An Investigation of Cathode Spots Functioning Process of a Composition Type Electrode

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Abstract — In this paper, the results of investigation devoted to the process of functioning of cathode spots on Zr-Y cathode are presented. The cathode spots evolution data with spatial and temporal resolution are included. The velocities of spots movement, as well as the spot mean current and current density have been measured. The regimes of arc burning with presence of drop fraction of the cathode material products are displayed.

1. Introduction

Nowadays due to hydrogen engineering advancement there is a need in development of effective fuel cells on the basis of thin-film solid oxide electrolytes [1, 2]. One of alternative paths to make such elements is vacuum ion-plasma thin-film deposition realized on the basis of an arc discharge in reactors with composition-type cathodes. As a rule, lifetime of such electric-arc reactors and coating quality depend on mode of cathode spot burning on composition-type cathodes. Should such spots function as if they are associated with definite places on cathode surface, then lifetime of such cathode, and, correspondingly, installation itself would be rather limited. Besides that, while using electric-arc method of thin-film deposition, an important problem is to prevent presence of drop fractions in plasma flow from arc evaporator.

This paper reports on the results of investigation devoted to cathode spots functioning process on Zr-Y cathode in operating gases argon and oxygen at arc current i=(100–200) A. The data of cathode spot evolution with spatial resolution l=0.1 mm and time resolution no worse than 100 ns are included. Rates of movement of spots, the values of the mean current and current density of a single spot have been measured. It has been shown that erosion of cathode surface is relatively uniform. The modes of arc burning, with which in plasma of a discharge column there is a considerable proportion of drop fraction of cathode material products, as well as the conditions under which microdrops number could be decreased have been investigated.

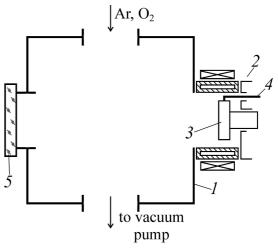


Fig. 1. The plasma chemical reactor scheme. 1 - vacuum chamber, 2 - arcing evaporator, 3 - Zr-Y cathode, 4 - trigger, 5 - observation window

2. Experimental arrangements and method of observation

For excitation of a low-pressure arc discharge, an electric-arc evaporator was used, which construction is described in a more detail in [3]. The ion-plasma installation, which included such electric-arc evaporator, consisted of a vacuum chamber 1 (Fig. 1), made of stainless steel, a gas system, and a system of electric ignition and control. The vacuum chamber sizes were 700×700×750 mm. of A roughing-down pump and a turbomolecular pump were used for vacuum chamber exhaust. Gas pressure in the chamber was $p=10^{-2}-10^{-4}$ Torr. Argon and oxygen were used as operating gases. The arc evaporator 2, providing plasma generation was placed at vacuum chamber lateral wall. The evaporator consisted of a water-cooled corpus and a cathode holder, isolated from each other electrically. A cylindrical composition-type cathode 3 with diameter $D_{ch}=120$ mm and height H_{ch} =25 mm was fixed on the cathode holder. Such cathode was made via method of electric-arc fusion of Zr and Y foils. The ratio of Zr and Y in electrode

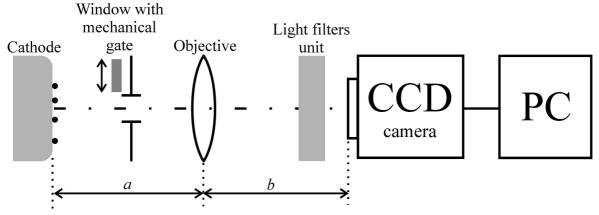


Fig. 2. Observation scheme of cathode spots with space-time resolution

material was 85:15. Next to the cathode, 5 mm distant from its side surface, there was placed a cylindrical shield, which prevented moving away of arc spots from the edge surface of the cathode.

A trigger electrode 4 located nearby the side surface of the cathode 3 initiated an arc discharge. In order to stabilize position of cathode spots, there was a solenoid settled on evaporator corpus. Besides that, there was also settled a system composed of 6 focusing coils, which allowed a configuration control of plasma flow. The arc evaporator provided stable ignition and stationary burning of an arc of low pressure with current of (80–250) A at combustion voltage of (20–40) V and operating vacuum of 10^{-3} Torr.

Cathode spots and their evolution observations were carried out through a window 5 by using a scheme presented in Fig. 2. Cathode surface image was projected to inlet matrix of a CCD camera by using an optical objective. Radiation was extracted through a quartz window, which was provided with a mechanical shutter to prevent cathode material vapors. Luminous flux attenuation was done by using diaphragms and neutral light filters. The spectral range of registration was $\Delta \lambda \approx (370 \times 800)$ nm.

For such image registration we used a SensiCam CCD camera with chip sizing of (8.6×6.9) mm, which contained 1280×1024 pixels with sizing of $(6.7\times6.7)\cdot10^{-3}$ mm. The brightness patterns of each pixel might be presented digitally. Besides that, the specially written program allowed an integration of such brightness patterns of the number of pixels required. That provided the necessary evaluation of relative brightness both of separate spots and groups of spots. The CCD camera provided shoot with exposure $t_{\rm exp}=100$ ns-100 ms as single pictures and as a continuity $N\le256$. In the last case time gap between shots was about 100 ms. Moreover, one shot might be superimposed with about ten sequential images with exposure $t_{\rm exp}=(0.1-1)$ μ s and a delay of (0.1-1) μ s.

For observation work, it was required that a registration system provided corresponding increasing of μ and spatial resolution Δl , that is related to selection

of a focusing element and distances *a* and *b* (Fig. 2). Inferences and calculations, based on which such selection has been done are presented below.

Lateral magnification of the system was selected out of consideration that the whole cathode image were placed on CCD camera chip. Taking into account that the maximal diameter of a cathode was $D_{ch} \approx 120$ mm, and lateral size of the chip was $l_{sh} \approx 6$ mm, the necessary magnification should be $\mu \approx l_{sh}/D_{ch} \approx 0.05$. In other words, our system should provide a twenty-fold decrease. Taking into consideration that the size of chip pixel was $\Delta l_p = 6.7 \cdot 10^{-3}$ mm, spatial resolution in perpendicular to optical axis direction was $\Delta l_p/\mu \approx 0.13$ mm. In order to calculate focal length f and distances a and b we employed the well-known set of equations for a thin lens:

$$\frac{1}{a} + \frac{1}{b} = \frac{1}{f}$$

$$\mu = \frac{b}{a}$$
(1)

It follows from (1) that:

$$b = \alpha \times \mu, \tag{2}$$

$$f = \alpha \times \mu / (1 + \mu). \tag{3}$$

In our experimental conditions the value a was restricted by the sizes of plasma reactor, being equal to a 100 cm. While selecting a=120 cm, and taking into account that μ =0.05 from (2) and (3), we can get $b\approx f$ =60 mm. The most correct lens with a focusing length f=58 mm might be Helios 44 objective, which was chosen by us for measurements to be done.

3. Results of observation and discussion

Experimental observations were done in the range of arc current i=(100-150) A. At the first stage, we took a continuity of several tens shots of cathode spots glowing image with exposure $t_{\rm exp}=0.5~\mu{\rm s}$ and time interval between shots $\Delta t=100$ ms. From such observations it was seen that the spots exist as groups on the cathode surface area with diameter $D_{\rm e} \le 3$ cm.

Such groups included from one to several spots. During arc burning, spots were observed with equal probability in various places on cathode surface that suggests chaotic motion of spots and uniform erosion of the cathode occurred. Table I presents the average number of groups N_g and the average number of spots N_{gg} calculated for various gases and arc current values.

Table I. The average number of groups N_g and the average number of spots N_{sp} at various values of arc current and operating gases

	Ar, <i>i</i> =105 A	Ar, <i>i</i> =150 A	O ₂ , <i>i</i> =150 A
N_g	1	2	5
N_{sp}	3	5	15

It is seen that the number of groups and the number of spots depend on gas type and arc current value. Thus, for oxygen at the given current value, the number of spots is about three times greater than that is in argon. Correspondingly, the mean current for a spot in oxygen is three times less, being equal to $i_{sp} \approx 10$ A/spot (at $i_{sp} \approx 30$ A/spot in argon). Such effect is probably connected with the great number of oxide films, presenting on cathode surface and initiating spots occurrence. For one and the same gas type, the number of spots increases with current rise, and at that the spot mean current is not noticeably changed.

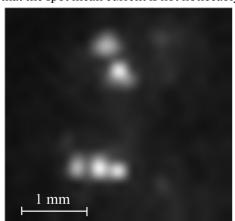


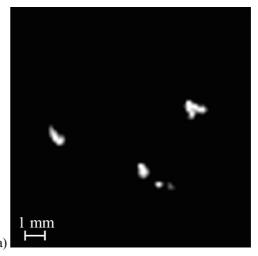
Fig. 3. Image of cathode spots. Ar, i=150 A, $t_{exp}=0.5 \mu s$

Figure 3 depicts enlarged image of a group of spots. Exposure is t_{exp} =0.5 μ s, operating gas is argon, arc current value is 150 A. The group consists of five spots. The average diameter of the spots, measured at the base of glowing area is $D_{sp} \approx 0.3$ mm. A spot has an area with 0.1 mm sizing, which glowing brightness is much greater than that of another areas that suggests a non-uniform distribution of current density in a spot. All said above allows an estimation of the average and maximal densities of current per spot, respectively, $j_{av} \approx 5.10^4 \text{ A/cm}^2$ and $j_{max} \ge 10^6 \text{ A/cm}^2$. Taking into consideration that in oxygen as compared with argon the mean spot current is approximately three times less, and the spots are of similar sizing, one may suppose that in oxygen the maximal and the mean densities of spot current will be somehow less too.

Table II. Dependence of integral intensity of cathode spots glowing I_{Σ} averaged by 10 measurements on exposure time. Ar, i=150 A

$t_{\rm exp}, \mu { m s}$	0,5	1	10
I_{Σ} , arb. units	55	120	750

It is seen in Fig. 3 that at exposure of $0.5~\mu s$ there is no light tracks, occuring on spots moving over cathode surface. The pattern is not essentially changed at exposure increasing up to $1~\mu s$. The average number of spots and their sizing do not practically change, and brightness of glowing is about proportionally increasing with exposure time (Table II). So, a conclusion can be drawn that at $\Delta t \le 1~\mu s$ the spots have no much motion $\Delta t \ge 0.1~mm$, and none of old spots die and no new spots birth occur.



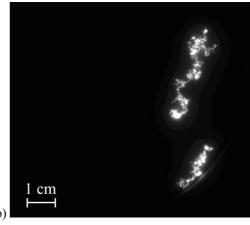
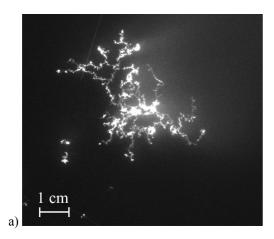


Fig. 4. Image of cathode spots. Ar, i=150 A, $t_{exp}=10 \mu s - (a)$, 10 ms - (b)

Another situation is observed at time interval increase up to $10~\mu s$ (Fig. 4, a). There are light tracks observed, and the number of spots increases. At that integral dependence of glowing increases disproportionately to exposure time (Table II). So, it is possible that at $\Delta t \approx 1-10~\mu s$ the spots move over the cathode surface within a distance $\Delta t \geq 0.3~mm$, and at the same time extinction of old spots and initiation of new spots occur.



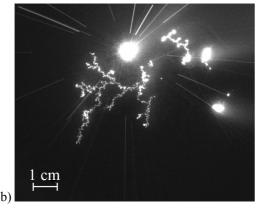


Fig. 5. Cathode spots glowing. O_2 , i=150 A. Operation time of the reactor $\Delta t > 10$ min (a), $\Delta t \le 10$ min (b)

It is possible to observe more distinctly the character of spots motion and processes of their extinction and birth at greater exposure values. Figure 4, b presents an image of spots glowing at exposure $t_{\rm exp}=10$ ms. The arc current i=150 A, gas argon was used. Remember that in these conditions there are mainly two groups of spots with the average number of spots $N_{\rm sp}\approx 5$. It is seen that there are light tracks, where at their background there are a great number of bright formations that suggests spots motion, their loss and birth. Velocity of their motion and direction vary, however it is possible to evaluate from the figure the mean velocity of spots motion as $V_{\rm ap} \approx 5 \cdot 10^2$ cm/s.

These findings are in agreement with the results obtained during observations of cathode spots motion on composite-type CuCr30 cathodes [4].

Figure 5, a demonstrates surface image in the case when argon was changed for oxygen. The estimations show that the mean velocity of spots moving was not essentially changed, however, it is seen that the spots as compared with the discharge in argon occupy the larger area of the cathode surface. As it was mentioned above, in other equal conditions, in oxygen there occur much more single spots.

On operation of arcing evaporator in argon, there were no microexplosions and light tracks on the cathode surface, which could be associated with presence of drop fraction. Nevertheless, in oxygen, during the first 10 min of evaporator operation there occurred another situation (Fig. 5, b). Based on light tracks, the velocities of drops moving were estimated as V~5 m/s. After 10 min of arcing evaporator operation, the bright plasma formations on the cathode surface and accompanying process of light tracks occurrence practically ceased (Fig. 5, a). Such an effect could be related to elimination of macroparticles of active yttrium in oxygen atmosphere. Such supposition is confirmed by decreasing of microdrops number after cathode aging during 10 min. These investigations allowed having an optimized mode of deposition of nanostructures films of zirconium dioxide stabilized by yttrium (YSZ), which could be used as an electrolyte in the cells of fuel elements.

References

- [1] S.C. Singhal, Solid State Ionics 135, 305 (2000).
- [2] Huayang Zhu and Robert J. Kee, Journal of Power Sources 117, 61 (2003).
- [3] Shygurov V.V., Strumilova N.V., Kolubaeva Yu.A, *in Proc. Ist Russian Conf. on Physic and chemistry of the high energy system*, 2005, pp. 346–348.
- [4] K.K. Zabello, Yu.A. Barinov, A.A. Logachev, S.M. Shkol'nik, in 21st Int. Symp. on Discharge and Electrical Insulation in Vacuum, 2004, pp. 280–283.