Method for measurement of emissivity and absorptivity of highly reflective surfaces from 20 K to room temperatures

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Abstract
We present a cryogenic method for the measurement of total hemispherical emissivity and absorptivity of various materials at temperatures from 320 K down to $\approx 20$ K. In absorptivity measurement the temperature of the examined sample is kept at $\approx 5$ K$–35$ K. Radiative heat flow between two plane parallel surfaces of 40 mm in diameter disk samples placed in a vacuum, a sample and a disk with reference surface, is absorbed by a colder sample and sinks into an LHe bath via a thermal resistor (heat flow meter). Heat flow is measured by substitution method, using thermal output of an electrical heater for heat flow meter calibration. A great deal of attention is paid to the estimation of uncertainties associated with this method. Capabilities of the instrument are demonstrated by the absorptivity and emissivity measurement of the pure aluminium sample. The expanded fractional uncertainty ($k = 2$) in emissivity $\varepsilon = 0.0041$ measured at $\approx 30$ K for pure aluminium is less than 11% and for values of emissivity $\varepsilon > 0.0053$ measured above 60 K the uncertainties are below 7%. The method was designed primarily for the measurement of highly reflective materials like pure metals, nevertheless high emissivity of the reference sample also enables the measurement of non-metallic materials with reasonable accuracy.

Keywords: absorptivity, emissivity, radiative heat transfer, metallic surfaces, cryogenics, uncertainty evaluation

(Some figures may appear in colour only in the online journal)
for investigation of highly reflective samples at temperatures higher than 80 K has been introduced [8] and applied for measuring stainless steel plates and the effect of silver coating [9].

On the other hand, calorimetric methods detect the heat which is lost or gained by the sample and measure solely total values of emissivity or absorptivity. In one approach the sample, positioned in a cavity of a given temperature, is thermally linked to surrounding walls by heat radiation while the other thermal links are negligibly weak. The sample is thrown out of equilibrium with a heater (electrical, light ray, e.g.) and let to relax. The value of emissivity can be evaluated from the relaxation time of the sample temperature, the known sample geometry and thermal capacity of the sample. For example a thermocouple was used as a thermometer in the work [10] while contactless measurements of sample thermal dilatation served for its temperature measurement in [11]. Another group of calorimetric methods, steady-state methods, uses a thermal link between the sample or radiative heat receiver on one side and a heat sink of constant temperature on the other side as a heat flow meter. Opposite the reference surface, the sample serves as a source of radiation (radiator) or as a receiver (absorber) in emissivity or absorptivity measurement, respectively. One of the simplest calorimetric approaches evaluates transferred radiative heat power and thus the emissivity/absorptivity from the boil-off rate of a cryogenic fluid [12, 13]. In more precise experiments a temperature drop on the thermal link gives the measured heat power. From a general point of view the receiver (absorber) together with the temperature sensor and thermal link thus create a bolometer, which alternatively can be calibrated using an electrical heater. The principle was used in measurements of absorptivity [14–16], emissivity [17] and of both properties [4].

Nevertheless, methods applied in the majority of the previous studies were designed for measurement in a relatively limited temperature range of thermal sources. They are also usually applicable for measuring either the sample emissivity or absorptivity. Our realization of the ‘calorimetric method’, utilizing the principle of bolometric measurement, is capable of covering the measurement of emissivity and absorptivity in a large range of temperatures of thermal radiation, spanning from cryogenic to room values. The instrument (‘Emister’) was presented for the first time in [18]. Since then, it has been utilized for the measurement of thermal radiative properties of a wide-range of materials such as various metals and alloys [4], copper surfaces with different treatments [5], DLC (diamond-like-carbon) coating on copper disc [19], water deposits on metal surface [16], as well as several other types of metallic surfaces intended for specific applications. On the basis of the apparatus presented here, the ‘Emister’, a new more precise instrument was developed [20] and used for the measurement of near-field enhancement of radiative heat transfer between plane-parallel surfaces [21].

In this study, along with the first comprehensive description of the apparatus and measuring process, we focus especially on the uncertainty analysis of emissivity/absorptivity of highly reflective samples measured with the presented method. Finally results on emissivity and absorptivity of pure aluminium are confronted with some results of other methods, both calorimetric and radiometric.

2. Method, system setup and measuring procedure

The instrument ‘Emister’ (figures 1 and 2) enables the measurement of both absorptivity and emissivity of a flat sample. The principle is based on measurement of radiative heat flow \(Q_R\) between two plane parallel surfaces, an examined sample surface and a reference surface with very high absorptivity \(\varepsilon_{REF}\) and emissivity \(\varepsilon_{EFF}\). The samples are concentric discs with a small gap between them. The total hemispherical absorptivity \(\alpha(T_A, T_R)\) is measured when the sample is cooled (position of absorber at temperature \(T_A\)) and irradiated with the heat emitted by the heated reference surface (position of radiator at temperature \(T_R\)). On the other hand, the sample is heated and the reference surface is cooled during the total hemispherical emissivity \(\varepsilon(T_R)\) measurements.

While emissivity is a material property, absorptivity depends both on the radiation and material temperatures,

\[
\varepsilon(T_R) = \frac{1}{\sigma T_R^4} \int_0^\infty \varepsilon(T_R, \lambda) \varepsilon(T_R, \lambda) d\lambda,
\]

\[
\alpha(T_A, T_R) = \frac{1}{\sigma T_R^4} \int_0^\infty \varepsilon(T_A, \lambda) \varepsilon(T_R, \lambda) d\lambda, \tag{1}
\]

where \(\sigma\) stays for Stefan–Boltzmann constant, \(T_R\) and \(T_A\) are respectively temperatures of radiation and material, \(\varepsilon(T, \lambda)\) hemispherical spectral emissivity and \(\varepsilon(T, \lambda)\) is spectral distribution of hemispherical emissive power of a black body.

Heat flux transferred between infinite plane parallel surfaces is [22]
The heat power $Q_R$ is transferred by thermal radiation from the radiator to the cold absorber where it is absorbed and further flows through a thermal resistor (heat flow meter) into a heat sink (LHe bath with stabilized pressure). The absorbed radiative heat power is evaluated from the temperature drop $T_A - T_B$ measured on the heat flow meter (the bottom of which is at stabilized temperature $T_B$). An insulation insert between the heat flow meter and the measuring chamber allows stabilization of $T_B$. The calibration of the heat flow meter and the measurement of emissivity or absorptivity are conducted in the same instrument setup.

Samples are in the form of the disk 40 mm in diameter and 1–5 mm thick. They are made of measured bulk material or consist of a substrate covered with a thin film, foil, thin sheet or paints. Thermal conductance of the sample, substrate and contact between the sample and substrate should ensure uniform temperature over the sample surface.

The gap between the surfaces of the radiator and the absorber cannot be set up smaller than 0.3 mm in the presented apparatus because of different thermal expansions of its parts. The gap $d_0 = 0.5 \text{mm}$ can be taken as a typical setup.

Temperature controller Lake Shore 340 is used for measurement of the temperatures $T_R$, $T_A$ and $T_B$, for changing setpoint of $T_R$ and for stabilization of $T_B$. A resolution of 0.1 mK in temperatures $T_A$ and $T_B$ is achieved with sensors LakeShore Cernox™ CX 1050. The radiator temperature $T_R$ is measured with the silicon diode LakeShore DT470 SD.

### 2.1. Calibration

In the calibration process the radiative heat flow is not present ($Q_R = 0, T_A \approx T_R$). The radiative heat flow $Q_R$ of the absorber is substituted by a power $Q_{HA}$ of a calibration electric heater which is incorporated in the absorber holder. The relation between the values $Q_{HA}$ and the measured temperature drop $T_{AB} = T_A - T_B$ is taken as a calibration curve of the heat flow meter.

In fact a non-zero difference $T_{AB} \approx (T_A - T_B)_{Q_R = 0} < 2 - 3 \text{mK}$ in readings of temperature sensors $T_A$, $T_B$ is observed when $Q_R = 0$. This originates mainly in the limited accuracy of individual calibration curves of the sensors and in limited repeatability of the sensors readings caused by thermal cycling. This difference $T_{AB0}$ is measured at the beginning $(T_A \approx T_B \approx T_R \approx 5 \text{K})$ of the calibration process and again before each emissivity measurement. A corrected temperature drop

$$T_{ABC} = (T_A - T_B) - T_{AB0}$$

is used both in calibration and in emissivity measurement procedure. The radiative heat flow $Q_R$ is evaluated from the calibration curve, $Q_R = Q_{HA}(T_{ABC})$. Before each measurement of temperature dependence of emissivity/absorptivity, the calibration procedure is conducted for values of $Q_{HA}$ at

$$\varepsilon(T_R) = C_\varepsilon \varepsilon_{RA}, \quad \alpha(T_A, T_R) = C_\alpha \varepsilon_{RA}. \quad (4)$$

The heat power $Q_R$ is used both in calibration and in emissivity measurement of radiative heat flow.

The gap $d_0$ can be taken as a typical setup.
several random temperatures $T_A$ to check calibration curve reproducibility.

The following effects on measurement result of radiative heat flow $Q_R$ are thus excluded:

- inaccuracy in the sensors calibration curves
- shift in sensors readings caused by thermal cycling between 320 K and 5 K
- systematic errors of the temperature controller inputs
- effect of sensors self-heating

Nevertheless the estimate of radiative heat flow $Q_R$ is still influenced by:

- stability of the temperature $T_B$ (below ±0.1 mK was observed for more than 5 h by Lake Shore 340)
- precision in reaching the steady state of the absorber temperature $T_A$ after a change of radiator temperature $T_R$ (a value of less than ±0.1 mK is achievable for $Q_R < 650 \mu W$ and ~1 mK for $Q_R > 650 \mu W$, nevertheless in real time measurement the stable value might be achieved with precision of 0.5% of $T_{ABC}$).
- accuracy of calibration points and interpolation between them
- leak of part of emitted radiation power from the gap between samples (see for correction of emissivity $\varepsilon_{RA}$ in section 3)

2.2. Heat flow measurement

2.2.1. Heat flow meter parameters. The resolution and the short term stability of $T_{ABC}$ measurement are better than 0.2 mK. With the sensitivity 25 $\mu W \text{ K}^{-1}$ of the thermal resistor it is possible to resolve heat flow down to ~50 nW. The range of the heat flow meter can be increased by one of two available thermal shunts added to the basic thermal resistor (sensitivity 90 $\mu W \text{ K}^{-1}$ or 1500 $\mu W \text{ K}^{-1}$), which also enables one to measure high values of emissivities up to room temperature. The absorber temperature $T_A$ increases above 5 K with increasing temperature $T_B$ in dependence on the absorbed heat flow and installed thermal shunt. Installation of the thermal shunt significantly lowers $T_A$ and also shortens time to reach thermal steady state of the heat flow meter.

2.2.2. Stabilization of temperature $T_B$. When measuring the lowest values of $Q_R$, the stability of the bottom temperature $T_B$ of the thermal resistor is important. The temperature $T_B$ is kept at 5 K with submilikelvin stability by a heater controlled by Lake Shore 340. Additionally, the pressure in the LHe Dewar vessel is stabilized to reduce the temperature fluctuations of the LHe bath below 10 mK. These precautions ensure that the temperature difference $T_{ABC}$ on the heat flow meter is caused only by the radiative heat flow between radiator and absorber. At higher heat flows ($Q_R > 650 \mu W$) where the thermal stabilization of $T_B$ is out of range, the stabilization of the pressure in the LHe bath influences measured heat flow by less than ~0.1%.

2.2.3. Radiator temperature range. An electrical heater attached to the radiator sets the temperature $T_R$ to values ranging from 10 K to about 320 K. Emission of radiation from the heater is suppressed by an aluminium shield. The heater is fed from the controller Lake Shore 340 in a closed loop.

2.2.4. Reference surfaces. Two types of ‘black’ reference samples are available for measurement of the emissivity or absorptivity (figure 3(b)). The first one, a flat reference sample, consists of an epoxy composite on a supporting copper disk of 40 mm in diameter and 1 mm in thickness (see upper picture of figure 3(b)). The composite itself is fabricated from four layers of polyester mesh impregnated by epoxy resin. The resultant organic layer is 0.38 mm thick. The second, a V-grooved reference sample (see lower picture of figure 3(b)) is a disk with a diameter 40 mm and 5 mm in thickness. The side facing the samples is shaped by 7 circular grooves coated with a layer of Fractal Black™ (Acktar Ltd.). The thickness of this layer varies within the range of 20–50 $\mu m$. For regular measurements the epoxy reference sample is used. The reason is its lower thermal capacity and higher mechanical resistance. Far infrared spectra of normal reflectivity of epoxy reference sample,
measured at wavelengths from 1 μm up to ~150 μm at room temperature, indicate nearly constant absorptivity \( \alpha_n \approx 96\% \) with small variations ±1%. By virtue of this result and moderate dependence of index of refraction on material temperature, we can suppose the epoxy reference as a grey sample within this region of wavelengths of radiation. Calculated hemispherical absorptivity \( \alpha_n = 96\% \) corresponding to the value \( \alpha_n \approx 96\% \) of normal absorptivity (for \( \alpha_n \) versus \( \alpha_h \) see sections 3–5.1 in [22]) is in good agreement with the hemispherical value \( \varepsilon_{REF} = 88\% \) (\( \varepsilon_{REF} \approx \varepsilon_{REF} \)) measured by the method described here, in which another epoxy sample and also V-grooved sample (figure 3) were used as a reference. From measurements we obtained equality between the total values \( \varepsilon_{REF}(T_A, T_R) \approx \varepsilon_{REF}(T_R) \), which is expected when dependence of optical properties of the reference sample on material temperature \( T_A \) is weak.

### Table 2. Measurement set up geometry and multiplying correction factors to be applied to \( \varepsilon_{RA} \).

| Quantity/Multiplication correction factor | Value [mm; %] | Expanded uncertainty \( (k = 2) \) [mm; %] |
|------------------------------------------|---------------|-----------------------------------|
| \( R \) | Samples radii | 20 mm | 0.05 |
| \( d \) | Gap between surfaces of the samples | 0.5 mm | 0.2 |
| \( e_m \) | Samples eccentricity | 0.2 | 0.2 |
| \( C_{\varepsilon, leak} \) | Leak of radiation in emissivity measurement \((7)\) | 1.074 | 3.7 + 3.3 \((d - d_0) + 8.9d\) | 5.5% |
| \( C_{\varepsilon, leak} \) | Leak of radiation in absorptivity measurement \((8)\) | 1.054 | 0.2 + 1.0 \((d - d_0) + 9d\) | 2.0% |
| \( C_{\varepsilon, exc} \) | Correction due to samples area eccentricity \((e_m = 0.2\ mm), (9)\) | 1.003 | \(0.014(e_m/d)^2(1 + (2d/d)^2)^{1/2}\) | 0.29% |
| \( C_{\varepsilon, exc} \) | Correction due to samples area eccentricity \((e_m = 0.2\ mm), (9)\) | 1.002 | \(0.008(e_m/d)^2(1 + (2d/d)^2)^{1/2}\) | 0.16% |
| \( C_{e, NG} \) | Non-grey surface of metallic sample, \( T_R/T_A > 3, (10b)\) | 1.000 | <0.001 | 0.03% |
| \( C_{e, REF} \) | Insufficient blackness, \((10a)\) and \((10b)\) \((n = 4/3),\) \( \varepsilon_{REF} = 88\%, \varepsilon < 10\%\) | 1.000–1.014 | <0.014 \(\varepsilon_{RA}\) | <0.14% |
| \( C_\varepsilon \) | Emissivity, final correction of \( \varepsilon_{RA} \) | \(C_{\varepsilon, leak}C_{\varepsilon, exc}C_{NG, REF}\) \(= 1.077 - 1.092\) | \(\approx \sqrt{\sum (\varepsilon_{exc})^2}\) | 5.9% |
| \( C_{\alpha} \) | Absorptivity, final correction of \( \varepsilon_{RA} \) | \(C_{\varepsilon, leak}C_{\varepsilon, exc}C_{NG, REF}\) \(= 1.056 - 1.071\) | \(\approx \sqrt{\sum (\alpha_{exc})^2}\) | 3.0% |

### 3. Corrections and uncertainties

To obtain an estimate of sample emissivity or absorptivity the values of measured mutual emissivity \( \varepsilon_{RA} \) are corrected (i) for leak of thermal radiation through the gap between samples, (ii) for samples’ eccentricity, (iii) for effect of non-grey surface of the measured sample, and (iv) for radiative properties of the used reference surface. The result of an emissivity \( \varepsilon \) or absorptivity \( \alpha \) measurement is then obtained by multiplication of measured mutual emissivity \( \varepsilon_{RA} \) by correction factors \((2b)\) by correction factors \((4)\), which are described in the following paragraphs and summarized in tables 1 and 2.

Uncertainties of all measured quantities and correction factors of \( \varepsilon_{RA} \) (tables 2 and 3) are combined into resulting uncertainty of the estimate of sample emissivity \( \varepsilon \) or absorptivity \( \alpha \) [23]. Two examples of corrected estimates of emissivity of
where $L$ stays for the fraction of leaked radiation. We suppose $L \ll 1$.

Radiation leak from the gap between a photons emitting Lambertian surface and fully absorbing reference surface is given by the configuration factor of two plane parallel concentric discs. For a typical setup with the gap $d_0 = 0.5 \text{ mm}$ and sample radius $R = 20 \text{ mm}$, a value of the configuration factor is 0.975 [22], which means a leak of 2.5% of transferred radiative heat power.

The actual amount of leaked radiation depends on the type of measured property (emissivity/absorptivity), on the values of emissivity/absorptivity of the samples, on the Lambertian versus non-Lambertian sample, on the type of non-Lambertian sample (dielectric/metal) and on surface morphology (specular/diffuse).

In the following text we assess correction factors for measurement of emissivity/absorptivity of metallic surfaces using the epoxy reference sample (figure 3). We calculated the leak numerically by Monte Carlo ray tracing, using real values of complex indexes of refraction of both opposite surfaces, distinguishing further between samples with diffuse and specular surfaces.

Extreme values of these distinguished leaks, $C_{\text{max}}$ and $C_{\text{min}}$ were applied to define correction factor and its uncertainty ($k = 2$) in the case of unclear effect of surface morphology on heat transfer:

$$C_{\text{leak}} = \frac{1}{2}(C_{\text{max}} + C_{\text{min}}) \pm \frac{1}{2}(C_{\text{max}} - C_{\text{min}}). \quad (6b)$$

In configuration of metallic sample versus epoxy reference sample having absorptivity 88% (figure 3) the leak between diffusion (Lambertian) surfaces with gap $d_0 = 0.5 \text{ mm}$ is about $L_{\alpha,d} = (3.0 \pm 0.2)\%$ and $L_{\alpha,d} = (5.4 \pm 0.2)\%$ in emissivity and absorptivity measurements, respectively. The difference is caused by the fact that in emissivity measurement most of the radiation is generated by the metallic sample (at least at $T_R > 20 \text{ K}$) and first impacts the reference where it is strongly absorbed, while in absorptivity measurement most of the radiation is nearly all reflected at the metal surface. Thus any additional leak after 1st, 2nd etc reflections is higher in the absorptivity measurement. Correspondingly we obtain the correction factors $C_{\alpha,d} = 1.030 + 0.056$ ($d - d_0$) and $C_{\alpha,d} = 1.054 + 0.100$ ($d - d_0$) where $d$ is in units of millimetres, see the 1st row in table 1. The last terms in these relations express variation of $C_{\text{leak}}$ due to the deviation of gap $d$ from the typical value $d_0$. In emissivity measurement of specular metallic samples (non-Lambertian) the calculated leak after all reflections is highest for metallic samples with the highest reflectivity, approaching the value $L_{\varepsilon,d} = (10.4 \pm 0.2)\%$ for the gap $d_0$ and radius $R = 20 \text{ mm}$. This leak $L_{\varepsilon,d}$ varies by 0.122 per 1 mm with varying gap $d$, thus the correction factor $C_{\varepsilon,d} \approx 1.104 + 0.122$ ($d - d_0$), see the 2nd row in table 1.

In the absorptivity measurement of a specular metallic sample the leak of radiation is slightly lower than in the case of diffuse surfaces. It results in a correction factor of $C_{\alpha,d} \approx 1.050 + 0.080$ ($d - d_0$). To obtain a correction factor taking into account all possible types of reflections from diffuse to specular, we use relation (6b) where we take into account uncertainty in the gap width $d$.

| Quantity | Quantity value | Expanded uncertainty ($k = 2$) | Contribution to uncertainty ($k = 2$) of emissivity—examples |
|----------|----------------|-------------------------------|----------------------------------------------------------|
| $T_R$    | Radiator temperature | 10–320 K | See (15) | $\varepsilon_{RA} = 0.384\%$ |
| $T_A$    | Absorber temperature | 5–30 K  | (0.001 $T_A + 0.005$) (K) | $T_R = 30 \text{ K}$ | 3.5% |
| $T_{ABC}$ | Corrected temperature | 0.001–25 K | (0.005 $T_{ABC} + 0.0002$) (K) | $T_R = 30 \text{ K}$ | 0.001% |
| $Q_R$    | Power measured by heat flow meter | $10^{-8}$–$10^{-2}$ W | $[(0.03)^2 + (n \delta T_{ABC}/T_{ABC})^2]^{1/2}$ | included in $Q_R$ | 7.8% |
| $C$      | Multiplying correction factor | 1.077–1.080 | See table 2 | included in $Q_R$ | 5.9% |
| $\delta$ | final fractional uncertainty ($k = 2$) of sample emissivity calculated in accord with equations (7) and (9)–(17) | 10.0% | 6.6% |
| $\varepsilon$ | result of measurement of sample emissivity (figure 4) includes corrections expressed by equations (7), (9) and (10a)–(10d) | $0.41 \pm 0.05\%$ | $1.89 \pm 0.13\%$ |
\[ C_{\text{max}} = C_{s,d} + 0.122 \delta d, \]
\[ C_{\text{min}} = C_{c,d} - 0.056 \delta d \text{ in emissivity measurement and} \]
\[ C_{\text{max}} = C_{d,d} + 0.100 \delta d, \]
\[ C_{\text{min}} = C_{o,s} - 0.080 \delta d \text{ in absorptivity measurement}. \]

Then the multiplying correction factor of mutual emissivity \( \varepsilon_{RA} \) which corrects leak of radiation from the gap and takes into account uncertainty in surface morphology (uncertainty in the type of reflections, specular or diffuse) results in accord with (6b) in following relation

\[ C_{c,\text{leak}} = 1.067 + 0.089 (d - d_0) + 0.033 \delta d \]
\[ \pm [0.037 + 0.033 (d - d_0) + 0.089 \delta d] \quad [1; \text{mm}] \]

(7)

for emissivity measurements of metallic samples and in

\[ C_{o,\text{leak}} = 1.052 + 0.090 (d - d_0) + 0.010 \delta d \]
\[ \pm [0.002 + 0.010 (d - d_0) + 0.090 \delta d] \quad [1; \text{mm}] \]

(8)

for absorptivity measurements of metallic samples. Hence the corrected emissivity and absorptivity are by \( \approx 7\% \) and \( \approx 5\% \) higher than the measured values.

3.2. Correction of \( \varepsilon_{RA} \) for samples eccentricity

Samples’ eccentricity \( e \) (distance between axes of the measured and reference samples) increases the leak of radiation analysed in previous section. We have evaluated its effect using configuration factors of plane parallel discs of slightly different radii. Using radii \( R_1 = R, R_2 = R + e \) and \( R_1 = R + e, R_2 = R \), and finding the difference between corresponding configuration factors, we have found the correction \( \approx 0.01(e/d)^2 \) for an increase in the leak when the diffusion (Lambertian) sample is opposite the 100\% black reference. We suppose that the percentage of this increase of total leak does not vary with variation of surface properties. Using this assumption and results from section 3.1, we obtained following corrections of eccentricity for discs of radius \( R = 20 \text{mm} \)

\[ C_{\text{exc}} \approx 1 + \left\{ \frac{\varepsilon_m}{d} \right\}^2 + \left\{ \frac{\varepsilon_m}{d} \right\}^2 \left( 1 + \left( 2 \frac{\delta d}{d} \right)^2 \right) \frac{1}{2}. \]

(9)

where \( c = 0.014 \) and \( c = 0.008 \) in emissivity and absorptivity measurement, respectively. We do not know the exact value of eccentricity, we assess its maximum as \( e_m = 0.2 \text{mm} \).

Emissivity \( \varepsilon_{RA} \) will be corrected by a multiplying factor \( 1 < C_{\text{exc}} < 1.0095 \) with uncertainty \( < 1\% \) \((k = 2)\) or \( 1 < C_{\text{exc}} < 1.0054 \) with uncertainty \( < 0.6\% \) \((k = 2)\). To calculate upper boundaries in these inequalities we used a minimum value of the gap, \( d = 0.3 \text{mm} \).

3.3. Correction for non-gray surface of the measured sample

For the temperatures above \( \sim 50 \text{K} \) the emissivity (absorptivity) of the flat reference and the V-grooved reference are \( \varepsilon_{\text{REF}}(T_A, T_R) \approx \varepsilon_{\text{REF}}(T_R) = 88\% \) and \( 95\% \), respectively, and exceed a value of 50\% above \( T_R > 15 \text{K} \) (figure 3(a)). This enables us to assume \( \varepsilon(T_R, \lambda)/\varepsilon_{\text{REF}}(T_A, \lambda) \ll 1 \). Then we can rewrite (2a) in the Taylor series in \( \varepsilon(T_R, \lambda) \) for the case of emissivity measurement.

\[ \frac{Q_R}{A} = \int_0^\infty \frac{\varepsilon(T_R, \lambda) - \varepsilon(T_A, \lambda)}{1 + \varepsilon_{\text{REF}}(T_A, \lambda)} \lambda d\lambda \]
\[ = \int_0^\infty \varepsilon(T_R, \lambda)[\varepsilon(T_R, \lambda) - \varepsilon(T_A, \lambda)] \lambda d\lambda \approx 1 + \varepsilon(T_R, \lambda) \left( \frac{1}{\varepsilon_{\text{REF}}(T_A, \lambda)} - 1 \right) \varepsilon(T_R, \lambda) \lambda d\lambda \]

(10a)

and analogically with \( \varepsilon(T_A, \lambda), \varepsilon_{\text{REF}}(T_R, \lambda) \) for measurement of absorptivity. The first term in the approximate relation in (10a) involves only property of the measured sample while the second one represents a correction for insufficient blackness of the reference sample.

Metals show weak wavelength dependence of spectral emissivity, \( \varepsilon(T, \lambda) \sim 1/\lambda^n \) with \( n \approx 0 - 2/3 \), in far infrared. For example absorptivity of gold, aluminium and copper weakly depends on wavelengths relevant to the maximum of emissive power of black-body at a temperature of about 300 K [7, 24]. On the other hand at cryogenic temperatures of radiation, with wavelengths in far infrared, a stronger dependence of up to \( n \approx 2/3 \) is expected, namely for metals proving anomalous skin effect [24]. Setting \( \sigma_{\text{REF}} = 100\% \) and \( \varepsilon(T, \lambda) \sim 1/\lambda^2 \) we obtain after integration of (10a)

\[ \frac{Q_R}{A} = \sigma \varepsilon(T_R) \left( T_A^4 - T_R^4 \left( \frac{T_A}{T_R} \right)^n \right) \]
\[ \frac{Q_R}{A} = \sigma \alpha(T_A, T_R) \left( T_A^4 - T_R^4 \left( \frac{T_A}{T_R} \right)^n \right) \]

and hence the correction factor

\[ C_{NG} = \frac{1 - (T_A/T_R)^4}{1 - (T_A/T_R)^{4+n}}. \]

(10b)

With the value \( n = 2/3 \), the correction factor \( C_{NG} \) gives an \( \varepsilon_{RA} \) correction of 2.5\% at \( T_R/T_A = 2 \) and is \(< 1\% \) for \( T_R/T_A \gg 3 \) (for example at \( T_R = 15 \text{K} \) and \( T_A = 5 \text{K} \)).

3.4. Correction for insufficient blackness of the reference sample

At first we suppose grey surfaces of both samples. From (10a) we obtain a correction for insufficient blackness of the reference sample (see (3))

\[ C_{\alpha, \text{REF}} \approx \frac{1}{1 - \varepsilon_{RA}(\varepsilon_{\text{REF}} - 1)} \]
\[ \pm \frac{\varepsilon_{RA}}{\varepsilon_{\text{REF}}} \left( \frac{\delta \varepsilon_{\text{REF}}}{\varepsilon_{\text{REF}}} \right)^2 + \left( 1 - \varepsilon_{\text{REF}} \right) \left( \frac{\delta \varepsilon_{RA}}{\varepsilon_{RA}} \right)^2. \]

(10c)
where we remember $\varepsilon_{\text{REF}} \approx \alpha_{\text{REF}}$ and suppose $\varepsilon_{\text{RA}} \approx \varepsilon$. For example, for measured values $\alpha \ll 1$ or $\varepsilon \ll 1$ and for the reference sample with absorptivity $\alpha_{\text{REF}} > 0.5$ (epoxy reference at $T_R > 15$ K, figure 3), this correction factor is close to unity.

$$1 < C_{\alpha,\text{REF}} < 1 + \varepsilon_{\text{RA}} \approx 1 + \varepsilon(T_R),$$
$$1 < C_{\varepsilon,\text{REF}} < 1 + \varepsilon_{\text{RA}} \approx 1 + \alpha(T_A, T_R).$$

with uncertainty much less than are fractional uncertainties $\partial \varepsilon_{\text{RA}} / \partial \varepsilon_{\text{RA}}$ and $\partial \varepsilon_{\text{REF}} / \partial \varepsilon_{\text{REF}}$.

As an example, for $\varepsilon$ or $\alpha = 0.1$ (10%) the corrections of $\varepsilon_{\text{RA}}$ for insufficient blackness of the reference surface are $C_{\varepsilon,\text{REF}}(\alpha_{\text{REF}} = 0.50) = 1.092$ and $C_{\varepsilon,\text{REF}}(\alpha_{\text{REF}} = 0.88) = 1.014$. Nevertheless for many metals, especially pure metals, usually characterized with emissivity/absorptivity of the order of magnitude 0.1%–1%, the correction and accompanying uncertainty are negligible or very small.

In the case of non-grey surfaces when the spectral emissivity of the reference sample depends modestly on wavelength of thermal radiation, the correction (10c) will be multiplied by a correction factor from (10b) using the exponent $4 + 2n$ instead of $4 + n$. We understand that (10c) will not be changed significantly for $T_R / T_A > 2$, and thus remains a good estimate of the effect of insufficient blackness of the reference surface in the measurement of emissivity/absorptivity of metallic surfaces.

3.5. Heat flow by residual gas

The vacuum in the apparatus is created by a turbomolecular pump before the cool down. During the measurement the vacuum is passively kept by cryopumping at a level lower than $10^{-7}$ Pa. Thus the parasitic heat flow conducted by residual gas is reduced below a measurable value.

3.6. Uncertainty in mutual emissivity $\varepsilon_{\text{RA}}$

The combined uncertainty of the estimated emissivity is calculated as a root of the sum of the squares of fractional uncertainties $\partial \varepsilon_{\text{RA}} / \varepsilon_{\text{RA}}$ of emissivity $\varepsilon_{\text{RA}}$ (2b) due to the uncertainties $\partial \varepsilon_{\text{RA}}$, $\partial T_A$, $\partial Q_R$, $\partial A$ of measured quantities and uncertainties of correction factors $C$ due to various effects (equations (7)–(10) and table 2).

$$\frac{\partial \varepsilon_{\text{RA}}}{\varepsilon_{\text{RA}}} = \sqrt{\left(\frac{\partial Q_R}{Q_R}\right)^2 + \left(\frac{\partial \varepsilon_{\text{RA}}}{\partial T_A} \frac{\partial T_A}{\varepsilon_{\text{RA}}}\right)^2 + \left(\frac{\partial \varepsilon_{\text{RA}}}{\partial A} \frac{\partial A}{\varepsilon_{\text{RA}}}\right)^2} + \sum \left(\frac{\partial C(\text{effect})}{C(\text{effect})}\right)^2$$

(11)

$$\frac{\partial \varepsilon_{\text{RA}}}{\partial T_R} \frac{\partial T_R}{\varepsilon_{\text{RA}}} \approx \frac{4}{T_R} \frac{\partial T_R}{T_R}, \quad \frac{\partial \varepsilon_{\text{RA}}}{\partial A} \frac{\partial A}{\varepsilon_{\text{RA}}} \approx \frac{4 T_R^4}{T_R} \frac{\partial A}{A} (T_R > T_A)$$

(12)

$$\delta A / A = \sqrt{(\epsilon_m / \pi R)^2 + (2 \epsilon R / R)^2}.$$  

(13)

Figure 4. Total hemispherical emissivity $\varepsilon(T_R)$ and absorptivity $\alpha(T_A, T_R)$ of a chemically polished aluminium sample in dependence on radiator temperature $T_R$. The temperature of the aluminium sample during the entire absorptivity measurement is kept at values $T_A = 5 \text{ K} - 35 \text{ K}$, which is within the temperature interval where absorptivity of this sample does not depend on the temperature of material, $\alpha(T_A, T_R) = \alpha(T_R)$. From the approximate equality $\varepsilon(T_R) \approx \alpha(T_A, T_R)$ at $T_R \leq 60 \text{ K}$ we can see that it is true up to $T_A \approx 60 \text{ K}$.

3.7 Uncertainty in temperatures $T_A$ and $T_R$

Absolute uncertainties of measured temperatures $T_R$ and $T_A$ are given by the parameters of the temperature controller Lake Shore 340 and of the sensors. The values derived from the LakeShore catalogue data were used for the calculation of the uncertainty associated with $\varepsilon_{\text{RA}}$. These LakeShore values are expanded uncertainty based upon a 95% confidence limit (coverage factor $k = 2$).

$$\delta T_A = 0.001 \ T_A + 0.005 \ (\text{K})$$

(14)

and $\delta T_R = 0.25 \ (\text{K})$ at $T_R < 60 \ (\text{K})$, respectively.

(15)

Equation (14) takes into account electronic accuracy of Lake Shore 340 temperature controller together with accuracy of calibrated Cernox™ sensor CX 1050 SD. The values in equation (15) are based on uncertainties of the Standard curve 10 at $T_R < 60 \ (\text{K})$ and 3-point SoftCal™ software calibration at $T_R > 60 \ (\text{K})$ for the used sensor DT 470 SD (class 11A).

3.8. Uncertainty in measured heat flow $Q_R$

The heat flow $Q_R$ is obtained by interpolation of the calibration curve $Q_{\text{HA}}(T_{\text{ABC}})$. An uncertainty less than 3% (coverage factor $k = 3$) in points $Q_{\text{HA}}(T_{\text{ABC}})$ and interpolation between them was obtained from repeated check calibrations performed at randomly chosen points $T_{\text{ABC}} > 10 \ (\text{mK})$. Within the uncertainties of measurement, the calibration curve is linear for values $T_{\text{ABC}} < 100 \ (\text{mK})$. Due to smoothness of temperature dependences of thermal conductivity of used materials we can suppose linear behaviour of the calibration curve also below $T_{\text{ABC}} \approx 10 \ (\text{mK})$ down to zero values of $Q_{\text{HA}}$ and $T_{\text{ABC}}$ with the same uncertainty, 3%.
The resulting uncertainty of heat power absorbed by the sample at temperature $T_A$ is a combination of the uncertainty of measured $T_{ABC}$ and of calibration values $Q_{HA}$.

As the exponent in relation for sensitivity $\frac{\partial Q_{HA}}{\partial T_{ABC}} = n \frac{Q_{HA}}{T_{ABC}}$ varies from $n \approx 1$ at values $T_{ABC} < 1$ K to $n \approx 2$ at the highest values of $T_{ABC}$, we can write

\[
\frac{\delta Q_R}{Q_R} = \sqrt{0.03^2 + \left( \frac{\partial Q_{HA}}{\partial T_{ABC}} \delta T_{ABC} \right)^2} \leq \sqrt{0.03^2 + \left( \frac{2 \delta T_{ABC}}{T_{ABC}} \right)^2},
\]

where

\[
\delta T_{ABC} = (0.005 T_{ABC} + 0.0002) \quad (K)
\]

is the standard uncertainty of the corrected temperature drop $T_{ABC}$ on the heat flow meter. The first term in (17) stays for uncertainty of the steady-state value of $T_{ABC} (\approx 0.5\%)$ in real time measurements. The second term, given by the 0.1 mK resolution of $T_A$ and $T_R$, dominates in measurements at low values of $T_{ABC} (<10$ mK).

4. Example of measurement and results verification

The capabilities and possible limitations of the presented method are demonstrated in figures 4–6. The measurement of total hemispherical emissivity $\varepsilon$ and absorptivity $\alpha$ (figure 4) of chemically polished aluminium 99.5% was conducted with a thermal shunt (bronze tape added to the basic thermal resistor of the heat flow meter) in the temperature range from 20 K to 320 K. A comparative measurement without the shunt, which is more sensitive at low temperatures, gave the same results at $T_R \geq 20$ K.

Error bars present expanded fractional uncertainties ($k = 2$) calculated in accordance with the section 3. Values are corrected for the leak of thermal radiation from the gap, samples’ eccentricity, non-greyness of sample surface and radiative properties of the reference sample.

To verify the method described here, we confront our current results on total hemispherical emissivity of pure Al sample with our measurement from 2005 [4] and with those obtained by other authors (figure 5) who used some of the calorimetric methods described here in the introduction. Figure 5 also includes two values of the total hemispherical emissivity at 300 K calculated from formerly published optical constants of aluminium [25, 26]. Tsujimoto et al [7] gained their results from spectral measurements of bulk Al. They used Al and Au standards for measurement of infrared spectra (wavelengths 1–20 μm) of normal reflectivity at temperatures between 16 and 306 K and calculated total hemispherical absorptivity and emissivity from a proposed model.

The Al samples compared in figure 5 were provided with different surface treatments such as chemical polishing (our measurement), electro-polishing [10], fine mechanical polishing followed by annealing [7] or evaporated Al layers [17, 25, 26]. Despite different approaches to emissivity measurement, there is generally a good match among presented experimental methods.

The value of total hemispherical absorptivity obtained at $T_R = 300$ K in our measurement ($\alpha = 1.2\%$; figures 4 and 6) is fairly consistent with $\alpha = 1.1\%$ achieved by Ramanathan [14] (figure 6). In his experiment the lower value of absorptivity may be ascribed to the losses of thermal radiation in the guiding tube and window between the source and the sample. The difference, up to ~25%, between ours and Tsujimoto et al [7] data on $\alpha$ and $\varepsilon$ is not surprising if we take into account the limited range of experimental wavelengths and accuracy.
5. Conclusions

We present a method for the measurement of temperature dependence of total hemispherical emissivity and absorptivity in dependence on the temperature of thermal radiation between ~ 20 K and 320 K. We performed a detailed analysis of corrections for possible disturbing effects together with a determination of uncertainty of the measurements done by a specific apparatus using this method. The method is especially suitable for the measurement of highly reflective materials like metals. We verified the method comparing the results for pure aluminium with published results gained within several other methods. Our data were corrected for leak of radiation due to sample configuration, the non-grey surface of the measured sample, and the insufficient blackness of the reference where the last two corrections and their uncertainties are negligible. The leak of radiation is mainly responsible for the total value of the multiplying correction factor 1.08 and 1.06 in emissivity and absorptivity measurements. Expanded fractional uncertainty (coverage factor $k = 2$), including the uncertainties of temperature measurements and correction factors, is less than 11% for our measurement of aluminium emissivity at $T_R \approx 30$ K and below 7% at temperatures above 60 K. In absorptivity measurement the values of uncertainty are slightly lower.

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Figure 6. Total hemispherical absorptivity of pure aluminium. Comparison of temperature dependence of our data (measured in 2005 and 2015 on the chemically polished sample) with those obtained by other methods. Samples’ characteristics: chemically polished rolled sheet of Al 99.5% (our measurements), Ramanathan’s value [14] was measured on Al 99.99% cut from a sheet and finished by electrolytic polishing. Tsujimoto et al [7] sample, see the caption of figure 5.

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