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TRANSFER OF EXCESS BARIUM IN AN OXIDE CATHODE

By

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RUSSIAN AND ENGLISH TRIGONOMETRIC FUNCTIONS

| Russian | English | Russian | English | Russian | English |
|---------|---------|---------|---------|----------|--------------------|
| sin | sin | sh | sinh | arc sh | sinh ⁻¹ |
| COS | COS | ch | cosh | arc ch | cosh |
| tg | tan | th | tanh | arc th | tanh |
| ctg | cot | cth | coth | arc cth | coth |
| sec | sec | sch | sech | arc sch | sech |
| cosec | csc | csch | csch | arc csch | csch ⁻¹ |

| Russian | English | | |
|---------|---------|--|--|
| rot | curl | | |
| lg | log | | |

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TRANSFER OF EXCESS BARIUM IN AN OXIDE CATHODE

V. P. Vasil'yev, I. M. Knyazeva, S. L. Payan, A. Ye. Chernov

Introduction

According to the views which exist at present excess barium is the main active component of an oxide cathode (OK). The thermionic emission of an OK depends on the amount of excess Ba. According to our data [1] there is a correlation within specific limits between the concentration of excess Ba in an OK and thermionic emission.

Excess Ba is formed both as a result of thermal activation (apparently Ba is distributed uniformly over the surface of the oxide grains) and due to chemical reactions with the metal of the core and active additives in it. A large part of the excess Ba is formed in the core area.

Up to now the mechanism of transfer of excess Ba has been vague. Here there is the possibility of volumetric diffusion of Ba through the oxide grains, migration of Ba over the surface of the grains, and Knudsen flow via the pores of the covering. In certain cases mixed mechanisms of Ba transfer are possible.

Experiments [2-4], where radioactive barium carbonate was applied in a thin layer on the surface of the core and covered from above with ordinary paste, did not give a clear answer on the mechanism of transfer of excess Ba. In these tests the transfer of the entire barium component took place (BaO - Ba), and, which is more significant, a considerable redistribution of the barium component took place during the breaking down of the carbonates and activation of the cathode.

We established that the spraying of Ba from above onto the surface of the OK, as this was done in [5], leads to a considerable penetration of the sprayed substance into the coating via the pores in the process of spraying.

Experimental Method

We developed a method for the transfer of excess Ba which excludes the influence of the process of activation. For this, on the end nickel core of the cathode, which has an opening in the middle, we pressed a nickel sponge, which by transillumination did not have through openings (specific weight of sponge 1 g/cm³, thickness 100 µm). The area of the sponge comprised 0.1 cm² (Figure 1). On the outside on the core an insuled preheater was put on. It was equipped with a heat screen. A coating of ternary carbonate on a binder 80-100 µm thick was applied to the sponge. The cathode was mounted together with the source of Ba, which was a niobium cylinder with a diaphragm, on the face of which there was a niobium sponge, where the BaCO, was rubbed in. The temperatures of the cathode and the source were measured with chromel-aluminum thermocouples. During the processing of the cathode and the source a screen was placed between them. The anode consisted of three molybdenum and one nickel leaves. A tungsten helix served for degasification of the anode (Figure 1).

The vacuum complex [6] consisted of one or two investigated tubes, a Bayard-Al'pert manometer, a titanium sprayed getter, and an ampoule with radioactive iodine, when this was possible.

The vacuum complex was evacuated at a temperature of 450° C and pressure of $1 \cdot 10^{-4}$ torr. Then the carbonates were dissolved in the source of Ba and the oxide cathode at 750-800°C, the device was annealed again in a furnace at a temperature of 400° C, the metallic parts were degasified by electron bombardment and the source and then the cathode were activated (the cathode in a regimen of

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 $850^{\circ}C - 3 \text{ min}$, $900^{\circ}C - 5 \text{ min}$). After this the vacuum complex was unsoldered from the installation.

Further evacuation to a superhigh vacuum was done with a titanium getter. Spraying of Ba onto the cold cathode was done in a vacuum of $(5-6) \cdot 10^{-9}$ torr and source temperature of $700-750^{\circ}C$ for several hours. First the initial effective work function of the OK φ was determined at the same temperature at which the diffusion heating was conducted subsequently.

During diffusion heating the work function (emission) of the OK was determined periodically. Heating was carried out up until a stable work function was obtained. Determination of migrated Ba was done with the help of I^{131} using the method from [16] or based on Ba¹⁴⁰. For this the cathode was paraffinized and cut into layers with a microtome. The analysis of radioactive substance was performed on a "Volna" counter.

Results and a Discussion of Them

A change in φ was observed in the first hours after the establishment of the operating temperature of the cathodes (Figure 2a). Sometimes jumps in emission were observed immediately after the spraying of Ba. This could be explained by thermal heating of the cathode during spraying of barium from the source. An increase of emission continued for tens of hours and virtually we were not able to notice its complete saturation. Figure 2a shows the effective values of work function φ on the time of heating from cathode temperatures $T_K 350-500^{\circ}C$. The main reduction of φ takes place in the first 2-3 hours. We were not able to observe regularities between the final and the initial work function.

If we evaluate the coefficient of diffusion D approximately, in our case from the relationship $I=2\sqrt{Dt}$, then for the conditions of the experiment 1=0.01 cm, $T=380^{\circ}C$ and time t=1 s, we obtain exceedingly high diffusion coefficients: $D=2.5\cdot10^{-5}$ cm/s. They can be compared with data from the literature [7]: for this temperature $D_{00} \sim 10^{-12}$ cm²/s, $D_{008} \sim 10^{-7}$ cm²/s.

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On the basis of this it can be concluded that the transfer of excess barium is realized by Knudsen flow, i.e., by molecular flow. In the tests there was no direct transit of Ba through the sponge and the oxide coating, which follows from data on evaporation on the anode in the case of spraying of Ba from a source and from data for cathodes, on which there was no thermal heating from the source = Figure 2b).

The Knudsen nature of Ba transfer also follows from Figure 4, where data are shown on the distribution of excess Ba, determined on Ba¹⁴⁰ (curve 2). This distribution has the nature of distribution of the density of the oxide coating (according to N. N. Yakimov [8] and our data). The distribution of the radioactive barium component, applied preliminarily in the form of a thin layer of carbonate on the core of the cathode and coated from above by the usual composition, is shown in curves 3-5. They confirm the independence of the mechanism of transfer on the composition of the OK. In this case only Knudsen flow of the barium component from the core to the surface is possible.

Data of Ba evaporation onto collectors show that if in the beginning the evaporation proceeds somewhat more slowly, then after a certain time it acquires a constant value, i.e., a specific flow of Ba is established (see Figure 2b). The ratio of the amount of ba on the collector and the cathode is equal to 3-4.

From 10^{-6} to 10^{-8} g of Ba was found in the oxide layer (with a weight of the oxide coating of $1 \cdot 10^{-3}$ g). However, even a large quantity of Ba did not lead to a significant improvement of work function, as this should be for a naturally activated cathode (thermal), which was investigated by us earlier. A systematic deviation from the correlation noted by us was observed [1]. These data are confirmed by the results of work [9], where it is also noted that sprayed excess Ba is not retained during the heating of OK.

This indicates that the process of an activated oxide cathode is connected not only with the simple mechanism of movement of Ba over the surface of grains, but also with its fixation in certain centers of the lattice surface. An approximate calculation of the surface of the grains with a grain radius of 3 nm for normal ther-

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mally activated cathodes shows that all the Ba can be found in several surface layers. This is also indicated by data on the rapid contamination by oxygen of OK [10] and BaO [11], and also the coincidence of values φ for external and interal grains of oxide, obtained by us by means of the successive section of layers of OK in a vacuum and measurement of emission [3], and also by the method of probes in a magnetic field [14].

From the data of Redington and Rittner [12], on the other hand, it follows that the grains of oxide at temperatures of 350-500°C should become saturated with Ba in several tens of hours. Apparently this does not take place as a result of the weak absorption of Ba on the surface of the grains, the greater part of it is evaporated [9]. A considerable part of the sprayed Ba is not an activator. In order that it would become an activator it is necessary to activate the cathode thermally at a high temperature or by some other method.

In this light the role of additives in the core is only technologically justified for poor vacuum conditions, insufficient purity of materials, etc. From the principle side additives are not needed for the operaion of an oxide cathode and are even harmful as a result of the formation of interelectrode conductivity, thermoelectric current of the grids, etc.

November 1968



Figure 1. Experimental device.

1 - oxide coating on nickel sponge; 2 - cathode heater; 3 - heat screen; 4 - diaphragm; 5 - thermocouple Ba source; 6 - electric lead-out of anode; 7 - rotating anode; 8 - tungsten helix; 9 - cathode thermocouple; 10 - gate; 11 - Ba source; 12 - heater for Ba source.



Figure 2. a - change of work function of OK from the time of heating T equal to: $1 - 350; 2 - 390; 3 - 420; 4 - 450; 5 - 500^{\circ}C;$ b - evaporation of sprayed Ba on the anode (T=350°C). Key: 1 - hours; 2 - eV.



Figure 3. Distribution of barium component in the oxide coating: 1 - density of oxide according to our data and [8]; 2 - excess Ba according to our data; 3 - (BaO+ Ba) according to [2]; 4 - (BaO+Ba), coating 9% Ba ? 91% SrCO₃ T=1350°K, t=4 hours (our data); 5 - (BaO+Ba), coating 68% BaCO₃ 32% SrCO, T=1350°K, t=4 hours (our data).

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