STATIONARY TEMPERATURE OF AN ANODE HEATED IN AQUEOUS ELECTROLYTES

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A review of the investigations of the heat exchange under anode heating in aqueous solutions of electrolytes has been conducted. Models of the calculation of the anode stationary temperature based on the heat release in the vapor-gas blanket due to the electric current flow through it have been considered. Comparison between the theoretical and experimental volt-ampere and volt-temperature characteristics of the anode heating has been made.

Anode heating of metals and alloys in aqueous electrolytes has been used in industry for two decades for thermochemical hardening of machine elements and tools and in the last few years — for electrochemical polishing of metals as well [1–4]. To date, many fundamental questions of the physicochemical mechanisms of the phenomenon itself have been elucidated, which makes it possible to conduct a review of the results achieved. The development of numerical models of the process for solving applied problems, in particular, modeling of technological regimes of treatment, is also becoming topical.

The high-temperature heating of a metal anode is a consequence of the local boiling of the electrolyte in the anode region. A continuous and stable vapor-gas blanket is formed around the anode if the anode surface is much smaller than the cathode surface [5, 6]. The critical voltages at which the electrolyte boils depend on its electrical conduction and the growth rate of voltage [7]. The liberation of gas at electrolysis does not affect the critical voltages but promotes the blanket formation due to the intensification of the heat removal from the anode region into the solution [8]. In practice, heating of an element is carried out by immersing it into the solution to which voltage has already been applied. Therefore, the judgment of the role of electrolysis and the estimates of the vapor-gas blanket formation time are only important for understanding the mechanism of the phenomenon. Upon the formation of an anode–blanket–solution system, anode heating to temperatures from 400 to 1000${}^{\circ}$C occurs.

The stability of the vapor-gas blanket is due to the heat release in it caused by the electric current traversing it. At a negative polarity of the element (cathode process of heating) a glow-type discharge is initiated around it, which has been convincingly shown in [9]. Attempts to explain the conduction of the anode vapor-gas blanket by the existence of electric discharges in it [10–12] or by the charge transfer by solution droplets [11–13] have received neither theoretical substantiation nor experimental confirmation.

The anode vapor-gas blanket conduction and the observed laws of heating were explained by the emission of the solution ions into the blanket and their transfer to the anode surface in the electric field [14, 15]. This mechanism was confirmed by the analysis of the anode electrochemical reactions [16, 17] and of the mobility and drift velocity of charge carriers [18] and by the absence of the spectral lines of blanket elements (except for alkali and alkali-earth metals with a low ionization potential if they were present in the solution) against the background of the heated body radiation [11, 19]. The power supply energy is released mainly in the blanket when the solution ions pass through the vapor-gas medium at atmospheric pressure. Under such conditions, the length of the free path of ions turns out to be much smaller than the vapor-gas layer thickness; therefore, heat is released exactly in the blanket and is subsequently transferred to the anode and electrolyte. The stability of the vapor–electrolyte interface is likely to be due to the same factors as in the case of film boiling.

The simplest dependence of the stationary temperature of the vertical cylindrical anode on the heating parameters was obtained from the heat equilibrium condition [20]. The flow of the electric current described by the Ohm law through the vapor-gas blanket provides the action in it of the thermal flow

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$$Q = \frac{2\pi Rh\sigma U^2}{\delta}.$$

Part of the heat flux released in the blanket is expended in heating the anode. In the stationary state, this heat is removed through the anode section to its projecting part, including the current lead with a mounting system, and further into the atmosphere by convective heat transfer. Here the familiar solution for the heat flow transferred by a semi-infinite rod to the environment was used [21]. The stationary temperature of the cylindrical anode is found from the equality of the heat flows, which, in the revised form (the coefficient given incorrectly in [20] has been corrected) is of the following form:

$$Q = \frac{2\pi Rhk\sigma U^2}{\delta} = \pi T_a \sqrt{2\alpha\lambda_a R^3},$$

whence

$$T_a = \frac{kh\sigma U^2}{\delta} \sqrt{\frac{2}{\alpha\lambda_a R}}. \tag{1}$$

The obtained relation permits explaining the dependences of the anode temperature on its dimensions. An increase in the immersion depth promotes the current growth and, consequently, the growth of the power released in the blanket. In so doing, the conditions for heat removal remain unaltered. An increase in the sample radius increases the heat removal section to a greater extent than the current strength in the chain, which leads to a decrease in the anode temperature. However, formula (1) is not a volt-temperature characteristic of the heating and cannot be used in practice. It contains the thickness of the vapor-gas blanket and the coefficient k depending on the voltage and other heating parameters.

A more complex model of the anode region includes a fourth element – a 100–700-μm-thick coating separating the metal anode from the vapor-gas blanket [13]. On the basis of this model the temperature distribution in the coating under constant temperature conditions at the boundary with the metal anode and in the presence of convective heat exchange at the boundary with the blanket was calculated [22]. The heat-transfer coefficient was a variable quantity depending on the vapor-gas blanket pulsations. The numerical solution of the heat-conduction equation made it possible to estimate the influence of the amplitude and frequency of blanket pulsations on the temperature distribution in the anode coating. The calculation did not involve the determination of the heated element temperature and its dependence on the treatment conditions and properties of the electrolyte. Moreover, under real heating conditions such thick coatings are only formed in heating in electrolytes containing tungsten, molybdenum, titanium, or vanadium [23]. For practically realized variants of chemicothermal treatment, this model is not realistic enough. However, it is the only model that considers the previously registered pulsations of the blanket and their influence on the heat-exchange parameters.

The calculation of the heated anode temperature by solving the equation of heat conduction in a thin vapor layer [24] appeared to be more fruitful. Such an approach was realized on a number of models with different boundary conditions and volume heat sources in the blanket. A thin vapor layer contained between the electrolyte solution and the metal anode and heated by the electric current flowing through it is considered. The heat release in the metal anode and in the electrolyte solution caused by the electric current flow through them was not taken into account, since current densities up to 1 A/cm^2 characteristic of anode heating cannot provide the observed heating rates of metals and solutions of strong electrolytes with a heat conductivity of the order of 0.1 Ω^{-1} cm^{-1}. As a heat source, only a vapor-gas blanket having a higher electrical resistance was considered. The heat losses associated with vapor evacuation into the atmosphere were also neglected. According to the experimental data, such losses do not exceed a few percent. We consider the conditions for heat release in the blanket and the heat exchange at its boundaries with the anode and the electrolyte solution.

Heat Exchange Between the Anode and the Vapor-Gas Blanket. The heat-exchange coefficient between the vapor-gas blanket and the metal anode was determined by the method of the regular thermal regime in heating and

cooling [25]. The obtained value was (1400 ± 200) W/(m^2·K) and its dependence on the solution velocity, the sample diameter, or the direction of the heat flow was not revealed. The possibility of using boundary conditions of the third kind in describing the heat exchange with the vapor-gas blanket was substantiated by the solution of the outer inverse problem of heat conduction under nonstationary heating of the cylindrical anode and comparison with the measurement data [26]. The obtained heat-exchange coefficient was (2500 ± 500) W/(m^2·K).

Formal description of the heat exchange between the anode and the vapor-gas blanket by means of boundary conditions of the third kind is deemed to be admissible, in principle, for the nonstationary stage of anode heating. Use of these conditions to calculate the stationary temperature of the anode becomes problematic if the convection in the vapor-gas blanket is neglected. The last suggestion is usually substantiated by its small thickness, measuring a few dozens of micrometers according to different estimates. Moreover, the experimental estimates of the thermal balance in the system make it possible to draw the quite expected conclusion that the major portion of the supply power is expended in heating the electrolyte. Such estimates were made by heating samples in a Dewar vessel (under the conditions of uncontrolled hydrodynamics) or by stabilizing the electrolyte by its cooling in the heat exchanger [25]. In the second case, a longitudinal flow of a vertically immersed element at a specified velocity realized in all anode heating facilities was provided. Measurements of the water temperature at the heat exchanger input and output have shown that from 80 to 90% of the source power is expended in heating the electrolyte and the heat flows into the solution amount to $(1.4–3.0) \cdot 10^6$ W/m^2 depending on the anode temperature determined by the applied voltage.

Because the thermal flow into the anode constitutes a few percent of the total heat release, it can be ignored in calculating the stationary temperature of a wide class of elements analogous to those investigated in [25]. Such a proposal is admissible if the anode contact area with the blanket noticeably exceeds the anode cross section at the level of the free surface of the electrolyte. It is through this cross section that heat removal into the part projecting from the electrolyte — the anode — is realized.

Heat Exchange Between the Vapor-Gas Blanket and the Electrolyte Solution. In the simplest models, the heat flow from the vapor-gas blanket into the solution was assumed to be constant and equal to the second critical flow providing stability of the solution–vapor-gas-blanket interface. Experimental measurements of the least possible thermal flow under anode heating were made in the following way [27]. The anode (steel 45) voltage was slowly decreased down to the condensation of a vapor-gas blanket, noted by a sharp decrease in the anode temperature. The density of the heat flow from the blanket into the solution at the instant the heating regime terminated was determined as the product of the current density being measured by the power with a coefficient of 0.9. It was assumed that 10% of the power consumption was expended in maintaining the anode stationary temperature and the electrolyte evaporation. Values of the minimum heat flow density of $(1.5\pm0.7) \cdot 10^6$ W/m^2 for a 10% water solution of ammonium nitrate and $(0.6\pm0.2) \cdot 10^6$ W/m^2 for a 30% solution of it were obtained.

The assumption of constancy of the thermal flow from the blanket can only be used as a zeroth approximation. Under real conditions of anode heating, the voltage can vary from 100 to 300 V, which leads to an almost doubled heat flow [27]. The stationary density of the heat flow from the blanket into the solution is approximated by the following dependence:

$$q_{liq} = q_0 + (\beta + pw)\, U\,,$$

where $q_0 = 0.9$ mW/m^2, $\beta = 3$ kA/m^2, and $p = 1.26 \cdot 10^5$ C/m^3. An expression was obtained for the heating of a cylindrical sample 8 mm in diameter immersed in a 15% solution of ammonium chloride to a depth of 90 mm at voltages from 140 to 270 V and longitudinal flow velocities from 0.34 to 1.42 cm/sec.

Moreover, the exponential dependence of the heat flow density from the blanket into the solution is known [28]:

$$q_{liq} = \frac{\lambda_{liq}}{h}\,(T_s - T_{liq}) \left\{ 1.43 - 1.3 \exp\left[-0.35 \cdot \frac{\lambda_v\,(T_a - T_s)}{\lambda_{liq}\,(T_s - T_{liq})} \right] \right\} \mathrm{Ra}^{1/3}\,, \tag{2}$$

where the Rayleigh number $\mathrm{Ra} = gh^3(T_s - T_{liq})/V_{liq}a_{liq}T_{liq}$.

Heat Sources in the Vapor-Gas Blanket. The volume power of the heat sources in a vapor gas blanket having a constant electrical conduction, i.e., when the Ohm law holds, is of the following form:

$$W = \frac{jU}{\delta} = \frac{\sigma U^2}{\delta^2} . \tag{3}$$

A more complex dependence is obtained by taking into account the role of the space charge in the vapor-gas blanket [15, 26, 28]. It is assumed that emission of ions from the solution begins at a field strength in the blanket not lower than the critical strength E_0. This value was found on the basis of the notions about the critical surface charge density (the Tonks–Frenkel problem) [29]. In the opinion of Ganchar and Dmitriev [28], in the blanket full screening of the phase-boundary potential by the summed potential of the space charge is usually realized. This leads to a simplified expression for the current density in the vapor-gas blanket:

$$j = \frac{9\varepsilon\varepsilon_0 \mu U^2}{8\delta^3} . \tag{4}$$

Model 1. The heat exchange at the anode–blanket interface was described by boundary conditions of the third kind and the current flow through the blanket — by the Ohm law, and the heat flow from the blanket into the solution was taken to be equal to the second critical one [24]. An increasing voltage dependence of the vapor-gas blanket thickness provided that the current was independent of the heating voltage was obtained. The numerical estimates made it possible to estimate the order of the vapor-gas blanket thickness — 10^{-5} m. Further attempts to develop this model did not lead to results comparable to the experimental ones. Obviously, this approach not only overestimates the heat flow from the blanket into the anode but also underestimates the heat flow from the blanket into the electrolyte solution.

Model 2. The method for calculating the stationary temperature of the anode being heated is based on the following assumptions [28]:

a) the heat flow from the blanket into the heated anode is neglected;

b) the dependence of the heat conductivity of vapor on its temperature is taken into account;

c) the dependence of the heat-flow density from the blanket into the solution on the anode temperature is taken into account according to (2);

d) the influence of the space charge in the vapor-gas blanket on the electric current flow through it is taken into account.

The dependence of the heat conductivity of vapor on its temperature was approximated by the linear function

$$\lambda(T) = \lambda_0 [1 + \gamma(T - T_s)] .$$

The temperature was calculated in the following way. First the equation of the heat conduction in the vapor layer

$$\lambda_0 \frac{d}{dx} \left\{ [1 + \gamma(T - T_s)] \frac{dT}{dx} \right\} + W = 0 \tag{5}$$

was solved. The x axis was directed perpendicular to the vertical surface of the anode, with $x = 0$ at the anode–blanket boundary and $x = \delta$ at the blanket–solution boundary. From the solution of (5) the anode temperature was determined:

$$T_a = T_s + \left(\frac{1}{\gamma^2} + \frac{W\delta^2}{\gamma\lambda_0} \right)^{1/2} - \frac{1}{\gamma} . \tag{6}$$

The specific power of the heat sources and the density of the flow into the solution were related by the following relation:

$$W\delta = jU = q_{liq} = \frac{9\varepsilon\varepsilon_0 \mu U^3}{8\delta^3} . \tag{7}$$
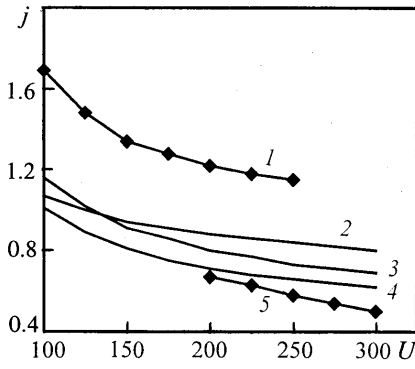
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Fig. 1. Experimental (1, 5) and calculated (2–4) volt-ampere characteristics of the anode heating: 1) data of [16]; 2) model 3; 3) model 4; 4) model 2; 5) data of [32]. j, A/cm^2; U, V.
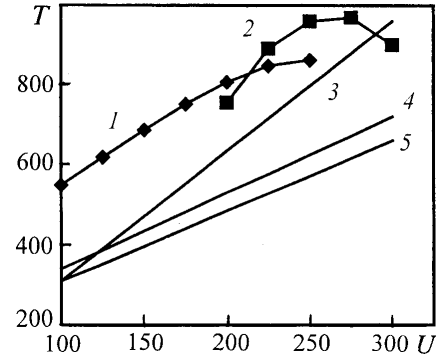
Fig. 2. Experimental (1, 2) and calculated (3–5) volt-temperature characteristics of the anode heating: 1) data of [16]; 2) data of [32]; 3) model 3; 4) model 4; 5) model 2. T, $^{\circ}$C; U, V.

By solving Eqs. (2), (6), and (7) for T_a, q_{liq}, and δ, the volt-temperature and volt-ampere characteristics of the heating process were obtained (Figs. 1 and 2). Note that the curves presented here somewhat differ from those published by Ganchar and Dmitriev [28]. One possible reason could be the different values of the process parameters used in plotting the characteristics. All the calculated dependences were obtained in this work for the following values: $c_{liq} = 4.22 \cdot 10^3$ J/(kg·K) [30], $E_0 = 2.4 \cdot 10^6$ V/m (calculation according to [29]), $T_{liq} = 30^{\circ}$C, $T_s = 100^{\circ}$C, $\gamma = 6.55 \cdot 10^{-3}$ K^{-1} [30], $\lambda_0 = 2 \cdot 10^{-2}$ W/(m·K) [30], $\lambda_{liq} = 0.68$ W/(m·K) [30], $\lambda_v = 7.34 \cdot 10^{-2}$ W/(m·K) [30], μ (for the Cl$^-$ ion) = $3.08 \cdot 10^{-4}$ m^{-2}/(V·sec) (calculation according to [31], $\nu_{liq} = 0.295 \cdot 10^{-6}$ m^2/sec [30], and $\rho_{liq} = 1.08 \cdot 10^3$ kg/m^3.

In our opinion, the realization of model 2 in [16, 28] is not correct for two reasons. Expression (4) is too rough a simplification of the accurate volt-ampere characteristic obtained for the vapor-gas blanket under the condition of its thickness stabilization in the implicit form [15, 28]

$$U = \frac{\mu \varepsilon_0 E_0^3}{3j} \left[\left(1 + \frac{2j\delta}{\mu \varepsilon_0 E_0^2} \right)^{3/2} - 1 \right]. \tag{8}$$

To estimate the competence of the approximation numerically, the above values of the necessary quantities as well as the experimental current density $j = 10^4$ A/m^2 and the estimated thickness of the vapor-gas blanket $\delta = 10^{-5}$ m were used. Moreover, in relation (7), instead of the x-coordinate-dependent specific power of the heat sources a constant appears. Actually, this means the replacement of the electric field strength $E(x)$ by its mean value equal to U/δ without any substantiation.

Model 3. This model differs from model 2 only in the refusal to take into account the role of the space charge. The solution of Eq. (5) in view of (3) permits obtaining an anode temperature in explicit form that does not contain the vapor-gas blanket thickness:

$$T_a = T_s - \frac{1}{\gamma} + \frac{1}{\gamma} \sqrt{\frac{\gamma \sigma U^2}{\lambda_0} + 1} . \tag{9}$$

This volt-temperature characteristic is given in Fig. 2 (curve 3) for a value of the blanket conductivity of $\sigma = 1.5 \cdot 10^{-3}$ $\Omega^{-1} \cdot$m^{-1}. The volt-ampere characteristic in this model given in Fig. 1 (curve 2) for the same value of the conductivity is obtained from relations (2) and (9):

$$j = \frac{q_{liq}}{U} = \frac{\lambda_{liq} (T_s - T_{liq})}{U} \left[\frac{c_{liq} \rho_{liq} g (T_s - T_{liq})}{T_{liq} \nu_{liq} \lambda_{liq}} \right]^{1/3} \left\{ 1.43 - 1.3 \exp \left[-\frac{0.35 \lambda_v}{\gamma \lambda_{liq} (T_s - T_{liq})} \left[\left(1 + \frac{\gamma \sigma U^2}{\lambda_0} \right)^{1/2} - 1 \right] \right] \right\}.$$

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Model 4. Sequential account of the role of the space charge. The exact solution of Eq. (5) with $W = jE(x)$ leads to a rather awkward expression. The calculation is simplified if it is assumed that the mean field strength in the vapor-gas blanket exceeds the threshold E_0 by at least three times. In such a case, the volume power of the heat sources in the blanket will be equal to

$$W(x) = \frac{\mu\varepsilon_0\sqrt{2}}{64\delta^4}\left\{\sqrt{(9U^2 - 12E_0^2\delta^2)^2 + 192E_0^3\delta^3(U - E_0\delta)} + 9U^2 - 12E_0^2\delta^2\right\}^{3/2}\left(1 - \frac{x}{\delta}\right)^{1/2}.$$

The numerical estimates show that the thus-simplified dependences do not differ from the exact solutions by more than 5%. The volt-ampere and volt-temperature characteristics of model 4 are also given in Figs. 1 and 2. The same figures also show the experimental dependences obtained under different conditions. Curves 1 (see Figs. 1 and 2) correspond to the heating of an St3 steel anode 1 mm in diameter immersed to a depth of 10 mm in a 13% solution of ammonium chloride; the flow velocity of the solution is not given [16]. Curve 5 in Fig. 1 and curve 2 in Fig. 2 pertain to the heating of a 0.8 kp steep anode 16 mm in diameter immersed to a depth of 15 mm in a solution containing 10% ammonium chloride and 6% sugar; the flow velocity of the solution was 0.37 cm/sec [32].

All theoretical volt-temperature characteristics agree with the experimental data only qualitatively in the 100–250-V range of voltages. At higher voltages the measured anode temperature decreases. This is likely to be due to the appearance of weak pulsed electric discharges causing a spraying of the solution and a cooling of the anode. At voltages above 300 V these discharges become stationary and the anode high-temperature heating ceases.

All calculated volt-ampere characteristics are decreasing, which also agrees with experimental data.

CONCLUSIONS

1. The anode stationary temperature can be determined by solving the equation of heat conduction in the anode vapor-gas layer in which volume heat sources associated with the electric current flow through it act. It is assumed that the heat released in the blanket is expended only in heating the solution. The proposed models describe the expansion of the blanket with increasing voltage, which leads to a decrease in the current but an increase in the input power. This appears as decreasing volt-ampere and increasing volt-temperature characteristics qualitatively agreeing with experimental data.

2. The account of the space charges in the vapor-gas blanket by the methods described gives lower values of the anode stationary temperature and approximately the same values of the density of the current flow through it.

NOTATION

a_{liq}, thermal diffusivity of solution, m^2/sec; c_{liq}, specific heat capacity of solution, J/(kg·K); E, electric field strength in the blanket, V/m; E_0, threshold electric field strength, V/m; g, gravitational acceleration, m/sec^2; h, depth of immersion of anode, m; j, density of current through the blanket, A/m^2; k, coefficient taking into account the portion of heat released from the blanket into the anode; p, coefficient in the dependence of the heat-flow density on the voltage and the solution velocity, C/m^3; q_0, density of the heat flow acting in the vapor-gas blanket, W/m^2; q_{liq}, density of the heat flow from the blanket into the solution, W/m^2; Q, heat flow acting in the vapor-gas blanket, W; R, anode radius, m; Ra, Rayleigh number; T, temperature in the vapor layer, K; T_a, temperature of the element—anode, K; T_{liq}, solution temperature, K; T_s, solution saturation temperature, K; U, heating voltage, V; w, electrolyte velocity, m/sec; W, specific power of heat sources, W/m^3; x, coordinate, m; α, coefficient of heat transfer into the atmosphere, W/(m^2·K); β, constant in the dependence of the heat-flow density on the voltage and the solution velocity; A/m^2; γ, coefficient in the temperature dependence of the vapor heat conduction, K^{-1}; δ, thickness of the vapor-gas blanket, m; ε, dielectric constant; ε_0, electric constant, Φ/m; λ, vapor heat conduction, W/(m·K); λ_a, heat conduction of anode material, W/(m·K); λ_v, mean heat conduction of vapor at the saturation temperature, W/(m·K); λ_{liq}, heat conduction of so-

lution, W/(m·K); μ, ionic mobility, m^2/(V·sec); ν_{liq}, kinematic viscosity of solution, m^2/sec; ρ_{liq}, solution density, kg/m^3; σ, blanket conductivity, $\Omega^{-1} \cdot$m^{-1}. Subscripts: liq, liquid; a, anode; s, saturation; v, vapor.

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