METHODS OF CALCULATING THE POTENTIAL AND CURRENT DISTRIBUTION IN ELECTROCHEMICAL AND CORROSION SYSTEMS (REVIEW)

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We present a review of theoretical works primarily concerned with the calculation of the potential and current distribution during corrosion. The basic purpose of the review is to give the reader an orientation in the specialized mathematical literature on this problem.

In recent years there was an increasing interest in calculations of the potential and current distribution during corrosion and electrodeposition of metals. In this review we will undertake to outline the present state of this problem.

The publications given in the bibliography presented below can be divided into review articles, papers devoted to the calculation of potential and current distributions (or to the solution of formally identical problems) by the methods of mathematical physics, and works based on the use of various qualitative and empirical relations.* From our point of view of great interest are the publications of the second group, which will receive the most attention in what follows.

As to the review articles, one should name among them the publications of Kadaner [46-48, 53, 54], Poddubnyi [84], Kronsbein [145], Wagner [185, 188, 193], and Posey [161] as well as Vagramyan [3], Izmailov [34], Mashovets [70], Fedot'ev, and Evstyukhin [100]. More or less detailed reviews of the state of the problem under consideration at its various stages were given by many other authors (see, for instance, [120, 136, 169, 197]) in the formulation of some concrete problem.

The publications [145, 185, 188, 193], which are devoted to reviewing investigations completed earlier, contain also bibliographies covering the particular questions of experimental investigations in the field of electrodeposition and corrosion of metals. A part of the papers listed deals with some particular methods of calculating the potential and current distribution. Thus, in [3, 70] the review of the possible methods of calculating the potential and current distribution is limited to mentioning approximate graphical methods of constructing the image of the field. A somewhat more detailed review of the methods of calculating the potential and current distribution in the problems of electrodeposition of metals and to some extent in the problems of corrosion, is given in the publications of Kadaner, and, especially, in the books [53, 54], which, however, consider only some simpler methods, similar to the method of calculating the electrostatic field. A wider range of calculation methods is given in the review articles [84] and [42].

It is well known that in a rigorous formulation the calculation of the potential and current distribution in the corrosion and electrodeposition of metals leads to the solution of the Laplace equation with boundary conditions determined by the electrochemical interaction of the metal with the surrounding medium. This is one of the special problems in potential theory, and therefore the general methods developed in mathematical physics have been applied since a long time to the solution of the problem of electrochemical corrosion and electrodeposition of metals. Unfortunately, many electrochemists and corrosion experts lack sufficient knowledge of these methods, and therefore they are relatively seldom applied in practice; for

* The bibliography does not fully cover the specific problems of drainage and cathode protection of extended installations and the design of grounding systems. On these questions see, for instance, [75, 76, 95].

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this same reason, in the electrochemical literature, along with many interesting publications of this type, there are sometimes also clearly erroneous results published (for instance, [73, 74]).

The possibility and efficiency of applying the methods of mathematical physics for the calculation of the potential and current distribution are determined by the geometry of the surface of the system under consideration and by the form of the given boundary conditions.

From the geometrical point of view the simplest boundary surface is, of course, the case of a one-dimensional problem, which yields for analytical calculations even with relatively complicated boundary conditions. The publications by Wagner [2], Daniel'-Bek [21], Ksenzhek and Stender [57-59], Mashovets and Fomichev [72], Frumkin [107], Kasper [136], Wagner [184-190], and Winsel as well as [66, 85, 102, 119, 124, 136, 146, 148, 161] are devoted to the consideration of the one-dimensional problem of calculating the potential and current distribution in the corrosion and electrodeposition of metals.

The first one of these publications considers the simplest calculation systems in the form of an infinitely extended plane, two infinite coaxial cylinders and concentric spheres; the polarization character of all these surfaces being assumed to be nonlinear. Similar calculation models with various boundary conditions for the polarization were considered by Drossbach [66], Gardam [122], Kronsbein [143] and in [136] and [184-190].

Another problem belonging to the simplest ones is that of the potential and current distribution along the internal surface of a pore, considered a long, thin pipe or slit. This problem becomes one-dimensional on the basis of the assumption that the changes of the potential and the current across the pore are negligible compared to their changes along it, where the only coordinate of the problem refers to. With this coordinate the calculation leads to the integration of an ordinary differential equation [21] whose form is determined by the boundary conditions on the wall of the pore. The most typical problems for cylindrical pores with various (including nonlinear) conditions for its polarization are treated in [21, 57-59, 107, 148, 161, 197].

The inverse problem, in the known sense, was considered by Weiselberg and Staff [196], who studied the potential and current distribution on the surface of a long, thin cylinder surrounded by an electrolyte layer. Taking similar assumptions into account, the authors brought these problems to a form essentially identical in structure with the calculation of the potential and current density on the surface of a cylindrical pore.

The group of publications dealing with the calculation of the potential and current distribution in two-dimensional, usually plane, systems is more numerous. Irrespective of the fact that for plane systems one can apply some special, quite efficient calculation methods (as, for instance, the method of complex potentials), the possibilities of doing calculations in this case are found much more limited than in the one-dimensional case. Therefore, in most published work analyzing plane systems various approximate (linear) boundary conditions are used.

One of these plane systems, considered in the problems of electrode position of metals, is the rectangular electrolyte cell, with various positions of plane linear electrodes in it. The following publications are devoted to the calculation of the potential and current distribution in such systems: [6, 11-14, 25, 26, 29, 55, 65, 67, 84-86, 119]. In [11, 65, 67, 94] the primary current distribution is considered in a slit cell. As a calculation model for this case, the authors use a plane system in the form of a strip cathode extending along the whole length of one of the walls of the rectangular cell, and a linear anode placed in one of the opposite (with respect to the cathode) corners of the cell. The case of linear cathode polarization is treated by Gnusin [12] and in [13, 85]; for small values of the specific polarizability of the cathode this problem leads to the previously discussed way of utilizing the method of characteristic surfaces [42].

In [65] a special method of constructing the coordinate functions, for the solution of such problems by the variation method, is proposed. Kadaner and Tsukernik [55] gave a calculation of the primary current distribution in a rectangular electrolyzer for arbitrary cathode width by means of the method of complex potential. The case of nonlinear polarization of the electrodes placed in a rectangular electrolyzer was considered in [29, 84]. The authors of these publications resort to numerical methods: in [84] the calculation leads to the solution of a nonlinear integral equation, whereas in [29] to the solution of the difference boundary value problem by the method of straight lines.* The latter method is also used in [25], where an arbitrary position of the anode and cathode is considered under the condition of their linear polarization.

* The same method is used in [27, 35].

The calculation of various concrete electrolyzer types led to the consideration in the literature of a large number of specific plane systems of particular form [31, 47-52, 84, 132, 133, 142], including those containing bipolar electrodes [101]. The same situation is observed in the literature devoted to electrochemical corrosion and the protection against it [36, 38, 39, 40-40-42, 142-144, 156] and in formally related problems [8, 28, 32, 37, 82, 91, 93, 123, 139, 167]. With the latter should be also listed the work of Vasil'ev [7, 9], whose results make it possible (using known analogies) to find the potential and current distribution on a linearly polarized metallic surface bent at a sharp angle and placed in the field of an infinitely long linear current source.

Interesting results were obtained by Kennard and Waber, who considered a semiinfinite rectangular electrolyzer, at the bottom of which are the linearly polarizing anode and cathode with different polarization parameters. The obtained results are, however, only applicable in the special case when the width of the cathode is twice the width of the anode.

From the methodological point of view the work of Breitner and Guggenberger is of interest. These authors considered the two-dimensional potential distribution between two polarizing flat electrodes of rectangular form, placed at the ends of a cell in the shape of an open ring segment. In the statement of this problem the nonlinear (Tafel) polarization law was assumed for the electrodes. Expanding the boundary conditions in the powers of a small parameter (the ratio of the current densities in an arbitrary point and some fixed point on the surface of the electrodes) the authors brought the problem to a linear approximation and, using the usual method of eigenfunctions, calculated the first two approximations.

One of the most typical plane calculation models considered in the problems of corrosion and electrodeposition of metals is the so-called strip system, that is, a system composed of an infinitely long anode (or cathode) of finite width, placed on an infinitely extended plane metallic surface. A large number of publications [2, 15, 22, 33, 38, 42-44, 61, 77, 84, 137, 138, 183] are devoted to the calculation of the potential and current distribution in such a system for various conditions of the polarization of the electrodes. Thus, in [33] and [42] formulas are obtained for the calculation of the primary current distribution in the strip system and the case is considered when both metallic surfaces are covered by some surface film. In the same papers the problem of the secondary potential and current distribution is solved under the condition that the specific polarizability of the strip electrode (anode) is smaller than the specific polarizability of the infinitely extended cathode. The solution of this latter problem is constructed by the method of characteristic surface, which, along with the method of coupled integral equations and the method of successive approximations, is used also in [43] for the solution of the analog problem for the inverted relation between the specific polarizabilities.

A more general problem of this type, which can be interpreted as the calculation of the potential and the current in the strip system for an arbitrary ratio of the specific polarizabilities of the metals and for a finite width of the medium covering them, was solved by Kuz'min [61]. The solution of this problem was obtained by the method of coupled integral equations and led to the solution of a second-order Fredholm equation. Some particular cases of the same problem were considered in [33, 68, 108]. Approximate calculations of the potential and current density are also given in [2, 23, 77]. In [77] an arbitrarily given potential was allowed on the surface of the strip electrode, while in the other two calculations a constant current density was taken under the condition that the infinitely extended surface of the metal is an equipotential surface. In [23] the solution by successive approximations is also given, taking into account a linear polarization of the strip electrode.*

The problem of the strip system with nonlinear boundary conditions is considered in [84] and [15], where it was assumed that the electrode is placed on an infinitely extended insulator (or, equivalently, on a metal with infinitely large polarization parameter), and the solution was taken to the consideration of a nonlinear integral equation. Some possibilities of taking into account the nonlinear character of the polarization follow also from the results of Strakhov [90-92].†

* Efficient approximate solutions by means of the method of successive approximations, and also the method of superposing solutions are obtained in [44] for the case when the conditions of linear polarization are given on the strip cathode.

† The method used in these works is based on an idea from Chestor's paper, "Relation of the third boundary value problem and the problem with first-order boundary conditions," J. Math. Phys., 40, No. 1, 68 (1961).

Another typical plane system is the one composed of two semiinfinite surfaces of different metals, which are in contact [39, 42, 109, 110, 140]. In [39, 42, 110] formulas are obtained, which make it possible to take into account the finite thickness of the layer of conducting medium covering the electrodes, and, in [39, 42], the presence of a thin layer of known conductivity on the surface of the metal as well. A more general problem of this type was considered by Khrustalev [109]. The solution found in this work makes it possible to characterize the case of nonlinear polarization of the metals under consideration, for arbitrary ratios of their specific polarizabilities. Unfortunately, the solution constructed in [109] (like, incidentally, the solutions of many other problems mentioned above taking into account polarization) is relatively complicated and its direct application for engineering calculations is difficult.

In the study of a number of problems of the corrosion and electrodeposition of metals many authors considered the calculation model comprising an infinitely extended plane cathode and, parallel with it, an infinitely long linear anode (or a system of such anodes), placed on parallel, infinitely extended insulating surfaces. The most interesting results for this case were obtained by Parrish and Newman [152, 153], who obtained solutions for the secondary current distribution for linear polarization of the electrodes, for their polarization according to Tafel [152], and for the current distribution obtained directly from the solution of the equation of diffusion kinetics [155].

Finally, the system of an infinite row of alternating strip anodes and cathodes lying in one plane also belongs to the typical plane calculation models. This system was used for the mathematical description of the corrosion of a microheterogeneous metal surface by Waber [176, 178, 181], Wagner [184, 187], Drossbach and Meggle [120], and in [45]. In [176] the simplest case of the primary potential and current distribution was considered for infinite thickness of the layer of the corroding medium, and in [45, 120, 179, 180] the linear polarization of the anode and cathode and the finite thickness of the layer of the corroding medium were taken into account. This system was considered in [184, 187] with a somewhat different formulation of the boundary conditions (on the surface of the anode constant potential, and on the surface of the cathode the normal component of the current density).

The list of publications dealing with the calculation of the potential and the current for another class of two-dimensional systems, that of the axisymmetric ones, is somewhat less extended. The majority of these publications considers only two typical systems, one of which is represented by a disk-shaped electrode placed on an infinit plane metal surface ("disk" system), and the other one by a point current source close to this surface. The first of these systems was first considered in its application to the problems of corrosion* by Levich and Frumkin [64], who constructed an efficient solution using a compressed spherical coordinate system for the case, when the current distribution on the cathode is uniform (small-size cathode) and the infinitely extended anode is an equipotential surface. Further in [24, 63] a more complicated problem was considered, in the formulation of which it was assumed that the current distribution is also uniform on a ring-shaped region of the anode surface adjacent to the cathode.

Other ways of specifying the boundary conditions in the disk system were considered in [22, 35, 42, 60, 78, 83, 149, 150, 147, 194]. Thus, in [42] the primary current distribution in the disk system was found, and the case when there is a thin film of given conductivity on the surface of each electrode was also considered. In [22] a general way of solving the mixed problem for the disk system, when for one of the electrodes the current distribution and on the other one the potential distribution is given, was presented. In [35] formulas were obtained for the current distribution of a disk anode with a ring-shaped insulating shield; Artykpaev [1] considered an even more general case, applied to the characterization of the potential and current distribution, taking linear polarization of the infinitely extended metal surface. The case of a linearly polarized disk system with an arbitrary ratio of the specific polarizabilities of the metals was considered in [60, 83]; in [60] the finite width of the electrolyte layer above the electrodes was also taken into account.†

* A formally similar problem was considered in detail also in the theory of elasticity in connection with the study of the indention of a round stamp into an elastic medium. This problem was apparently already solved in the classical works of Boussinesq (see, for instance, J. Boussinesq, Application des potentiels a l'étude de l'equilibre et du mouvement des solides elastiques, Paris (1885)). In this connection the work of Weber [194] should also be mentioned; he gave the solution of typical axially symmetric problems in the theory of stationary electric fields, an essentially for the first time made an attempt to take into account in the calculation the polarization of the electrodes.

† In the majority of the publications referred to, the method of coupled integral equations was used. The general theory of this method, as well as its bibliography, as applied to the axisymmetric problems of potential theory, is given in the monographs by Uflyand [99] and Sneddon [89, 168], and in the dissertation of Kuz'min [62].

Efficient approximate solutions were obtained in [44, 79] for the disk system with linear polarization and equipotential basis surface of the metal. The case of equipotential disk electrode placed on a polarizable plane metal surface was considered in [78].

Newman [149, 150] attempted to take into account the nonlinear character of the polarization of a disk placed on an insulating plane. The boundary conditions on the disk were formulated on the basis of the solution of the equation governing diffusion to the surface of a rotating disk electrode, and the solution was constructed by considering an infinite system of algebraic equations.

The second typical axisymmetric system mentioned above (a point-like current source near an infinitely extended plane metal surface) was considered in [16, 41, 87, 105, 138, 154, 194]. The problem of the primary current distribution in such a system has, as is known, an elementary solution. For the case of linear polarization of the metal surface the solution of this problem was already given by Weber [194]; it leads to an expression for the potential in terms of a quadrature of good convergence. There are also some publications (see, for instance, [41, 105]) which give various ways of constructing the solution for the field of a point-like anode placed near an infinitely extended polarizing cathode. In a number of publications more complicated cases also are investigated, when the polarization of a region of the cathode within a disk of arbitrary radius is taken into account, while the rest of the metal is taken to be equipotential [16], and also when under the point-like anode an insulating screen is placed [96]. On the basis of the solution of the problem mentioned above, Weber gave the calculation of the potential and current distribution for a more general case, when the disk anode is placed above a linearly polarized plane, but the conducting medium is limited by an insulating plane coinciding with the plane of the disk.* A formally similar problem was considered in [81].

Related formally axisymmetric problems were furthermore considered in [4, 5, 17, 18, 53, 80, 98, 111, 116-119, 141, 156-160]. In particular, Drossbach gave the solution of the internal problem of the potential and current distribution for the contact of two linearly polarized metals, one of which forms the end of a cylinder, and the other one the rest of the cylinder. For the primary current distribution in such a system, taking into account the ring-shaped insulating screen at the end of the cylinder, numerical results were given in [116]. A number of problems of the primary current distribution on the internal surface of cylindrical electrodes was considered in a series of papers by Plumpton and Wilson [156-160].

As to the other axisymmetric systems in the literature, the calculation of the potential and current is carried out only in a very limited number of cases, and it is based usually (see, for instance, [63]) on the use of results obtained previously for other industrial problems (for instance in electron optics). A full bibliography of publications on, and numerous solutions of the problems of potential theory for axisymmetric electrodes in toroidal and bispherical coordinates with mixed boundary conditions can be found in [88, 89].

The problems of potential and current distribution in three-dimensional systems are the least fully treated in the literature. The results published so far deal only with a small number of similar problems [41, 42, 127, 130, 166, 194, 195]. Thus, in [194] the solution is given to the problem of the potential distribution generated by two point-like current sources inside a cylindrical surface, on the surface of which they are placed. The more general case, when there is another, coaxial, cylinder within the first one, the surface of which is assumed to be linearly polarized, is considered in [195].

Another system belonging to those considered in the literature is the one consisting of an electrode in the form of a plane plate of arbitrary form placed on an infinitely extended metal surface. General results for the primary current in such a system are given in [42], where the case of the electrodes being covered by a thin surface film is also considered. A special case of this system, that of a rectangular electrode, is considered in [41].

In [127, 130] the solution is given to the problem of the primary potential distribution in the system given by an anode placed on an insulating rectangular screen and an infinitely extended cathode lying in the same plane. Here the cathode current distribution was taken to be piecewise uniform, which leads to the usual Neumann problem for the half space, and makes it possible to formulate the conditions of cathode protection by the method suggested earlier by Wagner [184].

In [166] numerous calculations are presented of the potential and current distribution in an electrolyte cell of the form of a rectangular parallelepiped, at the bottom of which is the rectangular anode, placed on the plane surface of the cathode, the same polarization parameters being taken for both electrodes.

* The boundary conditions in this formulation do not take into account the mutual effect of the electrodes.

A series of three-dimensional problems dealing with the primary current distribution at the surface of sphere segments, placed near a point-like anode, are considered in [88].

Turning to the publications based on the use of qualitative and empirical relations for the potential and current distribution, we remark that all of these are essentially based on the approximate way of constructing the "expected paths of current," well known in electrotechnics,* and rely on a quite approximate way of taking into account only the most general principles of field distribution (orthogonality of force and equipotential lines, bunching of current lines on protruding and sharp regions of the surface of electrodes, etc.).

Naturally, the accuracy of such a construction depends in an essential way in the intuition and experience of the person applying it, and also on how complicated the system under consideration is. For this reason the methods of graphical construction has no serious foundation whatsoever, and in the majority of cases make it only possible to give a qualitative description of the field image, especially in the solution of problems of corrosion and electrodeposition of metals, where it is imperative to take into account the polarizability of the electrodes.

Irrespective of what has been said, such methods prove very useful in the qualitative consideration of a problem, and, therefore, have been used in a relatively large number of publications ([3, 10, 19, 20, 49, 51, 69-71, 97, 104, 112, 113, 122, 169, 198]). Of these publications the paper by Sukhodskii [97] should be singled out, in which a graphical method was proposed for the calculation of the current distribution on the electrodes when the potential difference between them is known, the polarization curves of the metals are given, and the length and cross section of the current tubes originating from each point of the surface of the electrodes are also known ("Sukhodskii method"). Later on Mashovets and his co-workers [69-71] and Forsblom attempted to improve this method, and even to combine them with the rigorous methods of calculation [49, 51].

Finally, in a number of publications (see, for instance, [10, 19, 20]) the method of constructing the field were based on subdividing the surface into a number of squares on which the current distribution is uniform† and on taking approximately into account the resistance between such squares.

In conclusion we note that, naturally, the purpose of the present review was not to elucidate all publications on the problem under consideration (which is evidently impossible in the framework of one article), but to touch only upon those, which to a large extent characterize the tendencies appearing in the present approach to the problems of potential and current distribution in the electrochemical corrosion and electrodeposition of metals.

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* To these ways belong, in particular, the so-called Shmidel'–Roters method. A number of ways of constructing the expected paths of current is also given in the works of B. K. Bul, V. A. Govorkov, A. G. Slivinskii, Yu. S. Rusin, I. M. Grach, and some other authors.

† A similar method is also used in electrotechnics, where it is known under the name "method of squares."

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