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GALVANIC CORROSION RESISTANCE OF ALUMINUM AND ITS ALLOY IN GALVANIC COUPLES WITH STEELS WITH METALLIC COATINGS

The aim of this work is comparison of corrosion resistance of aluminum and its alloy D16 to galvanic corrosion in couples with carbon steel A284 and stainless steel type 321, on the surface of which various metallic coatings were deposited. Zinc, cadmium and aluminum used as the metal coatings on the steel. Coatings deposited by electroplating and vacuum ion-beam assisted deposition. Study of galvanic coupling of deposited coatings with aluminum and its alloy carried out by electrochemical methods in a 3% NaCl solution.

Zinc and cadmium coatings on steel, formed by both methods, have electrochemical characteristics of deposited metals. Zinc coatings in galvanic couples with aluminum and its alloy are anodes and aluminum coatings are cathodes. Compromise potentials of cadmium coatings approximately coincide with those for aluminum and its alloy.

Current density of the galvanic corrosion of aluminum and its alloy in galvanic couples with steel without coatings was $\sim 10^{-2}$ mA/cm², and in galvanic couples with aluminum coatings on steel $-\sim 10^{-3}$ mA/cm². Corrosion current density of zinc in coatings was $2 \cdot 10^{-2}$ mA/cm², and cadmium $-6 \cdot 10^{-3}$ mA/cm². Best galvanic compatibility with aluminum and its alloy D16 in 3% NaCl have cadmium coatings.

Key words: aluminum, alloy, steel, corrosion, deposition, zinc, cadmium.

Introduction. Aluminum and its alloys in contact with the steels may undergo intense corrosion due to the occurrence of corrosion galvanic cell in which aluminum having a more negative potential than steel, is the anode. In particular, this occurs during fixation of the structures based on aluminum with the help of steel fasteners in the construction industry [1, 2]. Contact of aluminum alloys with different steels inevitably occurs in shipbuilding [3, 4] and aircraft industry [5]. One of the methods of protection of aluminum and its alloys from corrosion is to put metal coating onto the steel parts, galvanically compatible with aluminum [6]. The most suitable are coatings of cadmium and zinc, which under many conditions have similar to aluminum electrode potentials. Galvanic deposition techniques of such coatings pose a serious environmental threat, especially in the case of cadmium. In this connection, it is reasonable to study the possibility of application of vacuum coating forming methods [7–10], alternative to galvanic. With the help of the methods of vacuum deposition it is possible to coat such kind of metals that is impossible or difficult to get deposited galvanically. For example, aluminum coating on steels will have excellent galvanically compatibility with aluminum and its alloys parts. However, galvanic deposition of aluminum is difficult, whereas the vacuum deposition of aluminum [8, 9] is the same as the deposition of other metals.

Main part. Aluminum A7, D16 aluminum alloy D16, carbon steel St3 and stainless steel 12X18H9T are chosen as objects for the research. The aim of this work is the comparison of the resistance to galvanic corrosion of aluminum and

its alloy in couples with steels, which surface contains with zinc, cadmium and aluminum coftings. For metal deposition, electroplating and vacuum ion-assisted deposition were used. Deposition of zinc and cadmium on the steel surface has been carried out by both methods, and deposition of aluminum – only by the second method, because galvanic deposition of aluminum from water solutions is impossible due to high negative value of standard potential of aluminum.

Electrochemical deposition of cadmium was conducted from ammonium solution [11] containing CdSO₄, (NH₄)₂SO₄ and H₃BO₃, deposition of zinc – from alkali solution comprising of ZnO, NaOH and brightener [12]. Coating deposition was conducted at a current density of 100 A/m². The estimated thickness of the coating was 12 µm. Actual thickness of the coatings was less and for different samples varied of about 10 µm, which corresponds to the output of current of the deposited metal - 80%. The obtained coatings were exposed to a passivation in the solution containing Cr (III) [13]. There was a preliminary processing of base surfaces with mechanical polishing and multi-step processes of chemical degreasing, etching, activation, as it is customary in the galvanic deposition coating technology [14]. For preparation of solutions of degreasing, etching, activation, electrochemical deposition and passivation reagents "hch" and "chda" were used.

Vacuum deposition of metals has been carried out by the ion assisting method (IBAD - ion beam assisted deposition) in a mode in which the deposition of metal and the mixing of the deposited layer with the atoms of the base surface by accelerated

voltage of 5 kV with ions of the same metal are carried out from the neutral fraction of vapor and ionized plasma of vacuum arc discharge [15]. Base surfaces have been exposed to mechanical polishing and degreasing in ethanol.

Analysis of the layers obtaining by ion-beam assisted deposition of cadmium and zinc on steels by electron probe microanalysis and Rutherford backscattering spectroscopy showed that their composition includes the deposited metal atoms, substrate material, and impurities of carbon and oxygen. According to data obtained in the processing of Rutherford backscattering spectra, the thickness of the modified layers is about 30-100 nm, and the integral content of the deposited metal atoms on the surface is $-10^{16}-10^{17}$ cm⁻².

Electrochemical diagnosis of galvanic compatibility of aluminum and its alloy D16 to the steel samples, as well as contact corrosion speed was carried out by registering of compromise potential and regestering of the polarization curves in a 3% solution of NaCl. To register compromise potential and the polarization curves potentiostat IPC Pro MF, connected to a PC with the software "IPC2000" was used. Electrochemical measurements were performed in a standard three-electrode cell. A saturated silver chloride electrode was used as a reference electrode, and platinum wire – as a counter electrode. Registration of compromise potentials was carried out whithin 20 min after putting the samples into solution. Then polarization curve has been registered in potentiodynamic mode at a scan rate of 1 mV/s. The initial value of the potential was chosen on 300 mV more cathodic than registered compromise potential after a 20-minute exposure. The final value of the polarization potential of the samples was determined on the achievement of anode current density of 10–40 mA/cm². Nonoperating part of the samples surface was isolated by nitrovarnish. The geometric area of the working part of the samples surface was determined on the images of their surface obtained with a scanner Mustek BearPaw 4,800 TA II with a high resolution of 1,200 dpi. To calculate the surface area ImageJ software was used.

Values of compromise potential of aluminum in a 3% NaCl solution ranged from -800 to -670 mV, of alloy D16 - from -690 to -600 mV. For the sample St3 stationary potential ranged from -470 to -420 mV, and for steel X18H9T - from -230 to -10 mV. Thus, aluminum and its alloy D16 in galvanic couples with carbon steel St3 and stainless steel 12X18H9T will have a negative charge, i. e. will be anodes in the occurring corrosion galvanic cell, and will be exposed to dissolution.

Compromise potential of zinc coatings (Fig. 1, a), obtained as by galvanic and by ion-beam assisted deposition is in the range from -960 to -1,000 mV.

It is obviously, that such values of the potentional are caused by zinc or more exactly by redox-processes between zinc, its surface oxide, hydrox-ide and chloride compounds forming while putting the samples into a 3% solution of NaCl. In corrosion galvanic cell composed of steel samples with Zn coating and aluminum or aluminum alloy D16, coating will have a negative charge, i. e. will be anodes and will be dissolved. Thus, zinc will provide sacrificial protection of aluminum and its alloy until it is dissolved itself.

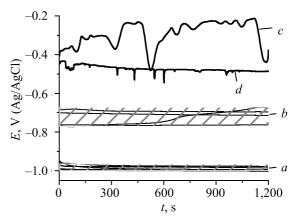


Fig. 1. Compromise potentials of steel samples with Zn (a), Cd (b) and Al coatings on steels St3 (c) and 12X18H9T (d) in a 3% NaCl solution

Cadmium coating on the steels obtained by the two methods (Fig. 1, b), shows the stationary potential in the range from -670 to -770 mV, due to the chemical nature of cadmium. Values of stationary potential of cadmium coatings are very close to the stationary potentials of aluminum and its alloy, so the cadmium coatings on steels will have a very good galvanic compatibility with aluminum and its alloy D16.

Compromise potentials of aluminum coatings on steel samples are close to the stationary potential of steel. For example stationary potential of Al coatings on steel St3 (Fig. 1, c) was ranged from -490 to -440 mV, which almost coincides with the stationary potential of the original steel St3. The stationary potential of Al coatings on stainless steel 12X18H9T was exibited to the significant fluctuations (Fig. 1, d) and changed at the ranges from -490 to -210 mV, which is close to the lower boundary of the stationary potential of the original stainless steel 12X18H9T. Proximity of stationary potentials of Al coatings on steels to stationary potentials of base surface indicates that Al coatings produced by vacuum ion-assisted deposition on steel St3 and 12X18H9T are not continuous. In corrosion galvanic cells formed with St3 and 12X18H9T steels with Al coatings produced by ion-beam assisted deposition, and aluminium or its alloy D16, the latter will have a negative potential and will therefore act as an anode and will be dissolved.

As a rate measure of contacted corrosion values of limiting current density of contacted corrosion were used. They were defined by crossing point of anodic and cathodic polarization curves of the materials that make up the galvanic couple (Fig. 2). Polarization curves were approximated with straight lines in semi-logarithmic coordinates.

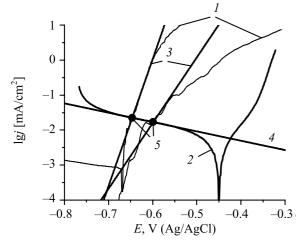


Fig. 2. Polarization curves (PC) of samples of alloy D16 (1), steel St3 (2); linear approximation of anode PC of alloy D16 (3) and cathode PC of steel St3 (4); corrosion parameters (5) (3% solution of NaCl, 1 mV/s)

The value of galvanic corrosion current density determined from the polarization curves is also called limiting current density of galvanic corrosion [16]. The actual current density in the corrosion galvanic cell will be smaller because of the ohmic voltage drop in electrolyte solution.

Determined values of the potentials and current densities of corrosion were averaged for different samples of the same materials composing of a galvanic couple and an interval estimate in the form of the radius of the confidence interval at a confidence level of 0.9 was calculated. Values of the obtained current densities and potentials of contact corrosion of aluminum in galvanic couples with the original steels and steels with Al coatings are presented in Table 1.

Table 1 Potentials (E_{cor}) and current density of aluminium contact corrosion (j_{cor})

Material	$E_{\rm cor}$, mV	$\lg j_{ m cor}$	$j_{\rm cor}$, mA/cm ²
St3	-540 ± 340	-1.93 ± 0.92	$1.2 \cdot 10^{-2}$
SS	-544 ± 39	-1.95 ± 0.22	$1.1 \cdot 10^{-2}$
St3\A1	-580 ± 220	-2.78 ± 0.70	$1.7 \cdot 10^{-3}$
SS\A1	-590 ± 210	-2.93 ± 0.66	$1.2 \cdot 10^{-3}$

Note. SS – stainless steel 12X18H9T.

Parameters of contact corrosion of alloy D16 in galvanic couples with original steels and steels with aluminium coating are presented in Table 2.

Table 2 Potentials ($E_{\rm cor}$) and current density of contact corrosion ($j_{\rm cor}$) of the alloy D16

Material	$E_{\rm cor}$, mV	$\lg j_{ m cor}$	$j_{\rm cor}$, mA/cm ²
St3	-620 ± 40	-1.72 ± 0.10	$1.9 \cdot 10^{-2}$
SS	-621 ± 12	-1.75 ± 0.11	$1.8 \cdot 10^{-2}$
St3\A1	-655 ± 24	-2.56 ± 0.08	$2.8 \cdot 10^{-3}$
SS\A1	-660 ± 21	-2.70 ± 0.07	$2.0 \cdot 10^{-3}$

The current on the cathode part of the curve is determined by the cathodic hydrogen evolution reaction and / or reduction of molecular oxygen. The hydrogen evolution reaction becomes thermodynamically possible at potentials more negative than the equilibrium potential of the hydrogen electrode, which depends on the pH-solution. In neutral environment, i. e. at pH = 7, the equilibrium potential of hydrogen release is ~ -640 mV relative to the saturated silver chloride reference electrode. Therefore, the currents at the cathode parts of polarization curves for steel samples 12X18H9T (Fig. 3) at potentials higher than -640 mV are likely caused by the reduction of molecular oxygen and at lower potentials – by both reactions. As the content of the dissolved oxygen in the solution is small ($\sim 10^{-4}$ mol/L [17]), diffusion limiting current density of oxygen reduction is low (approximately 10^{-3} – 10^{-2} mA/cm²). Upon reaching the values of limiting diffusion current density at the cathodic parts of the polarization curves, current will not depend on the potential. Therefore, for steel samples 12X18H9T at the current density corresponding to $lgj \sim -1.5$ current stops depending on the potentional (Fig. 3). This means that under these conditions, the cathodic process of oxygen reduction is in the diffusion mode. Therefore, the linear approximation was not carried out to determine the galvanic corrosion parameters of these samples. Parameters of galvanic corrosion were determined on the intersections of the experimental cathodic polarization curves of steel with aluminum anodic curves. According to Table 1 and 2 the average values of current density of galvanic corrosion of aluminum and its alloy in galvanic couples with steels St3 and 12X18H9T are about the same and constitute $(1-2) \cdot 10^{-2} \text{ mA/cm}^2$.

Cathode parts of polarization curves for the samples of steels with Al coating obtained by vacuum ion-assisted deposition, are characterized by a small angle, so that current densities of galvanic corrosion of aluminum and alloy are approximately one order less than in the case of initial steels (Table 1 and 2). This is due to a lower catalytic

activity to the oxygen reduction reaction of steel surface with Al coating, than the original steel.

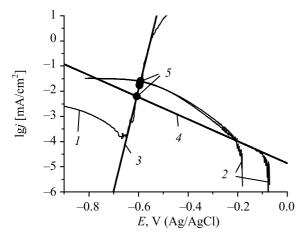


Fig. 3. Polarization curves (PC) of samples Al (1), steel 12X18H9T (2); linear pproximation of anode PC of Al (3) and cathode PC of steel (4) parameters of the corrosion (5) (3% solution of NaCl, 1 mV/s)

Potentials of corrosion of aluminum alloy D16 appeared to be about 100 mV less than aluminum. This is due to a steeper course of the anodic polarization curves for the alloy D16 than Al. Anodic dissolution of D16 alloy takes place at a higher rate than the dissolution of the Al. This is consistent with less corrosion resistance of the alloys containing copper, such as D16, in chloride solutions [18]. Current density of galvanic corrosion of aluminum and alloy D16 appeared to be about the same due to the fact that compromise potential of aluminum is slightly smaller (about by 50 mV) than compromise potential of the alloy D16.

Potentials of galvanic corrosion of aluminum alloy D16 in a galvanic couple with as the initial steels St3, 12X18H9T and steels after ion assisted deposition of Al are in the range from -600 to -680 mV, which is much closer to a compromise potential of alloy, than to compromise potentials of steel samples. In this case, the corrosion process takes place with the cathode control [19]. Diffusion of molecular oxygen to the cathode surface is a rate-limiting step that particularly clear performs at the polarization curves for stainless steels, at which the horozontal section of curve characteristic to the limiting diffusion current are clearly revealed (Fig. 3). In case of galvanic corrosion of aluminum with the same steel samples, the average corrosion potential is about 100 mV higher, however the confidence interval is very wide. Therefore, in this case we can not exclude that the corrosion process can take place under a mixed anodic-cathodic control, in which the corrosion rate is defined as by

the rate of diffusion of molecular oxygen to the cathode surface, and by the rate of anodic dissolution of aluminum. Mixed anode-cathode control characterizes the corrosion of passivated metals [19], which aluminum relates to.

Fig. 4 shows polarization curves for the samples of aluminum, its alloy D16 and Cd coatings formed by different methods on steels. According to the analysis of these curves it can be noted that potentials of corrosion of Cd are only a few millivolts less than potentials of corrosion of aluminum and its alloy, i. e. their potentials of corrosion are practically identical. Current density of corrosion of Cd coverings is more than one order higher than current density of corrosion of aluminum and its alloy D16. In galvanic couples of aluminum or its alloy with Cd coatings on steel aluminum should be a cathode and Cd covering – an anode and cadmium should be dissolved. However, due to nonsignificant difference of the potentials and very low currents on the cathodic branch of polarization curves for aluminum corrosion rate of cadmium must be small if we compare it with the rate of self-dissolution of cadmium, which is confirmed by the data in Table 3.

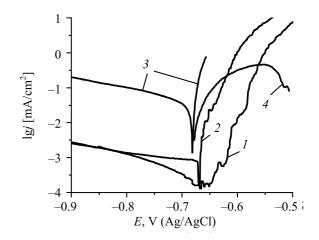


Fig. 4. Polarization curves of the samples of aluminum (*I*), alloy D16 (*I*) and Cd coatings on steel St3 obtained by galvanic (*3*) and ion beam assisted deposition (*4*) (3% solution of NaCl, 1 mV/s)

Table 3 Potentials ($E_{\rm cor}$) and current densities of corrosion ($j_{\rm cor}$) Cd, averaged on all Cd coatings

Corrosion type	$E_{\rm cor}$, mV	$\lg j_{ m cor}$	$j_{\rm cor}$, mA/cm ²
Self-dilution	-750 ± 50	-2.2 ± 0.6	$6 \cdot 10^{-3}$
Contact corrosion- with Al	-790 ± 20	-2.8 ± 0.2	$2 \cdot 10^{-3}$
Contact corrosion with D16	-780 ± 20	-2.6 ± 0.2	3 · 10 ⁻³

As the steel samples with Cd coatings obtained by both methods show approximately the same stationary potential, typical for cadmium, it can be assumed that potentials and corrosion current density for the sample of aluminium will be typical for cadmium. Therefore, it is possible to average all the potentials and corrosion current densities for steel samples coated with Cd and obtain values typical for cadmium. Table 3 shows the average values and 90% confidence intervals for potentials and current densities of self-dissolution and galvanic corrosion of Cd coatings. According to Table 3, the rate of self-dissolution of Cd is not less than a galvanic corrosion rate in galvanic couples with Al and its alloy D16.

Current density and corrosion potentials of Cd in the case of self-dissolution were determined as the intersection point of the anodic and cathodic polarization curves of Cd coatings, and in the case of contact corrosion of Cd – as intersection points of the anode curves of Cd coating with cathode curves of Al or its alloy D16.

Zn coatings on steels in galvanic couples with Al and its alloy will be anodes, their corrosion rate is to be higher than in the absence of galvanic couple. However, as cathode polarization curves of Al and its alloy (Fig. 5) are characterized by low slope and small currents, the contact corrosion rate of Zn may be less than its rate of self-dissolution as in the case of Cd coatings.

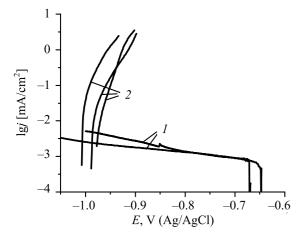


Fig. 5. Polarization curves of the alloy D16 (1) and Zn coatings on steels (2) (3% solution of NaCl; 1 mV/s)

Averaged on all Zn coatings values of current density of Zn self-dissolution appeared to be not

less than the current density of galvanic corrosion in galvanic couples with Al and its alloy D16 (Table 4). The similar situation was observed for Cd coatings (Table 3), but the self-dissolulation rate of Zn is about 30 times less than the rate of self-dissolution of Cd. Therefore, Cd coatings on steels are more preferred than Zn for protection from galvanic corrosion of Al and its alloys under the conditions of corrosion involving chloride ions, for e.g. in coastal and maritime climates.

Table 4 Potentials (E_{cor}) and current density of Zn corrosion (j_{cor}), averaged on all Zn coatings

Corrosion type	$E_{\rm cor}$, mV	$\lg j_{ m cor}$	j_{cor} , mA/cm ²
Self-delution	-1010 ± 30	-1.8 ± 0.3	$2 \cdot 10^{-2}$
Contact corrosion with Al	-1012 ± 12	$-2,0 \pm 0,1$	1 · 10 ⁻²
Contact corrosion with D16	-1018 ± 14	$-2,1 \pm 0,1$	8 · 10 ⁻³

Conclusion. The highest values of current density of galvanic corrosion (about $2 \cdot 10^{-2}$ mA/cm²) were observed in pairs of aluminum and its alloy D16 with the original carbon and stainless steels (aluminum or its alloy D16 is dissolved). The lowest values of current density of galvanic corrosion (about 10⁻³ mA/cm²) were observed in pairs of aluminum and its alloy D16 with Al coatings on steels, formed by vacuum ion-beam assisted deposition. Zinc and cadmium coatings on steels obtained by both methods exhibit compromise potential which is typical for metal coating and self-dissolution rate not exceeding the rate of galvanic corrosion in galvanic couples with aluminum and its alloy D16 in which these coatings are anodes. Self-dissolution rate of cadmium (corrosion current density of $6 \cdot 10^{-3}$ mA/cm²) is approximately three times less than the self-dilution rate of zinc $(2 \cdot 10^{-2} \text{ mA/cm}^2)$ in a 3% solution of NaCl. The nature of anodic and cathodic polarization curves indicates that galvanic corrosion of the alloy D16 occurs with the cathode control, whereas galvanic corrosion of aluminum may occur with mixed anodiccathodic control, especially in the case when aluminum forms a galvanic couple with carbon steel St3. The advantage of ion-beam assisted method in contrast to galvanic is that it is possible to deposit aluminum on the surface of steel and obtain a coating which can have a perfect galvanic compatibility with aluminum and its alloys.

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