

Review article

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Modern strategies for the creation of polymer coatings. Part III¹⁾

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ABSTRACT

Introduction. Coatings on hard materials are widely used in many industries. Coating technologies help prevent or reduce corrosion, contamination and biofouling, chemical and structural degradation, and wear and tear of external surfaces due to exposure to the elements and natural environments. The range of materials used for functional coatings is quite wide: from organic polymers to hybrid composites and inorganic nanoparticles, depending on the desired properties and functionality of the final product. Despite the excellent anti-corrosion characteristics of non-polymer coatings, their usage causes environmental damage. Organic coatings are among the most widely used. Such compositions are applied in liquid form; organic solvents are one of the main components. Environmental concerns have encouraged the development of alternative technologies. The main areas for development are availability of raw materials and the cost of environmentally friendly coatings. **Results and discussion.** The review substantiates the relevance of research on the development of multifunctional polymer-based coatings. The market for polymer coatings is presented. Methods of surface protection, types of coatings formed, their main components, features of the formation of coatings, the influence of various factors on the formation of polymer coatings, including methods of preparation and pre-treatment of the protected surface are presented. Methods for preventing corrosion are discussed in detail, as well as the main directions in the development of anti-corrosion coatings based on various protective mechanisms. The characteristics of the main components of protective coatings are given. The issue of destruction of polymer coatings depending on the operating environment is considered in detail. The types of media, their influence and mechanisms of action on protected objects are considered. Factors and mechanisms of destruction of polymer coatings, methods for preventing degradation of coatings are listed. The latest technologies for the formation of protective polymer coatings are highlighted. **Conclusion.** Currently, coatings provide a wide range of quality indicators. An important characteristic of modern coatings is minimal negative impact on the environment, which requires an integrated approach to the design and production of coatings.

KEY WORDS: adhesion, protection, corrosion, coating, polymer, solvent, thermosetting resin.

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3. DESTRUCTION OF POLYMER COATINGS

Deterioration of organic coatings can be superficial in the form of cosmetic defects and defects leading to corrosion. An important aspect of the development of highly effective anti-corrosion coating systems is the study of the interaction between coating components and the study of the fundamental physical and chemical mechanisms of coating degradation during operation [225].

Anti-corrosion coatings are exposed to various environments and factors: water, soil, atmosphere and ultraviolet radiation. The specific requirements for coating systems are determined mainly by environmental factors and elements interacting with the coating during operation, as well as the time of their exposure. For example, coatings in the industrial sector are exposed to chemicals and rain, while coatings buried in soil interact with bacteria and moisture.

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3.1. Classification of media

During operation, anti-corrosion coatings are exposed to environments with varying aggressiveness. The classification of environmental conditions by type of exposure is given in state standard 34667.5-2021 (ISO 12944-5:2019) “Paint and varnish materials. Protection of steel structures from corrosion using paint and varnish systems” [226]. The standard describes the environmental effects on steel structures that are exposed to the atmosphere, submerged in water or buried in the ground. The standard provides a classification system based on corrosivity categories for different environmental conditions. Classification of environmental conditions is necessary for the selection of protective systems.

Atmospheric corrosion is a process that occurs on a metal surface under the influence of air moisture. The corrosion rate increases under the influence of the following factors:

- with increasing air humidity;
- when condensation forms;
- with an increase in the amount of pollution in the atmosphere, aggressive pollutants react with the substrate and form deposits on the surface.

Corrosion develops intensively at relative air humidity above 80% and temperatures above 0 °C. If air pollution is high or hygroscopic salts are present, corrosion also occurs at lower humidity.

Climate determines air humidity and temperature. In cold or dry climates, corrosion processes are slowed down. The highest corrosion activity is observed in hot, humid and maritime climates. The determining factor is the humidity period – the time the structure is exposed to high humidity.

Corrosion processes are affected by the placement of structural elements. Outdoor structures experience different climatic conditions and cycles: rain, sun, pollutants in the form of gases, aerosols, etc. The presence of shelter smoothes out the impact of climatic factors. Indoors there is virtually no atmospheric pollution, but the rate of corrosion can increase due to insufficient ventilation, high air humidity and condensation.

In environmental conditions, the corrosiveness is quite low compared to the industrial atmosphere and marine climate. Industrial environments are characterized by high concentrations of atmospheric particulates, soot, sand and sulphate salts. Rain with high levels of sulfur dioxide in the atmosphere forms acid rain and the coating becomes acidic. The marine atmosphere is characterized by a very high content of aggressive Cl⁻ ions, which cause pitting corrosion.

The rate of corrosion processes in water is affected by:

- type of water – fresh, brackish or salty;
- oxygen concentration in water;
- type and concentration of substances dissolved in water;
- water temperature.

Underwater flora and fauna accelerate corrosion.

There are three different zones for immersion of structures in water:

- underwater zone;
- intermediate (or level-fluctuating) zone, the water level on the structure changes due to natural or artificial factors that increase corrosion due to the combined effects of water and the atmosphere;
- zone of splashes and variable wetting, the structure wetted by waves and splashes experiences high corrosion stress, especially when exposed to sea water.

The splash zone is an extremely hostile environment, combining an oxygen-rich atmosphere with a continuous spray of electrolytes from the sea. The degradation of coatings in the splash zone is accelerated by ultraviolet radiation and mechanical stress caused by constant alternation of periods of wet and dry conditions.

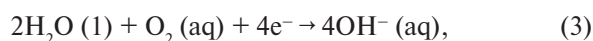
Corrosion in the soil depends on the type of soil and is determined by the content of minerals, their types, the presence of organic substances in the soil, the content of water and oxygen, the degree of aeration, and the presence of bacteria.

Structures submerged in water or buried in soil may be damaged by sand, gravel or rocks, biofouling.

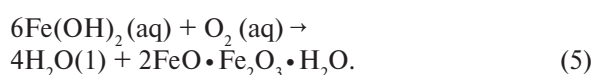
3.2. Mechanisms of destruction

Corrosion is a process that occurs during the physical and chemical interaction of a metal with the environment. For corrosion processes to occur, an electrolyte solution and a metal conductor are required between two separate areas with different potentials, that is, the anode and the cathode. Besides iron and steel, other examples of corrosion products are green patina on copper and white rust on zinc.

As a result of corrosion processes, anodic and cathodic regions are formed on the steel substrate. At the cathode, oxygen is reduced on the catalytically active surface of the oxidized metal predominantly to hydroxide ions (reaction 3), but other reaction products such as peroxides, superoxides and radicals can also be formed. Several corrosion reactions occur at the anode, resulting in the formation of Fe²⁺ ions and electrons (reaction 4).



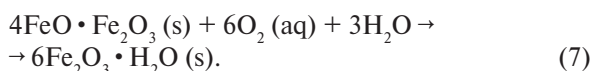
Iron (II) hydroxide then oxidizes into iron oxides to form green hydrated magnetite: FeO·Fe₂O₃·H₂O:



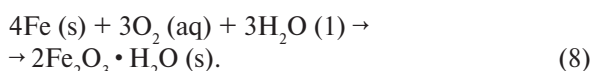
Hydrated magnetite is unstable and decomposes into black magnetite:



In the presence of oxygen, black magnetite oxidizes into a stable red-brown hydrated hematite ($\text{Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}$) called rust:



General reaction



The driving force for corrosion (Figure 14) for steel is the potential difference between the anodic and cathodic sites. The overall electrical equilibrium potential of a galvanic cell is determined by the difference in standard potentials between the anodic and cathodic reactions of the half-cell: E_{Ox}^0 and E_{Red}^0 , respectively.

3.3. Factors of destruction

Under atmospheric conditions, the coating is exposed to various factors, which leads to the formation of cosmetic defects: loss of gloss, color change and chalking.

Most cosmetic defects are caused by degradation of the binder under the influence of UV radiation [227–228]. More serious problems are cracks in the coating and loss of adhesion.

Since adhesion reduction occurs before corrosion occurs, it is important to know the mechanisms responsible for the reduction of adhesion between an organic coating and a metal substrate [229]. For example, in atmospheric conditions, zinc coatings can peel off due to poor curing conditions, which subsequently leads to compromised corrosion protection. The mechanism responsible for the decrease in adhesion is “wet adhesion”, i.e. adhesion of a coating to a metal substrate in the presence of water [229]. Water vapor always eventually penetrates the coating. Water molecules at the interface between the coating and the substrate bind hydrogen ions and neutralize the adhesion forces between the coating and metal oxides [230]. A small number of polymer-metal bonds are suitable to withstand hydrolysis over a long period of time. Thus, in a humid environment, adhesion weakens, an electrolyte solution is formed at the interface, and as a result, corrosion processes are initiated. Using FTIR-MIR methods, the thickness of the safe water layer was determined, and it was also discovered that this layer accumulates with increasing potential. Several layers of coating are required to inhibit water accumulation [231–233]. The process of

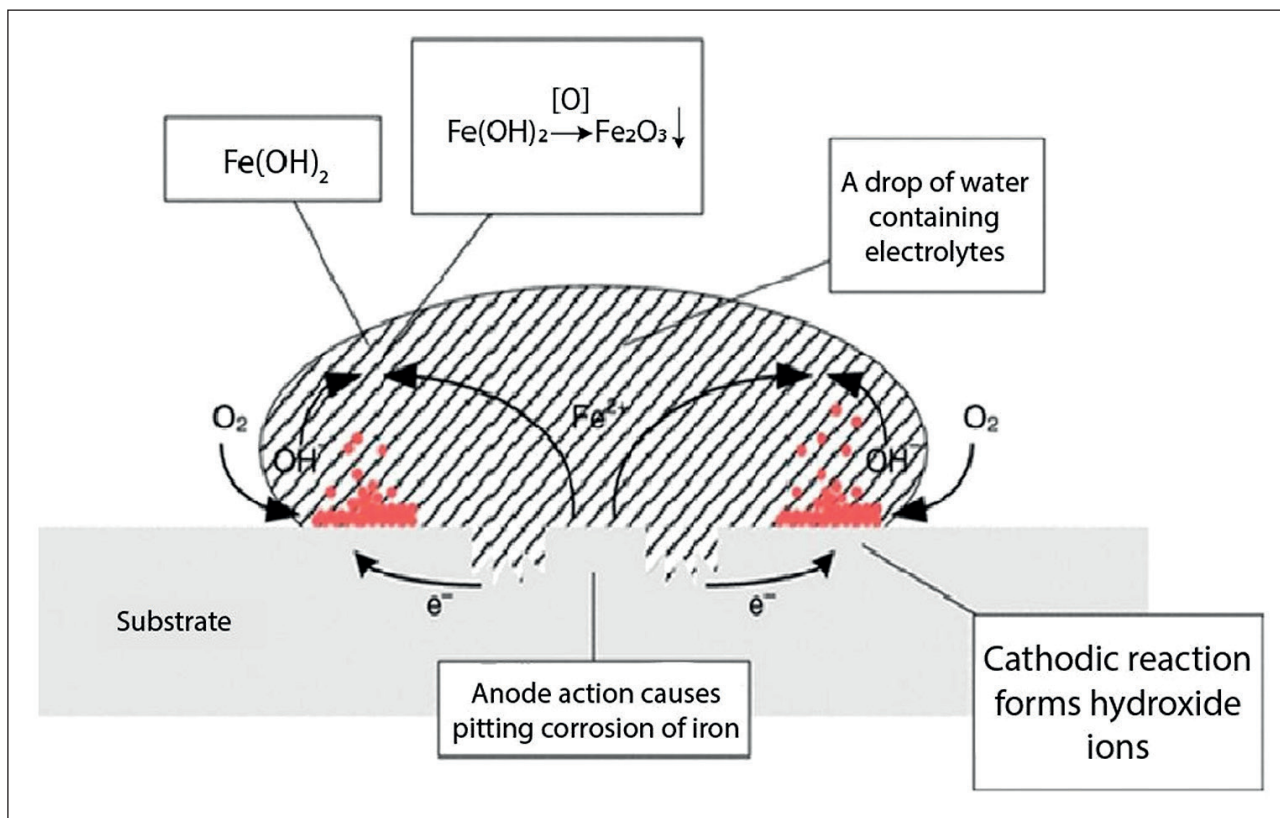


Fig. 14. Scheme of corrosion processes on steel

water accumulation at the coating-substrate interface is confirmed by studies of water absorption in adjacent and free films. Water absorption in adjacent areas of coatings is higher than in the corresponding free films [234]. Based on the magnitudes of metal-adhesion interaction energies (in the range of about 25 kJ/mol or lower) and metal-water interactions (binding energies in the range of 40–65 kJ/mol), water displacement of the coating is likely [235].

3.4. Types of destruction

Filiform corrosion

In humid atmosphere conditions, filamentous corrosion is a relevant mechanism for the degradation of organically coated metals. Although filamentary corrosion is often observed on organically coated aluminum parts, magnesium and cold-rolled steel are no exception. Filiform corrosion usually occurs on microscopic scratches or coating defects. Filiform corrosion is usually superficial and consists of thin filamentous deposits, especially progressing under the coating along the surface. Corrosion threads can be thought of as a corrosion head followed by a tail, which are porous corrosion products. Oxygen acts exclusively on the active corrosion head [236]. Differential aeration is observed as oxygen and water enter the corrosion head through diffusion through the porous tail [237]. The only effective method of preventing filament corrosion is to dehydrate the filament head by reducing the relative humidity to approximately 60%. Multiple layers of coating, low water vapor transmission and inhibitors slow but do not prevent filamentous corrosion in environments where humidity cannot be controlled.

Degradation of submerged coatings

In systems with embedded organic coatings, the most important and common forms of visible failure are blis-

tering and delamination (Figure 15). The differential interaction of hydroxyl ions with the metal substrate leads to either cathodic swelling or cathodic delamination. The formation of bubbles is the result of osmotic pressure due to the high solubility of the cathodic reaction products in water. As a result of the resulting alkaline environment, due to the presence of cathode reaction products, bonds are broken at the coating-metal interface, which leads to delamination.

The alkaline environment under the peeling coating and in the cathode areas is associated with the main corrosion processes of steel in seawater. The presence of defects in the protective coating contributes to a more active impact of the environment on the steel. This is how a galvanic cell with anode and cathode regions is formed. The steel dissolves in the anodic region. The oxidized metals then catalyze cathodic reactions, the anodic reaction being balanced by the cathodic reaction. Under natural conditions, the cathodic reaction will involve the reduction of oxygen [238].

Cathode stratification

In the case of an anti-corrosion coating with defects, in practice, corrosion begins much faster than for a coating without defects. The basic principles of cathode dissection are generally described [239]. It is believed that the cause of delamination is the so-called cathodic polarization caused by the action of hydroxyl ions or chemical reactions. Oxygen and water necessary for depolarization of the cathode region penetrate through the coating or along the coating-substrate interface. Then oxygen diffusing to the damaged areas enters into the oxidation reaction $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$.

Researchers have paid much attention to studying the transport of water, oxygen and cations through organic coatings onto a metal substrate. It has been shown that typical organic coatings are sufficiently permeable to water and oxygen, and therefore are not able to inhibit delami-

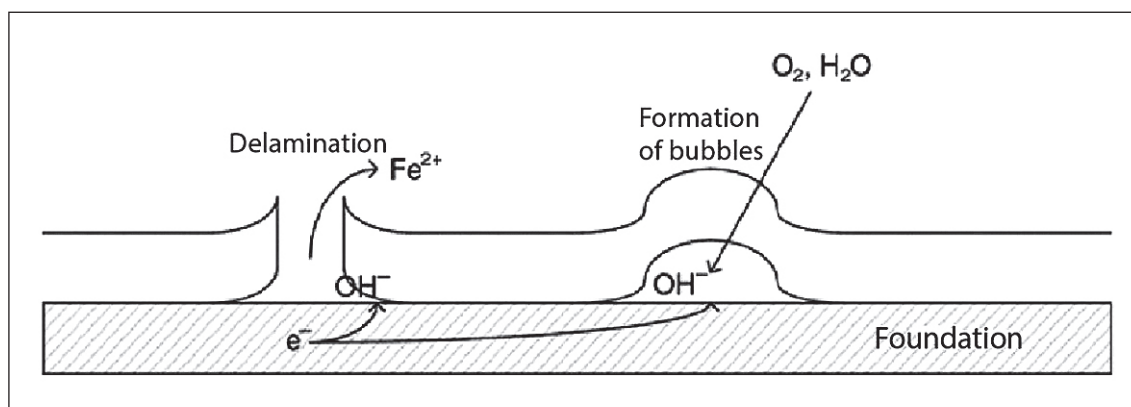


Fig. 15. Sketch of delamination and swelling of the coating

nation [240–241]. The transfer of ions from the environment to the metal surface is carried out through discrete channels with low resistance in the coating. The presence of alkali metal ions and OH ions leads to the formation of an alkaline environment under the coating and in the cathode region the pH can reach 12–14 [242–243]. Diffusion of cations to neutralize the charge of hydroxyl ions is usually the rate-limiting step. It has been proven that the transport of cations along the coating-steel interface occurs much faster than through the coating. The delamination of the coating is controlled by the diffusion of cations [244–245]. This hypothesis is confirmed by the dependence of the separation rate on the hydrated size of the cations. However, a slight change in the rate of delamination in a more stable silane-modified epoxy coating showed that interfacial transport itself does not determine the rate of cathodic segregation.

In cathode blisters under the coating, high alkalinity causes degradation of the coating. However, the exact mechanism has not yet been described. According to [246], the mechanisms of loss of adhesion between the substrate and the coating in an alkaline environment can be divided into three groups: “reduction of the oxide layer, chemical degradation of the polymer, interfacial fracture.” The first way of forming areas of delamination is preferable [246]. Based on this mechanism, the degradation of epoxy and thermosetting acrylic and polybutadiene coatings on cathode-protected steel is described. According to the diagram, such processes are quite predictable for iron at pH = 14 and at cathode potentials less than

960 mV SHE. In situ X-ray photoelectron spectroscopy measurements confirmed that, in an alkaline environment, significant reduction of iron oxide is only possible at cathode potentials less than 800 mV SHE [247–248].

During operation, the polymer matrix of the coating undergoes chemical decomposition; at elevated pH, saponification occurs. Similar cases have been described with coatings based on polyethylene, polybutadiene and epoxy resins on cathode-protected steel [249]. For example, degradation by saponification in the case of polybutadiene coatings is accelerated at pH > 11.8. Studies have shown that the area of initial destruction of the polymer coating is located near the metal surface [250]. The radicals HO²⁻, OH and O²⁻ formed during the reduction of oxygen are the main factor in the degradation processes of the organic layer [251].

Schemes of cathode stratification mechanisms based on literature data are shown in the Figure 16.

In the area of the defect, due to the presence of water, oxygen, and electrolyte ions, a galvanic cell is installed. The resulting hydroxyl ions lead to high alkalinity in the area and reduced adhesion between the substrate and coating. Internal stresses in coatings, temperature and humidity changes that arise during the curing of the resin accelerate corrosion processes. The rate of separation in alkaline solutions [252–253] is significantly lower.

Damage and subsequent detachment of the organic coating from the metal surface occurs in various ways: in the form of cathode dissection, in the form of filamentous corrosion, or when these processes occur together.

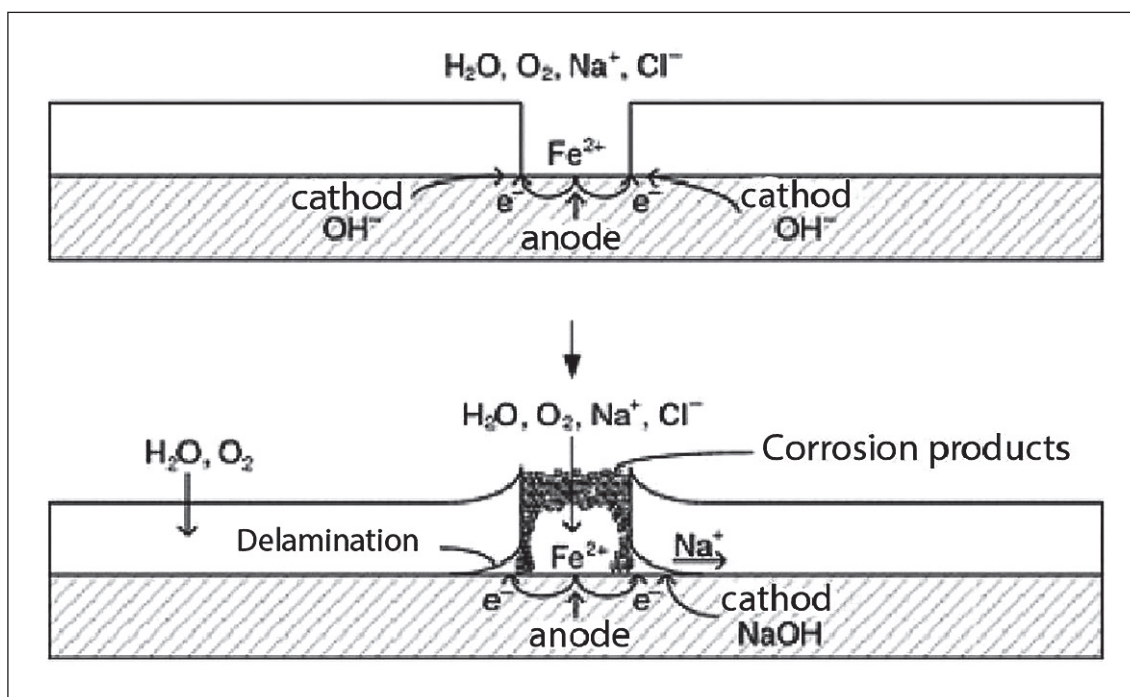


Fig. 16. Delamination of the coating after damage

Delamination of organic coatings on surfaces treated with chromate or phosphate is a consequence of dissolution of the conversion layer. It has been proven that delamination of epoxy coatings on chromated steel is caused by deterioration of the conversion coating itself. Cathodic conditions under the epoxy coating reduce $\text{Cr}^{6+} \rightarrow \text{Cr}^{3+}$. In the case of phosphate conversion coatings, similar processes occur [254]. Experimental data confirm a reduction in coating peeling under cathodic conditions in the presence of phosphate-containing pigments [255]. However, the characteristics of phosphate-pigmented coatings under cathodic conditions ensure that the binder is resistant to alkali displacement at the coating-steel interface. The decrease in the rate of delamination in the presence of phosphate-containing pigments is explained by the deposition of a phosphate layer on the cathode sites and the polarization of the cathode reaction [256]. Among the factors of cathodic stratification of coatings, one can also highlight the degradation of polymers as a result of oxidative processes and interaction with active intermediate products, such as H_2O_2 and HO_2 , and OH radicals. It is stated that their action is more destructive to the coating than the resulting alkaline environment.

Cathode bubble formation

The formation of blisters is the first visible sign of inadequate protection of the anti-corrosion organic coating. The formation of bubbles on defect-free coatings is described by the following stages: swelling, expansion due to swelling, gas formation and subsequent osmotic processes [257]. It is water-soluble contaminants on the surface of the substrate that are responsible for the formation of osmotic bubbles. Research has been carried out to determine the maximum permissible concentrations of pollutants; in research studies there are significant discrepancies in the limits of the permissible amount of water-soluble pollutants on the substrate before applying coatings.

The fundamental model for the degradation of defect-free organic coatings on steel in a neutral electrolyte assumes the formation of cathode swelling after the transfer of ions along conductive paths. It is assumed that water-soluble cations diffuse through the coating in low cross-linked or low molecular weight regions. The degradation of a defect-free coating under the action of a neutral electrolyte can be summarized in the following stages (in Figure 17, the number in the circle corresponds to the stage number):

Stage 1 – pathways develop under the influence of water in hydrophilic regions or regions with low cross-linking density;

Stage 2 – electrolyte ions diffuse along conductive paths to the surface of the substrate;

Stage 3 – formation of anodic areas on the surface of the substrate;

Stage 4 – formation of cathode sections on the periphery of the tracks;

Stage 5 – Na^+ ions diffuse to the cathode sites to neutralize the charge along the coating-substrate interface;

Stage 6 – alkalinity of the environment promotes cathode stratification;

Stage 7 – water passes through the coating to the cathode areas due to the hygroscopicity of the material in the cathode areas;

Stage 8 – bubbles enlarge and merge.

However, studies have not established the mechanism and reasons for the formation of cathode bubbles, their relationship with certain areas at the coating-substrate interface, such as interfacial microvoids or defects in the structure of the substrate surface. Cathode bubbles can form near the exposed area and on damaged areas of the coating. The damaged area may be a coating defect (pores and microvoids) or an area with low cross-link density. The liquid in neutral blisters is slightly acidic or neutral, while the liquid in cathode blisters is strongly alkaline.

CONCLUSION

Protective coatings are one of the most effective, cost-effective and simple strategies for preventing damage. In particular, anti-corrosion protection of coatings is based on barrier properties, self-healing, active corrosion inhibition, anodic passivation and cathodic protection. Polymer coatings protect a larger tonnage of metal compared to other anti-corrosion methods. While coatings were originally developed solely to protect metals and alloys from corrosion, there is now a need to develop coatings that provide a wide range of quality properties and minimal negative impact on the environment, which requires an integrated approach to the design and production of coatings. In combination with this, it is necessary to develop modern methods for determining the characteristics of coatings by their morphology, chemical and physical properties and corrosion behavior. In addition, the vast majority of research in coating development has been conducted *ex situ*. In this regard, to fully investigate the properties and mechanisms of coatings, ideally there should be a synergistic use of methods to study the properties and mechanisms of coatings, as well as to obtain a deep mechanistic understanding at different length scales – studying the behavior of the coating *in situ*. Future research should focus on investigating the cause-and-effect relationships between metal surface properties, moisture penetration, and corrosion rates to develop a multiscale, quantitative, and simulation model to evaluate the anticorrosion properties and durability of organic coatings.

Nanotechnology has great potential for creating highly effective anti-corrosion coatings: increasing water adsorption/absorption, thermomechanical proper-

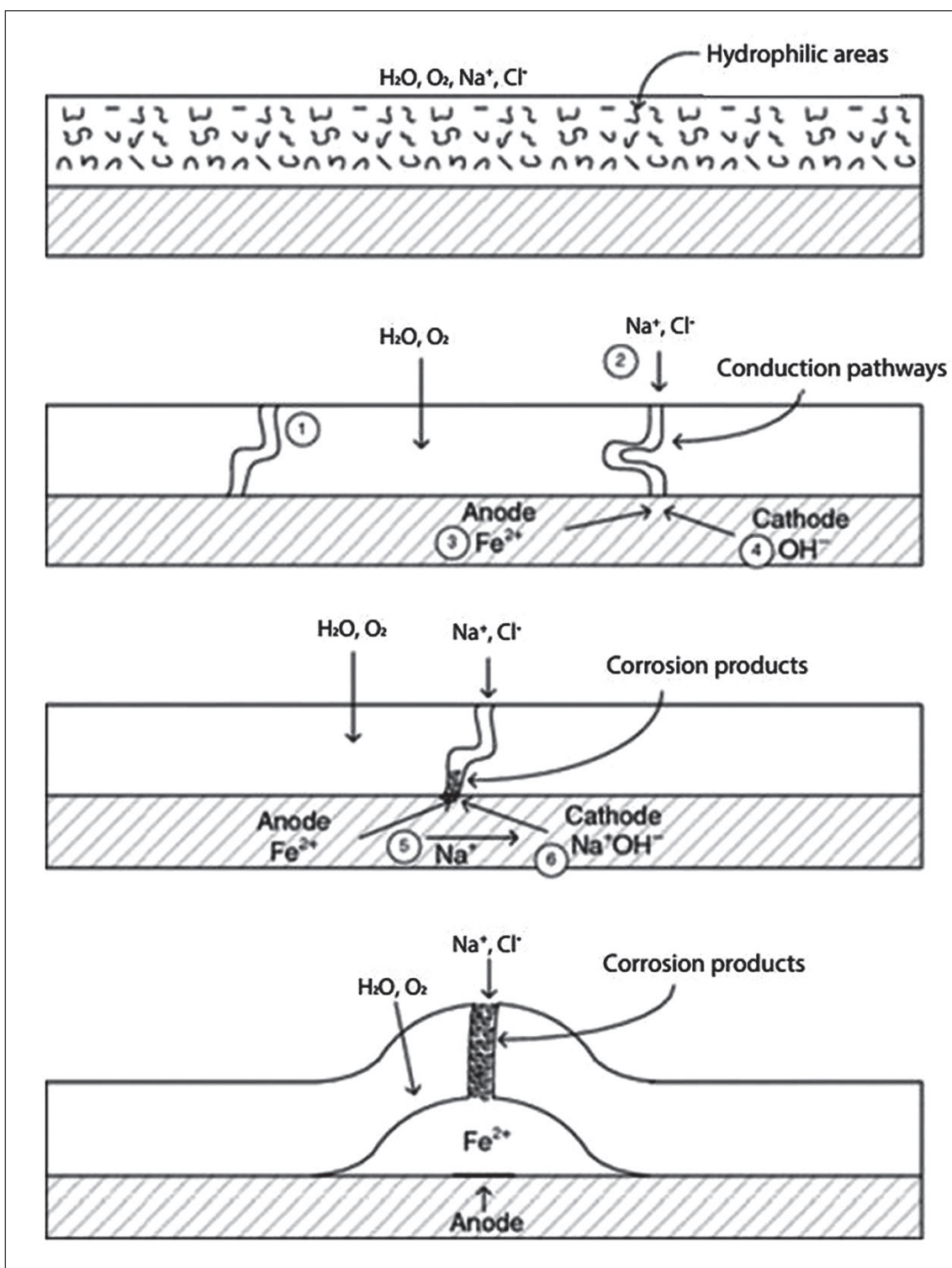


Fig. 17. Mechanism of destruction of defect-free organic coatings on steel in a neutral electrolyte environment

ties, adhesion to the substrate, ultraviolet resistance and chemical resistance, wear resistance, electrical conductivity, cross-linking density, uniformity and biocompatibility. The use of nanomaterials in organic coatings significantly improves barrier properties by re-

ducing porosity and increasing the tortuosity path for corrosives. The development of environmentally friendly polymers, nanoparticles and corrosion inhibitors is a promising solution for creating more environmentally friendly nanocomposite organic coatings.

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AUTHORS CONTRIBUTION

Irina N. Vikhareva – research concept; development of methodology; writing the preliminary text.

Vyacheslav E. Antipin – conclusions.

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