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RADIATIVE PROPERTIES OF THERMALLY PROTECTIVE MATERIALS BASED ON PHENOLIC ASBESTOS-PLASTICS

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A study was made of the radiative characteristics of composites based on asbestos-capron and asbestos-polyformaldehyde as a function of wavelength, temperature, and heating regimes.

It is necessary to study the radiative properties of thermally protective materials (TPM) and their dependence on wavelength, temperature, and heating parameters in order to develop formulas for thermally protective coatings for use under given conditions.

One of the most promising groups of TPM's contains materials based on composites of asbestos-bearing fillers and phenolformaldehyde resin. The available literature data on the radiative characteristics of phenolic plastics is very limited and applies mainly to coke residues of phenolic carbon and phenolic nylon at T > 2500°K [1]. At the same time, study of the radiative properties of phenolic asbestos-plastics in the temperature range 1100-2000°K is of considerable interest. Here, it should be noted that the radiative properties of the composite in this temperature range are determined not only by the characteristics of the coke residue of the binder, but also to a significant extent by the filler. Depending on the temperature and the heating parameters in the surface layer, it is possible to have different relationships between the filler and coke residue and, thus, a significant difference in the optical properties of composites.

Here we report results of measurements of the normal hemispherical reflectivity $\rho_{\lambda n, 2\pi}$ of a TPM at the wavelength 0.58 μm and of spectral normal emissivity $\varepsilon_{\lambda n}$ in the range 1-9.5 μm. The TPM was a composite based on asbestos-capron, asbestos-polyformaldehyde, and phenolformaldehyde resin. The TPM was tested in the working-temperature range. The relative error of $\varepsilon_{\lambda n}$ in the range 1-9.5 μm and of $\varepsilon_{\lambda n} = 1 - \rho_{\lambda n, 2\pi}$ at 0.58 μm was no greater than 4%, with a confidence level no lower than 0.95. The measurements were made with a negative pressure of 13-1.3 Pa (10^{-1}-10^{-2} mm Hg).

Normal hemispherical reflectivity was determined by comparing reflected radiation from the specimen and a standard — magnesium oxide — by means of the relation

$$\rho_{\lambda n, 2\pi} = \left(\frac{J^{s}_{\lambda n}}{J^{st}_{\lambda n}} - \frac{J^{sp}_{\lambda n}}{J^{st}_{\lambda n}} \right) \rho^{st}_{\lambda n, 2\pi}, \quad (1)$$

where $J^{s}_{\lambda n}$, $J^{sp}_{\lambda n}$, and $J^{st}_{\lambda n}$ are the spectral luminance of the total (characteristics and reflected) radiation, the characteristic radiation of the specimen, and the luminance of the standard; $\rho_{\lambda n, 2\pi}$ is the normal hemispherical reflectivity of the standard. The specimen and standard were placed at the center of an integrating sphere. The radiation sources used were high-pressure quartz mercury lamps with a linear spectrum. The sources were located in the rear of the sphere relative to the direction of sighting of the half behind the specimen. Repeated

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reflection ensured diffuse hemispherical illumination of the test material. The specimen was heated by beams from continuous CO_2 lasers with a working wavelength of 10.6 μm. The luminance of the total (characteristic and reflected) radiation of the specimen $J^s_{\lambda n}$ and the luminance of the standard $J^s_{\lambda n}$ were measured at a wavelength $\lambda_i = 0.58$ μm (in the radiation lines of mercury vapors $\lambda = 0.577$ μm, 0.579 μm), while the intensity of the characteristic radiation was designated as $J^{sp}_{\lambda n}$ at the wavelength $\lambda_j = 0.60$ μm. Here, reflected radiation could be ignored due to the linear character of the radiation spectrum of the sources. The setup of the measuring instrument allowed us to directly determine the ratios $J^s_{\lambda n}/J^{st}_{\lambda n}$ and $J^{sp}_{\lambda n}/J^{st}_{\lambda n}$ and then to compute $\rho_{\lambda n, 2\pi}$. Here, we assumed that due to the closeness of λ_i and λ_j, the equality $\varepsilon_{\lambda n}(\lambda_i) = \varepsilon_{\lambda n}(\lambda_j)$ was valid. The reflectivity of the standard $\rho^{st}_{\lambda n, 2\pi}$ was measured beforehand. A detailed description of the measurement method and unit was given in [2, 3].

Figure 1 presents representative results of determination of $\rho_{\lambda n, 2\pi}$ for the investigated materials after they were subjected to a plasma flow. It follows from Fig. 1a that $\rho_{\lambda n, 2\pi}$ of both composites based on asbestos-capron and asbestos-polyformaldehyde is nearly independent of temperature in the ranges 1100-1500 and 1750-1950°K, while the difference in reflectivity for both materials is small. Curves 1 and 2 in Fig. 1b show results of measurements of $\rho_{\lambda n, 2\pi}$ of composites exposed to a plasma flow for 60 sec, where the heat flux on the specimen surface was 30 W/cm². It is evident from a comparison of Fig. 1, a and b, that at T < 1500°K, the reflectivity of specimens subjected to heating for 60 sec is considerably lower than the values of $\rho_{\lambda n, 2\pi}$ for a material exposed to the treatment for 120 sec. The change in the reflectivity of the composite with an increase in the duration of thermal loading can be explained as follows. When a gas-plasma flow acts on a composite, the organic components of the latter decompose, and carbon in the surface layer undergoes partial combustion. As a result, there is a change in the relationship between the filler and binder on the surface. An increase in the contribution of the filler to the optical properties of the surface layer leads to higher values of reflectivity, since the asbestos-bearing filler has a lower emissivity than the coke residue of the phenolformaldehyde resin in the visible region of the spectrum. An increase in thermal loading time leads to more substantial carbon combustion in the surface layer and, thus, to a further increase in the role of the characteristics of the filler in determining the optical properties of the composite. This in turn results in higher values of reflectivity. Similarly, an increase in heat flux to the specimen surface leads to an increase in $\rho_{\lambda n, 2\pi}$ in the temperature range 1550-1900°K (see curves 3 and 4 in Fig. 1b). Here, in the temperature range 1400-1600°K° the values of $\rho_{\lambda n, 2\pi}$ for the investigated composites are nearly the same at different heat fluxes.

The data obtained on normal hemispherical reflectivity can be used to calculate normal spectral emissivity at the wavelength 0.58 μm and to subsequently pyrometrically determine the temperature of the thermally protective coatings. The data also makes it possible to evaluate the behavior of the optical characteristics of composites in the visible region with a change in temperature. Since the emissivities of the structural components of TPM's (the coke residue of the organic components — carbon and asbestos) do not significantly change with wavelength in the visible part of the spectrum, it is natural to suggest that composites based on these components are also nearly gray in the visible region.

Most of the characteristic thermal radiation of the coating occurs in the interval 0.6-9.5 μm at temperatures of 1100-2000°K. In connection with this, data on $\varepsilon_{\lambda n}$ in this spectral range can be used to calculate the characteristic radiation associated with thermal protection. This radiation plays an important role in balancing heat flows on working surfaces in a number of cases. Such information is also of interest for calculating convective-radiative heating of thermally stressed elements of hot equipment.

Normal spectral emissivity was determined by comparing the radiation from a specimen of the test material and a blackbody at the same temperature in accordance with the relation

$$\varepsilon_{\lambda n} = \frac{J^{sp}_{\lambda n}}{J^{b.b}_{\lambda n}}, \tag{2}$$

where $J^{sp}_{\lambda n}$ and $J^{b.b}_{\lambda n}$ are the normal spectral luminances of the specimen and blackbody, respectively.

We measured the true temperature of the test surface of the specimen by using values of $\varepsilon_{\lambda n}$ at 0.58 μm calculated in accordance with Kirchhoff's law from the formula

$$\varepsilon_{\lambda n} = 1 - \rho_{\lambda n, 2\pi} \tag{3}$$

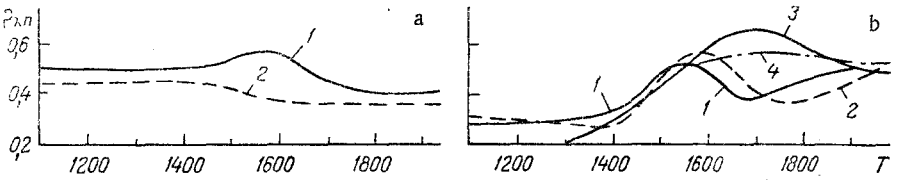

Fig. 1. Normal hemispherical reflectivity $\rho_{\lambda n, 2\pi}$ at the wave length 0.58 μm for a TPM based on asbestos-capron (AKP) and asbestos-polyformaldehyde (APF): a) τ = 120 sec, q = 30 W/cm²: 1) AKP; 2) APF; b) τ = 60 sec: 1) AKP, q = 30 W/cm²; 2) APF, q = 30 W/cm²; 3) AKP, q = 210 W/cm²; 4) APF, q = 210 W/cm². T, °K.

on the basis of data on $\rho_{\lambda n, 2\pi}$. The blackbody was in the form of a graphite tube heated by an electric current and having a rectangular slit on its lateral surface. A description of the equipment and method used to measure $\varepsilon_{\lambda n}$ was given in [4].

Figure 2 shows results of determination of $\varepsilon_{\lambda n}$ for a TPM based on asbestos-capron with different heating regimes. In each case there is a monotonic increase in spectral emissivity with an increase in wavelength in the region 1-6 μm. Here, the values of $\varepsilon_{\lambda n}$ vary within a very broad range, from 0.47 at λ = 1 μm, q = 210 W/cm², τ = 60 sec, and T = 1700°K (curve 4 in Fig. 2c) to 0.91 at q = 30 W/cm², λ = 6 μm, τ = 60 sec, and T = 1700°K (curve 4 in Fig. 2a). The value of $\varepsilon_{\lambda n}$ generally decreases with an increase in wavelength in the region 6-9.5 μm. The same tendencies are exhibited by the emissivity of the TPM based on asbestos-polyformaldehyde (Fig. 3). With an increase in heating time (τ = 120 sec) or in the case of high heat fluxes (210 W/cm²), the curves $\varepsilon_{\lambda n}(\lambda)$ of this composite have a "plateau" in the region λ = 2-4.5 μm on which $\varepsilon_{\lambda n}$ changes little.

The above features of change in $\varepsilon_{\lambda n}$ with an increase in wavelength can be explained as follows. The optical properties of the composite are determined both by the characteristics of the binder and by the properties of the filler. The contribution of different structural components to the properties of the surface layer of the composite is dissimilar due to decomposition of organic components and partial combustion of carbon after exposure to the plasma flow and, in several cases, due to melting of the asbestos. The radiative characteristics of the components of the investigated materials change differently with a change in wavelength. Asbestos has low values of emissivity in the visible region, and its emissivity increases with an increase in wavelength in the infrared region. Conversely, the coke residues of phenol-formaldehyde resin have a high emissivity in the visible region but decrease with an increase in wavelength — particularly on the section from 2 to 4 μm [5, 6]. With asbestos having a significant effect on the properties of the surface layer of the composite, there is a characteristic (for asbestos) increase in emissivity with an increase in wavelength in the region 1-6 μm. This increase is manifest to the grestest extent in a TPM based on asbestos-capron (see Fig. 2). With a reduction in the amount of asbestos in the composite, as in a TPM based on asbestos-polyformaldehyde, and with more intensive thermal loads (Fig. 3b and c), the interaction of the opposing tendencies of the change in $\varepsilon_{\lambda n}$ of the structural components of the composite (an increase in $\varepsilon_{\lambda n}$ for asbestos and a decrease in $\varepsilon_{\lambda n}$ for coke residues of phenol-formaldehyde resin) leads to their compensation, and a "plateau" characterized by a slight change in $\varepsilon_{\lambda n}$ with wavelength is formed. The reduction in $\varepsilon_{\lambda n}$ with an increase in wavelength noted for most curves $\varepsilon_{\lambda n}(\lambda)$ in Figs. 2 and 3 on the section 6-10 μm may also be due to the effect of the optical properties of the coke residue of the resin, particularly with the reduction in its emissivity with an increase in λ in this spectral range. The combination of this factor and the increase in $\varepsilon_{\lambda n}$ of the filler in the region 1-6 μm can probably explain the presence of the maximum of $\varepsilon_{\lambda n}$ on the section 6-7 μm for many curves $\varepsilon_{\lambda n}(\lambda)$ in Figs. 2 and 3.

Figure 4 shows data on the partial normal integral emissivity of the materials $\varepsilon_{\Sigma n}$. These values were calculated from measurements of $\varepsilon_{\lambda n}$ in the range 1-9.5 μm, with the use of data on $\varepsilon_{\lambda n}$ at 0.58 μm and approximation of the values of $\varepsilon_{\lambda n}$ on the section 0.58-1 μm. From 92% (at T = 1100°K) to 98% (at T = 2000°K) of the blackbody radiation is concentrated in the spectral range 0.58-9.5 μm. Here, from 0.1% (at T = 1100°K) to 6% (at T = 2000°K) of its radiant flux is found on the section 0.58-1 μm. Thus, the corrected values of $\varepsilon_{\Sigma n}$ give a good representation of the total normal integral emissivity.

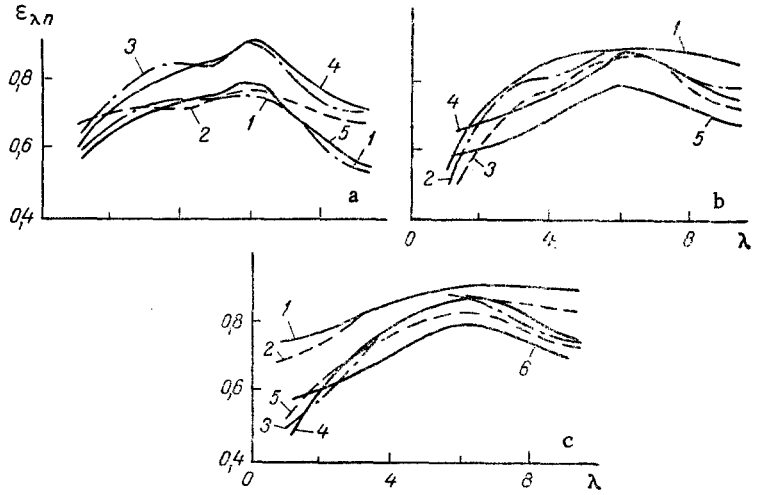

Fig. 2. Normal spectral emissivity of a TPM based on asbestos-capron: a) τ = 60 sec, q = 30 W/cm^2; 1) 1130, 2) 1300, 3) 1410, 4) 1700, 5) 1830 K; b) τ = 120 sec, q = 30 W/cm^2; 1) 1230, 2) 1470, 3) 1650, 4) 1720, 5) 1890 K; c) τ = 60 sec, q = 210 W/cm^2; 1) 1200, 2) 1420, 3) 1590, 4) 1700, 5) 1790, 6) 1900 K. λ, μm.

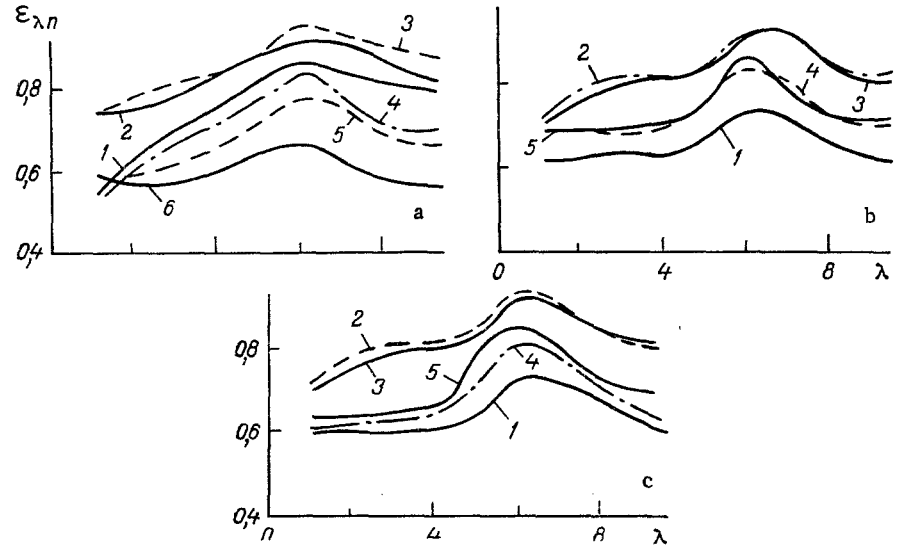

Fig. 3. Normal spectral emissivity of a TPM based on asbestos-polyformaldehyde: a) τ = 60 sec, q = 30 W/cm^2; 1) 1160, 2) 1300, 3) 1440, 4) 1580, 5) 1740, 6) 1920 K; b) τ = 120 sec, q = 30 W/cm^2; 1) 1200, 2) 1320, 3) 1470, 4) 1740, 5) 1970 K; c) τ = 60 sec, q = 210 W/cm^2; 1) 1230, 2) 1340, 3) 1450, 4) 1720, 5) 1940 K.

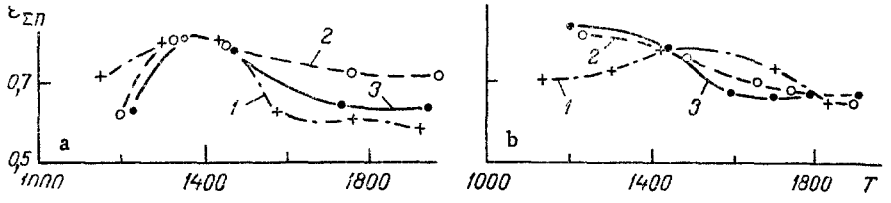

Fig. 4. Normal integral emissivity of a TPM based on APF and AKP: a) APF: 1) q = 30 W/cm^2, τ = 60 sec; 2) 30 and 120; 3) 210 and 60; b) AKP: 1) q = 30 W/cm^2, τ = 60 sec; 2) 30 and 120; 3) 210 and 60.

It can be seen from Fig. 4 that at T > 1400°K, the integral emissivity of both materials decreases with an increase in temperature. An increase in ε_{Σ_n} with an increase in temperature is seen for the composite based on asbestos-polyformaldehyde in the range 1100-1400°K. The heating regimes also have a significant effect on $\varepsilon_{\Sigma n}$. Here, the partial normal integral emissivity of the investigated TPM's is concentrated mainly in the range 0.6-0.8.

The emissivity of a material, $\varepsilon = 0.8$, without consideration of the wavelength and temperature dependence of emissivity (see [7], for example), is often important in problems of convective-radiative heat transfer when calculating the heating of thermally protective coatings based on asbestos cloth and phenolformaldehyde resin. As follows from the above results, such an approach is excessively simplified. The emissivity of a TPM based on phenolic asbestos-plastics in the temperature range 1100-2000°K depends heavily on wavelength and, to a somewhat lesser extent, on temperature and heating regime. This fact must be taken into account when calculating thermal protection.

NOTATION

q, heat flux; τ, heating time; λ, wavelength; T, absolute temperature; $\rho_{\lambda n, 2\pi}$, normal hemispherical reflectivity; $\varepsilon_{\lambda n}$, normal spectral emissivity; ε_{Σ_n}, partial normal integral emissivity; $J_{\lambda n}$, normal spectral luminance; λ_i, radiation line of mercury vapors; $\lambda_i = 0.60$ μm. Indices: s, total characteristic and reflected radiation; sp, specimen; st, standard; b.b, blackbody; TPM, thermally protective materials.

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