

Radiative Heat Exchange Method for Thermal Conductivity Measurement Applying a Perpendicular Heat Flow to a Thin-Plate Sample: Principle and Apparatus

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To measure thermal conductivity of materials of low conductivity (0.1 to $1 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), a method using a specimen of small size ($2 \times 25 \times 25 \text{ mm}$) has been developed. This method applies a well-defined, steady, and uniform heat flux perpendicular to the surface of a small plate sample of polymers or ceramics jointly by means of radiative heat exchange as well as by an areal heater on the sample surface and allows a reasonably rapid (5-min) measurement of thermal conductivity. This method of measuring conductivity is an absolute and direct measurement method which does not need any standard reference materials or information about heat capacity. The principle of the method has been demonstrated by constructing a measurement apparatus and measuring thermal conductivity of a few materials. The thermal conductivities of silicone rubber and Pyrex (Corning 7740) glass measured by the present method between 30 and 90°C are compared with recommended values.

KEY WORDS: low-conductivity materials; radiative heat exchange; radiation thermometry; thermal conductivity.

1. INTRODUCTION

The thermophysical properties of materials which can be supplied only in small sizes, such as new experimental materials, thin-film materials, or samples heavily irradiated by neutrons, have become more important; and methods for measuring thermophysical properties are needed which are applicable to specimens of small size. Concerning thermal conductivity

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measurements, the guarded hotplate method has been representative of steady-state methods for materials of low conductivity [1]. However, the sample size needed in this method is relatively large, for example, about 1–2 cm in thickness and 20 cm in diameter. Moreover, as a consequence of the large sample size it can take a long time (i.e., as long as a half- or a full day) to make one measurement at one stable temperature.

Thermal diffusivity is measured by non-steady-state methods. The laser flash method, a representative non-steady-state method, allows measurements to be made rapidly with a sample of small size (about 1 mm in thickness and 10 mm in diameter) [1]. This method, where the thermal conductivity of the sample is derived from the thermal diffusivity with the knowledge of heat capacity and density, is complementary to the steady-state method.

The authors have measured thermal conductivity using a radiative heat exchange method, wherein the heat flows parallel to the surface of thin-plate samples of high-conductivity materials, such as metals and diamond films [2–4]. To extend the capability of the radiative heat exchange method to materials of low conductivity, such as polymers or ceramics, a measurement technique applying a heat flow perpendicular to the surface of a thin-plate sample was developed. This method sets up a steady and uniform heat flux perpendicular to the sample surface using radiative heat exchange in a vacuum between the sample surface and the environment, supplemented by heat from an areal heater adhered to the outer surface of the sample.

2. PRINCIPLE OF METHOD

The thermal conductivity is defined as the proportionality coefficient between the heat flux passing through a material and the temperature gradient in the material resulting from the causative heat flux. Based on this definition, various measurement methods have been developed. In most of the methods, a given amount of electrically supplied heat flows through a well-defined area of the sample; the heat flux is the supplied heat divided by the area. One of the critical requirements of these measurement methods is that all of the supplied heat must flow through the defined area of the sample without any gain or loss, in order to determine the heat flux accurately. Another requirement is that the quantity of heat, from whatever source, producing the measured temperature gradient must be known. Failure to achieve these conditions has often been a major source of measurement error in these methods.

The basic approach of the present measurement method is different from the above methods in that a given amount of heat flux is directly lost

from or supplied to the surface of a plate sample by means of radiative heat exchange. If the heat flux is supplied solely by radiation, then the heat flux at the center of the sample is directly determined by a precise knowledge of emissivity and temperature of the sample surface, using the Stefan–Boltzmann law. So, if the emissivity is constant and well-known, then at a small area at the middle of the sample, where the effect of transverse heat flux is negligible, only the measurements of local surface temperature and temperature gradient perpendicular to the sample surface are needed to obtain the thermal conductivity. An evaluation of heat flow over the total area of the sample is not necessary. Following this principle of measurement, heat loss or gain at the perimeter of the sample does not lead to a critical measurement error, and so, complicated guard heaters are not necessary. This approach offers a possibility of avoiding a major source of measurement error and complicated measurement procedures in the steady state method.

In practical measurements using this method, the emissivity of the sample surface can sometimes be unknown or not reproducible. In addition, measurements near the environmental temperature are difficult because the amount of the radiative heat exchange is small. To make a simultaneous measurement of the emissivity and cause an additional heat flux near the environmental temperature, supplementary electrical resistive heating of the sample surface with an areal heater may be employed, if necessary. In this case, although heat generation per unit area at the areal heater must then be known to obtain the thermal conductivity and the emissivity, not all of the heat generated by this heater is required to flow through the sample area.

Figure 1 illustrates a schematic cross-sectional view of the sample assembly and temperature distributions on the sample surface in the present method. A plate sample (1) is embedded and adhered on a metal block (2) of high thermal conductivity, forming a flat surface with the block. A thin-film areal heater (3) which can supply a uniform areal heat is adhered on the outer surface of the sample only. The surfaces of the metal block and the areal heater are coated with black paint (4) of a high and constant emissivity.

Figure 2 shows the scheme of the measurements. The metal block is kept at a constant temperature higher than that of the environment in a vacuum chamber, and if necessary, the areal heater is heated by direct current. Heat is exchanged between the sample surface and the environment by means of radiation from the surface directly to the environment and by conduction from the surface through the sample to the metal block. In this state, a temperature difference, ΔT_1 , between the surfaces of the sample area and the metal block is set up as a consequence of the difference

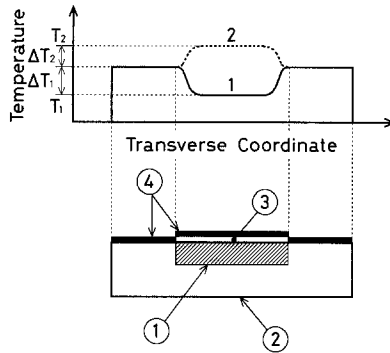


Fig. 1. Schematic illustrations of the cross section and the surface temperature distributions of the sample assembly. The temperature distributions designated by numbers 1 and 2 correspond to two different heating rates of the areal heater. 1, Sample; 2, metal block; 3, areal heater; 4, black paint.

in thermal conductivity between the metal and the sample, as indicated by Line 1 in Fig. 1. This surface temperature difference is measured remotely without any thermal disturbances by a thermograph through an optical window of the vacuum chamber as illustrated in Fig. 2.

Assuming that (i) the thermal conductivity of the metal block is much higher than that of the sample, (ii) the block is at a uniform temperature, and (iii) the temperature drops across the black paint and the areal heater are negligible compared to that across the sample, then the temperature

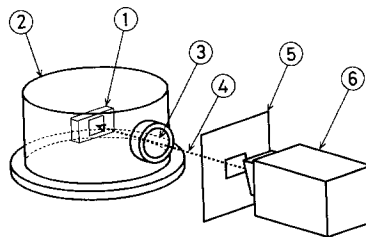


Fig. 2. Schematic view of the scheme of the measurements. 1, Sample assembly; 2, water-cooled vacuum chamber; 3, optical window; 4, sight of the thermograph; 5, water-cooled radiation shield; 6, thermograph.

difference between the outer surface of the sample and the metal block is equal to the temperature difference between the two large surfaces of the sample assembly. The temperature gradient perpendicular to the sample surface is obtained by dividing this temperature difference by the sample thickness.

If the current supplied to the areal heater is increased, while keeping the metal block at the same temperature, another temperature difference, ΔT_2 , is set up, as indicated by Line 2 in Fig. 1. Assuming that (iv) the heat flux is directed perpendicular to the sample surface at the middle of the sample area, (v) the environment has hemispherical total emissivity which equals unity, and (vi) the environment is at a uniform temperature (does not change due to increased rate of heating of sample), then the thermal conductivity of the sample, λ , is expressed by Eq. (1) by setting $i=1$ and 2 for the states of Lines 1 and 2, respectively.

$$\lambda = d \{ H_i - \varepsilon_{\text{ht}} \sigma (T_i^4 - T_e^4) \} / \Delta T_i \quad (i=1 \text{ or } 2) \quad (1)$$

where d is the thickness of the sample, H_i is the heat generation rate per unit area at the areal heater, ε_{ht} is the hemispherical total emissivity of the black paint, σ is the Stefan-Boltzmann constant, T_i is the surface temperature at the middle of the sample area, T_e is the environmental temperature, and ΔT_i is the temperature difference between the surface at the middle of the sample area and the surface of the metal block neighboring the sample area.

Expressing Eq. (1) in the states of the Lines 1 ($i=1$) and 2 ($i=2$) and solving for the thermal conductivity and the emissivity, they are represented by Eqs. (2) and (3), respectively.

$$\lambda = d \frac{\phi_1 H_2 - \phi_2 H_1}{\phi_1 \Delta T_2 - \phi_2 \Delta T_1} \quad (2)$$

$$\varepsilon_{\text{ht}} = \frac{H_1 \Delta T_2 - H_2 \Delta T_1}{\sigma (\phi_1 \Delta T_2 - \phi_2 \Delta T_1)} \quad (3)$$

where

$$\phi_i = T_i^4 - T_e^4 \quad (i=1 \text{ or } 2). \quad (4)$$

In practical measurements of materials of relatively high thermal conductivity (more than $1 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), measured values must be corrected for the thermal resistance of adhesive. The corrected thermal conductivity, λ_c , is expressed by Eq. (5).

$$\lambda_c = d \left/ \left[\frac{\phi_1 \Delta T_2 - \phi_2 \Delta T_1}{\phi_1 H_2 - \phi_2 H_1} - \frac{d_a}{\lambda_a} \right] \right. \quad (5)$$

where d_a and λ_a are the total thickness and the thermal conductivity of the adhesive, respectively.

3. MEASUREMENT APPARATUS

Figure 3 shows a detailed view of the sample assembly, whose quarter section is cut away. The sample size used in the present measurements was $2 \times 25 \times 25$ mm. Copper was used as the metal block and a stainless-steel foil ($10 \mu\text{m}$ in thickness, made of SUS 304) was used as the areal heater. Epoxy adhesive (Araldite Standard, made by Ciba-Geigy Ltd.; about $4 \mu\text{m}$ in thickness) was used to adhere the sample to the metal block and the areal heater to the sample surface.

On the top and bottom sides of the metal block a pair of plate insulators made of fluorocarbon polymer was attached. The stainless-steel foil was bent around opposite ends of the sample at the edges of the insulators. A pair of blade-shaped electrodes and voltage probes was contacted to the surface of the stainless steel foil on the surfaces of the insulators, and uniform current was supplied through the electrodes. The areal heat generation rate, H , at the areal heater was obtained by Eq. (6).

$$H = \frac{VI}{WL} \quad (6)$$

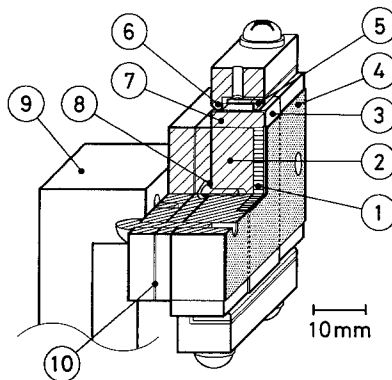


Fig. 3. Detailed view of the sample assembly used in the present measurements whose quarter section is cut away. 1, Sample ($2 \times 25 \times 25$ mm); 2, metal block; 3, areal heater; 4, black paint; 5, voltage probe; 6, electrode; 7, insulator; 8, thermocouple; 9, sample support; 10, main heater.

where V is the voltage drop between the voltage probes, I is the current passing through the stainless-steel foil, W is the width of the foil in the direction across the current, and L is the distance between the voltage probes along the foil. The distance, L , was evaluated from the ratio of the voltages measured by the voltage probes and standard probes of fixed distance contacted to the surface of the stainless-steel foil when a constant current was supplied. The current, I , was measured from the voltage drop at a standard resistor inserted in series with the circuit.

On the rear surface of the metal block, a type K thermocouple (0.3 mm in diameter) was fixed using a screw (2 mm in diameter) to measure the temperature of the metal block. The metal block was attached to a main heater to keep the block at a constant temperature. This main heater was designed to realize a uniform temperature distribution and consisted of a heater of stainless-steel foil sandwiched by a pair of insulators and copper plates. The main heater was supported by a support, the other end of which was fixed on the water-cooled base plate of the vacuum chamber. The metal block was kept at a constant temperature by controlling a direct current supplied to the main heater using the thermocouple and a temperature controller.

The surfaces of the areal heater and the metal block were coated with black paint (heat resistant, black matte, made by Asahipen Co.). The emissivity and the thickness of the black paint were about 0.9 and 50 μm , respectively. To realize the proper condition of radiative heat exchange expressed by Eq. (1), all of the inner surfaces of the vacuum chamber visible from the sample surface were also coated with the same black paint and cooled by water. A calcium fluoride window of 64 mm in diameter and 12 mm in thickness was used as the optical window (3, Fig. 2). A water-cooled radiation shield with an aperture was placed between the optical window and the thermograph in order to cover the outside of the optical window with a surface of uniform temperature equal to that of the inner surface of the vacuum chamber. The inside of the vacuum chamber was kept at a pressure lower than 2×10^{-5} Torr during the measurements to eliminate the effect of heat losses by convection and gas conduction from the sample surface.

An infrared thermograph (Model JTG-IB, made by Japan Electron Optical Laboratory Ltd.), whose operating wavelength range was from 3.4 to 5.6 μm , was used to measure the temperatures at different points along a line across the center of the surface of the sample assembly. The one-dimensional temperature distribution along the center line on the surface of the sample assembly was measured every 8.3 ms while stopping the vertical scanning of the thermograph. Although the temperature imprecision of the thermograph was about 0.2 K at 30°C for one scanning, the imprecision

was reduced up to about 5 mK by averaging the output signals of the thermograph 2000 times using a signal averager. The averaged output signals were transferred to a microcomputer and converted into temperature distributions using a calibration equation relating the temperature to the output signals of the thermograph.

Before a series of measurements, the thermograph was calibrated against a variable-temperature blackbody furnace whose temperature was given by a platinum resistance thermometer. The calibration equation between the temperature of the blackbody furnace, T_b , and the voltage of output signals of the thermograph, $V_b(T_b)$, is given by Eq. (7).

$$V_b(T_b) = C \exp\{-c_2/(AT_b + B)\} + D \quad (7)$$

where A , B , and C are coefficients characteristic of the thermograph, D is the offset voltage of the thermograph, and c_2 is the second radiation constant. The coefficients A , B , C , and D are determined from the measured values of V_b and T_b using the least-squares method.

In practical temperature measurements using the thermograph, the spectral emissivity of the black paint on the sample assembly and the transmittance of the optical window were not unity and the effects of reflected or emitted environmental radiation could not be neglected near the environmental temperature. Assuming that the surfaces of the black paint and the optical window reflect the blackbody radiation of the same environmental temperature, the resultant output voltage of the thermograph, V' , is expressed by Eq. (8).

$$V' = \varepsilon_s V_b(T_s) + (1 - \varepsilon_s) V_b(T_e) \quad (8)$$

where T_s is the surface temperature of the sample or the metal block, and ε_s is the product of the spectral emissivity of the black paint and the transmittance of the optical window.

Strictly speaking, the value of ε_s can be different from paint to paint and the offset voltage, D , can change with the environmental temperature. In order to avoid these effects, the resultant output voltages of the thermograph viewing the surface of the metal block whose temperature was given by the thermocouple were measured at several temperatures before each measurement. The values of ε_s and D were determined from these data using the least-squares method. After the parameters were determined the true surface temperature of the sample assembly was obtained using Eqs. (7) and (8) from the resultant output voltage of the thermograph.

Figure 4 shows an example of temperature deviations of the surface of the metal block obtained by the thermograph from the indication of the thermocouple fixed on the metal block. In this example, although the tem-

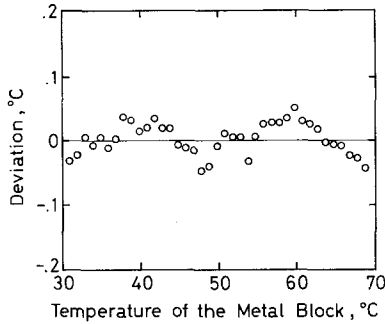


Fig. 4. Temperature deviations of the surface of the metal block obtained by the thermograph from the indication of the thermocouple fixed on the rear surface of the metal block.

perature deviations were within a range of $\pm 0.1^\circ\text{C}$, a small systematic deviation of the temperature scale every 10°C was observed. The output voltages of the thermocouple were converted into the values of temperature using a linear approximation of the output voltages every 10°C . The small systematic deviation in Fig. 4 may be caused by this linear approximation of the output voltages. Even if any deviations existed, they would not lead to a critical measurement error because the effect of the measurement error of the metal block temperature on the total measurement error of the thermal conductivity was estimated to be much smaller than that of the measurement error of the temperature difference.

In the present measurements of the thermal conductivity, the thermograph was used only to measure the temperature difference between the surfaces of the sample and the metal block. The absolute temperature of the sample surface was obtained by adding this temperature difference to the temperature of the metal block measured by the thermocouple under the assumption that the metal block was all at a uniform temperature. The environmental temperature was measured by a thermocouple fixed on the water-cooled base plate in the vacuum chamber. The output voltages of the thermocouples and the voltage drops at the areal heater and the standard shunt resistor were measured by a digital voltmeter and transferred to the microcomputer.

The measurements of temperature, temperature difference, and voltage drops were performed twice while varying the current supplied to the areal heater at each temperature of the metal block. The thermal conductivity of the sample at each temperature was calculated from these data by the microcomputer using Eq. (5).

4. RESULTS AND DISCUSSION

Figure 5 shows some examples of one-dimensional transverse temperature distributions on the surface of the sample assembly when a thermal conductivity measurement was performed. Line 1 in Fig. 5 indicates a temperature distribution without heating the areal heater. Supplying a current to the heater, the temperature of the sample surface rises according to the increase of the current as indicated by Lines 2 and 3. In these temperature distributions, flat regions of temperature are observed in the middle of the sample. This shows that at the middle of the sample the heat flux is directed perpendicular to the sample surface and that the effect of any transverse heat flux on the thermal conductivity measurements is negligible at the middle of the sample.

Figure 6 shows an example of hemispherical total emissivity of the black paint obtained from Eq. (3) when a thermal conductivity measurement was performed, and the solid line indicates a fitted line. In this example, scattering of the emissivity becomes larger near the environmental temperature. This may be caused by the lowered precision of emissivity measurement due to the decrease in radiative heat exchange where the temperature of the sample is close to that of the environment. In addition, the emissivity decreases slightly (by 5%) as the temperature increases from 30 to 80°C. This temperature dependency was not reproducible. The cause of this nonreproducible temperature dependency was not fully understood,

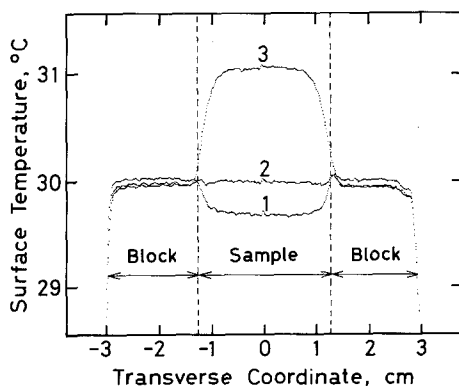


Fig. 5. Transverse temperature distributions on the surface of the sample assembly whose middle portion corresponds to the sample area. Line 1 indicates the temperature distribution without a current heating of the areal heater. The temperature of the sample surface rises according to the increase in the current as indicated by Lines 2 and 3.

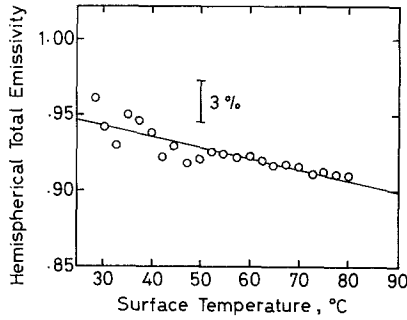


Fig. 6. Hemispherical total emissivity of the black paint coated on the sample assembly measured simultaneously [calculated from Eq. (3)] when a thermal conductivity measurement was performed. The solid line indicates the least-squares fit to the measured values.

but its effect on the measurement of the thermal conductivity is quite small in the present method.

Figure 7 shows the measured values of thermal conductivity of a silicone rubber standard reference material. The solid line indicates the recommended value of the same sample measured by the direct guarded hotplate method [5], and the dashed line indicates a line fitted to the presently measured values. It took about 5 min to obtain each data point in Fig. 7. In this result, most of the measured values were higher than the

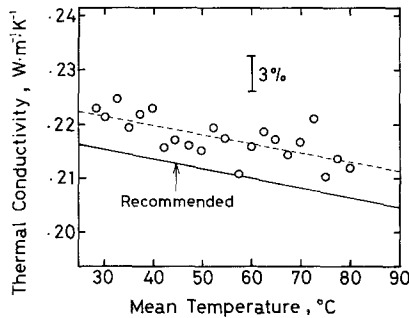


Fig. 7. Thermal conductivity of the silicone rubber standard reference material. A solid line indicates the recommended value measured by the direct guarded hotplate method [5], and a dashed line indicates a least-squares fit to the measured values.

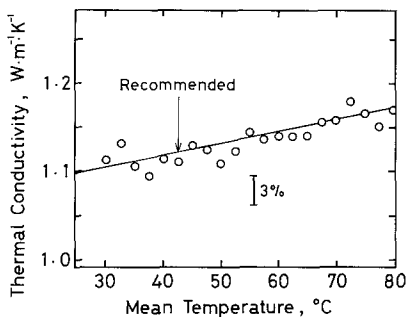


Fig. 8. Thermal conductivity of Pyrex (Corning 7740) glass. The solid line indicates the recommended value [6].

recommended value. The average deviation from the recommended value was 3%, and the standard deviation from the fitted (dashed) line was 1.3%. Considering that the accuracy of the recommended value was evaluated to be $\pm 5\%$ [5], the measured values are in agreement with the recommended value within the experimental uncertainties of both sets of measurements.

Figure 8 shows the measured values of the thermal conductivity of Pyrex (Corning 7740) glass, and the solid line indicates a recommended value [6]. Because the thermal conductivity of the Pyrex glass was larger than that of adhesive, the measured values were corrected for the thermal resistance of the adhesive. To correct the measured values, the thickness and the thermal conductivity of the adhesive must be known. Using an electric micrometer, the thickness of the adhesive was measured to be about $4 \mu\text{m}$ for each surface of the sample by removing the sample after the thermal conductivity measurement. The measured thermal conductivity of the adhesive was $0.22 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ using the present measurement method with a separate sample of the same lot of adhesive used in the present thermal conductivity measurements.

For the data shown in Fig. 8, the average deviation of the measured values from the recommended value was 0.4% and the standard deviation from the fitted line was 1%. Considering that the accuracy of the recommended value was reported to be $\pm 5\%$ for the Pyrex glass [6], the measured values are in good agreement with the recommended value.

5. SUMMARY

A radiative heat exchange method was developed which applies a perpendicular heat flow to a small thin-plate sample of low conductivity

material. The principle of this method was demonstrated by constructing a measurement apparatus and measuring a few materials of low conductivity whose thermal conductivity had been independently evaluated.

The present method allowed thermal conductivity to be measured in a steady state with a sample size much smaller than those needed by the previous methods, i.e., about 10^{-3} of the volume needed for specimens in the guarded hotplate method. Because the sample size was small, the time required to make one measurement was much shorter than those needed by the previous methods. Due to the technique of high-precision radiation thermometry a small temperature difference across the sample thickness, i.e., about 1°C , was enough to make a measurement. Such a small value of temperature difference provides a good approximation to a uniform gradient within the sample. This measurement method can be applied to materials whose thermal conductivity has a large temperature dependence. It should be noted that this is an absolute and direct measurement method of thermal conductivity, so that any reference materials or knowledge about the heat capacity of the sample are not needed.

If the hemispherical total emissivity of the black paint is well-known and reproducible, a passive measurement method without the electrical heating of the sample surface ($H_i = 0$) will be possible according to Eq. (1). In the passive method, the thermal conductivity is obtained by only one temperature measurement of the sample surface, and the areal heater on the sample surface can be eliminated. This passive method may be appropriate for simplified measurements of thermal conductivity by the radiative heat exchange method and for measurements at such high temperatures that adhesion of the areal heater onto the sample surface is not possible or reliable.

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