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Research paper

Investigation of corrosion damage of hydration aluminium alloys at full-scale accelerated tests

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Abstract

The results of the study of hydrogen penetration in corrosion of aluminum alloys of eight alloying systems are presented. Tests were carried out for four years with additional irrigation with chloride solutions in a moderately warm climate. It is shown that intensive hydrogenation of the alloy occurs during corrosion. The amount of hydrogen in the alloy is determined by the type of corrosion - with intercrystalline corrosion, hydrogen penetration is more intense than with pitting and delaminating corrosion. The mechanism of hydrogenation of alloys is proposed. The change in the energy of the aluminum crystal in the occlusion of atomic hydrogen and the formation of hydrogen molecules is determined by quantum chemical calculations. The relationship between the plasticity of the tested alloys of aluminum alloys and the intensity of hydrogen saturation is shown.

Keywords: *Corrosion; Aluminum Alloys; Full-Scale Accelerated Tests.*

1. Introduction

The results of the study of hydrogen penetration in corrosion of aluminum alloys of eight alloying systems are presented. Tests were carried out for four years with additional irrigation with chloride solutions in a moderately warm climate. It is shown that intensive hydrogenation of the alloy occurs during corrosion. The amount of hydrogen in the alloy is determined by the type of corrosion - with intercrystalline corrosion, hydrogen penetration is more intense than with pitting and delaminating corrosion. The mechanism of hydrogenation of alloys is proposed. The change in the energy of the aluminum crystal in the occlusion of atomic hydrogen and the formation of hydrogen molecules is determined by quantum chemical calculations. The relationship between the plasticity of the tested alloys of aluminum alloys and the intensity of hydrogen saturation is shown.
Key words: corrosion, aluminum alloys, full-scale accelerated tests, hydrogenation.
The investigation of aviation materials protective properties in aggressive environment conditions and researching of corrosion destruction mechanisms are important fields of materials and technologies development in our time [1], [2].
The exposition in high humidity and chloride-ions intensive influence conditions has most aggressive conditions for nature climatic tests and it is more typically for coastal areas. The aluminum alloys corrosion tests were provided by Gelendzhik center for climate research named after G.V. Akimov, which is based on Black seaside; climate characterization – moderately warm with mild winter.
To obtain full data about corrosion processes and to set the critical state requires a comprehensive parameter which characterizing general, pitting, delamination and intercrystalline aluminum alloy corrosion. Metals destruction mechanism and velocity with the fast-test may be obtain at 0,5 – 1 year [3], and forecast time of metal safe using is understate and corrosion destruction mechanism is changed.
Mechanical strength is the most important characteristic of bearing structure material design. Corrosion processes have significant influence on strength and fatigue characteristics of metallic materials. If common corrosion which is inherent for carbon steels in aquatic environment predictably reduces bearing section, then local corrosion, which aluminum alloys is exposed in atmospheric environment may be cause avalanches to reduce the strength of the material and the structure at all.
The work aim is speed-nature in conditions of canopy at the Gelendzhik center for climate research named after G.V. Akimov new generation aluminum alloys 1370T1, 1424T1, B-1461T1, 1913T, B-1341T1, В96ц3пчТ12, B-1469T1 tests result analysis.

2. Materials and method

A number of works [4-6] devoted to investigation of materials corrosion durability under speed-nature tests.
The conditions of speed-nature tests are:

- Installation of samples in a horizontal position on an atmospheric stand under a canopy;
- The exhibition of samples without and with increase of chlorides concentration on the surface of samples.

Increase of chlorides concentration realized by spraying fine solution of sea salt in a concentration of 5 g / l using a spray 1 time a day in the evening [8].
For testing were used samples from sheets of wrought aluminum alloys 1424T, B-1341T1, В96ц3пчТ12, 1913T3, B-1461T1, B-1469T1, 1370T1 thickness of ~ 2 mm (Table. 1).

Table 1: Characteristics of Aluminum Alloys, Which were used for Tests

Alloy	Prefabricated, thickness	System
B-1461T1	List 2,0 mm	Al-2,8Cu-Li

1424T1	List 1,6 mm	Al-5Mg-1,5Li
1370T1	List 1,6 mm	Al-1,4Mg-1,2Si-1,4Cu
B-1341T1	List 1,5 mm	Al-0,7Mg-Si
В96ц3пчТ12	List 1,8 mm	Al-4,5Zn-2,3Mg-2Cu
1913T3	List 2,5 mm	Al-2Mg-4,5Zn
B-1469T1	List 2,4 mm	Al-1,8Cu-Li-Mg

2.1. Cracks formation theory in the metals

Destruction of metal materials under all types of loading (static, dynamic, cyclic, etc.) connected with the processes of plastic deformation, origin and propagation of cracks, which in turn are characterized by a number of stages. For the first time in 1935 A. V. Stepanov found that the stress - strain diagram Of NaCl crystals [9] is similar to the three – stage hardening curves of ICC (intercrystalline corrosion) - metals single crystals: the stage of light sliding, linear hardening and the stage of parabolic hardening [10] (in a number of works on this diagram there is also a “zero” stage; in [11] the stage of passage of The Luders-Chernov front is adopted for the zero stage). A detailed analysis of the stages of plastic deformation of single crystals is given in articles [10 - 12].
In the works [13], [14] it was shown that in flexible metals (Fe, Ni, Al and Cu) micro cracks of 80-500 a wide and up to 5 microns long arise under deformation of 7-10% under conditions of active tension at room temperature and threshold stress at which such cracks appear on metal surface was isolated. L. M. Rybakova [16] analyzed stages of destruction.
When considering the process metal materials destruction (whether static deformation or any more complex type of loading-fatigue, creep, etc.), it is customary to divide the whole process of deformation accumulation and destruction into two main periods - the period of origin and the period of crack propagation [16, 17]. With static tension, it seems possible to flexible deformation and damage accumulated before the neck formation is classified as the period of origin of cracks, and the neck formation with subsequent destruction as the period of cracks propagation. Scheme of nucleation of microcracks by Cottrell: inside the grains; the grain boundary. During this reaction, the elastic energy is reduced, so that all subsequent dislocations are poured into the newly formed dislocation and form a gap that acts on the crystal as a wedge and with sufficient length and applied voltage leads to the destruction of the material. In the case of flexible metals and alloys containing non-metallic inclusions, viscous fracture often has a patchy character [18]. In this case, microcracks are formed at the interface of the inclusion - matrix, and in this case, the origin of microcracks along grain boundaries under tensile stress in corrosion [18] environment aggressive environment makes a great specificity in the formation of microcracks. Microcracks originate mainly on grain boundaries as a result of joint action of applied stress and corrosive environment influence [18].
Hydrogen-containing medium also facilitates the emergence of microcracks in structural steels due to the flow of physico-chemical processes and electrochemical effects. [19].
For the origin and propagation of cracks in deformable (flexible) materials and the associated occurrence of plastic deformed zones, energy is required, the amount of which is several orders of magnitude higher than the specific surface energy. If atomic hydrogen penetrates into the crystal lattice, the processes of physico-chemical and electrochemical effects become similar: in both cases, atomic hydrogen diffuses into critical areas. The latter can be internal defects, on which molecular hydrogen is formed as a recombination result at the internal boundaries of the phases and high pressure occurs [20].
This failure occurs at high temperatures and low strain rates or low stresses. Wedge cracks are usually formed at the junction of the borders of three grains and spread in the direction of the grain boundary perpendicular to the axis of loading.

2.2. Surface layer condition influence

The influence of roughness [21], [22] is most pronounced for higher-strength materials. If the maximum depth of unevenness R is less than a certain critical value, which depends on the material, then there is no change in the fatigue limit with the improvement of surface treatment quality. If R is greater than this critical value, the fatigue limit decreases with increasing R, and in some cases this dependence is close to linear in the coordinates "the ratio of the fatigue limit for a given roughness to the fatigue limit for a polished surface - lg R".
The stress concentrators presence (for example, from rough machining), residual tensile stresses, corrosion damage to the surface under the aggressive environment influence and other factors number leads to a decrease in the endurance limit.
Hydrogen is the only gas that is appreciably soluble in aluminum and its alloys. The hydrogen solubility in aluminum changes in proportion to the temperature and the square root of the pressure and increases sharply with the increase in temperature above the liquidus. Hydrogen solubility is much greater in molten than in the solid state of aluminum. The values of the limit concentration of hydrogen in molten and solid pure aluminum just above and below the solidus have a content of 0.69 cm^3 / 100 g and 0.04 cm^3 / 100 g, respectively. These values are slightly different for most casting alloys.
As shown in [23], hydrogen concentration increasing at aluminum alloy surface due to corrosion leads to a certain increase in hardness, reducing the concentration – to a decrease. The content of hydrogen depends on the temperature, with temperature increasing; the yield of hydrogen molecules from the crystal lattice of the aluminum alloy is more likely. This is confirmed by the data [24], in which the hardness values were shown depending on the aging time, that is, at low mobility of the crystal lattice, hydrogen accumulates in the surface layer of the alloy, then due to the restructuring of the surface structure of the metal, the equilibrium and hardness (hydrogen concentration) returns to the initial value.

3. Results and discussion

3.1. Evaluation of changes in the mechanical properties of materials

After full-scale exposure to the samples of the above thicknesses were carried out standard tests for axial tension at a temperature of +200 C (GOST 1497), as well as tests for low-cycle fatigue (ICC) at a temperature of +200 C, the coefficient of asymmetry R = 0,1, the frequency f = 35 Hz (GOST 25.502-79).
During the tests with PX there was a significant decrease in the relative elongation for almost all alloys; values σ_b and $\sigma_{0,2}$ decreased by 1.5-2 times during the exposure period from 1 to 4 years (Table 2).
Summary data on the aluminum alloys in situ-accelerated tests of for 4 years results is shown at table 2, from which it can be concluded that the greatest loss of alloys mechanical properties characteristic that have shown a high tendency to ICC, delaminating corrosion (DLC) and pitting corrosion, which form stress concentrators in mechanical and fatigue tests.

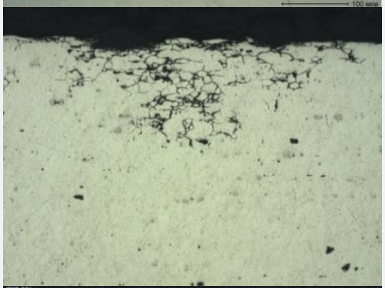
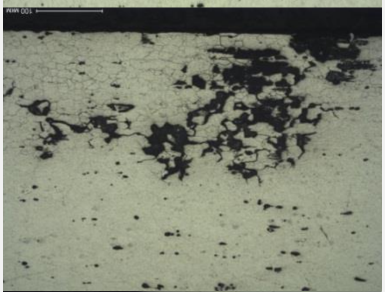
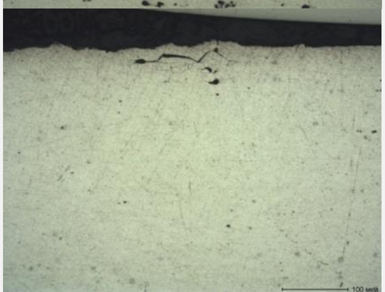
Table 2: After 4 Years of Full-Scale Accelerated Tests Aluminum Alloys Corrosion Destruction Studies Results

Alloy	V_{corr}, g/m^2day	ICC, mm	Pitting depth, mkm	DLC	Loss $\sigma_в$, %	Loss δ, %
B-1461T1	0,0327	0,13	228	2	10,1	45,1
1424T1	0,0082	No	159	2	13,9	78,2
1370T1	0,0217	0,4	273	2	21,1	89,2
B-1341T1	0,0177	0,14	149	6	14,2	73,2
В96ц3пчТ12	0,0349	0,21	360	8	12,1	89,4
1913T3	0,0164	No	291	2	5,6	42,5

B-1469T1	0,0314	0,19	216	2	13,5	64,7

Thus, the best corrosion durability according to the results of a comprehensive assessment 1424T1 have alloys (Al-5Mg-1,5 Li), 1913T3 (Al-2Mg-4,5 Zn), medium – 1469T1 (Al-1,8 Cu-Li-Mg), B-1341T1 (Al-0.7 Mg-Si), 1469T1 (Al-1,8 Cu-Li-Mg), B96ц3пчТ12 (Al-Zn 4,5-2,3 Mg-2Cu), 1370T1 (Al-1,4 Mg-Si 1,2-1,4 Cu). It is necessary to pay attention that the greatest corrosion causes the maximum fall of plasticity of metal.
To determine the reasons for the significant reduction in the ductility of aluminum alloys in the course of climatic tests, the plates were manufactured on equipment manufactured by Struers (Denmark). With the help of cutting machine Labotom cut cross-section samples. Then the samples were pressed into the polystyrene at the CitoPress installation and prepared micro–plates on the polishing machine Tegramin.
Image shooting is performed on the optical complex of the company "Leica", computer processing is carried out with the help of image analyzer "Image Expert Pro 3x" of the company «Nexsys» (Russia).
And the measurement of hydrogen content in samples according to GOST 21132.1 Aluminum and aluminum alloys. Hydrogen determination method in solid metal by vacuum heating/ Vacuum heating method with mass spectrometric analyzer.
In table 3 shows the results of measurement of hydrogen content in the aluminum alloy samples after the environmental tests.

Table 3: Hydrogen Content in Aluminum Alloy Depending on the Type of Corrosion Damage

Alloy	Prefabricated, thickness	System	Hydrogen content, % mass (cm3 / 100 g)	The appearance of the cone
1424	Plate 1,6 mm	Al-5Mg-1,5Li	0,000300 (3,4)	
1370	Plate 1,6 mm	Al-1,4Mg-1,2Si-1,4Cu	0,000643 (7,22)	
B-1341	Plate 1,5 mm	Al-0,7Mg-Si	0,000373 (4,19)	
1913	Plate 2,5 mm	Al-2Mg-4,5Zn	0,000106 (1,19)	
B-1461	Plate 2,0 mm	Al-2,8Cu-Li	0,000452 (5,08)	

В96ц3пч	Plate 1,8 mm	Al-4,5Zn-2,3Mg-2Cu	0,000406 (4,56)	
В-1469	Plate 2,4 mm	Al-1,8Cu-Li-Mg	0,000406 (4,56)	

It can be seen that the type of corrosion damage has a decisive influence on the hydrogen content in the sample from table 3. Because the sample 1370, cut from a sheet thickness of 1.6 mm and a doping of Al Mg Si -1,4 -1,2 -1,4 Cu contains the maximum number of hydrogen - 0,000643 % wt. (7,22 cm^3 / 100 g), which is associated with a specific type alloy corrosion damage, practically, only ICC. The amount of hydrogen contained in the crystal aluminum alloy lattice depends on the degree and corrosion damage type: if the minimum thickness of the corrosion damage, the amount of hydrogen as much as possible, in large cavities, etched by corrosion in the metal, the amount of hydrogen is somewhat lower, that is, when the difficulty of the diffusion step of the removal of hydrogen through the narrow gaps at the ICC hydrogen is mainly directed into the crystal lattice of aluminum. The hydrogen content in the samples after the field-accelerated tests is 5-10 times higher than the maximum possible in the smelting of aluminum alloys, which clearly indicates hydrogen saturation in the corrosion of aluminum alloy. The results of the analysis of the literature and experiments made it possible to formulate the mechanism of corrosion of aluminum alloys in conditions of high concentrations of chloride ions on the surface.

Multistage corrosive [23], [25] process of metal electrochemical dissolution and corrosion rate depends on depolarizer ions to the metal surface transfer time, corrosive metal ion from the crystal lattice at the anode site release time and hydrogen recovery time at the cathode site. At ICC, the dissolution rate of the intermetallic phase at grain boundaries significantly exceeds the dissolution rate of aluminum grains, which can act as a cathode in relation to the anode of the intermetallic intergranular phase [3]. Arrangement of the corrosion process development in aluminum alloy is shown at fig. 3.

Common areas – anode at the crack bottom and cathode, as ever, under aluminum oxide film are shown at fig.3 too. Anode half reaction is aluminum dissolution due to surface oxide film loosening and potential surface in the anode area displacement. Cathode half-reaction is hydrogen recovery from hydroxide ions occurs.

Oxide films optimized structure results on the surface of aluminum by quantum chemical calculations using HyperChem program, MINDO3 method [26-28] are shown at Fig. 4. The calculation result determined bonds lengths in aluminum and aluminum oxide crystals.

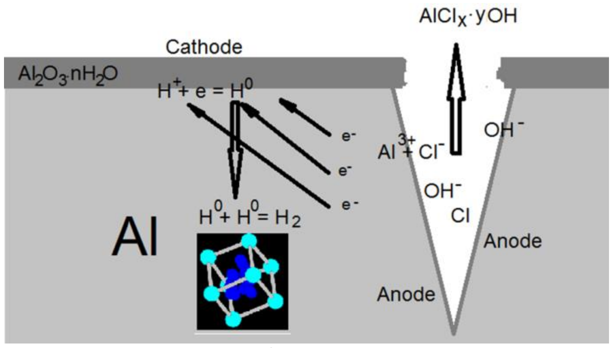


Fig. 3: Hydration Mechanism of Aluminum Alloy Corrosion in Chloride Ions Presence.

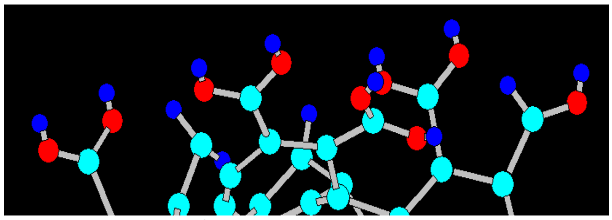


Fig. 4: External View of the Aluminum Crystal Aluminum Oxide Layer Formed.

Thus, the atomic hydrogen formed on the aluminum – aluminum oxide boundary is more likely to rush not through a dense oxide film (0.23-0.40 nm) not outwards, but into the more rarefied crystal structure of the aluminum crystal (0.45-0.63 nm). Hydrogen occlusion occurs in aluminum.

Aluminum alloys are not able to absorb an infinite amount of hydrogen and after the onset of the limit (7-15 molecules) there is a rupture, bonds rearrangement in the crystal lattice and the hydrogen molecules yield to the outside.

It is known [29]; the fragility has a significant influence on the fracture aluminum alloys toughness. On the chart (Fig. 5) shows the various fracture toughness change K_c alloys of the applied load of $\sigma_{0.2}$. It is seen that with the fracture increase the toughness strength is greatly reduced.

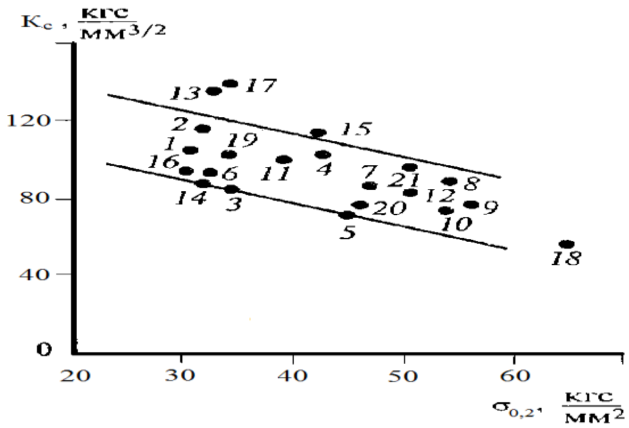


Fig. 5: Aluminum Alloys Crack Resistance Characteristics Dependence on Strength.

1-D16T, 2-D16pch, 3-D16T1, 4-D16T1pch, 5-2024-T851, 6-AK4-1T1, 7-V95T1, 8-V96T1, 9-V96cT1, 10-7075-T651, 11-7075-T7361, 12-7079-Kzt650., 13-01911T1, 14-01420T1, 15-AK8T1, 16-6061-T651, 17-ED-1, 18-7178-T6, 19-7039-Kzt61, 20-2014-T651, 21-7178-T651, 22 al-23-1.

Aluminum alloys research results comparison after field and field-accelerated testing in the marine atmosphere of Noumea city when exposed to elevated concentrations of chlorides by methods such as physical-mechanical tests, microstructure analysis, hydrogen content-vacuum-heating with a mass spectrometric analyzer determination and literature analysis resulted in important, from authors point of view, conclusions about the corrosion cracking mechanisms of aluminum alloys.

4. Conclusion

1) During aluminum samples exposure in black sea coast environments is an intensive hydrogen absorption in a surface metal layer.
2) Hydrogen absorption is greater than is less than the geometric sizes corrosion damage – the maximum hydrogen absorption exposed alloy with the ICC and the DLC in the corrosion propagation along the grain boundaries and delamination from the closed direct corrosive environment cavities access. The minimum hydrogen absorption is observed in pitting and ulcer corrosion case, forming a "broad" corrosion cavity, enabling the free release of the atoms and molecules of hydrogen out.
3) Hydrogen occlusion of into the metal crystal lattice not only causes its embrittlement, and leads to the stratification of samples (alloys В96ц3пчТ12, В-1341Т1).
4) Alloys containing copper are the most susceptible to hydrogen embrittlement.
5) Hydrogen embrittlement of alloys, changes in the surface relief, the formation of stress concentrators in the form of pitting and cracks ICC leads to a significant decrease in the strength and plasticity of aluminum alloy samples.
6) The most significant factor in strength reduction during aluminum alloys corrosion is hydrogen occlusion by the ICC and the DLC, therefore, the change of hydrogen content is the most indicative value characterizing the general condition of the aluminum alloy and its crack resistance in static terms and fatigue loading in corrosive black sea coast environments.

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