EFFECT OF EVAPORATION OF HIGH-TEMPERATURE CHAMBER ELEMENTS ON ELECTROPHYSICAL PROPERTIES OF GASEOUS MEDIA CONTAINED THEREIN

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UDC 536.423+533.9

The content and composition of vapors generated from elements of high-temperature equipment are studied. Their effects on the electrophysical properties of gaseous media in such devices are evaluated, with special reference to electrical conductivity.

A unique feature of high-temperature devices is that their gaseous operating media contain impurities in the form of vapors formed from the construction elements. The generation and quantity of such vapors and their effects on the medium are of interest, since as a rule, they have a high ionization potential $\varphi_i \approx$ (4-8) V and a partial pressure p_i which increases significantly with increase in temperature T. In connection with this, sufficiently intense heating can significantly change the overall characteristics of gaseous media, foremost, their electrophysical properties, as well as the parameters of the device in which they are used. For example, it is known that evaporation of a solid dielectric may decrease the surface electrical strength by a factor of several times [1], while an insignificant amount of impurity ($p_i \sim 10^{-4}$-10^{-3} Pa) with low ionization potential encourages Penning reactions [2] which produce a decrease in breakdown voltages within the gas. It follows from this that knowledge of the effect of evaporation of dielectric and metallic vessels at high temperatures on the gaseous working media or gases being studied therein is necessary. The present study will consider dielectric vessels of aluminum oxide, which is widely used as a construction material for high-temperature devices. The goal of the study is to investigate the qualitative and quantitative composition of vapors from the vessel wall and their effect upon the electrical conductivity of the contained gas.

The experimental arrangement is shown in Fig. 1. Tubular vessels 3 were used, made of high-purity ceramic with chemical composition: Al_2O_3, 99.8%; MgO, 0.08%; SiO_2, 0.06%; Fe_2O_3, 0.04%; CaO, 0.01%; TiO_2, 0.01%; Na_2O, 0.005%. The platinum–rhodium wire heater 4 was coated with thermal insulation 5. Radiation from the vapor in the tube passed through monochromator 7 (MDR-2) to photomultiplier 8 and was recorded by chart recorder 9. A type S18-200U incandescent lamp 1 was used as a reference source, and lenses 2, 6 were used to focus the light.

Considering the composition of the tube material, one would expect an abundance of lines in the spectrum. However, the experiments performed revealed that beginning at T ~ 1300°K lines from sodium excited to the first level with wavelengths λ_1 = 588.9 nm and λ_2 = 589.6 nm began to appear. No other lines or molecular bands, in particular, those of the main wall material, Al_2O_3, were observed in the visible spectrum up to 1900°K. This can be explained in the following manner. The intensity I of radiation of any component of the vapor is $I \sim p_s \cdot \exp(-e\varphi_e/kT)$. In the temperature range studied the partial pressure of saturated sodium vapor p_s is 6-10 orders of magnitude higher than the pressure of the other components of the tube material [3], while its excitation potential is the smallest ($\varphi_e \approx 2.1$ V). Therefore, despite the very small amount of Na_2O in the ceramic wall ($\leqslant 0.005\%$), the atomic concentration and radiation intensity I of sodium are the highest.

To quantitatively analyze the content of sodium vapor produced from the tube wall the absolute radiation intensity method was used [4]. As is well known, given thermodynamic equilibrium in a plasma, the concentration of atoms in the excited state is determined by the Boltzmann expression

$$n_к = \frac{g_к}{g_0} n_0 \exp\left(-\frac{e\varphi_e}{kT}\right). \quad (1)$$

Electrodynamics Institute, Academy of Sciences of the Ukrainian SSR, Kiev. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 50, No. 4, pp. 592-597, April, 1986. Original article submitted February 4, 1985.

0022-0841/86/5004-0419 $12.50

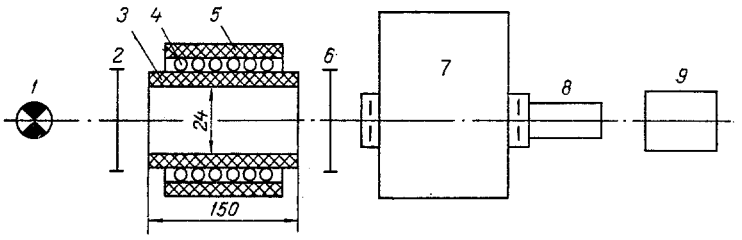

Fig. 1. Experimental arrangement.

The intensity of the spectral line corresponding to transition of an atom from the upper to the lower level i (i = 0) is

$$I = n_{\text{к}} A_{\text{к}i} h\nu_{\text{к}i} l = n_0 \frac{g_{\text{к}}}{g_0} \exp\left(-\frac{e\varphi_e}{kT}\right) A_{\text{к}0} h\nu_{\text{к}0} l. \quad (2)$$

The constants on the right side of Eq. (2) are known for the majority of elements. Therefore, by measuring the absolute line intensity I, the unknown concentration n_0 can be determined. However, at sufficiently high values of n_0 it is necessary to consider self-absorption in the radiation source, which can significantly reduce the measured value of I. In connection with this, it is necessary to check the line for reabsorption and introduce a corresponding correction. In the present study this checking was done by the method described in [5]. Measurements revealed that the level of reabsorption did not exceed 10%.

Results of the study of concentration n_0 of Na atoms using the line λ_1 = 588.9 nm with consideration of self-absorption are shown in Fig. 2. The radiation intensity at the center of the tube was determined, and then compared to the intensity of light from the reference lamp at the wavelength λ_1, after which Eq. (2) was used to calculate the concentration of sodium vapor n_0. As is evident from Fig. 2, with increase in T the value of n_0 increases from 10^{11} to 10^{12} cm^{-3}. Under the same conditions, the vapor concentration at the tube walls is approximately twice as high. Figure 2 also shows the partial sodium pressure p_0 in the chamber

$$p_0 = n_0 kT, \quad (3)$$

which over the temperature range T = 1500-1900°K comprises $p_0 = 4 \cdot 10^{-3}$-$6 \cdot 10^{-2}$ Pa.

Thus, if we consider that the gas pressure in the tube is atmospheric, the impurity content reaches ~10^{-5}%. However, even such a slight addition can have a significant effect on the electrophysical characteristics of the gaseous medium at high temperature. This appears in the values of electrical strength and electrical conductivity of air and nitrogen when they are studied in an aluminum oxide chamber [6, 7]. The studies of [6] showed that even at T ~ 1600°K evaporation leads to a marked reduction in breakdown voltages compared to values determined under clean conditions. At T ~ 2000°K this reduction reaches 70%. The vapors have an especially strong effect on the electrical conductivity of the gaseous medium σ, which is usually ignored at such temperatures, being assumed negligibly low. However, when the value of σ was measured beginning at T ~ 1000°K, due to the presence of the easily ionized impurity its value was 7-8 orders of magnitude above that calculated with the Sach equation for the pure gas [7]. Calculated values of the electrical conductivity of the chamber gas σ_c based on results of spectral studies of the vapor composition and concentration agree well with experimental values of the electrical conductivity σ. Measurement of σ was performed by the two-electrode method [7]. Experimental dependences σ = f(T) for several gases at atmospheric pressure (He, Ar, air) are shown in Fig. 3 by curves 1-3. These curves are the result of averaging experimental data with a scattering of 30%.

To calculate σ_c of a thermally ionized gas one may use the expression

$$\sigma_c = e\mu_e n_e + e\mu_i n_i. \quad (4)$$

From the Sach equation the concentrations n_e, n_i are equal to

$$n_e = n_i = \left[n_0 \left(\frac{2\pi mkT}{h^2}\right)^{3/2} \frac{2g_i}{g_0} \exp\left(-\frac{e\varphi_i}{kT}\right)\right]^{1/2}. \quad (5)$$

Since air contains an electronegative gas, oxygen, in the general case it is necessary to consider the effect on conductivity of the electron capture process: $O_2 + e \rightarrow O_2^-$. However, as is shown by calculation with the expression of [2]

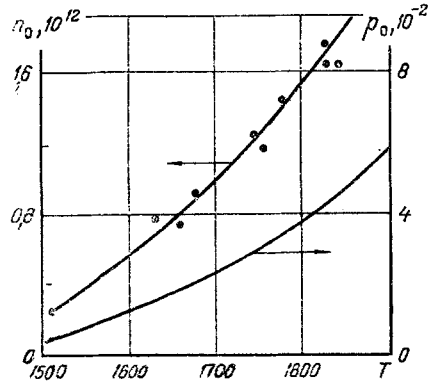

Fig. 2

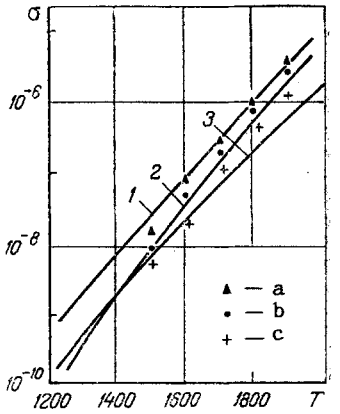

Fig. 3

Fig. 2. Concentration n_0 and pressure p_0 of sodium vapor vs temperature T. n_0, 10^{12} cm^{-3}; p_0, 10^{-2} Pa; T, °K.

Fig. 3. Electrical conductivity σ of helium (1), argon (2), and air (3) vs temperature: curves, experiment; a, b, c, calculated points for helium, argon, and air, obtained from results of spectral measurements.

$$\frac{n_i^-}{n_e} = n_a \left(\frac{h^2}{2\pi mkT}\right)^{3/2} \frac{g_0'}{g_i'} \exp\left(\frac{e\varphi_a}{kT}\right), \tag{6}$$

at $T \geqslant 1500$°K the effect of this process on the value of σ_c may be neglected. At T = 1500°K the ratio n_i^-/n_e is equal to 0.23, while with increase in temperature to 1900°K it decreases to 0.08.

Since the ionic component of the conductivity in Eq. (4) is so small ($\mu_i \ll \mu_e$), then

$$\sigma_c = e\mu_e n_e. \tag{7}$$

For the conditions under which the conductivity was measured ($E/p \sim 10^{-2}$ $V \cdot cm^{-1} \cdot Pa^{-1}$), electron mobility in air $\mu_e = 3 \cdot 10^3$ $cm^2 \cdot V^{-1} \cdot sec^{-1}$; in helium, $\mu_e = 5 \cdot 10^3$ $cm^2 \cdot V^{-1} \cdot sec^{-1}$; and in argon, $\mu_e = 3.2 \cdot 10^3$ $cm^2 \cdot V^{-1} \cdot sec^{-1}$ [2]. Results of calculating σ_c with Eqs. (5), (7) are shown in Fig. 3. The accuracy of the σ_c values obtained is 25%, determined by the uncertainty of the spectral method of measuring vapor concentration n_0, and as is well known [2], by the relatively low precision of the mobility values μ_e. As is evident from the figure, within the limits of the calculation uncertainty the values of σ_c coincide with the experimental σ values.

The values of σ and σ_c were compared by a somewhat different method. The measured values of σ together with Eqs. (5) and (7) were used to reestablish n_0. The ionization potential φ_i required to do this was calculated from the experimentally obtained $\sigma = f(T)$. For this purpose Eq. (5) was substituted in Eq. (7) and the log of the resulting expression taken:

$$\lg \sigma_p = \lg A + \lg n_0^{1/2} T^{3/4} - 0{,}217 \frac{e\varphi_i}{kT}, \tag{8}$$

where $A = e\mu_e \left(\frac{2g_i}{g_0}\right)^{1/2} \left(\frac{2\pi mk}{h^2}\right)^{3/4}$ is a constant quantity. If we initially assume that with increase in temperature the concentration n_0 of the impurity responsible for the conductivity changes only slightly, then the increase in σ will be determined mainly by the last term of Eq. (8). Then, drawing a graph of the function $\sigma = f(T^{-1})$ in semilogarithmic coordinates, the slope of that graph can be used to determine some averaged ionization potential of the medium

$$\varphi_i' \approx 3{,}97 \cdot 10^{-4} \frac{\Delta(\lg \sigma)}{\Delta(T^{-1})}. \tag{9}$$

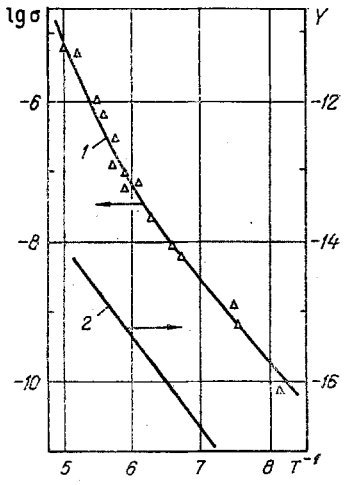

Fig. 4. Electrical conductivity σ (1) and quantity $Y = \log \sigma - \log n_0^{1/2}T^{3/4}$ (2) vs inverse temperature T^{-1} for argon. Points, experimentally determined conductivity values. σ, $(\Omega\cdot cm)^{-1}$; n_0, cm^{-3}; T, °K; T^{-1}, 10^{-4} K^{-1}.

As an example, Fig. 4 shows such a dependence for argon. It consists of two linear sections. The potential φ_i' calculated with Eq. (9) for the first segment $T^{-1} = (6.5\text{-}8.5)\cdot 10^{-4}$ K^{-1} (T = 1540-1175°K) is equal to $\varphi_{i1}' \approx 5$ V, while on the second $T^{-1} = (5\text{-}6.3)\cdot 10^{-4}$ K^{-1} (T = 2000-1600°K) – $\varphi_{i2}' \approx 10$ V. The values obtained for φ_{i1}' are close to the first ionization potential of sodium $\varphi_i = 5.1$ V and correspond to the qualitative composition of vapor in the tube as determined spectrally. The value φ_{i2}' is too large and cannot characterize the ionization potential of the medium, since if $\varphi_i \sim 10$ V, then gas ionization at $T \leqslant 2000$°K should be negligibly small. The increase in φ_i' from 5 to 10 V indicates a significant increase in the term $\log n_0^{1/2}T^{3/4}$ in Eq. (8), caused by an increase in the impurity concentration n_0 with chamber heating above 1600°K.

Now, knowing the approximate value of the impurity ionization potential $\varphi_{i1}' \approx 5$ V, we refine that value by iterative calculation. Substituting in Eq. (8) $\varphi_i = \varphi_{i1}'$, we calculate the concentration n_{01} and construct the graph of the quantity $Y = \log \sigma - \log n_{01}^{1/2}T^{3/4}$ vs inverse temperature T^{-1}. This dependence is shown by curve 2 of Fig. 4. In analogy to Eq. (9) the recalculated ionization potential is equal to

$$\varphi_i'' = 3{,}97\cdot 10^{-4}\frac{\Delta(\lg\sigma - \lg n_{01}^{1/2}T^{3/4})}{\Delta(T^{-1})}. \qquad (9')$$

Calculation with Eq. (9') gives $\phi_i'' \approx 5.1$ V. Subsequent iterations refine this value no further, so that we take $\phi_i = 5.1$ V.

Thus, the coincidence of calculated and experimental values of electrical conductivity confirms the reliability of the results of the spectral studies of vapor composition and content in the chamber. The dominant effect on electrophysical properties of the gaseous atmosphere is exerted by the components of the chamber material with the highest saturated vapor tension and lowest ionization potential. The relative content of such components in the chamber material may even be very small ($\leqslant$0.001%). In particular, for vessels of aluminum oxide, Na_2O acts as such an impurity. Upon evaporation from the wall of such a vessel, the Na partial pressure above the surface is significant at a very low Na_2O concentration in the chamber material ($\leqslant$0.005%) and with increase in temperature over the range T = 1500-1900°K increases from $5\cdot 10^{-3}$ to $6\cdot 10^{-2}$ Pa. One of the electrophysical parameters of the gaseous medium most sensitive to the presence of vapor is its electrical conductivity, which under the experimental conditions changed over three orders of magnitude: from 10^{-8} to 10^{-5} $(\Omega\cdot cm)^{-1}$ with a temperature increase of only 500°K. Conductivity measurements can be used to evaluate introduction of vapor into the atmosphere of high-temperature equipment.

NOTATION

φ_i, ionization potential; T, temperature; p_i, partial pressure of impurity; I, radiation intensity; e, m, charge and mass of the electron; φ_e, excitation potential; k, Boltzmann's constant; n_0, g_0, concentration and statistical weight of atoms in normal state; n_k and g_k, concentration and statistical weight of atoms in excited state; A_{k0}, probability of atomic transition from excited to normal state; h, Planck's constant; λ, wavelength; ν_{k0}, frequency of quantum emitted upon transition of atom from excited to normal state; ℓ, length of scintillating gas column; p_0, sodium partial pressure in tube; σ, electrical conductivity; σ_c,

calculated conductivity; g_i, statistical weight of Na^+ ion; μ_e, electron mobility; μ_i, ion mobility; n_e, n_i, electron and ion concentrations; n_i, g_i', concentration and statistical weight of O_2^- ions; n_a, g_0', concentration and statistical weight of O_2 molecules; affinity of O_2 to an electron; E, electric field strength.

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COMBINED USE OF QUALITATIVE AND QUANTITATIVE REFRACTOMETRIC METHODS

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UDC 681.795.2:536.5

The results of an analysis of the operation of multichannel and scanning laser refractometers as well as systems enabling simultaneous employment of schlieren and laser refractometric methods in hydroaerodynamic experiments are presented.

In heat- and mass-transfer problems, studies of the distributions of the density, concentration, temperature, and other parameters, giving rise to the presence of a gradient in the index of refraction, are of great significance. The gradient of the index of refraction can be measured by different optical methods [1, 2], but in experimental practice only three are most widely used: interferometric, schlieren, and direct-schlieren methods.

A convenient quantitative refractometric method in practice is the method based on recording in the form of an electric signal the instantaneous value of the angle of deflection of the probing laser beam [3], which yields operational quantitative information on the local value of the gradient of the quantity under study with high spatial resolution.

Quantitative information on the spatial structure of a nonuniform medium can be obtained with a multichannel laser refractometer, first proposed in [4]. In this case the medium under study is probed with a collection of parallel narrow beams of light and the angles of deflection of all beams are measured simultaneously.

In the study of a wide class of phenomena in hydro- and aerodynamics the simultaneous employment of shadow methods, which give qualitative information about the behavior of the quantity under study in a large spatial volume, and of a laser refractometer, which records local quantitative information, imparts to the measuring system qualitatively new properties, since it enables separating, for carrying out quantitative measurements, the most informative sections of the field under study and visually monitoring the development of the process under study.

To study dynamic phenomena in stratified media we developed and investigated laser refractometers: single-channel, multichannel, scanning, and their combination with the IAB-458 schlieren apparatus.

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Translated from Inzhernerno-Fizicheskii Zhurnal, Vol. 50, No. 4, pp. 597-604, April, 1986. Original article submitted January 14, 1985.

0022-0841/86/5004-0423$12.50