first, before the platinum was vaporized onto the beryllia surface.)

A circular piece of gold foil (10 mg in.$^{-2}$) was cut to the same diameter as the beryllia sample. A strip about 0.2 cm wide was cut from the center of this disk. The remaining two segments

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[6] R. E. Stephens, J. Opt. Soc. Am. 29, 158 (1939).

were rubbed into intimate contact with the beryllia. The segments were placed so as to leave exposed a 0.2-cm strip of clean beryllia surface. The gold foil adhered to the polished beryllia fairly well.

The beryllia surface was heated to the melting point of gold in the apparatus shown in Fig. 2. The brightness temperature of the clean beryllia surface was determined at the first appearance of holes in the gold foil. This procedure was repeated at least twice with each disk. The observed brightness temperatures of the beryllia surface at the melting point of gold seldom differed by as much as 2 deg. for a given sample. (It was found necessary to cool the sample rapidly as soon as melting of the gold was observed, and then to examine the disk under a low power microscope to verify that melting had occurred. If melting had occurred, small globules of gold adhered to the beryllia surface. If the observed holes resulted from volatilization of the gold during a long period of heating, no globules of gold were present.)

The true temperature of the beryllia surface at the instant the melting of the gold foil was observed was assumed to be the melting point of gold, 1336°K. This temperature together with the observed brightness temperature of the beryllia surface permitted the calculation of the spectral emissivity of the beryllia at 1336°K by Eq. (3).

TABLE I. Brightness temperature (T_B), spectral emissivity ($\epsilon_{0.665}$), and total emissivity (ϵ_t) of three types of fabricated beryllium oxide.

	Type 1			Types 2 and 3		Type 2	Type 3
T°K	T_B°K	$\epsilon_{0.665}$	ϵ_t	T_B°K	$\epsilon_{0.665}$	ϵ_t	ϵ_t
1200	1160.4	0.542	0.665	1104.6	0.212	0.351	0.336
1300	1253.7	0.543	0.706	1187.9	0.209	0.383	0.361
1400	1347.0	0.546	0.746	1271.0	0.210	0.405	0.392
1500	1440.4	0.552	0.785	1354.2	0.213	0.425	0.420
1600	1533.8	0.559	0.819	1437.1	0.217	0.447	0.439
1700	1627.3	0.568	0.843	1519.8	0.222	0.474	0.453
1800	1720.9	0.577	0.867	1602.4	0.228	0.499	0.463
1900	1814.7	0.587	0.894	1684.9	0.235	0.513	0.470
2000	1908.5		0.931	1767.2		0.517	0.474
2100				1849.6		0.514	0.475
2150				1890.7		0.509	0.475
Melting point of gold							
1336	$1288._5$	0.55_2		1216	0.20_4	(Type 2)	
				1217	0.20_7	(Type 3)	
Melting point of potassium sulfate							
1342	1293	0.54_4					

The determination of the spectral emissivity of Type 1 beryllia at the melting point of potassium sulfate was made in the apparatus shown in Fig. 1. A few crystals of anhydrous potassium sulfate were melted so as to cover about half of the upper surface of the beryllia disk. The disk was then heated to constant temperature and the brightness temperature of its surface measured at a point covered with potassium sulfate and at a point free of potassium sulfate. Measurements were made at the same points on the surface at various increasing temperatures. The transmissivity of the potassium sulfate layer, $\tau_{0.665}$, was calculated by the equation

$$\log_{10}\tau_{0.665} = 9352[(1/T_B) - (1/T_A)], \tag{4}$$

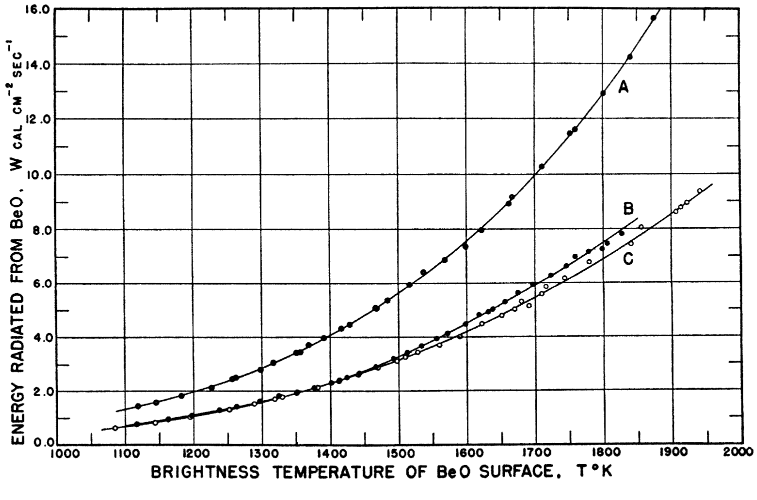

FIG. 5. Curve A: Type 1 beryllia. Curve B: Type 2 beryllia. Curve C: Type 3 beryllia.

TABLE II. Liebmann's values for the spectral emissivity of sintered 0.5μ beryllia.

T°K	1300	1400	1500	1600	1700	1800	1900	2000	2100	2200
$\epsilon_{0.665}$	0.057	0.068	0.080	0.091	0.102	0.113	0.124	0.135	0.146	0.157

where T_B is the measured brightness temperature of the clean beryllia surface and T_A is the apparent brightness temperature of the beryllia surface as viewed through the layer of potassium sulfate. The values of $\tau_{0.665}$ are shown in Fig. 3 as a function of T_B. The layer of potassium sulfate no doubt became thinner during the course of measurements due to evaporation and further creepage over the beryllia surface. This would increase the transmissivity. The observed decrease in transmissivity with increasing temperature resulted from the predominant effect of temperature. Within the experimental error of the measurements the data could be represented by two intersecting straight lines. The point of intersection (Fig. 3) was considered to lie at the melting point of potassium sulfate, 1342°K. The corresponding brightness temperature of the beryllia at the intersection, 1293°K, permitted the calculation of the spectral emissivity of the beryllia surface at 1342°K by Eq. (3).

III. RESULTS

Figure 4 shows the relation between the brightness temperatures of the beryllia surface and of the adhering platinum surface for each of the three types of beryllia used in this investigation. Within experimental error, Type 2 and Type 3 beryllia gave identical results. In Table I are given the spectral emissivities, $\epsilon_{0.665}$, calculated from the curves in Fig. 4 and Stephens'[6] values of the brightness temperature of platinum.

Figure 5 shows the relation between the energy radiated from the beryllia surfaces and the brightness temperatures as measured with the three types of beryllia. In Table I are given the total emissivities, ϵ_t, calculated from the curves in Fig. 5 by use of Eq. (1). The values given for temperatures above 1900°K involve extrapolations both of Stephens' results and of the curves in Fig. 4. The differences in the values for Type 2 and Type 3 beryllia are probably no greater than the variation which might be found between different samples of either one of the types. However, there is considerable difference between the dark Type 1 beryllia and the white Types 2 and 3, both in spectral and total emissivity.

The values of spectral emissivity determined at the melting point of gold and the melting point of potassium sulfate are also given in Table I.

The sample disk of Type 2 beryllia used for the measurements given in Table I had two small grey spots on the surface which was used for the radiation measurements. These spots were about 0.1 cm in diameter and could not be removed by heating the sample in air. The spots were near the edge of the disk and therefore did not affect the measurements of total radiation. Measurements were made of the brightness temperature of the spots each time a corresponding measurement was made on the white surface and the ratio of the spectral emissivities of grey spots and white surface was calculated. The spectral emissivity of the grey spots was approximately twice that of the white surface in the temperature range 1200°K to 2000°K. The higher spectral emissivity of the grey spots and also of the black Type 1 beryllia may be due not only to the impurity present which imparts the color to the fabricated beryllia (carbon, carbide, or suboxide), but also to the crystal size of the beryllia. When viewed with a low power microscope the beryllia crystals present in the grey spots of Type 2 appear to be considerably larger than the crystals present in the white surface adjoining the grey spot.

Other investigators have measured the spectral emissivity of beryllia. Since the beryllia surfaces used in these earlier measurements were fabricated differently than the surfaces used in the measurements reported herein, agreement of results is not to be expected.

Burgess and Waltenberg[2] report a spectral emissivity of 0.37 ($\lambda = 0.650\mu$) for beryllia at 1740°K. They do not report the nature of the beryllia surface which was used in making this measurement. The value 0.37 is based on the value of the spectral emissivity of platinum taken to be 0.33 at all temperatures. Based on the more recent and complete measurements of the emissivity of platinum[6] which were used in calculating the values reported in Table I, the value given by Burgess and Waltenberg for the emissivity of beryllia would be 0.33 at 1755°K.

Liebmann[3] measured the spectral emissivity of a beryllia surface which had been slip-cast on a Nernst glower. The grain size of the beryllia was 0.5μ. His results are given in Table II.

Discussion of Errors

The values given in Table I are reported in accordance with the precision of measurement of the various experimental quantities. However, the accuracy of the values obtained for the spectral and total emissivities is not determined by the precision of measurement of the experimental quantities.

The temperature of all surfaces surrounding the sample during measurement should be so low that the radiant flux from the surrounding surfaces is insignificant compared to that of the hot beryllia surface. In the apparatus shown in Fig. 2 the surfaces which could radiate to the beryllia surface were always colder than necessary to decrease the error in the spectral emissivity from this source to less than 0.1 percent. However, in the measurement of the total emissivity a much greater error resulted from failure to maintain neighboring surfaces at a sufficiently low temperature. There was partial compensation of errors since the Rayotube was calibrated *in situ* and there was a corresponding error in the measurement of the brightness temperature of the beryllia surface. It was estimated that the error in the total emissivity resulting from this source was less than 5 percent.

There is another possible source of error in the measurement of the spectral emissivities and therefore also in the calculated values of total emissivity. While good thermal contact was obtained between the platinum and the beryllia, the temperatures of the two surfaces may not have been identical due to the difference in total emissivity of platinum[7] and beryllia surfaces. The platinum reference surface may therefore have been at a higher temperature than the beryllia surfaces. The magnitude of the error from this source is minimized by the relatively high heat conductivity of beryllia (as compared with other oxides) and by the fact that heat was lost from the platinum and beryllia surfaces by conduction and convection as well as radiation.

The spectral emissivities as measured in this investigation are normal spectral emissivities. This is also true of Stephens'[6] values for platinum. However, there is some uncertainty as to whether the values given by Stephens, determined in vacuum, may be applied to the platinum surfaces used in this investigation, in air. Stephens' results agree well with those obtained by Worthing,[8] who measured the spectral emissivity of platinum in an argon atmosphere. However, there has been a wide variation in the measurements of the spectral emissivity of platinum as made by various investigators.[9] Worthing[9] suggests that these variations may be due in part to the atmosphere used in each case.

The spectral emissivity of the beryllia surfaces was determined by three independent methods which gave values in good agreement. Since it is not probable that the net error inherent in each of the three methods was the same, the total error resulting from the above sources must not have been very great. It is estimated that the values of the spectral emissivity given in Table I are accurate to well within 10 percent and that the values of total emissivity are correct to within 20 percent.

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[7] C. Davisson and J. R. Weeks, J. Opt. Soc. Am. **8**, 581 (1924).

[8] A. G. Worthing, Phys. Rev. **28**, 174 (1926).

[9] American Institute of Physics, *Temperature, Its Measurement and Control in Science and Industry* (Reinhold Publishing Corporation, New York, 1941), p. 1164.