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Measurement of Emittance of Metal Interface in Molten Salt

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A new technique for measuring the total normal emittance of a metal in a semi-transparent liquid has been proposed and this technique has been applied to measure the emittance of stainless steel (SUS304), nickel, and gold in molten potassium nitrate KNO_3. These emittance data are indispensable to analyzing the radiative heat transfer between a metal and a semitransparent liquid, such as a molten salt.

KEY WORDS: emittance; metal surface; molten salt; radiative heat transfer; radiative properties; reflectance.

1. INTRODUCTION

While a significant amount of data for thermal radiation characteristics of solid materials exists in many compilations, the data are not always useful in radiation heat transfer evaluation, particularly in the estimation of radiation characteristics of metals. This is due to the facts that radiation depends on the characteristics of the surfaces and that most of the real surfaces of metals are roughened and oxidized in actual industrial and/or natural environments. It is substantially impossible to compare and identify the surface of a particular material in use with that of the surface in a data book. Additionally, there is the more essential problem that values of emittance and/or reflectance in data books are usually those for surfaces in vacuum or in air. That is, the data books do not offer any knowledge for emittance or reflectance of a surface in a liquid whose refractive index

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cannot be approximated to be unity. The emittance of a metal in a liquid depends not only on the optical property of the metal but also on that of the liquid.

Such situations are encountered, for example, in the cases of the emittance and/or reflectance of a metal surface in water, in liquid helium, and in molten salt at high temperatures. These emittance and/or reflectance values are important not only in the radiation heat transfer evaluation but also in the radiation heat transfer correction for thermal diffusivity measurement system using a three-layered liquid cell [1, 2]. It is desirable to develop an experimental technique for measuring the emittance of a metal in a liquid, because it is impractical to expect that the emittance data for various kinds of metal surfaces in various kinds of liquid are included in a data book.

In this paper, a new technique for measuring the emittance of a metal in a liquid is presented. This technique has been used to measure the total normal emittance of stainless steel SUS304, nickel, and gold in a molten salt, such as potassium nitrate KNO_3. While the proposed technique does not give the exact total hemispherical emittance, i.e., the most useful property for evaluating radiation heat transfer, it does enable to estimate a more probable value for the radiation heat transfer evaluation to be estimated.

2. PRINCIPLE OF THE MEASUREMENT TECHNIQUE

2.1. Real Surface in Liquid

The most practical surface is an oxidized roughened surface, and this is distinguished from a clean optically smooth and flat surface. However, for this method the real surface is considered to be an ideal one which is optically smooth and flat and does not scatter radiation. The surface of the metal may be oxidized and covered with a thin film layer, but it is not roughened. Radiation characteristics of such a system of a specular surface possibly attached with a parallel surface film can be described in principle by an electromagnetic wave theory. However, even in this simplest case, calculation to obtain the emittance of the metal in a liquid is not easy, because the spectral values of complex index of refraction of the metal and film at high temperature have to be known, and the thickness of the surface film has to be measured. Obtaining the data for these properties at high temperatures is not easy even for a pure metal and is more difficult for various kinds of metal alloys. In addition, another kind of measurement is needed for the composition and thickness of the film grown on the metal.

Then a good experimental technique to measure the emittance of a metal in a liquid needs to be developed.

2.2. Physical Model of the Specimen System

A parallel liquid layer is formed on a plane metal substrate and the upper surface of the liquid layer is bounded on an infinite space of air to form a "specimen system" of air, liquid, and metal. Figure 1 shows the physical model of this specimen system, where the liquid layer is substituted by a molten salt layer, dealt with in the experiment in Sections 3 and 4. The metal in Fig. 1 absorbs radiation strongly, and all the radiation phenomena occur in the vicinity of the metal surface. Although the metal surface may be oxidized and have a film layer, the surface region including the surface film is considered to be the metal surface having the unknown radiation characteristics. The liquid is treated as a semitransparent medium, in which radiation is not scattered. The temperature over the liquid layer is uniform and is equal to that of the metal substrate. Optical properties of the liquid are known. The air layer in this model is equivalent to vacuum, that is, the air is treated as a nonabsorbing medium of radiation, and the refractive index is unity over the entire wavelength region. The total normal emittance of the specimen system is measured from the air side, and a series of measured values of the emittance is analyzed to determine the unknown value of the total normal emittance of the metal in the liquid.

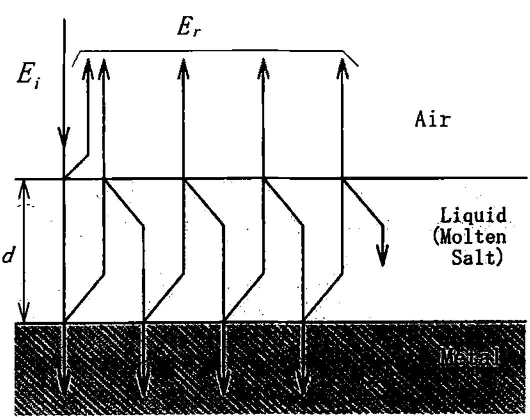

Fig. 1. Model of the specimen system.

2.3. Spectral Emittance of the Specimen System

Under the thermal equilibrium condition, Kirchhoff's law is valid for spectral quantities. The spectral normal emittance ε_{sys} of the specimen system is related to the spectral normal reflectance r_{sys} of the system by

$$\varepsilon_{sys} = 1 - r_{sys} \tag{1}$$

An experimental system is generally not under thermal equilibrium conditions, but applying the Kirchhoff's law to the system does not lead to a serious problem.

When the radiation of intensity E_i is incident to the specimen system in the normal direction, the reflected normal radiation intensity E_r is expressed as the sum of the infinite geometric series and the reflectance r_{sys} of the specimen system can be described as follows:

$$r_{sys} = E_r/E_i = r_i + \frac{(1-r_1)^2 \, r_2 \exp(-2Kd)}{1 - r_1 r_2 \exp(-2Kd)} \tag{2}$$

where d is the thickness of the liquid layer. The symbols r_1 and r_2 denote the normal reflectance of the air–liquid interface 1 and the reflectance of the metal–liquid interface 2, respectively, where radiation can be incident normally on either of the upper and lower sides of the interfaces. Reflectance r_1 of the interface 1 is written by Fresnel's equation,

$$r_1 = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \tag{3}$$

where n and k are, respectively, the real and imaginary parts of the complex index of reflection of the liquid, which are expressed as follows:

$$\hat{n} = n - ik \tag{4}$$

The real part is the refractive index, and the imaginary part is the absorption index. The symbol K in Eq. (2) denotes the absorption coefficient of the liquid. Provided the absorption of the liquid is not as strong as that of a metal, the absorption coefficient K is related approximately to the absorption index k by

$$K = 4\pi k/\lambda \tag{5}$$

where λ is the wavelength of radiation in vacuum. The spectral normal emittance ε_{sys} of the specimen system is written as

$$\varepsilon_{sys}(\lambda) = \varepsilon_{sys}(d, \lambda, n(\lambda), k(\lambda), r_2(\lambda)) \tag{6}$$

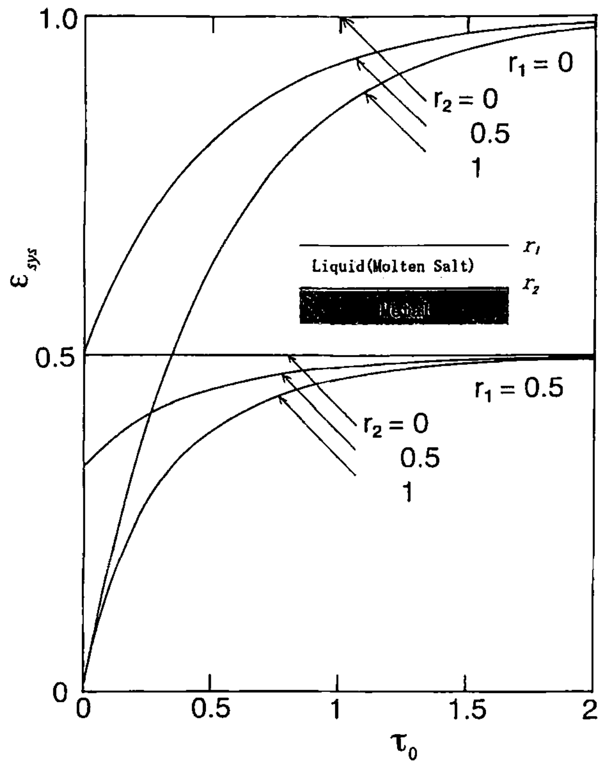

Fig. 2. Calculated values of normal emittance of the specimen system.

where the unknown quantity is the spectrum of reflectance r_2, and all other quantities are known.

Figure 2 shows calculated values of ε_{sys} for various values of r_2 and thickness d. The reflectance r_1 of the interface 1 is fixed at a high value of $r_1 = 0.5$ as an example and at the lowest value of $r_1 = 0$. The emittance ε_{sys} of the specimen system increases with increasing optical thickness $\tau_0(=Kd)$ and converges to the limit of $(1 - r_1)$.

2.4. Total Emittance of the Specimen System

In order to interpret spectral quantities to a spectrally integrated total quantity, the characteristics are presumed to be constant over each localized region. This assumption is called the local gray approximation. Many narrow spectral regions are prepared for a rigorous radiation heat transfer evaluation, but the entire spectral region is divided into m regions with comparatively wide spetral band widths. That is, the total normal emittance $\varepsilon_{sys}^{total}$ of the specimen system of liquid layer thickness d_i is evaluated by

$$\varepsilon_{sys}^{total} = \sum_{j=1}^{m} \varepsilon_{sys}^{j}(d_i)\{F(\lambda^j, T) - F(\lambda^{j-1}, T)\} \tag{7}$$

where ε_{sys}^{j} is the normal emittance of the specimen system in a band region j. The blackbody radiation function $F(\lambda^j, T)$ is the energy ratio of the radiation of wavelength $0 \sim \lambda^j$ to the total energy emitted by the blackbody at temperature T. Provided the local gray approximation is made properly on spectral quantities, Eq. (7) reduces to

$$\varepsilon_{sys}^{j} = \varepsilon_{sys}^{j}(d_i, n^j, k^j, r_2^j) \tag{8}$$

where the unknown quantities are the reflctance r_2^j ($j = 1, 2,..., m$) of the metal in the liquid for as many as m spectral band regions. A numerical technique is needed to determine a set of most probable values of r_2^j.

2.5. Processing of Measured Data

The measured total normal emittances of the specimen systems with N kinds of liquid layer thickness d_i ($i = 1, 2,..., N$) are analyzed to obtain m kinds of unknown values of r_{2j} ($j = 1, 2,..., m$). The number N should be greater than the number m of the spectral band regions. A least-squares processing technique is adopted so that a set of r_2^j values can minimize an objective function,

$$f = \sum_{i=1}^{N} w_i \{\varepsilon_{sys}^{total|calc}(d_i) - \varepsilon_{sys}^{total|exp}(d_i)\}^2 \tag{9}$$

That is, an equation,

$$\delta f = 0 \tag{10}$$

is solved. The superscripts "calc" and "exp" in Eq. (9) are calculated and experimental values, respectively. The weight function w_i is given by

$$w_1 = \frac{2(d_2 - d_1)}{3(d_N - d_1) + (d_{N-1} - d_2)} \tag{11}$$

$$w_i = \frac{d_{i+1} - d_{i-1}}{3(d_N - d_1) + (d_{N-1} + d_2)} \tag{12}$$

$$w_N = \frac{2(d_N - d_{N-1})}{3(d_N - d_1) + (d_{N-1} - d_2)} \tag{13}$$

The w_i values are proportional to the spacing of experimental points for thickness d_i and are normalized to be

$$\sum_{i=1}^{N} w_i = 1 \quad (14)$$

In the above inversion analysis, a lattice point search technique is used.

The band emittance ε_2^j of the metal in the liquid can be determined with the obtained results of r_2^j and Kirchhoff's law,

$$\varepsilon_2^j = 1 - r_2^j \quad (15)$$

The total normal emittance $\varepsilon_2^{\text{total}}$ of the metal in the liquid is then readily calculated using Eq. (7).

3. APPARATUS

A schematic diagram of the apparatus is shown in Fig. 3. A powdered salt in a vessel (5) is melted by an electric heater (6). A metal plate specimen (2) is placed horizontally in the bottom of the vessel and the molten salt layer (1) is formed with uniform thickness on the metal. For eliminating the effect of background radiation from the circumstances, the outside of the vessel is covered with a multilayered radiation shield (7) and a water-cooled jacket (8).

The surface temperature of the metal specimen is measured by a chromel–alumel thermocouple of 0.1 mm in diameter. Two terminals of the

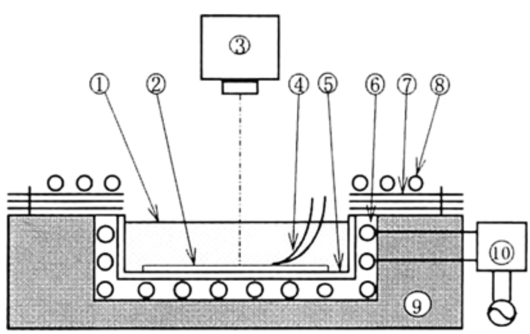

Fig. 3. Schematic diagram of measuring apparatus. (1) Molten salt; (2) metal plate as a specimen; (3) total radiation flux meter; (4) thermocouple; (5) vessel; (6) heater; (7) radiation shield; (8) water-cooled jacket; (9) insulation brick; (10) voltage transformer.

hot junction are spot-welded on the specimen surface separately with a 1-mm distance from each other. The accuracy of the temperature measurement is evaluated to be within ±1.0% at 700 K. Radiation energy emitted from the specimen system is measured by a total radiation flux meter (3) faced vertically downward above the specimen system.

The radiation flux meter contains a pyroelectric sensor and can detect infrared radiation substantially are the entire spectral region. The output of the meter is expressed by the following equation:

$$V = \alpha(\varepsilon_{sys} T_e^4 - \varepsilon T_c^4) \qquad (16)$$

where α is the apparatus constant including the Stefan–Boltzmann's constant and the shape factor, T_e is the absolute temperature of the specimen system, and T_c and ε_c are the absolute temperature and the emittance of an optical chopper in the meter. As the value of α is calibrated with a black-body furnace and $\varepsilon_c \approx 1$ for practical purposes, the emittance of the specimen system ε_{sys} is then obtained from the measured values of T_e, T_c, and V. The temperature T_e is almost the same as the room temperature and the accuracy of measuring V is evaluated to be within ±2%.

In order to verify the performance of the measuring system, the emittance of the specimen system consisting of a water layer on the metal surface (SUS304) was measured and the values were compared with the calculated values. The results are shown in Fig. 4. The calculated values

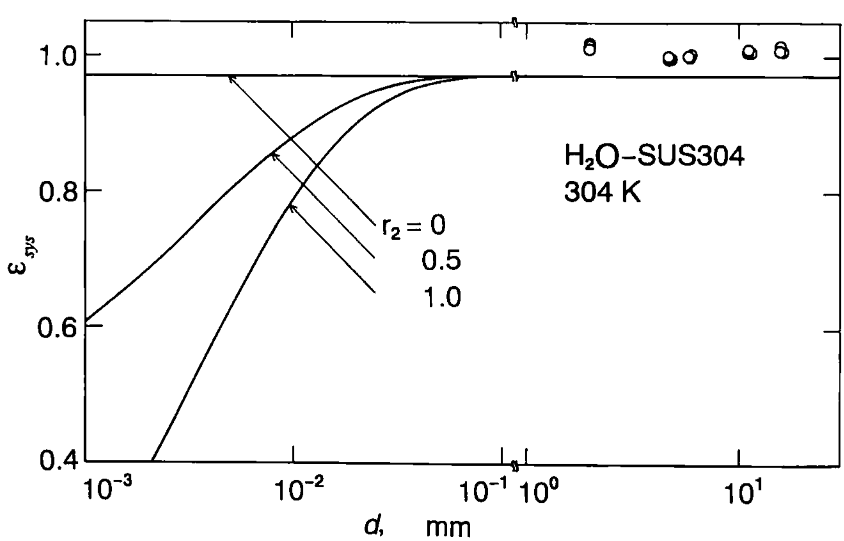

Fig. 4. Total normal emittance of the specimen system with water layer on a metal surface (SUS304).

were approximately constant at 0.97 for the range of the thickness above 0.1 mm regardless of the values of r_2, while the measured values were around 1, thus showing good agreement.

Systems containing of molten potassium nitrate KNO_3 and metal surfaces of SUS304 stainless steel, nickel, and gold-plated nickel were then measured. The absorption coefficient of KNO_3 [3] and its bands approximation are shown in Fig. 5. In this band approximation about 30% of the blackbody radiation energy at 700 K was included in the wavelength region of 0 to 3.2 μm. Furthermore, this region was divided into two bands at 2.5 μm each with the same blackbody radiation energy. The mean value of the adsorption coefficient for each band was calculated using Planck's method.

The reflectance r_1 of KNO_3 interfaced with air, calculated using the measured values of n and k [3] and its band approximation are shown in Fig. 6. In order to determine the reflctance r_2^j to minimize the objective function f in Eq. (9), a curve-fitting technique was used.

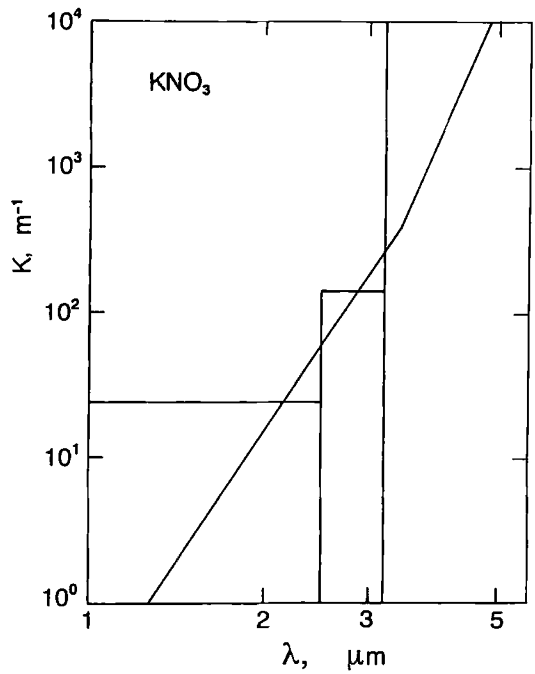

Fig. 5. Absorption coefficient of KNO_3 and its band approximation.

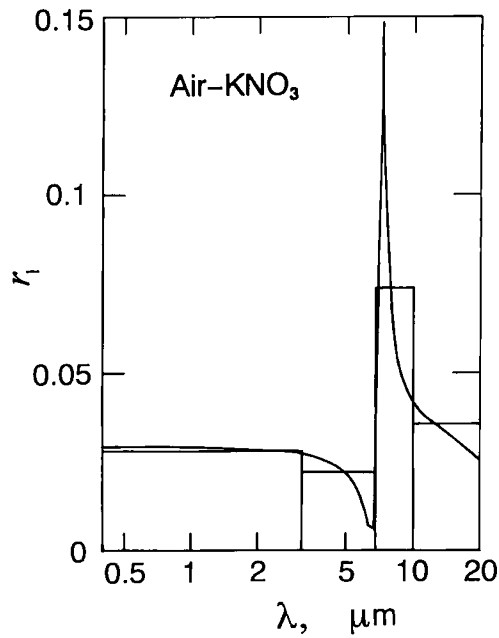

Fig. 6. Spectral normal reflectance of KNO_3 interfaced with air and its band approximation.

4. RESULTS AND DISCUSSION

Symbols ○ and △ in Figs. 7, 8, and 9 present the experimental results of the total normal emittance ε_{sys} of the specimen system for the three kinds of metal surfaces at two temperature levels around 630 and 735 K. The measurements were carried out on various thicknesses d of the KNO_3 layer.

In every metal interface at every temperature, the emittance ε_{sys} increases with increasing the thickness d of the salt layer. The thickness dependence is due to a significant increase in emission of the layer with increasing d. The emittance ε_{sys} decreases with increasing the temperature level. The temperature dependence is weak in comparison with the thickness dependence. Since the spectral radiative properties of the salt and metals depend weakly on the temperature [3], the temperature dependence of total emittance ε_{sys} is considered to be caused by a coupled effect of spectral characteristics of the absorption coefficient of the salt and the temperature shift of the Planckian distribution.

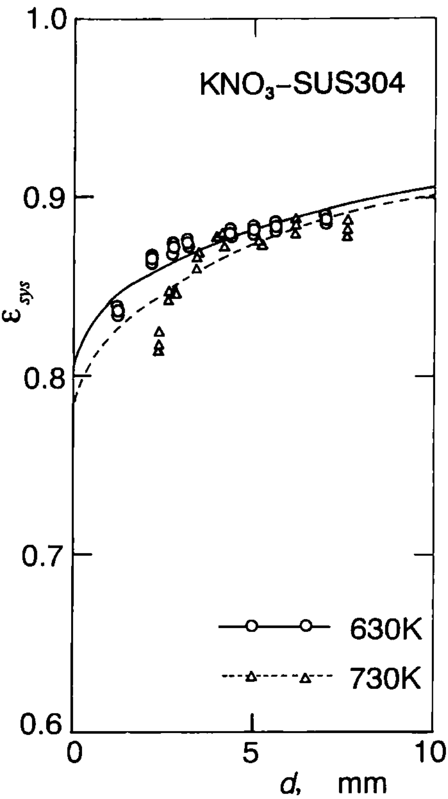

Fig. 7. Total normal emittance of the specimen system with SUS304 in molten KNO_3.

Table I. Experimental Results of Band and Total Normal Emittance of Metals in Molten KNO_3

	ε_2^j					
	$\lambda_{0-2.5\,\mu m}$		$\lambda_{2.5-3.2\,\mu m}$		ε_2^{total}	
System	630 K	730–740 K	630 K	730–740 K	630 K	730–740 K
KNO_3–SUS304	0.030	0.39	0.66	0.58	0.38	0.48
KNO_3–Ni	0	0.24	0.34	0.35	0.19	0.29
KNO_3–Au	0	0.18	0.17	0	0.098	0.095

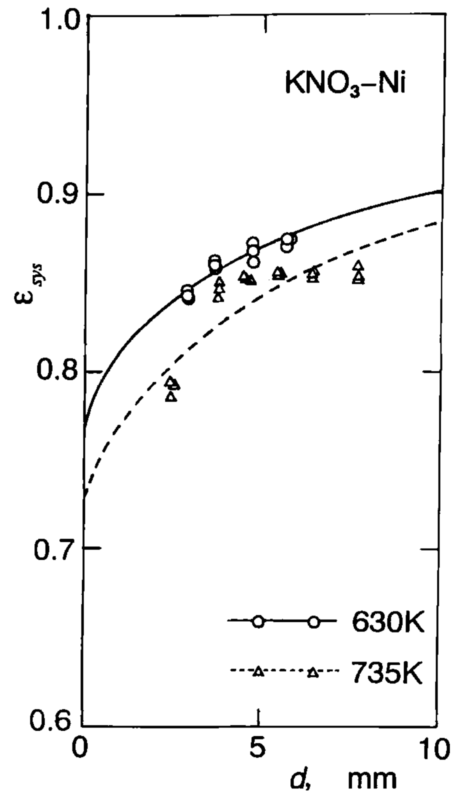

Fig. 8. Total normal emittance of the specimen system with Ni in molten KNO_3.

The solid and dashed curves of ε_{sys} in Figs. 7–9 were drawn by using the curve-fitting method to determine the band emittances ε_2^j of metal interfaces in the molten salt. These curves describe the behavior of the experimental points of ε_{sys} within a deviation of $\pm 10\%$. The obtained values of ε_2^j each metal at each temperature are shown in Table I. It is natural that the uncertainty in the determined values of ε_2^j themselves are reasonably less than that in the ε_{sys} curves. However, these determined values of ε_2^j can be used to evaluate the radiative heat transfer in laboratory and engineering systems including metal interfaces in semitransparent media of radiation.

The values of the total normal emittances ε_2^{total} of the metals in the salt, for comparison, which are obtained by using Eq. (7) with the values of the band emittance ε_2^j, are shown in Table I. Table II shows directly

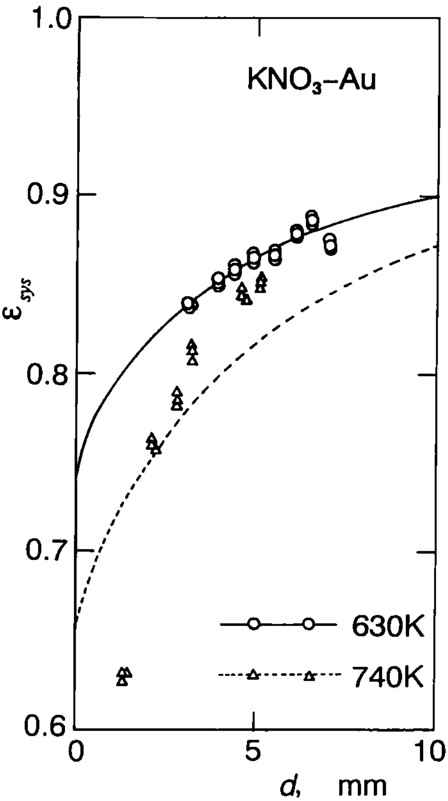

Fig. 9. Total normal emittance of the specimen system with Au in molten KNO_3.

measured values of total normal emittances of metals in air. The total emittance of every metal in the salt is higher than that of the same metal in air.

Table II. Experimental Results of Total Normal Emittance of Metals in Air

	ε_2^{total}	
System	620–630 K	730–740 K
Air–SUS304	0.19	0.19
Air–Ni	0.13	0.13
Air–Au	0.091	0.071

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