INVESTIGATION OF THE ELECTRODE POTENTIAL DROP AT A MOLYBDENUM ELECTRODE IN A FLOW OF ARGON SEEDED WITH POTASSIUM

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The results of an experimental investigation of the operation of an electrode in argon containing 0.15% of potassium at temperatures from 1400 to 1800°C and a pressure of 1 atmosphere when the gas and the electrode are in thermal equilibrium are presented. The current-voltage characteristics obtained are compared with the theory of electrode processes developed previously [1, 2]. Good agreement is obtained between the theoretical and experimental data.

Experimental Apparatus

The apparatus used is shown in Fig. 1. Argon is supplied at the rate of 1.25 g/sec to a plasmotron, where it is heated to the required temperature, and introduced into the evaporator 2, made of porous tungsten. A liquid alloy consisting of 80% K and 20% Na is also supplied to the evaporator. The flow rate of potassium is 0.15% of the flow rate of the argon. The heated argon, containing potassium and sodium vapors, was fed to the operating channel 6, made of aluminum oxide. The operating chamber 4, in the form of a vessel made of aluminum oxide, is placed inside the channel. The gas which is fed into the channel circulates around the operating chamber from the outside. In this way thermal equilibrium was attained in the operating chamber – the walls, the gas, and the operating electrode had the same temperature to within approximately 50°C. For thermal insulation the channel was placed inside a filling of corundum powder 3.

The operating electrode 5 was made of molybdenum in the form of a bead 6 mm in diameter with a current-conducting support. The electrode did not touch the walls of the channel in the hot region. To avoid condensation of the potassium on the cold parts of the support and on its mounting and also to prevent a discharge occurring in the heated part of the support, a small amount of argon not seeded with potassium (0.1-0.5% of the main flow) was blown along the support. The second electrode 7, also made of molybdenum, was placed at the end of the channel. The probe 8, made of 1-mm-diameter tungsten wire, was placed in the exit tube, without touching the wall. The distance between the end of the probe and the electrode surface was approximately 5 mm.

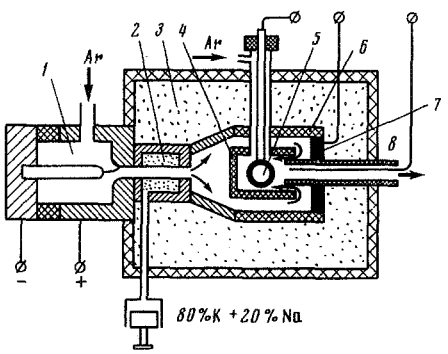

Fig. 1

The procedure used to make the measurements is similar to that described in [3]. This enabled us to obtain photographs on an oscilloscope of the current-voltage characteristics of the discharge gap between the electrode and the probe.

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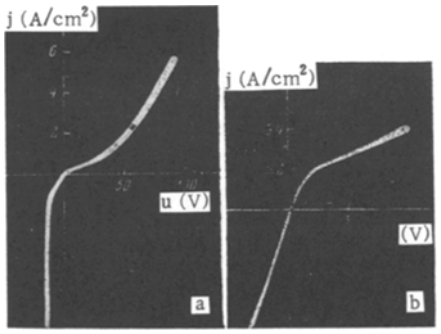

Fig. 2

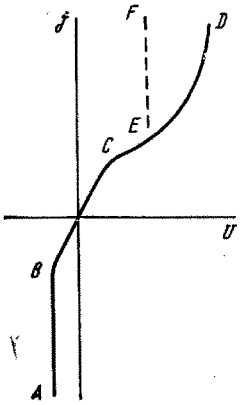

Fig. 3

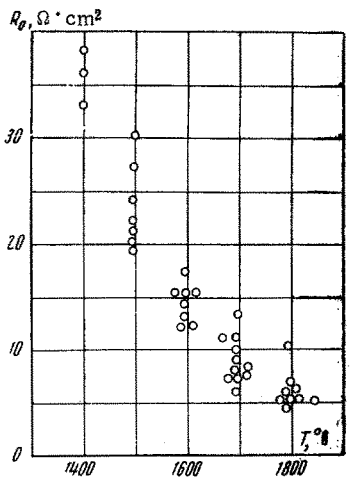

Fig. 4

The temperature of the electrode was measured with an optical pyrometer through the exit tube.

Interpretation of the Experiments

Figure 2a shows a typical current-voltage characteristic, and Fig. 2b shows its central portion on an amplified scale. The characteristic consists of three main parts (Fig. 3), each of which represents the specific behavior of the electrode drop in potential over a given range of current densities. Along the section AB, which refers to the anode branch, the potential drop is independent of the current. Along the section BC there is a linear relation between j and U, which does not change when changing from anode to cathode operation. Along the section CD the current increases linearly with the potential drop. The following empirical relations can be used to describe the three parts of the characteristic:

$$U = U_a \quad \text{section } AB \tag{1}$$

$$j = U / R_0 \quad \text{section } BC \tag{2}$$

$$j = j^* \exp\{a\sqrt{U}\}(1 + cU) \quad \text{section } CD \tag{3}$$

Here U is the electrode drop in potential, j is the current density which flows through the electrode, and U_a, R_0, j^*, a, and c are parameters of the characteristic which are independent of the current and the potential drop.

Processing of the experimental data reduces to selecting the values of the parameters for each individual current-voltage characteristic obtained in the experiments.

The graphs of Figs. 4-7 show the values of the corresponding parameters as a function of the temperature. (The temperature was adjusted during the experiments while all the remaining conditions were maintained constant.)

The crosses denote the values of the parameters obtained during preliminary experiments, when the system for supplying the potassium did not enable the potassium density in the flow to be calculated.

Discussion of the Results

The measured potential difference between the electrode and the probe consists of the potential drop in the space-charge region close to the electrode, the potential drop in the ambipolar diffusion region, and the change in potential in the unperturbed plasma* outside the diffusion region [2].

The length of the space-charge region under these conditions is of the order of several mean free paths, so that when treating the results of the experiment later we will assume that in this region the particles move without collisions.

*The term "unperturbed plasma" relates to the flow region where there are no charged-particle concentration gradients.

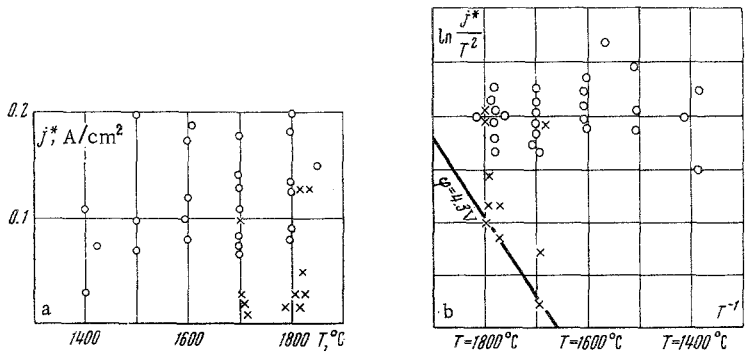

Fig. 5

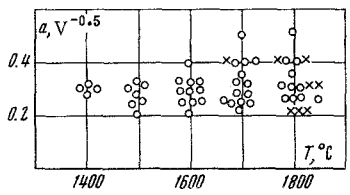

Fig. 6

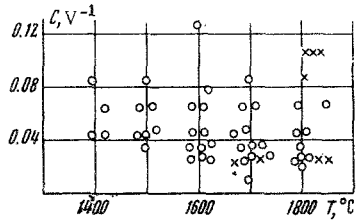

Fig. 7

The potential drop in the diffusion region, as in the unperturbed region, depends on the current and differs very little for the cathode and anode regions, when there is no difference between the temperature of the electrons and the heavy particles [2]. In this connection we might expect that under these conditions (argon with potassium) for small currents the characteristic should be linear, and its slope should be the same for the cathode and anode branches of the characteristic. The experimental curves (see Eq. (2) and Fig. 2) confirm this conclusion.

Note that R_0 in Eq. (2), which represents the total resistance of the diffusion and unperturbed region, differs from the resistance of the layer of unperturbed plasma between the electrode and the probe due to the large resistance of the diffusion region, resulting from the decrease in the charged-particle density in this region [2]. When the current increases, the resistance of the plasma between the probe and the electrode should fall both due to the increase in the charged particle density in the diffusion region and due to the increase in the electrical conductivity, resulting from the increase in the electron temperature in the argon-potassium plasma.

The experimental data on the potential drop in the region of the anode agree qualitatively with this conclusion (see Eq. (1) and Fig. 2). On the other hand, the increase in current above the electrode emission current is due to the increase in potential in the space-charge layer near the cathode, which leads to an increase in the effective resistance of the plasma between the electrode and the probe under cathode operating conditions. This is reflected in the break in the characteristic at the point C (Fig. 3). The qualitative form of the overall characteristic under cathode operating conditions close to $j = j_e$, which represents both of the processes described above, depends on the relation between j_e, j_i, and R_0. Under the experimental conditions described, the charged particle density outside the diffusion region when there is no current flowing $n_e = n_i \sim 10^{12}$ cm^{-3}. Because of the neutralization of the ions on the electrode surface, the charged particle density near the electrode surface is approximately two orders less than in the unperturbed plasma [2]. Consequently, for small currents the ion current at the electrode $j_i \sim 10^{-3}$ A/cm^2. This value is much less than the value corresponding to the point of inflection C on the current-voltage characteristics. Hence, we might expect that under these conditions the point of inflection C corresponds to the electron emission current.

The same characteristics were interpreted differently in [4]. The current at the point D, i.e., the maximum current level attained in the distributed discharge, is taken as the emission current (a further increase in current leads to the appearance of cathode spots and the occurrence of an arc discharge). However, on this assumption it is difficult to explain the quite large potential drop at the point D (15-20 V obtained in [4] and 50-100 V in our experiments).

Despite the considerable difference between the experimental data, we can definitely state that the value of the emission current from the electrode in the flow $j_e \approx j^* \approx j_c$ under these conditions was considerably higher than the emission current for molybdenum in a vacuum. For comparison, in Fig. 5b, where the experimental values of j^* are plotted in Richardson coordinates, we show the theoretical straight line for the thermionic-emission-current density for a work function of 4.3 eV. This fact may explain the "activation" of the electrode surface due to chemical reaction with the gas flow or film deposition. The very weak temperature dependence of the emission current (see Fig. 5a) is obviously due to the fact that the effective work function of the surface layer of the electrode is a function of temperature. This may be due to a change in the composition of the surface layer when the temperature changes. Unfortunately, the small temperature range over which measurements were made does not enable us to draw any more definite conclusions.

The main part of the cathode branch of the current-voltage characteristic is the section CD. Its characteristic feature is the fact that a current considerably greater than the emission current is drawn from the electrode. Due to the small potential drop outside the space-charge layer for large currents (see above) this part of the characteristic describes the change in potential in the space-charge region close to the cathode.

The physical processes which give rise to an increase in current above the emission current can be divided into two groups. The first includes the Schottky effect and thermoautoelectron emission, due to which the electron component of the current increases, and the second includes impact ionization (direct and multistage), which causes an increase in the ionic component. A detailed consideration of the combined action of these two factors when the electron and ion temperatures are the same and when there are no collisions in the space-charge layer has been given in [1, 2]. If we neglect the random ion current density, the corresponding relation for calculating the potential drop in the space-charge layer is

$$j = j_e \exp\left\{\frac{4.39}{T}\sqrt{E}\right\}[1 + C_a(U - U_{ia}) + C_k(U - U_{ik})] \quad (4)$$

Here U_{ia} and U_{ik} are the ionization potentials of argon and potassium, C_a and C_k are quantities which depend on the probability of impact ionization and the argon and potassium densities, and E is the electric field strength at the electrode surface. It is shown from general considerations in [1] that E can be represented in the form

$$E = \beta \frac{U}{d}\left(\frac{eU}{kT}\right)^{\alpha} \quad (5)$$

where α, β are constants defined by the properties of the electrode surface, and d is the effective thickness of the space-charge layer, of the same order of magnitude as the Debye length, calculated from the parameters in the region of the electrode surface.

The difference between the temperature of the electron gas and the temperature of the heavy particles leads to an increase in the charged-particle density. The random ion current from the plasma will increase compared with the case when $T_e = T_g$, and the effective thickness of the space-charge layer will decrease. If we assume that the increase in the electron component of the current density occurs more rapidly than the increase in j_i and that d depends only slightly on the current density, then using (4) and (5) we can suggest the approximation equation (3) to describe the section CD of the cathode branch of the current-voltage characteristic. Equation (3) is obtained from (4) and (5) by putting $\alpha = 0$, $\beta = 1$, $4.39/T\sqrt{d} = a$ and also simplifying the expression taking impact ionization into account. Instead of separately taking into account the results of ionization of argon and potassium in (3), the parameter c occurs which represents the total effective impact ionization of all the gas components.

The assumptions made above require a more thorough experimental proof, but the fact that (3) is a good approximation to the actual characteristic is evidence in favor of these assumptions.

Figure 6 shows that the quantity a in Eq. (3) is practically independent of the temperature. Hence we can conclude that the thickness of the space-charge layer d depends only slightly on temperature under these conditions. The value of d, calculated from a, is of the order of 10^{-4} cm, which agrees with the assumption that the thickness of the space-charge layer is small compared with the mean free path.

In order to estimate a possible value for the impact ionization constant in Eq. (4), we used data on the ionization probability p_i for argon and potassium. From the data given in [5, 6] it is easy to obtain that for low electron energies ($U \lesssim 2U_i$), when the relation for the current has the form (4),

$$p_{ia} \approx 0.02\,(U - U_{ia}), \qquad p_{ik} \approx 0.01\,(U - U_{ik}) \tag{6}$$

At energies $U < 15$ V only potassium atoms can be ionized, but the probability of a collision with these is small due to their small concentration. Nevertheless, since electrons lose a small portion of their energy in elastic collisions with argon atoms, we might expect a certain increase in the ion current due to ionization of the scattered electrons. Obviously in this case in Eq. (4) $C_k \leqslant 0.01$. For potential drops of the order of $15 \leqslant U \leqslant 30$ V argon is ionized, and the ionization probability is calculated from (6), and consequently the corresponding value in Eq. (4) is $C_a \geqslant 0.02$. For electron energies $100 > U > 30$ V the ionization probability depends only slightly on the energy, and $p_i \sim 0.3$. For large potential drops (of the order of several ionization potentials) the electron possesses enough energy to ionize U/U_i atoms, and in view of the high ionization probability all the ionization events occur due to a few collisions. If all the ions so formed arrive at an electrode, the corresponding increase in the ion current will be

$$I_i = i_e U/U_i = i_e C_a U, \qquad C_a \sim 0.06 \tag{7}$$

This expression has the same form as the corresponding terms in Eq. (4), though for this process $C_a \leqslant 0.06$. The experimental current-voltage characteristics are approximated over the section CD by expression (3) in the region of high electrode potential drops. Hence the value of c in Eq. (3) must lie in the limits $0.02 \leqslant c \leqslant 0.06$. The experimental values obtained (see Fig. 7) are in satisfactory agreement with this conclusion.

LITERATURE CITED

1. G. A. Lyubimov, "Electrode layers with a sharp change in potential at hot electrodes," Teplofiz. Vysok. Temp., 4, No. 1 (1966).
2. G. A. Lyubimov and V. N. Mikhailov, "Analysis of the perturbation region of a plasma in the electrode region," Izv. Akad. Nauk SSSR, Mekh. Zhidkosti i Gaza, No. 3 (1968).
3. V. O. German, G. A. Lyubimov, and B. V. Parfenov, "Experimental investigation of the operation of an electrode in a current of air seeded with potassium," Teplofiz. Vysok Temp., 5, No. 3 (1967).
4. O. Biblarz and R. H. Eustis, "Discharge characteristics in a seeded plasma flow," Electricity from MHD, Vol. 1, Vienna (1968).
5. V. L. Granovskii, Electric Current in Gases [in Russian], Gostekhizdat, Moscow-Leningrad (1952).
6. G. Francis, Ionization Phenomena in Gases [Russian translation], Atomizdat, Moscow (1964).