AN EXPERIMENTAL INVESTIGATION OF THE EFFECT
OF OXYGEN ON THE EROSION OF A MULTICHANNEL
TUNGSTEN CATHODE

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UDC 533.9.07

The problem of the preservation of electrodes and the reduction of their depletion to a minimum has decisive significance among the practical questions of maintaining the efficiency of steady sources and plasma accelerators during an extended period of operation. For this purpose electrodes are made, as a rule, out of the refractory metals in familiar designs. In particular, tungsten, which has the highest phase-transition temperatures among all the metals, thus permitting an appreciable reduction in the evaporation rate of the material under the thermally stressed operating conditions of these devices, is attractive. However, there exist other factors besides evaporation which affect the integrity of hot tungsten electrodes and their ablation (chemical processes, erosion in microarcs, ion bombardment, and so on). One of these factors requiring special investigation is the chemical interaction of tungsten with the oxygen contained in the working medium as an industrial impurity. Such an impurity can, for example, be present in industrially pure alkali and alkali-earth metals used as the working media. In addition, these metals can be contaminated by the oxygen of the air in the process of different industrial operations, in particular, in connection with the servicing of the supply system. The entrance into the working material of oxygen liberated from the structural elements of the supply route and the operating cavities of the plasma source is not excluded. In practice it is impossible to eliminate oxygen, even when using contemporary methods of deep cleaning of lithium, due to its great affinity for oxygen. The actual role of the oxygen impurity and the necessity for taking it into account in connection with the creation and operation of plasma devices can only be revealed as a result of direct experimental investigation on samples having an electrode geometry similar to the class of sources or accelerators in question. Results are presented in this article of an experimental investigation of the erosion of a multichannel tungsten electrode [1-4] and a coaxial source of lithium plasma [5]. The use of lithium as the main plasma-forming material ensured the maintainance of a discharge distributed over the electrodes and having a small value of the potential drop next to the cathode, which lies below the threshold of cathode erosion upon bombardment of the surface of polycrystalline tungsten by lithium ions [6]. Thus, the phenomena of cathode destruction associated with microarcs and ion bombardment could be exluded from consideration in practice. Thus evaporation and chemical ablation due to interaction with the oxygen of the working medium remain the principal competing processes contributing to the erosion of the cathode mass. Therefore, in order to obtain reliable quantitative relationships which characterize differential erosion due to oxidation and evaporation, the oxygen content in the working medium was varied in the experiments described within limits extending beyond the framework of the usual industrial impurity, which does not exceed tenths of a percent. One should note that in the process of investigating the role of an industrial oxygen impurity in causing the erosion of a tungsten cathode in a lithium plasma its direct effect was revealed on the anomalous current of a hollow cathode, which is characterized by the recorded average densities of the discharge current exceeding significantly the values of the emission current given at the same surface temperature by the Richardson—Schottky equation [7-9].

1. Experimental Procedure. For the experimental investigations which were undertaken a steady coaxial source of a lithium plasma in an open version (Fig. 1) was used which is described in [5] with a multichannel tungsten cathode similar to that of [1-4]. The vapors of the working medium entering into the multi-
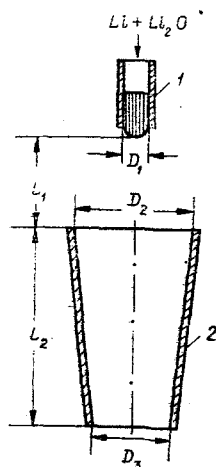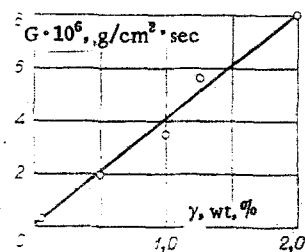
Fig. 1

Fig. 2

channel tungsten cathode 1 and the interelectrode space are ionized, maintain the operation of the discharge, and flow out in the form of a plasma jet through the molybdenum anode 2, which is a tapered truncated cone ($D_2 = 85$ mm, $D_3 = 60$ mm) of comparatively large length ($L_2 = 175$ mm). The diameter of the working part of the cathode is $D_1 = 18$ mm, and the interelectrode distance is $L_1 = 80$ mm. A source consisting of a cathode and anode subassembly with the necessary current supplies was mounted on a special holder and attached to the flange of a vacuum chamber ~ 20 m^3 in volume cooled by water.

In order to produce in the working medium a specified oxygen concentration which is higher than an industrial impurity, the specific additive lithium oxide Li_2O, whose solubility in liquid lithium has a quite definite temperature dependence [10], was introduced into the working medium. In the experiments a saturated solution of lithium oxide in liquid lithium produced in advance was introduced into the source from the supply system at a fixed temperature corresponding to the required oxygen concentration in the working medium. Thus, an oxygen concentration of 0.5% by weight was maintained in the working medium by the production of a saturated solution of lithium oxide in liquid in lithium at $T = 759°K$ (0.935% by weight of Li_2O), 1% by weight at $T = 831°K$ (1.87% by weight of Li_2O), 1.5% by weight at $T = 893°K$ (2.81% by weight of Li_2O), and 2% by weight at $T = 953°K$ (3.75% by weight of Li_2O).

The effective vacuum under the conditions of an operating source was usually $\sim 10^{-4}$ mm Hg. During the entire operation of the plasma device the electrical parameters of the discharge (current, voltage), the flow rate of the working medium, and the temperature of the electrodes measured with the help of an OPPIR-017 optical pyrometer were recorded by an NOO4M loop oscillograph. The parameters of the interelectrode plasma (concentrations and temperatures of the neutral and charged particles) were determined in the necessary cases by probe and spectroscopic methods [5].

The removal of material of the polycrystalline tungsten elements of the cathode during operation of the discharge was found as the difference of the values obtained by weighing the cathode on an analytical balance prior to triggering the source and after the conclusion of its continuous operation, which continued up to 2 h and more in the experiments. The procedure applied provided for a careful cleaning of the cathode after an experiment of contamination by lithium and its compounds by means of treatment with boiling distilled water for several hours, flushing in a solvent, and subsequent complete drying in a desiccator adjusted periodically by controlled weighing of the cathode on an analytical balance.

While the source was operating, a continuous record was also made by an oscillograph of the heating of the subassembly for the evaporation of the working substance and of the temperature of the liquid solution in the supply system.

2. Characteristics of the Discharge Operation. Both when industrial lithium was supplied to the plasma source and lithium artificially enriched with lithium oxide was supplied, the discharge was easily triggered with a preliminary warming up of the multichannel cathode to temperatures of $\sim 1000-1500°K$ and upon the appearance in the interelectrode space of vapors of the working substance. The voltage necessary for this was usually less than ~ 60 V. The difference of the potentials upon the operation of the discharge with an average mass flow rate of the working medium of $\dot{m} = 0.01$ g/sec and $I_d = 800$ A was $\sim 20-22$ V. Optical investigation of the behavior of the discharge at the cathode of an operating source in the subcritical region permitted establishing

the nature of its operation with a distribution over all channels; in the basic operating mode with $\dot{m} = 0.01$ g/sec and $I_d = 800$ A all the channels were "lit up," i.e., appeared active in emission and plasma formation processes. No contraction of the discharge and the microspot was observed, and the measured maximum temperature of the cathode surface was at the level of 2700-2800°K.

According to the Richardson—Deshman equation, the saturation thermoelectron current density j_e from polycrystalline tungsten ($\varphi_W = 4.54$ eV), which is equal to ~3-6 A/cm², corresponds to this value of the temperature. Actually, the density of the discharge current realized in the cathode under discussion was appreciably higher.

As the dismantling of cathodes which have operated several hours at discharge currents up to 1000 A ($\dot{m} = 0.01$ g/sec) and the visual analysis of the state of their tungsten elements have shown, quite definite traces of burning of the discharge were observed on the inner surface of the channels, which reached a depth $\delta \sim$ 2-4 mm (1-2) wire diameters). The appearance of the cathode elements indicated a localization of the discharge in the indicated regions both by the color of the metal in them in comparison with the remaining surface and by the erosional alteration of the structure of the tungsten surface. Based on this factual data, the estimated average density of the discharge current with the cathode surface emitting and also with the end surface of its elements taken into account was $\gtrsim 36$ A/cm² (36-62 A/cm², depending on the value of δ) for $I_d = 800$ A.

As follows from the optical observations, the surface which effectively emits is evidently determined mainly by the walls of the cathode cavities. In this case the discharge current density realized from the cathode is higher than the estimate made; in the case under discussion it is ~45-90 A/cm², depending on the assumed penetration depth of the discharge into the channels. Thus the comparison cited above of the thermo-electron possibilities of pure polycrystalline tungsten and the current densities produced in practice from a tungsten multichannel cathode in the distributed-burning mode reveals their rather convincing discrepancy. One should note that the Schottky correction does not rectify the situation; for the electric field intensity occurring in the Langmuir layer ($E_W < 10^5$ V/cm) it is comparatively small ($\Delta j_e < 50\%$).

Thanks to the use of a special optical system for observing behind the cathode surface [11], it was possible to investigate the physical state of the lithium plasma in the exit section of the channels and to determine its parameters. The results obtained permitted estimating for an average mass flow rate of hundredths of a gram per second the values of the fluxes of heavy lithium particles (ions and atoms) from the plasma to the emitting surface. According to this estimate, the value of the flux ν can amount to ~10^{22} particles/cm²·sec under the experimental conditions. According to the existing data [12], the degree of covering θ of the surface of the polycrystalline tungsten by adsorbed lithium particles ($T_W = 2500$-2800°K) is comparatively small and has a weak effect on the work function of its surface. However, the presence of oxygen in the cathode plasma makes the adsorption two-component in nature and appreciably alters the degree of covering of the tungsten surface. When lithium and oxygen particles are simultaneously falling onto the surface, a double molecular film is formed which is characterized by high-temperature strength and effectiveness in reducing the work function.

It is of interest to estimate the role of oxygen as an admixture to lithium in the process of adsorption and emission on the working surface of a tungsten cathode. With a weight content up to ~0.1% by weight of oxygen in lithium (industrial lithium) the adsorption of oxygen of polycrystalline tungsten at $T_W = 2500$-2800°K in the bound β_1 state (the first layer of chemisorbed oxygen atoms, which is characterized by the strongest bond to the backing [13]) causes, according to a calculation based on the procedure of [13], the existence of a covering with $\theta_1 \approx 1$ (θ_1 is the degree of coverage of the first layer). According to the experimental data of [14], an increase in the work function of tungsten by $\Delta\varphi \approx 0.5$-0.6 eV corresponds to this covering in the case of the (100) face. Bearing in mind that the adsorbed characteristics of the (100) face and polycrystalline tungsten are similar to each other, we obtain a qualitative characteristic of the emission of tungsten with oxygen for $\Delta\varphi \simeq 0.5$ eV by using for the estimate the experimental results on the adsorption of lithium on tungsten ((100) face) covered by a film of oxygen [15]. Experimental values of the work function of the electrons φ_e and the heat of desorption of lithium atoms from an oxidized tungsten surface as a function of the concentration of adsorbed lithium particles were used in calculations based on the equations of the adsorption isotherm and those of Richardson—Deshman. The density obtained for the thermoelectron emission j for $T_W = 2500$-2800°K and $\nu \approx 10^{22}$-10^{23} particles/cm²·sec lie in the range ~40-400 A/cm².

The effect of oxygen on the parameters of a lithium discharge due to a change in the surface characteristics of the cathode was observed especially clearly in tests associated with a special increase of the pressure in the vacuum chamber to 1 mm Hg by admitting outside air into it. This increase in the pressure was accompanied by an appreciable reduction of the discharge starting voltage (up to ~25-30%). When argon was ad-

mitted into the chamber, no change occurred in the volt—ampere characteristics of the lithium discharge in the model. In addition, the use of argon and nitrogen as the working medium in a plasma source similar to the one described here led to enhanced destruction of the tungsten cathode at lower operating conditions with respect to the current. After ignition of the discharge due to the increased contamination of the first portions of lithium by oxygen (because of oxidation in the process of servicing the supply system and in the supply system itself, and also as a result of gas liberation from the structural materials upon heating up) the voltage of the discharge is set lower than in the case of extended operation of the source, when the vacuum conditions and purity of the lithium improve.

Thus, the estimates made indicate that an admixture of oxygen to the working medium can play an important role under the specified conditions in the emission mechanism of electrons, which actually occurs from the surface of the multichannel cathode of sources and accelerators of lithium plasma.

3. Chemical Erosion of the Cathode. An experimental investigation of erosion was performed with practically constant discharge parameters ($I_d = 800$ A, $U_d = 22$-24 V) and various measured amounts of oxygen in the lithium supplied to the source (lithium flow rate ~0.01 g/sec). A series of experiments was set up with the goal of obtaining the value of the erosion rate upon the use of industrial lithium, which was further used to prepare the combined working medium. The oxygen content in the industrial lithium of the batch used was ~0.06% by weight according to the data of the laboratory analysis conducted. As a result of the dismantling of the multichannel cathode, it was established that the surface changes (color, structure) and the traces of destruction are observed over the entire length of its elements; however, the sections adjacent to the interelectrode space were subjected to the greatest erosion. The area occupied by the discharge is, according to the inspection carried out, evidently determined mainly by the lateral surface of the filling elements of the cathode to a depth of up to ~4 mm; the ends of the elements introduce a certain correction (this area amounts to approximately one-fifth the total area of the elements). The pattern of the erosion became more significant upon inspection of details after operation of the cathode in a medium with an increased oxygen content. In connection with the ablation pattern indicated above, the entire surface of the cathode elements (wires) was taken in connection with finding the average specific rate of tungsten removal. The maximum cathode temperature in the experiments measured with the help of an optical pyrometer was ~2700-2800°K. As a result the use of the experimental procedure described above for the investigation of the chemical erosion of a tungsten cathode, it was found that an increase in the amount of the oxygen admixture in lithium to 2% by weight is accompanied by an increase in the removal of mass from the cathode elements of more than an order of magnitude from ~$3.3 \cdot 10^{-7}$ g/cm$^2 \cdot$ sec for ~0.06% by weight of oxygen (industrial lithium) to ~$8.0 \cdot 10^{-6}$ g/cm$^2 \cdot$ sec (lithium containing 2% by weight of oxygen). The obtained dependence of the specific erosion G on the amount of oxygen γ is given in Fig. 2, where it is satisfactorily approximated by the relationship

$$G = G_0 + \alpha\gamma, \tag{3.1}$$

where $G_0 = 0.1 \cdot 10^{-6}$ g/cm$^2 \cdot$ sec corresponds to the mass removal during operation on pure lithium ($\gamma = 0$); $\alpha = 3.83 \cdot 10^{-6}$ g/cm$^2 \cdot$ sec, and γ is the weight %.

Essentially, the second term in (3.1) gives the contribution of chemical erosion due to oxidation and removal of tungsten oxides. According to methodical research conducted earlier with the use of special indicators and an optical system in combination with an ÉOP-66 electronic optical pyrometer [11], it was shown that an appreciable temperature gradient occurs in the working region of the multichannel cathode along the surface of its elements. Taking this fact into account, one can assume that the term G_0 in (3.1) gives the average value over the elements of the specific erosion of tungsten caused by its evaporation. Thus when industrial lithium is used, the average rate of specific erosion of the polycrystalline tungsten cathode is $G \approx 0.33 \cdot 10^{-6}$ g/cm$^2 \cdot$ sec. The results obtained show that chemical ablation of the cathode when industrial lithium is used can predominate over erosion due to evaporation; in this investigation the fraction of chemical ablation was ~70% of the total mass removal.

In connection with the experimental results obtained, it is interesting to estimate on the basis of contemporary ideas the rate of oxidation of polycrystalline tungsten in the range of temperatures and low partial pressures of oxygen that are characteristic of the operation of a multichannel cathode with lithium having an admixture of oxygen. Adsorption and dissociation of molecular oxygen in the two different bound states β_1 and β_2 (the degrees of covering are θ_1 and θ_2, respectively) precede oxidation of the tungsten [13]. The formation of the oxides WO_2 and WO_3 becomes significant after the first layer (β_1 state) is filled by atomic oxygen and after an increase in the covering θ_2 of the second layer (β_2 state). Desorption of the atomic oxygen from the first layer and of molecular oxygen from the second layer occurs simultaneously with the formation of these oxides (as well as the polymers W_2O_6 and W_3O_9 in small amounts). One can neglect evaporation of atomic oxy-

gen from the second layer in comparison with the departure of O_2. One can obtain for the physical model of adsorption and oxidation of tungsten based on the theory of active centers [13] relationships for the rate of formation of tungsten oxides with the partial dissociation of molecular oxygen in the gaseous phase $(0 < \alpha_d < 1)$ taken into account.

According to these relationships for the cases $\alpha_d = 0$ and 1, a calculation is made of the rate of oxidation of polycrystalline tungsten at partial pressures of oxygen of $p_\Sigma = 1$ and 10 μbars, which characterizes the realizable region of pressures in a multichannel cathode when industrial lithium is used. The experimental result on the chemical erosion of a multichannel cathode lies in the range of ablation values given by a theoretical calculation in the crude approximation that all the oxides formed are carried away by the flow.

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