SOME EXPERIMENTAL DATA ON A GAS DISCHARGE BETWEEN A FLOWING ELECTROLYTE AND A METAL ELECTRODE

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Experimental data on a gas discharge obtained for comparatively high currents (4–11 A), powers (5–15 kW), and current density at a liquid cathode (0.8–1.0 A/cm²) are presented. As the electrolyte, a solution of sodium salt in distilled water was used. The losses on the liquid cathode were significantly diminished by decreasing its electric resistance. Regimes in which the thermal efficiency of a discharge apparatus is equal to ~80% have been determined.

Introduction. Gas discharges between a liquid electrolyte and a metal are unique in having a volumetric (diffuse) form at atmospheric pressure. The characteristics of such gas discharges are substantially dependent on the chemical composition and concentration of the electrolyte. Discharges arising between a high-concentration electrolyte (with a mass concentration of $\sim 10\%$ or higher) and a metal are used for heating of metals in the process of their thermal and mechanical treatment [1]. Discharges initiated with the use of a low-concentration electrolyte hold considerable promise for modification of metal surfaces and purification of gas flows [2]. However, the gas discharge itself remains not clearly understood. The majority of investigations were carried out for the case of a stationary electrolyte, where many parameters of a discharge (its duration, power, current density, and others), which are important from the practical standpoint, are restricted by the ohmic heating of the electrolyte. The present work is devoted to investigating a discharge arising under the conditions of forced removal of Joule heat from an electrolyte by pumping it through the discharge-action zone. It should be noted that a flowing heat electrolyte was also used earlier, e.g., in [3]. Investigations with two flowing liquid electrolytes were also carried out in [4]. The works mentioned were carried out at comparatively low currents (tens and hundreds of milliamperes) and current densities at a liquid cathode (no more than 0.2 A/cm²). Under these conditions, a very small amount of the electrolyte evaporates, which prevents the formation of a plasma flow that could be used in practice. This has generated a need to investigate the influence of an increase in the above-indicated parameters of a discharge between a flowing electrolyte and a metal electrode (discharge current and its density) on the intensity of electrolyte evaporation.

Experimental. A diagram of a discharge unit in section is shown in Fig. 1. Discharge 1 was initiated between an electrolyte (liquid cathode) 2 and a metal anode 3. The velocity of an electrolyte flow 2 through the discharge-action zone was changed from 0.1 to 5.9 cm/sec. Anode 3 was made from grey cast iron in the form of a rod having a square cross section of size 9×9 mm with ends bent up at an angle of 45° . The lengths *s* of the horizontal portion was equal to 36 mm. The anode contained a space for a cooling liquid (tap water) with a round cross section of diameter 3 mm, which extended along its axis. The cathode was switched to a power source through a current lead 4 made from a technological graphite in the form of a plate of size 100×100 mm. The anode and the current lead were positioned at a distance h = 15 mm from each other. The thickness of the electrolyte layer h_e was changed from 10 to 14 mm. Since the surface of the liquid electrolytic cathode deflected inward under the action of the reactive vapor forces, the length of the discharge gap l was larger than the difference between h and h_e .

Electric power was supplied from a three-phase, full-wave rectifier connected to the secondary winding of a step-up transformer. The rectified current was controlled by continuously changing the voltage across the primary winding of the transformer. The negative terminal of the power source was grounded. The discharge power-supply cir-

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Fig. 1. Diagram of a discharge unit.

cuit did not contain a balance resistor. Investigations were carried out at currents of 4-11 A. The current I and the voltage U were measured by pointer devices of accuracy class 0.2.

A hydraulic pump provided the circulation of the electrolyte around a closed path including the discharge unit and a heat exchanger. The velocity of the electrolyte flow v in the discharge zone was estimated as the ratio between the mass rate of the electrolyte flow through the hydraulic pump and the cross-section area of the channel in the discharge unit. The instrumental error in measuring the velocity v, calculated by the method of processing the results of indirect measurements [3] at a confidence coefficient of 0.9, was ± 0.05 cm/sec.

The heat losses on the anode Q_a and the liquid cathode Q_c were determined by the calorimetric method. It was assumed that Q_c is equal to the amount of heat carried away from the heat exchanger by the cooling liquid.

The voltage drop across the liquid electrolyte cathode ΔU_c was determined by two different methods. The first of them was as follows. A special electrode-anode, in which the area of the working face was equal to the area of the conjugation of a discharge to the liquid cathode, was fabricated. The working face of this electrode was brought into contact with the liquid cathode and a low voltage was supplied from the power source. The voltage was increased as long as the current was equal to the current of the discharge initiated in the experiment. The voltage determined in this way was taken as the desired voltage drop across the liquid cathode. This method finds use in experimental practice in the case where both electrodes are liquids. The electrode closing the discharge gap is fabricated with account for the sizes of the conjugations of a discharge to the cathode and the anode [4, 6].

Since the area of the conjunction of a discharge to the cathode depends on the current and the electric conduction of the electrolyte σ , several special electrodes-anodes with different working faces were fabricated. As experiments have shown, at currents of ~8 A and higher, in the contact zone, gas is vigorously released and a discharge arises. An increase in the velocity of the electrolyte flow increased the range of currents at which a discharge is absent. However, this caused a dependence of σ on the temperature: the temperature of the electrolyte decreased with increase in its velocity because, in this case, it has no time to heat, and the ohmic resistance of the cold electrolyte was much higher than that of the heated one. Thus, the voltage drop determined at high currents was overstated because the electrolyte was heated during the discharge, and it was possible to measure ΔU_c only for the cold electrolyte.

The second method was free from the above-indicated disadvantage. It was based on the measurement of the potential distribution inside the electrolyte. For this purpose, we used a probe, the shape and dimensions of whose tip are presented in Fig. 2. The probe as a whole represented a tungsten wire 1 inside tube 2 of quartz glass. A lip located above the bulging part of the wire protected the wire from direct shorting by the discharge at the instant the probe is at the surface of the electrolyte. The error in measuring φ in the bulk of the electrolyte was smaller than that at its surface: it increased as the probe moved to the electrolyte surface. It seems likely that the main reason for this is the presence of vapor bubbles. The spread of values of φ at the electrolyte surface reached 5 V.

The temperature in a plasma flow was measured by a PR-30/6 platinum-rhodium thermocouple that was moved in three mutually perpendicular directions with the use of a traverse gear. The temperature in the discharge re-



Fig. 2. Diagram of a probe.

gion between the liquid electrolytic cathode and the anode was higher than the upper limit of the thermocouple throughout the range of currents investigated.

The current density was calculated as the ratio between the current strength and the area of the electrolyte surface in the region of conjugation of a discharge to the liquid cathode. The dimensions of this region were determined by the video image and photographs, taking into account the "pit" formed in the liquid cathode during the discharge. The depth of the "pit" was estimated by the values of the potential measured inside the electrolyte. Photographic camera 5 was mounted at an angle to the surface of the liquid cathode, as shown in Fig. 1.

It was assumed that the mass rate of electrolyte evaporation is equal to the mass rate of a fresh-electrolyte flow added into the discharge unit on condition that the level of filling of the unit remained unchanged i.e., at $h_e = \text{const.}$

The electrolyte conduction increased with increase in the discharge time, which was supported by the measurements of σ before and after the experiment at a constant electrolyte temperature. The value of σ measured after the experiment was larger than its initial value; therefore, distilled water was used as the fresh electrolyte. Because of this, σ increased by a smaller value and did not exceed 10% of its initial value after one hour of a continuous discharge. (Hereinafter we deal with the values of σ measured at room temperature.)

Results and Discussion. The surface of the electrolyte deformed under the action of a discharge. It deflected inward with the formation of a depression. The depth of the "pit" in the electrolyte under the anode increased with increase in the current. Thus, at high currents the electrolyte evaporated from the concave surface. For this reason, a cumulative effect arose and the plasma flow, formed as a result of the electrolyte evaporation, was focused in the vertical direction. The height of the visible plasma torch reached 10–15 cm. Clearly this configuration of the plasma flow of a gas discharge initiated with the use of a liquid electrolytic cathode will aid in increasing the field of its practical use.

Since the anode was rigidly fixed, the interelectrode spacing l increased with increase in the current. Experiments have shown that, at high currents, the maximum value of l at which a stable glow discharge free of spark channels arises does not exceed 5–6 mm. It should be noted that the appearance of conjugated discharge channels is one of the conditions limiting the current. Another condition is associated with the thermal stability of the anode. In experiments, the priority of taking into account these conditions was determined by the degree of filling of the discharge unit with the electrolyte. In the case where the difference between h and h_e was 3–5 mm, the current was restricted to prevent the breakdown of the discharge gap by spark channels (first condition). If this difference is smaller than 3 mm, the second condition should be fulfilled, i.e., the current should be restricted to prevent the appearance of melting sites on the sharp-pointed edges and the melting of the anode material (in the range of currents at which the investigations were conducted, the anode was practically not subjected to erosion).

The dependence of l on I is clearly demonstrated by the volt-ampere characteristic of the discharge unit. All volt-ampere characteristics are growing because l increased in the current range studied (Fig. 3). Thus, it may be suggested that a growing volt-ampere characteristic is a distinguishing feature of a gas discharge between a flowing electrolytic cathode and a metal anode. Owing to this feature, the indicated discharge is stable even though a ballast resistor is not used in its circuit; this resistor is a practically obligatory element in the electric-power circuit of a glow discharge between metal electrodes. In the case of metal electrodes, the volume of a glow discharge is increased by dividing the cathode into sections in which individual ballast resistors are installed [7, 8]. The ohmic resistance of an electrolyte R_e plays the role of such resistors. It seems likely that, for this reason, a discharge initiated with the use of a liquid electrolytic cathode is volumetric. The presence of R_e causes an increase in U when I increases. It is natural to assume that this provides the initiation of a stable discharge. However, the ohmic resistance was low when the



Fig. 3. Volt-ampere characteristics of a discharge: $\sigma = (2.5-2.7) \cdot 10^{-3}$ (1, 4), $(1.8-1.9) \cdot 10^{-3}$ (2), and $(0.9-1.0) \cdot 10^{-3} \cdot (\Omega \cdot \text{cm})^{-1}$ (3, 5) ($\nu = 0.1$ (1, 2, 3) and 4.5 cm/sec (4, 5)). U, V; I, A.

electrolyte flow was weak. In particular, it was equal to $\sim 10 \ \Omega$ at v = 0.1 cm/sec. Clearly, the volt-ampere characteristic obtained in the experiments cannot be formed at such small values of R_e . Consequently, a stable discharge is maintained mainly due to the dependence of l on I. Experiments have shown that the surface of an electrolyte is deformed under the action of the reactive forces of the vapor only at high current densities and a stable discharge arises under these conditions only in the case where the electrolyte flows through the discharge-action region. Thus, the volt-ampere characteristics of a gas discharge will be growing only in the case where this discharge is excited with the use of a flowing electrolyte.

The voltage U across the discharge-unit terminals located between the current lead and the anode represents the sum of the voltage across the discharge gap and the voltage drop across the electrolyte ΔU_c . The ohmic resistance of the electrolyte decreases with increase in σ ; in this case, ΔU_c is small and, consequently, the value of U decreases. Therefore, the volt-ampere characteristics obtained for electrolytes possessing a high electric conduction lie lower (Fig. 3, curves 1, 2, and 3) than the analogous characteristics of electrolytes with a lower electric conduction. At an increased velocity v of the electrolyte flow, the volt-ampere characteristics lie higher (Fig. 3, curves 4 and 5), which is also due to a change in σ because, in this case, σ depends on the temperature t_e. The higher the velocity of the electrolyte, the lower the temperature to which it is heated in the zone of discharge action. For example, at v = 0.1 cm/sec and I = 8-9 A, the highest electrolyte layers were heated to $\sim 100^{\circ}$ C and the temperature of the lower layers reached only 70°C. At v = 4.5 cm/sec and the same values of the current, the temperatures of all of the layers were equal to $\sim 50^{\circ}$ C. We present numerical values of the parameters for the concrete electrolyte, the specific resistance of which changed depending on the temperature in the following way: at $t_e = 20, 30, 40, 50, 60, 70, 80, 90$, and 100° C, it was equal to 1.00, 1.35, 1.67, 2.00, 2.31, 2.62, 2.94, 3.22, and 3.55 (in $10^{-3} (\Omega \text{ cm})^{-1}$) respectively. It is seen that when t_e increases from 50 to 100°C, σ increases by approximately two times. Consequently, the above-indicated change in the temperature of the electrolytic electrode leads to a marked change in ΔU_c and is mainly responsible for the increase in U arising when the velocity of the electrolyte flow increases.

Measurement of the potential distribution in a liquid cathode in the direction from the current lead to the electrolyte surface made it possible to determine the interelectrode distance l. The potential φ in the bulk of the electrolyte changed practically linearly (Fig. 4). When the probe cropped out at the surface of the electrolyte (this was observed visually), φ increased more rapidly. The boundary between the liquid cathode and the gas discharge is shown in the figures by a dashed line. The probe located above this boundary was further wetted by the flowing electrolyte, i.e., it remained in contact with the liquid cathode. Therefore, an abrupt change in the cathode potential was not observed in this case. Comparison of the corresponding curves in Fig. 4 shows that the electrolyte level under the discharge zone decreases with increase in the current (the coordinate z_c shifts to the left) and, consequently, the discharge extends in the vertical direction. In the range of plasma-generator parameters investigated, the maximum value to which the electrolyte level was lowered under the action of a discharge was 3 mm. The difference between h and z_c was equal to the interelectrode distance $l = h - z_c$.



Fig. 4. Distribution of the potential in the bulk of the electrolyte: I = 6 (a) and 8 A (b) [v = 4.5 (1) and 0.1 cm/sec (2, 3); $\sigma = (0.9-1.0) \cdot 10^{-3}$ (1, 2) and (2.4–2.6) $\cdot 10^{-3}$ ($\Omega \cdot \text{cm}$)⁻¹ (3)]. φ , V; *z*, mm.



The value of φ at the electrode surface is the voltage drop across the liquid cathode ΔU_c . In Fig. 4b, $\Delta U_c =$ 400 V at the point K₁ and $\Delta U_c =$ 140 V at the point K₂. It follows from the indicated numerical data that, when v decreased from 4.5 to 0.1 cm/sec, ΔU_c decreased by more than two times. As was noted above, σ changes by the same number of times under these conditions. Thus, the experimental data supported the fact that ΔU_c decreases with increase in the temperature of an electrolyte. It was established that ΔU_c also decreases with increase in the electric condition of an electrolyte (Fig. 4, points K₃). A similar dependence of ΔU_c on σ was obtained when ΔU_c was measured by the contact method with the use of an electrode–anode.

The voltage drop across a cathode is one of the most important (from the practical standpoint) parameters because the Joule-heat release in the bulk of an electrolyte decreases with decrease in the value of this parameter. This means that the heat losses by the heating of the liquid cathode will decrease and the thermal efficiency of a discharge apparatus will increase with decrease in ΔU_c . Therefore, it is advantageous, from the energy-consumption standpoint, to provide small values of ΔU_c .

According to Ohm's law, ΔU_c is proportional to the ohmic resistance of a liquid electrolytic cathode at a constant value of the current ΔU_c . The values of R_e calculated by this law were comparatively small. For example, to the point K₁ in Fig. 4b corresponds $R_e = 50 \Omega$, and to the points K₂ and K₃ corresponds $R_e = 17.5$ and 7.5 Ω .

Figure 5 presents the dependences of Q_c and Q_a on the current. The solid lines approximate experimental data and demonstrate directly the proportional dependences of these quantities on the current. The heating of a liquid cathode depends on the velocity of the electrolyte flow. At small values of v, the heat losses on the cathode were much



Fig. 6. Mass rate of electrolyte evaporation. Designations are identical to those in Fig. 5. G, g/sec; I, A.

Fig. 7. Temperature distribution along the plasma jet (the dotted curve represents the interpolation to a range higher than the upper limit of the thermocouple). v = 0.1 cm/sec; I = 11 A, $\sigma = (2.5-2.6) \cdot 10^{-3} (\Omega \cdot \text{cm})^{-1}$. t, ^oC; z, mm.

smaller than those at high velocities, which appears at first glance to be wrong. One would think that the lower the velocity of an electrolyte flow, the higher its temperature and the larger the amount of heat expended for its heating should be. Actually, the temperature of an electrolyte increased under these conditions. However, this caused a decrease in the Joule heat because R_e substantially decreased with increase in t_e . For the same reason (i.e., because of the change in R_e), the heat losses on the cathode decreased further with increase in σ (Fig. 5a, points 3). Thus, the value of Q_c is substantially dependent on the heat Q_J . It should be noted that the experimental values of Q_c were smaller than the calculated values of Q_J . We did not analyze the reasons for this in detail in the present work.

The changes in the kinematic parameters of an electrolyte flow and in its electric properties practically had no influence on Q_a (Fig. 5b). In the energy balance of the discharge unit, the heat losses on the anode comprised 10–20% and the fraction of Q_c was smaller (5–6% or less). Thus, the thermal efficiency of the plasma generator reached ~80%.

The electrolyte evaporated intensively under the heat action of a discharge. The mass rate of electrolyte evaporation G increased in direct proportion to the current (Fig. 6). The quantity G depended insignificantly on v and σ . The influence of these two parameters on G is, in all probability, due to the change in the temperature of the surface layer of the liquid cathode. At small values of v, this temperature is close to the boiling temperature of the electrolyte and, therefore, the electrolyte evaporates from the whole free surface of the cathode. When v increases, the temperature of the electrolyte decreases and the vapor formation at the open (not occupied by discharge) surface of the cathode becomes less intensive. This leads to a decrease in G; the same takes place when σ increases.

One of the main parameters characterizing the intensity of the processes on the surface of the electrolyte in the discharge zone is the current density at the cathode j_c . In the plasma generator studied, the current density j_c changed from 0.8 to 1.0 A/cm². The value of j_c increased markedly with increase in the current from 4 to 6 A and remained practically unchanged when the current j_c further increased. This can be explained by the fact that the area of conjugation of a discharge to the liquid cathode increases with increase in this current.

The heating of the gas in a discharge depends on the current density. Since the value of j_c changed insignificantly in the experiments, it may be suggested that the temperature of the gas in the discharge zone is practically constant. Its value can be estimated by interpolating the temperature distribution in a vertical plasma flow to the discharge gap (Fig. 7). As is seen, the gas inside the discharge is heated to ~2000°C.

Conclusions. The organization of a forced flow of an electrolyte through the region of action of a gas discharge made it possible to substantially increase the current density at a liquid cathode when a fairly high power (~10 kW) was supplied to the discharge unit of a plasma generator. Experiments were conducted at upper limits of the current density at the electrodes. Under these conditions, the surface of the liquid electrolytic cathode deflected inward under the action of the reactive forces of the vapor. The depth of the "pit" formed increased with increase in the current. Owing to this, first, the volt-ampere characteristics were growing and a stable discharge was initiated without re-

course to a ballast resistor and, second, there arose a cumulative effect that was favorable for the formation of a plasma flow in the strictly vertical direction. The temperature in the plasma flow remained fairly high ($\sim 1000^{\circ}$ C) at a large distance (100 mm or more) from the anode. The heat losses on the liquid cathode were minimized by varying the velocity of the electrolyte flow, which made it possible to increase the thermal efficiency of the plasma generator to 80%.

Thus, our experimental investigations have shown that the gas discharge between a flowing electrolyte and a metal electrode can serve as a source of plasma flows with fairly high energy characteristics comparable with the analogous technological parameters of plasma generators of other types.

NOTATION

G, mass rate of electrolyte evaporation, g/sec; *h*, distance between the current lead of the cathode and the anode, mm; h_e , thickness of the electrolyte layer in the discharge unit, mm; *I*, current, A; j_c , density of the current at the cathode, A/cm²; *l*, interelectrode distance, mm; Q_a and Q_c , heat losses on the anode and cathode, kW; Q_J , Joule heat released in the liquid cathode; kW; R_e , ohmic resistance of the electrolyte, Ω ; *s*, length of the anode; *t*, temperature, ^oC; *U*, voltage across the discharge unit, V; ΔU_c , voltage drop across the cathode, V; *z*, coordinate along the vertical plasma jet, mm; *v*, velocity of the electrolyte flowing through the discharge-action zone, cm/sec; z_c , coordinate of the upper level of the electrolyte in the discharge zone, mm; σ , electric conductivity of the electrolyte, ($\Omega \cdot cm$)⁻¹; φ , potential of the electric field in the bulk of the electrolyte, V. Subscripts: a, anode; e, electrolyte; c, cathode.

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