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Behavior of Thermal Radiation Properties of Oxide Ceramics Under Subsecond Laser Heating[1]

G. A. Kobzev[2] and V. A. Petrov[2]

It is shown that the data on spectral emissivity and reflectivity obtained in subsecond laser heating experiments differ from values which are obtained by the conventional methods. Owing to the semitransparency of oxides, these thermal radiation properties depend on the temperature distribution in near-surface layers and cannot be related to a definite temperature value. The condensation of the products of oxide evaporation at high temperatures, which takes place in air surrounding a sample, may affect the measurement of temperature, reflectivity, and emissivity.

KEY WORDS: absorption coefficient; ceramics; emissivity; laser heating; reflectivity; high temperatures.

1. INTRODUCTION

Refractory oxide ceramics are used in various high-temperature equipment as thermal insulating and heat protective materials. They quite often surround an oxidizing medium under intensive heating when the temperature reaches 3500–4000 K and an ablative layer of a melt on their surface exists. Information on the thermal radiation characteristics of ceramics is required to calculate the radiation and combined radiation and conduction heat transfer in the thermal insulation of various high-temperature apparatus.

The majority of the materials used in high-temperature technologies that are fabricated from refractory oxides is polycrystalline ceramic materials with a certain porosity. They relate to the class of semitransparent volume scattering materials. When a transport equation is used to describe the

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[2] Institute for High Temperatures, Russian Academy of Sciences, Moscow 127412, Russia.

propagation of the radiation, one has to use the entire set of optical properties including absorption coefficient k, refractive index n, scattering coefficient β, and phase function γ of volume scattering. The magnitude of the latter depends on the direction of the incident and scattered radiation. The temperature fields and energy fluxes in such a materials can be calculated by using both the radiation transfer equation and the energy conservation equation if one knows all four optical parameters indicated above as a function of the temperature and wavelength.

At present, however, such information is rarely available for materials. Practically, the calculations can be performed only for one-dimensional cases, and even here many simplifying assumptions must be introduced [1]. In practice, the averaged thermal radiation characteristics are used as a rule, namely, the reflectivity and emissivity. But both quantities can be obtained only experimentally, and this means that studies must be conducted at temperatures up to 3500–4000 K in an oxidizing atmosphere. Until recently, such high temperatures were obtained only in solar or arc image furnaces. In these cases, however, the study of the radiation characteristics is hindered by the high reflectivity of the oxides in the region of the spectrum of solar and arc radiation. This makes it difficult to register the flux of the intrinsic radiation of the material being studied and lowers the upper temperatures. In this connection, the use of the radiation of CO_2 lasers with a wavelength of about 10.6 μm for heating is very promising. This radiation belongs to the region of opacity (the region of high absorption with $k \propto 10^3\ \mathrm{cm}^{-1}$) of virtually all refractory oxides. At present, there is no problem in obtaining several kilowatts of power for such lasers under laboratory conditions, and this power can be utilized very efficiently because of the low magnitude of reflection at 10.6 μm (several percent).

In spite of the great possibilities and obvious advantages of using CO_2 laser radiation for heating oxides, we cannot assume that the procedure of studying the radiation characteristics under the conditions of such heating is completely developed. The first experiments involving the use of laser subsecond heating for studying the radiation characteristics of oxides were apparently conducted by Bober et al. in 1977 [2]. This work was further developed also by Bober et al. [3]. But the results of these studies were treated without considering the influence of the peculiar character of the temperature field formation in refractory oxides under intensive laser heating conditions and, therefore, contain a number of errors. This was pointed out for the first time by Petrov and Chernyshev [4].

2. TEMPERATURE DISTRIBUTION IN OXIDE CERAMICS UNDER LASER HEATING

Let us consider, as an example, the results of calculating the temperature distribution when alumina ceramics are heated in vacuum by CO_2 laser radiation. The physical and mathematical models describing the processes of heating ceramics by laser radiation are described in detail by Galaktionov and Stepanov [5].

The model considers fusion and ablation of the material, combined radiative and conductive heat transfer in the ceramics, and dependence of the optical and thermophysical properties on temperature and porosity. The model includes the energy equation, the radiation diffusion equation for the thermal radiation, the radiation transfer equation for the directional component of the laser radiation, the radiation diffusion equation for the scattered component, and several additional relations and initial conditions.

Figure 1 represents curves showing the calculated distribution of the

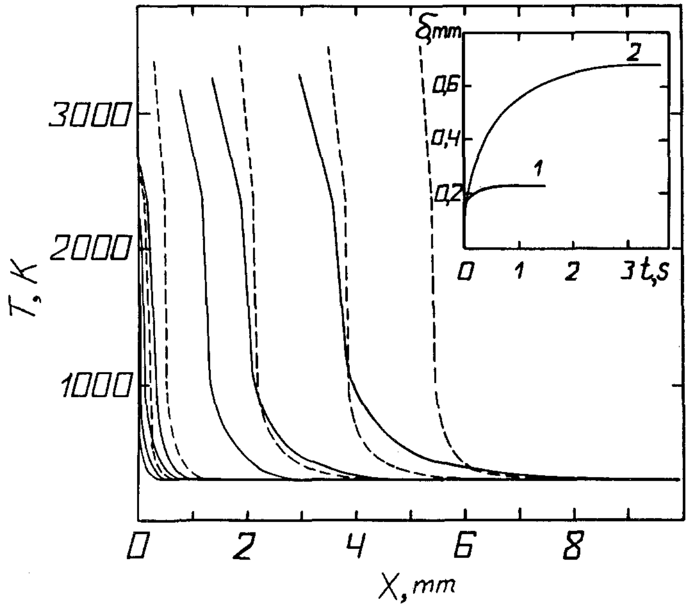

Fig. 1. Temperature distribution in a layer of alumina ceramics when heating with various q at different times t. Solid lines: $q = 2\ \text{kW}\cdot\text{cm}^{-2}$; $t = 3.2\times10^{-3}$; 9.2×10^{-3}, 0.023, 0.051, 0.4, 0.8, and 2.0 s. Dashed lines: $q = 6\ \text{kW}\cdot\text{cm}^{-2}$; $t = 9.2\times10^{-3}$, 0.05, 0.4, 0.8, and 1.2 s. The inset shows the time dependence of the melt thickness δ: $q = 6\ \text{kW}\cdot\text{cm}^{-2}$ (curve 1); $q = 2\ \text{kW}\cdot\text{cm}^{-2}$ (curve 2).

temperatures in ceramics with a density of 1.1 $g \cdot cm^{-3}$ (a porosity of 73%) at different times when heating with two different fluxes: $q_1 = 2$ and $q_2 = 6\ kW \cdot cm^{-2}$. It was assumed that the initial thickness of the ceramics layer is 1 cm and that its right-hand boundary is adiabatically insulated. It can be seen that the temperature of the irradiated surface grows very rapidly. The melting point (2320 K) is reached in the first and second cases in 2.3×10^{-2} and 4.8×10^{-3} s, respectively. The temperature profile in the near-surface layer is a very steep. The temperature gradients at the surface before melting are 8360 and 13,000 $K \cdot mm^{-1}$.

Though the density of molten aluminium oxide is about 30% lower than the density of a single crystal, in melting the surface moves to the right because of closing up of the pores. The surface begins to move rapidly when the temperature reaches the value at which the rate of evaporation becomes noticeable. Finally, some time after the beginning of heating, the speed of the front boundary becomes virtually constant, and the temperature field becomes close to a quasi-steady one.

It can be seen from Fig. 1 that at different values of q both before and after melting the temperature gradients in the near-surface layer appreciably differ. The melt thickness also differs. In the quasi-steady state it is 0.67 and 0.23 mm, respectively. The temperature curves show that at the same surface temperature we obtain different temperature distributions adjacent to the surface layer, and this should result in differences in the energy irradiated by the material. Obviously there must also be the corresponding differences in the reflectivity because the temperature will greatly affect the optical properties.

3. REFLECTIVITY AND EMISSIVITY OF CERAMICS UNDER LASER HEATING

To establish what thickness of the near-surface layer determines the radiation characteristics, we must have the optical properties of the material. For the alumina ceramics being considered, the absorption coefficient in the region of high transparency (e.g., at a wavelength of 1.15 μm) in the solid phase near the melting point $k = 0.16\ cm^{-1}$, while the scattering coefficient $\beta = 250\ cm^{-1}$; in the melt, on the other hand, the absorption coefficient is about 300 cm^{-1}. It means that the thickness of the layer determining the radiation characteristics will be 0.17 mm for the indicated values of the optical properties in a melt and over 1.2 mm in the solid phase. The last value has been obtained by extrapolating the dependence of the emissivity on the optical thickness for a scattering albedo of $\omega = 0.99$ given in Ref. 6. We obtained $\omega = 0.99936$ provided that the entire layer has a temperature of 2300 K. With a view to the lowering of the temperature with depth, ω will be still greater.

If we make an estimate of the thickness of the emitting (and reflecting) layer and compare it with the results of calculating the temperature distribution in the course of heating (Fig. 1), we should be able to see that up to melting at a surface temperature of 2300 K, the radiation characteristics are determined by the layer whose thickness is greater than that of the heated layer at both values of q.

The temperature difference is about 240 and 860 K in a melt with heating from $q_1 = 2$ and $q_2 = 6\ \mathrm{kW \cdot cm^{-2}}$, respectively, at a thickness of 0.17 mm under quasi-steady-state conditions. This is substantially lower than the temperature difference at the thickness of the emitting layer in the solid phase with a surface temperature close to the melting point.

The radiation characteristics (reflectivity and emissivity) in the heating of oxide ceramics by laser radiation are not fixed physical constants that can be related to the surface temperature but depend on the flux of the heating radiation. This means that if we refer to the magnitude of the reflectivity R but we do not know the temperature to which this quantity should be related, we cannot define the emissivity ε in its conventional sense because what we have here is the irradiation from a nonisothermal layer.

The same relates to the values of the temperature T measured by optical pyrometers. The measured value of the temperature, which we call the effective one T_{ef}, corresponds to the temperature of a blackbody whose radiation at the wavelength of the pyrometer has the same intensity as the radiation reaching the pyrometer from a layer of infinite optical thickness adjoining the surface of the sample being studied. All this introduces major complications into the use of laser heating for determining the radiation characteristics of refractory oxide.

Let us consider the influence of the flux q in greater detail. Figure 2 gives, as an example, the results of our experiments involving the measurement of the normal-hemispherical reflectivity R when heating samples of polycrystalline magnesium oxide of commercial purity mixed with 1% of calcium oxide in air by CO_2 laser radiation. The reflection was measured at a wavelength of 1.15 μm belonging to the region of high transparency, while the reduced values of the effective temperature $T_{1\mathrm{ef}}$ correspond to the radiation at a wavelength of 0.55 μm registered by a two-wavelength pyrometer. The second wavelength was equal 0.72 μm and the corresponding temperature was designated as $T_{2\mathrm{ef}}$. For heating up to $T_{1\mathrm{ef}} = 2000$ K, the values of reflectivity for all q's are close and are lower by only 2–4% than its value at room temperature. Reflection diminishes quite rapidly beginning at a definite value of the temperature $T_{1\mathrm{ef}}$. This tendency

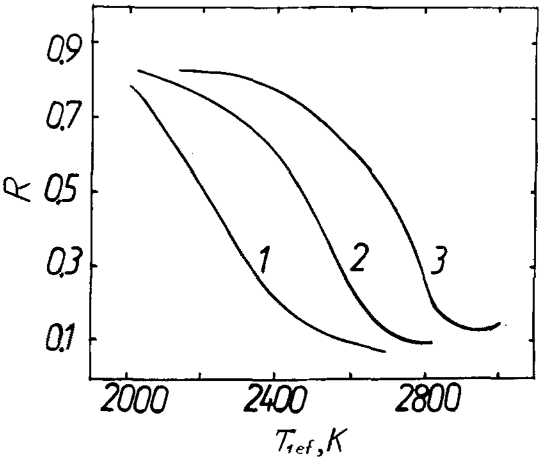

Fig. 2. Dependence of reflectivity of MgO for $\lambda = 1.15$ μm on T_{1ef} at various q's: $q = 870\ \mathrm{W \cdot cm^{-2}}$ (curve 1); $q = 1100\ \mathrm{W \cdot cm^{-2}}$ (curve 2); $q = 1370\ \mathrm{W \cdot cm^{-2}}$ (curve 3).

accentuates when the value of the flux is greater. It is exactly this behavior of R that is affected by the specific features of semitransparent scattering ceramics.

Analysis shows that, for instance, to achieve a melting point of 3100 K on the surface at $q_1 = 1370\ \mathrm{W \cdot cm^{-2}}$, a time of 1.04 s is needed, and at $q_2 = 945\ \mathrm{W \cdot cm^{-2}}$ this time is 3.3 s. The temperature differences at a distance of 0.3 mm from the surface are 420 and 320 K, respectively. Since $T = f(x)$ for q_1 is lower at any values of x, the absorption coefficient is also lower at each corresponding point. The scattering coefficient, determined mainly by the porosity and refractive index, depends only slightly on temperature. This is why at the same surface temperature with larger values of q, a material at any wavelength, including that of the pyrometer, 0.55 μm, emits less, while its reflectivity is larger. For the intensity of radiation at 0.55 μm at q_1 to become equal to that at q_2, heating must be continued. It must be kept in mind, however, that elevation of the temperature is accompanied not only by an exponential increase in Planck's radiation intensity $I_p = f(T)$, but also by an increase in the absorption coefficient k by a certain law. Since the spectral volumetric emission coefficient of radiation $j = k(T)\, n^2 I_p(T)$, to ensure the equality of T_{1ef}, a smaller increment of the surface temperature is needed in heating with higher values of q than to ensure the equality of R because the temperature field affects the decrease in R only by growth of the absorption coefficient k. This is why with equality of T_{1ef} during heating with various values of q, the reflectivity will be higher with larger q's (Fig. 2).

Consequently, neither the reflectivity nor the emissivity of ceramics in heating by laser radiation is related unambiguously to the temperature being measured, but they depend on the flux q. The measured value of the temperature at the same surface temperature, in turn, also depends on q. If, on the other hand, we wait for a stable quasi-steady temperature distribution, the dependence of the radiation characteristics on an effective temperature, e.g., on T_{1ef} will be a stable value for each q that will grow with increasing q. It is exactly these values as a function of q that can be employed for calculating heat transfer in apparatus using oxide thermal insulation if, naturally, steady-state conditions also set in there with surface, e.g., convective heating.

4. INFLUENCE OF VAPOR CONDENSATION NEAR A SAMPLE ON THE RESULTS OF STUDYING THE THERMAL RADIATION CHARACTERISTICS

Studying of the thermal radiation characteristics of refractory oxides at the extreme temperatures is hindered by evaporation of the oxides. When the latter are heated by CO_2 laser radiation in the air, the evaporation products condense near the surface being heated. Condensation causes nuclei to grow into droplets which are then carried away by convective flows. The concentration of the droplets and their size change within the confines of the plume of evaporation products emerging from the oxide surface being heated.

The possible influence of the condensate particles near the surface of oxide ceramics was noted when heated by a flux of laser radiation on the results of measuring the radiation characteristics at the extreme temperatures [4]. This phenomenon has been studied in greater detail in Ref. 7.

Figure 3 shows the results of two experiments involving the measurement of the intensity of radiation of a vertically standing magnesium oxide ceramic sample when its central part was heated by the flux q. The emission spectra were registered under a quasi-steady state and were recalculated to the effective temperature of a blackbody with the same radiation intensity. The measurements were made in two different ways: one (curve 2 in Fig. 3) with air blowing over the specimen and another without. It can be seen that in the first case, higher values of the measured effective temperatures and a qualitatively different nature of their spectral dependence were obtained. In the second case in the visible region, T_{ef} also decreased with decreasing λ because of scattering by the condensate particles, which grew with the decreasing λ. This does not correlate with the nature of the change in the absorption coefficient of magnesium oxide, because near the electron

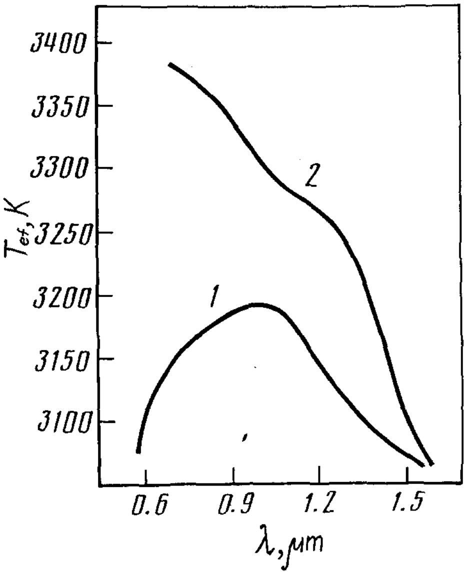

Fig. 3. Dependence of the effective temperature T_{ef} of magnesia ceramic on the wavelength when heating in air with a flux density of 2000 W · cm^{-2}: without blowing air over the sample (curve 1); with blowing (curve 2).

edge of absorption it should grow with decreasing λ. Blowing away of the condensate particles by a stream of air yielded a more plausible relation $T_{ef} = f(\lambda)$.

The space in which condensation occurs and in which the formed droplets migrate depends on the formation of convective streams near the sample. The amount of formed vapor depends on the temperature of the surface and the time. The size of the particles also depends on the flux of the CO_2 laser radiation because the droplets are heated by the radiation, and the latter limits their growth. For example, when heating a vertical flat magnesium oxide sample using a flux of about 2000 W · cm^{-2} and with an irradiation spot 3 mm in diameter, the size of the condensate particles with a change in the distance to the surface from 0.5 to 2.5 mm changed from hundredths of a micrometer to about 0.25 μm. The values of mean radius of particles was estimated by using the Mie theory on the base of measurement of plume transmission and scattering of probe laser radiation as described in Ref. 7. There were no particles at a distance over 3.5 mm (Fig. 4). It is exactly such a particle size that underlay the dependence of the effective temperature on the wavelength shown in Fig. 3 because it

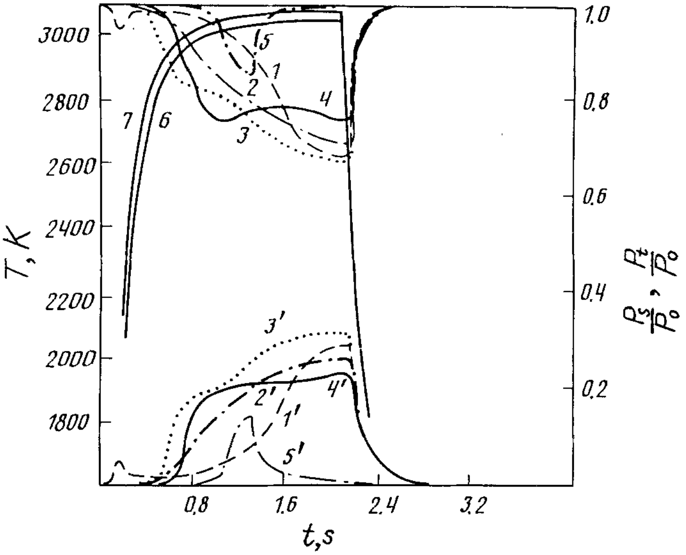

Fig. 4. Transmission P_t/P_0 (curves 1–5) and scattering P_s/P_0 (curves 1′–5′) of the plume with condensate droplets of a MgO sample heated by a radiation flux of 1600 W · cm^{-2} for a probing laser radiation beam ($\lambda = 0.48$ μm) at various distances *a* from the sample surface: 0.5 mm (curves 1 and 1′); 1.0 mm (curves 2 and 2′); 2.0 mm (curves 3 and 3′); 2.5 mm (curves 4 and 4′); 3.0 mm (curves 5 and 5′).

resulted in a sharp reduction in scattering in a wavelength-dependent manner; it was reduced to almost zero in the infrared region beginning at $\lambda = 1.5$ μm.

When silicon dioxide ceramic samples were heated under similar conditions (about 2000 W · cm^{-2}) in the quasi-steady state, no particles formed near the heated spot in the beam of the CO_2 laser (Fig. 5). But at a lower flux, approximately 500 W · cm^{-2}, quite intensive scattering on the condensate droplets was observed. In the first case, the mass of the carried off SiO_2 was greater, but no scattering was observed in the near-surface layer because of the restricted growth of the particles as a result of their being heated by the CO_2 laser radiation.

The difference between the behavior of SiO_2 and that of MgO is apparently due to the difference in the magnitude of the absorption index $\kappa = k\lambda/4\pi$ at a wavelength of 10.6 μm, which is 0.025 for MgO according to Ref. 8 and 0.33 for SiO_2 according to Ref. 9. Blowing away of the condensate particles enables one to obtain more correct information on the radiation characteristics and optical properties of the melt, but it is not always possible to establish its effectiveness exactly. Conducting experiments in

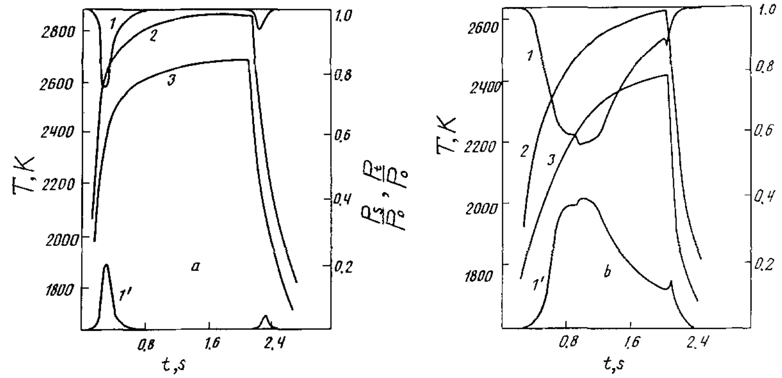

Fig. 5. Probing laser radiation beam ($\lambda = 0.48$) transmission P_t/P_0 of the plume and its scattering P_s/P_0 at a distance of 1.0 mm from the surface of the silica sample. (a) $q = 1600\ \mathrm{W \cdot cm^{-2}}$; (b) $q = 500\ \mathrm{W \cdot cm^{-2}}$. Transmission (curve 1); scattering (curve 1'); $T_{1\mathrm{ef}}$ (curve 2); $T_{2\mathrm{ef}}$ (curve 3).

good vacuum would eliminate the errors associated with the influence of the condensate, but the heating of oxides in vacuum often leads to a change in their optical properties because of incongruent evaporation.

What has been said above indicates that the formation of condensate particles near a material being studied may very greatly distort the results of measuring the thermal radiation characteristics at the temperatures where intensive evaporation occurs, and this must be given serious attention.

5. CONCLUSION

Though the use of CO_2 lasers for subsecond heating oxides in air has removed virtually all restrictions on the maximum temperatures that can be reached to study thermal radiation characteristics, the specific nature of intensive surface heating may significantly affect the accuracy of the obtained results. An individual approach must be used for every substance being studied, and the heating and measurement conditions must be selected accordingly.

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