OPTICAL THERMOPHYSICAL CHARACTERISTICS OF THE CONDENSED PHASE OF COMBUSTION PRODUCTS AND HEATPROOF MATERIALS

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An unconventional unit for experimentally studying the optical thermophysical properties of materials over a wide temperature range is described. Results are presented of studying the temperature function and dispersion of the absorption index for the condensed phase of the combustion products of a metalbearing fuel and of the emittance of fiber fireproof materials.

One of the ways of improving the efficiency, energy capacity and economy of high-temperature thermal-power units is to increase the temperature in the combustion chambers, which, in turn, requires the solution of some challenging problems of heat transfer both within the working medium and between the working medium and the chamber walls, as well as to produce new heatproof materials able to operate in aggressive high-temperature media. The use of metal-bearing fuels necessitates experimental investigations of the optical thermophysical characteristics of the substance of condensed combustion products of such fuels and of the materials of the chamber walls.

At present, the experimental determination of the radiative and optical properties of materials involves a variety of methods based on the measurement of reflectance, absorptance, and emittance, and on the analysis of the degree of polarization of the reflected radiation [1-3]. The choice of a specific method is dictated by the type of material considered, its temperature state, and other factors.

The condensed combustion products of aluminized fuels investigated by the present authors consists of more than 90% of the α phase of aluminum oxide and about 1.5% of iron oxide and 2% of silicon oxide. A spectral analysis of the samples revealed the presence of compounds of carbon, copper, nickel, bismuth, chromium, magnesium, manganese, and other elements. Plane-parallel plates of thickness from 43 to 120 μ m were fabricated from the condensate melt. Surfaces of the plates were polished to the 11-12th grades of roughness.

Preliminary experimental studies of the optical properties of condensed combustion products in a temperature range up to 1200 K revealed insufficient accuracy of the reflection methods because of semitransparency of the samples. Moreover, it is well known that emitting (absorptive) properties of the substance of dielectrics are determined mainly by the absorption coefficient (index) and studies of clean polycrystalline samples of aluminum oxide show a slight linear temperature dependence of the refractive index. Therefore, for studying real combustion products we selected the transmission method. Here, we used an amplitude modulation of the probing radiation to tune out the emitted radiation of the sample in conditions of high temperatures.

In order to experimentally study the transmission coefficient for semitransparent samples of the substance of the combustion products under conditions of high temperatures (up to 2300 K), we designed a high-temperature experimental setup, whose optical circuit is given in Fig. 1. The substance sample in the form of a $7 \times 7 \text{ mm}^2$ plane-parallel plate was located in the central channel (with \emptyset 4.5 mm) of a graphite cylinder installed in a single-coil inductor of a VChG4-25/0.44 high-frequency generator. The heating temperature of the sample was measured by a VR 5/20 tungsten—rhenium thermocouple. The inductor with the sample was placed in a pressurized water-cooled chamber with fluorite windows, which is filled with an inert gas (argon) having little excess pressure. As the source of probing radiation we employed a glass-carbon flat plate of SU-2000 grade, which was heated by alternating current up to a temperature of 2300 K. The

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Fig. 1. Basic circuit of the experimental setup: 1) source of probing radiation; 2, 9) CaF_2 windows; 3) quartz window; 4) emitting glass-carbon plate; 5) LOP-72 pyrometer; 6) diaphragms; 7) modulator; 8) pressurized cooled chamber; 10) sample considered; 11) graphite cylinder; 12) inductor; 13) cassette with sets of light filters; 14) optical acoustic receiver.

temperature of the plate surface was controlled by the an LOP-72 optical pyrometer. The radiation from the heated glasscarbon plate was modulated by a mechanical modulator and directed to the sample considered. Having passed through a monochromator representing a block of limiting apertures and rotary drums with sets of light filters, the radiation transmitted by the sample reached the entrance part of an OAP-5M optical acoustic receiver, whose signal was amplified and brought, in the form of standard signal up to 10 mV, to the digital voltmeter. The current study used a set of interference and dispersion filters with transmission bands in the spectral range 0.63-6.1 μ m, which was stipulated, according to the Planck distribution, by the main contribution of the radiation of this spectral range to the radiative heat transfer at the realized temperature levels. Band-pass reflecting light filters were employed to suppress the background radiation.

The experiment began with a measurement of the intensity of the probing radiation in the absence of the sample from the channel of the graphite cylinder, after which the sample was placed in the cylinder channel and a run of measurements of the temperature (up to 2300 K) dependence for the spectral transmittance was performed. The surface temperature of the emitting glass-carbon plate in the source of the probing radiation was maintained constant throughout the measurement run.

It is well known [4] that the method of determining the spectral absorption coefficients K_{λ} and, correspondingly, the absorption indexes which are correlated as $\varkappa_{\lambda} = \lambda \cdot K_{\lambda}/4\pi$ has the greatest precision with the condition $K_{\lambda}d = 1$ met, where d is the thickness of the semitransparent layer, and the error of determining the spectral absorption coefficient K_{λ} within $0.1 < K_{\lambda}d < 10$ does not exceed 10%. Analyzing the experimental conditions and errors of the measuring apparatus with a view to the fact that the measured values of the spectral transmission coefficients D_{λ} of 5-15% indicate agreement with the criterion of the range of minimal error ($K_{\lambda} \sim 10^2 \text{ cm}^{-1}$, $d \sim 10^{-2} \text{ cm}$, and $K_{\lambda}d \sim 1$) and the possibility of ruling out the effect of multiple reflections in the bulk of the semitransparent plate permits us to evaluate the total relative error of measurements within 10% of the absorption coefficient.



Fig. 2. Spectral absorption indexes for a condensed phase of combustion products at T = 1900 K for samples of various thicknesses: 1) d = 43.3 μ m; 2) 66.2; 3) 76; 4) 78; 5) 81; 6) 80.3 μ . λ , μ m.

Figure 2 plots results for the dispersion of the absorption index for the condensed phase of the combustion products over a wavelength range of 0.63-6.1 µm at a temperature of 1900 K. The absorption indexes obtained for the condensed phase of the combustion products in the considered temperature and wavelength intervals range from $2 \cdot 10^{-3}$ to $3 \cdot 10^{-2}$, i.e. are about two orders of magnitude greater than the corresponding absorption indexes for leucosapphire [5]. Such a distinction is due to the fact that, apart from aluminum oxide, the samples contain up to 8% of impurities. The results given in Fig. 2 also show that with increasing thickness of the sample considered, the measured values of the absorption indexes of the condensed phase decrease, this relationship being observed over the entire temperature range of 300-2300 K studied by the authors. Such dependence of the obtained absorption indexes on the sample thickness may be attributed to the presence on the samples of surface layers having a larger absorption index than the basic material of the sample. As the sample thickness decreases the relative contribution of the surface layer to the measurement result is enhanced. The formation of such surface layers is likely related to the surface grinding and polishing in manufacturing the samples from the condensate melt. It has been known that in abrasive treatment (grinding and polishing) of the glass, a disrupted layer consisting of relief and crack layers forms on its surface. The thickness of the disrupted layer is comparable with the average grain sizes of abrasive powders [6]. In the sample polishing we used diamond pastes with granularity ranging from 5/3 to 1/0 and, therefore, the thickness of the disrupted surface layer could amount to 3-5 μ m. During polishing, the microcracks can be clogged up with the disintegration products both of the polished and abrasive substances, i.e., the structure and the optical properties of the substances of the surface and main layers may differ. Since the polishing technology was identical for all samples, it may assumed that the thicknesses and the optical properties of the surface layers for all samples are the same.

If the absorption index for the substance of the main layer of the samples is represented by \varkappa_0 , the absorption index of the substance in the surface layer will vary from a certain value \varkappa_s on the outer boundary to \varkappa_0 on the boundary between the surface and main layers of the sample (Fig. 3). A character of variation in the absorption index across the thickness of the surface layer was prescribed by the exponential relation

$$\varkappa(x) = \varkappa_0 \exp\left[a\left(d_s - x\right)\right],\tag{1}$$

where x is the distance from the outer boundary of the surface layer and a is a constant.

In accordance with the Bouguer law, on incidence of a monochromatic light flux of intensity I_0 on a homogeneous plane sample of thickness d, the intensity at the sample exit will decrease and equal

$$I_{\lambda} = I_{0\lambda} \exp\left(-\frac{4\pi\varkappa_{\lambda}}{\lambda} d\right), \qquad (2)$$



Fig. 3. Model of variation in the absorption index of the substance across the thickness of the samples considered (I_0 , intensity of probing radiation; 1, intensity of radiation transmitted by a sample).

where λ is the radiation wavelength and \varkappa_{λ} is the spectral absorption index of the substance. For the sample depicted in Fig. 3, the radiation intensity at the sample exit is defined by the relationship

$$I_{\lambda} = I_{0\lambda} \exp\left[-\frac{4\pi\varkappa_{0\lambda}}{\lambda}(d-2d_{s})\right] \exp\left[-2\frac{4\pi}{\lambda}\int_{0}^{d_{s}}\varkappa(x)\,dx\right].$$
(3)

With consideration of the chosen law of variations in the absorption index across the thickness of the surface layer (1), expression (3) rearranges to the form

$$I_{\lambda} = I_{0\lambda} \exp\left\{-\frac{4\pi\varkappa_{0\lambda}}{\lambda}\left[(d-2d_{\rm s}) - \frac{2}{a} + \frac{2}{a}\exp\left(ad_{\rm s}\right)\right]\right\}.$$
(4)

On expanding the function $exp(ad_s)$ into a series

$$\exp(ad_{s}) = 1 + ad_{s} + \frac{a^{2}d_{s}^{2}}{2!} + \ldots + \frac{a^{n}d_{s}^{n}}{n!} + \ldots$$

and on substituting the first three terms of the series into Eq. (4), we obtain

$$I_{\lambda} = I_{0\lambda} \exp\left[-\frac{4\pi\kappa_{0\lambda}}{\lambda} \left(d + ad_{s}^{2}\right)\right], \qquad (5)$$

$$\ln \frac{I_{\lambda}}{I_{0\lambda}} = \ln D_{\lambda} = -\frac{4\pi\varkappa_{0\lambda}}{\lambda} (d + ad_{s}^{2}), \qquad (6)$$

$$\varkappa_{0\lambda}d + \varkappa_{0\lambda}ad_{s}^{2} = -\frac{\lambda}{4\pi}\ln D_{\lambda}.$$
(7)

Having assumed that the quantities $\varkappa_{0\lambda}$, a, and d_s are independent of the sample thickness and have the same values, and having written Eq. (7) for two samples of thicknesses d_i and d_j, we derive a formula for calculating the absorption index for the substance of the main layer of the samples considered

$$\varkappa_{0\lambda} = \frac{\lambda}{4\pi (d_i - d_j)} \ln \frac{D_j}{D_i} \,. \tag{8}$$



Fig. 4. Temperature dependences of the spectral absorption index for the substance of the main layer of samples: 1) λ = 0.63 μ m; 2) 1.81; 3) 2.04; 4) 2.49; 5) 3.0; 6) 3.5; 7) 4.8; 8) 5.4; 9) 6.1 μ m. T, K.

Fig. 5. Temperature dependences for the integral emissivity of fiber refractories based on mullitosilicone: 1) binder of pure silica ($AI_2O_3 - 51\%$; $SiO_2 - 49\%$); 2) binder of silica with NH₂Cl additive ($AI_2O_3 - 51\%$; $SiO_2 - 48.3\%$; NH₄Cl - 0.7\%); 3) binder of silica with (NH₄)₂SO₄ additive ($AI_2O_3 - 51\%$; $SiO_2 - 48.3\%$; (NH₄)₂SO₄ - 0.7\%); 4) combined binder ($AI_2O_3 - 31.5\%$; $SiO_2 - 45\%$; MgO - 8%; CaO - 14.5%; Fe₂O₃ - 1%).

The calculated values of $\varkappa_{0\lambda}$ are about 1.5-2 times smaller than the corresponding values of the spectral absorption index for the condensed phase, obtained without regard for the effect of the surface layer. The values of \varkappa were calculated in both cases with allowance for multiple reflections of radiation in the samples.

The temperature dependences of the spectral absorption index for the condensate $\varkappa_{0\lambda}$ (Fig. 4) are appreciably weaker than for leucosapphire. Apparently, this is associated with the fact that, rather than consisting of pure aluminum oxide the samples contain up to 8% of an impurity. Study [7] notes a similar impact of contamination on the temperature dependence for the spectral emittance of refractories based on magnesitochromite. This dependence is pronounced for a pure refractory and weak for a contaminated one.

To study the temperature (750-1200 K) effect and the composition of special fiber refractory materials on their total emittance, we designed a device wherein the measurement was performed in an inert atmosphere by comparing the intensities of heat radiation from the sample considered with the standard (polished tungsten); in this case a thermal element with a potassium bromide window served as the radiation receiver. The temperature of the sample surface was measured by a tungsten-rhenium VR 5/20 thermocouple. In conformity with the assessments of the error of determining the emittance, the major contribution to the error is introduced by a fallacy in the measurement of the temperature of the sample surface, and the total error of the obtained values of the total emittance is 25% for a temperature of 800 K and 11% for 1200 K.

Figure 5 presents experimental results for the effect of the filler temperature and composition on the total emittance of fiber refractories. We considered four samples based on mullitosilica fiber with a silica binder. We established that the total emittance ranges within 0.18-0.35 with the temperature varying from 700 to 1200 K and increases actually by a linear law as the temperature rises. As for the influence of the chemical composition of the filler on the integral emissivity, it is insignificant and lies within the experimental error. It should be pointed out that, with low temperatures (of about 770 K), the emittance of the ceramic fiber plates in question is in favorable agreement with the data for the SiO₂ powder at 300 K ($\epsilon \approx 0.3$) and for the Al₂O₃ powder ($\epsilon \approx 0.16$). For crystalline silica, $\epsilon \approx 0.8$ at T = 773 K, whereas, for aluminum oxide, $\epsilon \approx 0.7$. In this connection, we should also remark that the emittance of wires is lower than that of a flat surface because of violation of the Lambert law, and this leads, to some extent, to a reduction in the emissivity of the fiber material in comparison with a flat surface. However, a more prominent role in the decrease of the effective emittance both for the fiber and for the powders is played by the fact that they have a lower thermal conductivity as compared with monolith, and the radiation is formed in a certain layer Δd rather than on the surface. In this layer, with a significant temperature drop between the "inner" and "outer" planes (of 200 K mm⁻¹), part of the radiation is absorbed and scattered, which can result in a marked reduction in the emittance as compared with massive samples.

DEDUCTION

1. We have created a unique device permitting one to study the optical thermomechanical properties of materials over wide ranges of wavelengths and temperatures.

2. We have derived data on the temperature dependence and dispersion of the absorption index for the substance of the condensed phase of an aluminized fuel in a wavelength interval of 0.63-6.3 μ m and with temperatures ranging from 300 to 2300 K. The effect of the surface layer of the semitransparent samples considered on the measurement results has been demonstrated.

3. We have obtained temperature dependences for the total emittance of fiber refractory materials based on mullitosilica.

NOTATION

 K_{λ} , spectral absorption coefficient; D_{λ} , spectral transmission coefficient; \varkappa_{λ} , spectral absorption index; λ , wavelength; d, sample thickness; d_s, surface layer thickness; I_{λ}, spectral intensity of radiation; ϵ_{λ} , ϵ , spectral and total emittances.

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