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Modern strategies for the creation of polymer coatings. Part I

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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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INTRODUCTION

In many areas of everyday life, the use of coatings plays an important role. In response to global warming and climate change, green technologies have been identified as a key goal, leading to more efficient consumption of energy and water resources, reduction of waste, pollution and carbon footprint, and protection of the environment and human health [1–3]. Functional coatings are also integral components of "green" technologies. The simplest form of coating is paint, which not only serves aesthetic purposes, but also has additional functions: solar reflectivity, antimicrobial properties and other characteristics. Likewise, other functional coatings, especially those used on exterior building facades such as walls, roofs and windows, can perform important functions: self-cleaning, sunlight filtration, light and heat regulation.

Functional coating formulations are based on various materials and range from organic polymers to hybrid composites and inorganic nanoparticles depending on the desired properties and functionality of these materials [4]. In particular, the rapid development of nanotechnology in recent years has led to a significant increase in innovation in coatings using nanostructured materials (0.1–100 nm).

For example, inorganic nanoparticles, quantum dots, fullerenes, carbon nanotubes. Compared to bulk materials, nanomaterials have a higher surface area to volume ratio, higher surface energy, and less imperfection, resulting in completely different properties and performance of the resulting materials. The introduction of nanotechnology can introduce new properties and functionality and improve the performance of coating materials compared to conventional fillers. As a result, the development of coating technology has contributed to the emergence of new developments in the field of functional coatings, such as anti-fouling, anti-reflective and fire retardant coatings [5–7]. Thus, the development of functional coatings and application technologies is driven by high demand and economic feasibility of their use.

1. POLYMER COATINGS

Polymer coatings on solid materials play an important role in many industries. Universal coatings are coatings that modify a wide range of material surfaces and are stable under certain conditions. Ideally, the coating is independent of the chemical composition and physical characteristics (e.g., topology and stiffness) of the substrates. To develop coatings, it is important to design the interactions between the polymer and the substrate. Existing irradiation technologies are capable of activating many types of surfaces, but the efficiency and density of active sites on some surfaces is relatively low. To obtain dense surface coatings, it is important to strengthen the interactions by compensatory methods, for example, polymerization. On the other hand, interactions of non-covalent nature, such as electrostatic interaction, hydrogen bonding, hydrophobic and van der Waals interactions, are used at almost all types of interfaces. Multiple non-covalent interactions can be considered the driving forces for the formation of polymer coatings on various types of surfaces. But in general, noncovalent interactions between interfaces are not strong enough to produce polymer coatings. Therefore, to increase the stability of the coating, additional intralayer interactions are used: physical and chemical cross-linking.

Crosslinking can be initiated in situ during coating application or in stages after the formation of each layer of the precast coating. In situ coating application is easy and quick. But spontaneous cross-linking can lead to aggregation of polymer modifiers and the process of forming surface morphology is less controlled. Therefore, when obtaining a layer-by-layer coating, additional cross-linking procedures are required, such as heating or irradiation.

Secondary functionalization of universal coatings is typically required to achieve specific surface characteristics. But in this case, a sufficient number of active groups must remain in the coating formula for further modification.

1.1. Classification of coatings

A wide range of coatings are now developed to meet a variety of needs, from food and pharmaceuticals to devices and consumer products, industrial and equipment, automotive and construction products. Accordingly, the functionality of coatings also varies widely: coatings are applied as an outer layer of film to protect, enhance, and/or impart additional functionality, surface properties of an object, or impart characteristics to bulk materials. Coating technologies help prevent or reduce corrosion, fouling and biofouling, chemical and structural degradation, and wear of external surfaces due to various factors, including environmental conditions such as weather, humidity, UV radiation, etc. Coating provides stability, durability, increases the service life of objects or surfaces, and also gives surfaces additional properties and functions: antimicrobial properties, superhydrophobicity and superhydrophilicity, self-cleaning. Functional coatings are used in food and medicine for taste and odor masking, protection and stabilization in the physiological environment, targeted release in the body, etc.

Coatings are classified according to the following basic principles: by purpose, by physical or chemical characteristics, by the nature of the elements included in the composition, by the nature of the phases in the surface layer.

The protective function is performed by reflective, heat-resistant, wear-resistant, corrosion-resistant, electrical insulating, heat-protective coatings.

Structural coatings and films are used for structural elements in products in the production of products in various fields: instrument making, radio electronics, integrated circuits, turbojet engines.

To simplify technological processes, technological coatings are used. For example, applying solders during soldering; production of semi-finished products in the process of high-temperature deformation.

Decorative coatings are important in the production of household products, jewelry, prosthetics in medicine, and improving the aesthetics of industrial installations and devices.

Restorative coatings are used to reduce costs: when restoring worn surfaces of products, such as propeller shafts; crankshaft journals of internal combustion engines; blades in turbine engines; various cutting and pressing tools.

Reflectivity is reduced by surface geometry in optical coatings.

1.2. Substrate surface preparation methods

The characteristics of coatings are significantly influenced by the initial state of the substrate surface. A typical coating on a well-prepared surface performs a better protective function than a high-quality coating applied to a substrate with a poorly prepared surface [8]. The du-

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rability of the coating and adhesive strength completely depend on the preparation of the surface of the product material. That is, surface preparation is an important technological operation in the production and application of coatings. For this, it is important to carry out high-quality preparatory stages: removing contaminants from the surface, including adsorbed substances, and activating the surface.

In the technological process, preparatory operations can be carried out jointly or separately. More often they resort to separate preparation in two or three stages. The surface is activated at the last stage. Surface preparation methods include: washing with water; degreasing; etching; mechanical processing; thermal and chemical-thermal treatment; electrophysical processing; processing with light fluxes; drying [9–10]. Sandblasting is the most effective way to prepare the surface of a metal substrate before coating. However, in situations where sandblasting is not possible due to safety or environmental concerns, the use of a rust remover is recommended [11].

1.3. Coating Methods

Depending on the purpose of the coating, various methods for their preparation are used [12–14]. Coating methods are described in detail in the table 1.

Table 1
Coating formation methods

Method formation	Advantages of the method	Disadvantages of the method
Vapor condensation spraying (PVD)	– variability of work with various solid elements and materials; – application of coatings of any thickness (5–260 microns); – variations of the method are possible.	– processing exclusively the exposed part of the substrate; – low spray quality; – expensive equipment.
Chemical vapor deposition (CVD)	– surface treatment with chemically active elements and compounds that are chemically active in the vapor state; – good spray quality; – thickness 5–260 microns.	– the heating source is important; – higher temperatures than in the PVD method; – difficulty in regulating substrate heating; – probability of unclaimed direct deposition.
Diffusion deposition from solid phase	– good homogeneity; – possibility of processing small sizes; – high economic efficiency; – increased coating hardness; – thickness 5–80 microns.	– limited substrate sizes; – suitable only for heat-stable materials; – excessively thin coatings; – increased brittleness of coatings.
Sputtering	– the ability to vary spraying conditions and the quality of the applied material during the process; – the possibility of obtaining thick, uniform coatings; – thickness 75–400 microns.	– quality depends on the operator's skills; – resistance of the substrate to heat and impact; – porous coatings with a rough surface and possible inclusions.
Cladding	– possibility of applying thick coatings; – processing of large areas; – thickness 5–10% of the substrate thickness.	– warping of the substrate is possible; – for hard substrates.
Electrodeposition (including chemical and electrophoresis)	– cost-effective in case of electrolyte solutions; – the possibility of applying precious metals and refractory coatings from molten salts; – the possibility of industrial production of cermets; – thickness 0.25 – 250 microns.	– special equipment for good dispersive power; – strict control of moisture and oxidation exclusion; – harmful vapors above the melt; – porous coatings and in a stressed state; – special areas of high temperatures – applicable only for some elements and types of substrates.
Hot dipping	– possibility of applying thick coatings; – coating application speed – thickness 25–130 microns.	– limited to application of A1 for high temperature coatings; – porosity and discontinuity

In addition to the considered methods for producing nanocoatings, layer-by-layer assembly and self-assembly of coatings are currently used [15–17]. The decisive factors in the formation of nanocoatings are: synthesis method, substrate material, structure of nanoparticles, grain size, thickness, microstructure.

2. ANTI-CORROSION COATINGS

Currently, both organic and inorganic coatings are widely used to protect metals from corrosion, and significant progress has been made in coating technologies. For example, the development of new "corrosion-resistant" alloys makes it possible to operate critical process equipment in highly corrosive environments under an increasingly wide range of conditions. However, despite significant advances in coating technologies, there are still some challenges in long-term protection of metals from aggressive environments. The diversity and complexity of the coating-substrate system, as well as some factors that determine the performance and durability of the coating, limit the number of highly effective and reliable anti-corrosion coating systems. The performance characteristics and service life of coatings are determined by the main components of the formulation, substrate material, preparation of the substrate surface, curing mechanism, film thickness, adhesion processes in the coating-substrate system, and environmental parameters [18–22]. To effectively protect an anti-corrosion coating, internal strength, adhesion to the substrate, and sufficient flexibility are required.

2.1. Corrosion Prevention Methods

Methods for preventing corrosion are quite varied and are discussed in detail in the Figure 1.

2.2. Anti-corrosion coatings market

The global market for protective coatings in 2018 was estimated at 26.5 billion euros, the market volume in 2023 is 9.4 million tons (Fig. 2) [23–24]. Demand is largely driven by spending in the infrastructure, power, automotive, transportation and oil and gas sectors.

The cost of paint and varnish materials for major repairs ranges from 5 to 21% of the total costs, surface preparation

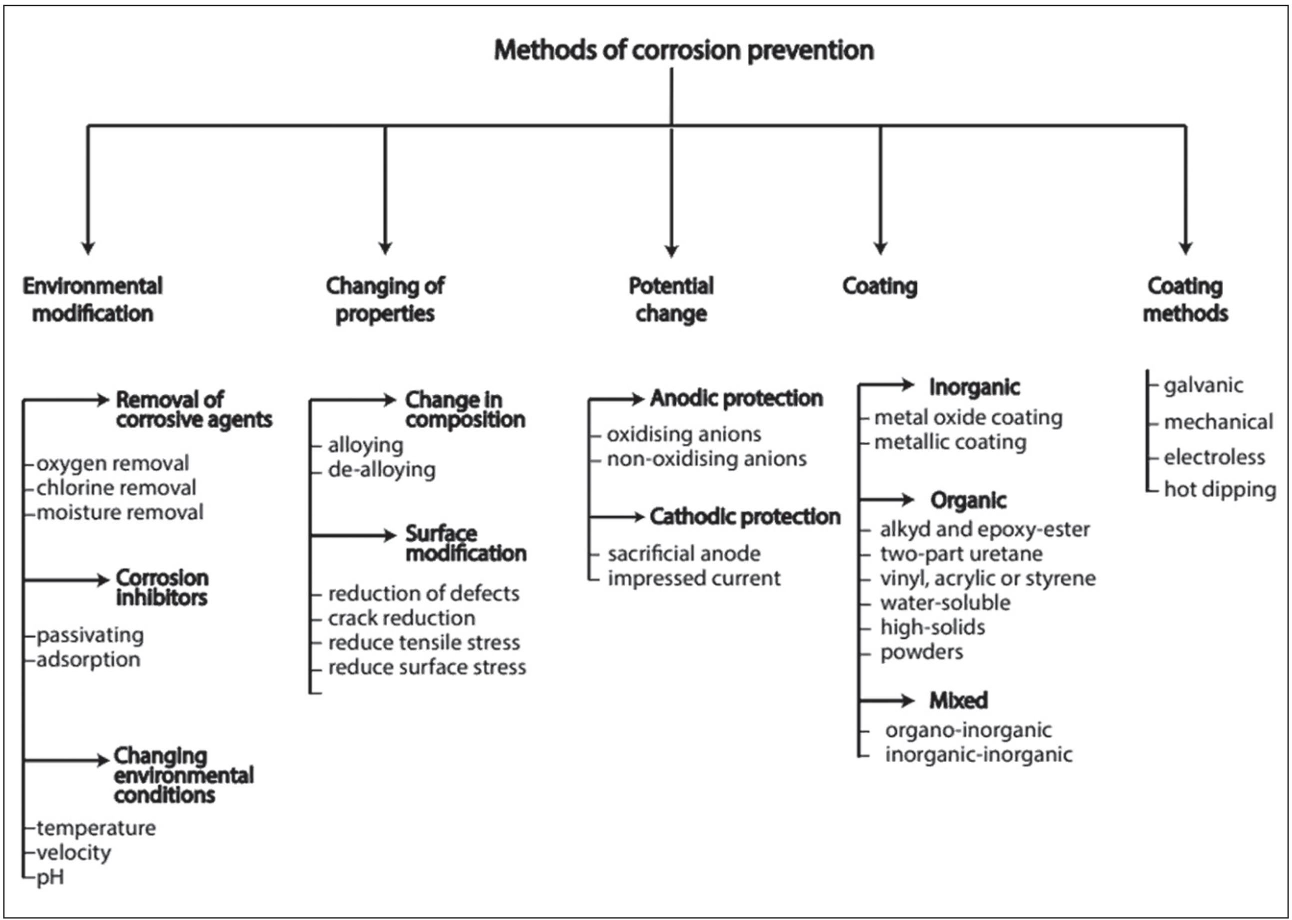

Fig. 1. **Corrosion prevention methods**

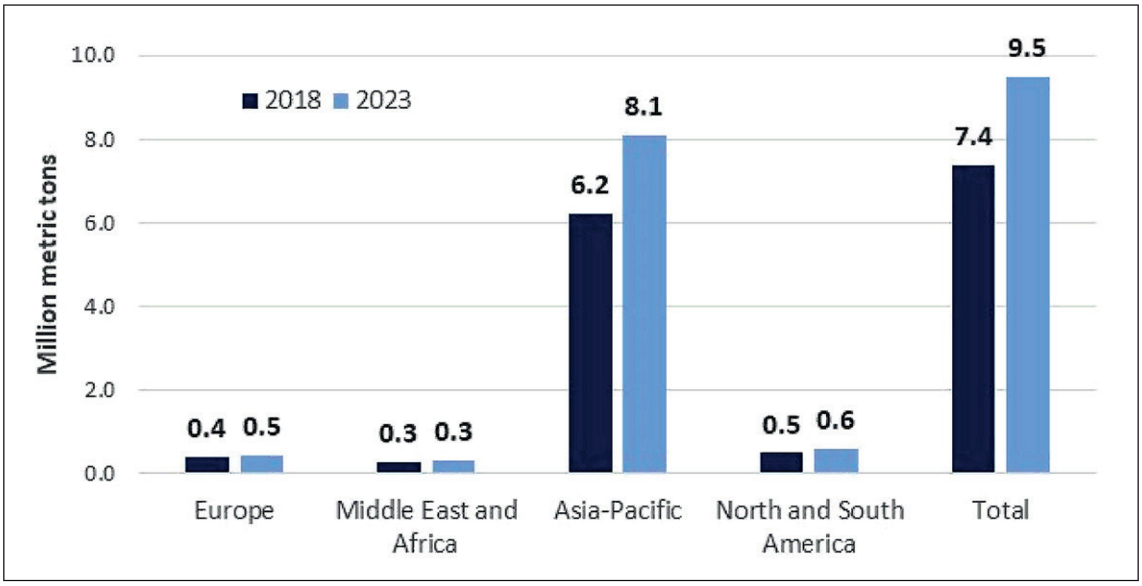

Fig. 2. **Market volume of anti-corrosion materials**

accounts for about 45% of the total amount. Although expensive, high-performance anti-corrosion coating systems such as epoxy, polyamide, urethane or zinc can provide savings of almost 40% over a 10-year service life [25].

For anti-corrosion protection of metal materials, polymer coatings are most widely used, especially in the transport and infrastructure sectors (pipelines, bridges, buildings) [26]. However, non-polymer solutions based on phosphates [27], chromates [28], and silicates [29] still remain relevant in industry for anti-corrosion protection; as well as metal coatings obtained by anodizing, galvanizing, plating, galvanic coatings, thermal spraying [30–32]. They exhibit excellent anti-corrosion properties, but they need to be replaced by more environmentally friendly technologies without the formation of sediment and the use of toxic substances. For example, the elimination of hexavalent chromium and heavy metals (lead, mercury) from coating formulations and coating processes remains an important topic for the anti-corrosion coatings sector and especially for the aerospace industry [33]. In this regard, industry and scientific organizations are developing inorganic coatings using sol-gel technologies and with pre-treatment with hybrid organic-inorganic compounds, for example, coatings such as nanoceramics using more environmentally friendly alternatives: titanium [34], zirconium [35], rare earth metals [36], silicate and molybdates [37]. However, such formulations are prepared in very dilute aqueous solutions. Large volumes of wastewater must be treated to the standards established by environmental quality standards before discharge [38].

Depending on the type of binder, organic coatings are produced based on epoxy resin, polyurethane, acrylic, alkyd or polyester formulation. When applied, polymer coatings are mainly used in the form of solutions in organic solvents. Therefore, limiting emissions of volatile organic compounds promotes the development of new formulations with low organic solvent content [39]. Several potential avenues have been identified to address this challenge: high solids coatings [40], waterborne [41], and ultraviolet (UV) light-curing coatings [42]. In order to reduce the negative impact of toxic substances in the production of polyurethane, the search and implementation of alternative substitutes remains relevant [43–46]. The authors of the article are working to reduce the negative impact of the resulting polymers in the direction of developing non-isocyanate polyurethanes. Using carbon dioxide under catalysis with tetrabutylammonium bromide, the main components for the synthesis of non-isocyanate polyurethanes were obtained – cyclocarbonates of epoxidized soybean oil and ED-20 and ED-16 resins [43]. The chosen synthesis method makes it possible to successfully obtain valuable chemical products of organic synthesis, but also to attract carbon dioxide and, thus, help reduce the carbon footprint of the resulting polymers. In addition, the implementation of the REACH (Registration, Evaluation, Authorization and Restriction of Chemicals) regulation promotes the development of greener and more sustainable solutions in the field of anti-corrosion coatings using bio-based raw materials: binder [47], solvent [48], additives [49].

2.3. Main directions of development of anti-corrosion coatings

The scope of application of anti-corrosion coatings includes:

1) anti-fouling paints, used in conjunction with anti-corrosion protective coatings as fuel-saving coatings for ships and mesh cages in aquaculture [50];

2) coatings immersed in water, used in river installations for fresh water (hydroelectric power plants); in port facilities to protect metal structures (locks, sluice gates, berths); in maritime areas;

3) buried in the soil, such as buried tanks, steel piles and pipes.

The best options for anti-corrosion coatings are produced for almost any purpose. Currently, among environmentally friendly anti-corrosion marine coating technologies, particularly important areas of development are:

1) solvent-free, high solids and low VOC coatings,

2) UV-curable coatings [51],

3) waterborne coatings,

4) based on polymer resins from plant raw materials [52] and natural compounds [53–54];

5) based on non-toxic compounds, such as non-isocyanates [55], polyurethane ureas [56], graphene [57], magnesium salts [58–60];

6) biodegradable coatings [61].

A range of low VOC anti-corrosion coatings are widely available on the market, particularly for shipbuilding and offshore applications. Epoxy resin coatings with solids contents in the range of 70–100% are more often used for primers and intermediate coats in shipbuilding [62], wind energy structures [63], and oil and gas pipelines [64]. High solids polyurethane coatings have excellent UV resistance and are preferred as weathering finishes. Subsea finishes are used to prevent biofouling, especially on ship hulls, and can minimize drag and fuel consumption [62]. Biofouling is associated with microbial-dependent corrosion [65], which is a serious problem in pipelines and submersible tanks [66]. Attachment of marine organisms also affects the performance of anti-corrosion coatings [67–69].

Solvent-free powder coating is showing rapid growth in the protective coatings market. The resulting coating is usually harder and more durable than liquid paints. Epoxy powder coatings provide excellent barrier protection, wear resistance and are characterized by high adhesion to metal surfaces, and mainly provide protection for immersed pipelines [70]. The author of the review also carried out work to reduce the hazard class of epoxy binders [71–73]. However, such high-solids, solvent-free technologies have a higher cost than traditional solvent-based technologies. Therefore, the main objective is to increase the durability of the coating system during operation, which will increase the durability of structures and reduce operating and maintenance costs.

Water-soluble paints are used to protect low-aggressive environments [74–75]. For highly aggressive environments, innovative thin-film [76], acrylic [77], zinc-containing [78] and silane [79] primers are commercially available; finishing epoxy, urethane, acrylic coatings [80]. Areas of their application: industrial (containers), transport [81], construction [82] and heavy-duty coatings (bridges, offshore and marine) [83], in places where long-term protection with a reduced impact on the environment is required.

2.4. Protective mechanism of anti-corrosion coatings

In accordance with the mechanisms of the protective action of metal products against corrosion, coatings are classified as shown in Figure 3 (Table 2).

Typically, the protective coating is not limited to one layer: a primer is applied to the metal, followed by intermediate layers, the final coating is exposed to the external environment [84].

Adhesion of the coating to the substrate is important for anti-corrosion coatings. Adhesion is an interfacial phenomenon that occurs at the interface under the influence of physical and chemical forces. Low adhesion contributes to the destruction of the coating and exposes the metal to an aggressive environment. When developing an organic coating formulation for a metal surface, adhesion processes are considered using the theory of wetting [85] and acid-base interactions [86].

Mechanical adhesion is considered in studies of large surface areas interacting with a large number of contact points. The sealant penetrates into existing surface pits and provides mechanical adhesion to the substrate. Mechanical adhesion is observed and taken into account when working with primed and porous metal surfaces.

Chemical adhesion. Interactions at the interface are provided by three types of chemical bonds: covalent or ionic bonds with energies from 40 to 400 kJ/mol; dispersion forces, or dipole interactions or van der Waals forces with energy from 4 to 8 kJ/mol; hydrogen bonds with binding energies from 8 to 35 kJ/mol.

It is generally accepted that the adhesion of polymer coatings to metal surfaces is explained by the formation of hydrogen bonds (Fig. 4).

The top layer of metal surfaces is usually a thin oxide layer [87]. The adhesion of the polymer coating and the metal occurs due to the formation of hydrogen bonds. Therefore, binders with polar groups exhibit excellent adhesive properties. The adhesion of epoxy resins to steel directly depends on the number of OH groups, but this type of adhesion is not the main one and is common for binders such as zinc silicates and epoxy resins [88]. The proposed adhesion mechanism is not able to fully explain the different bond strengths between resins and different metals, and the abnormal adhesion of epoxy resins in comparison with other polymers with an equal number of hydroxyl groups. In the course of research, it was discovered that epoxy compounds dissociate through the bond between phenoxy oxygen and aliphatic carbon (Fig. 5) [89].

Previously, according to the proposed mechanism, it was believed that adhesive interactions are carried out

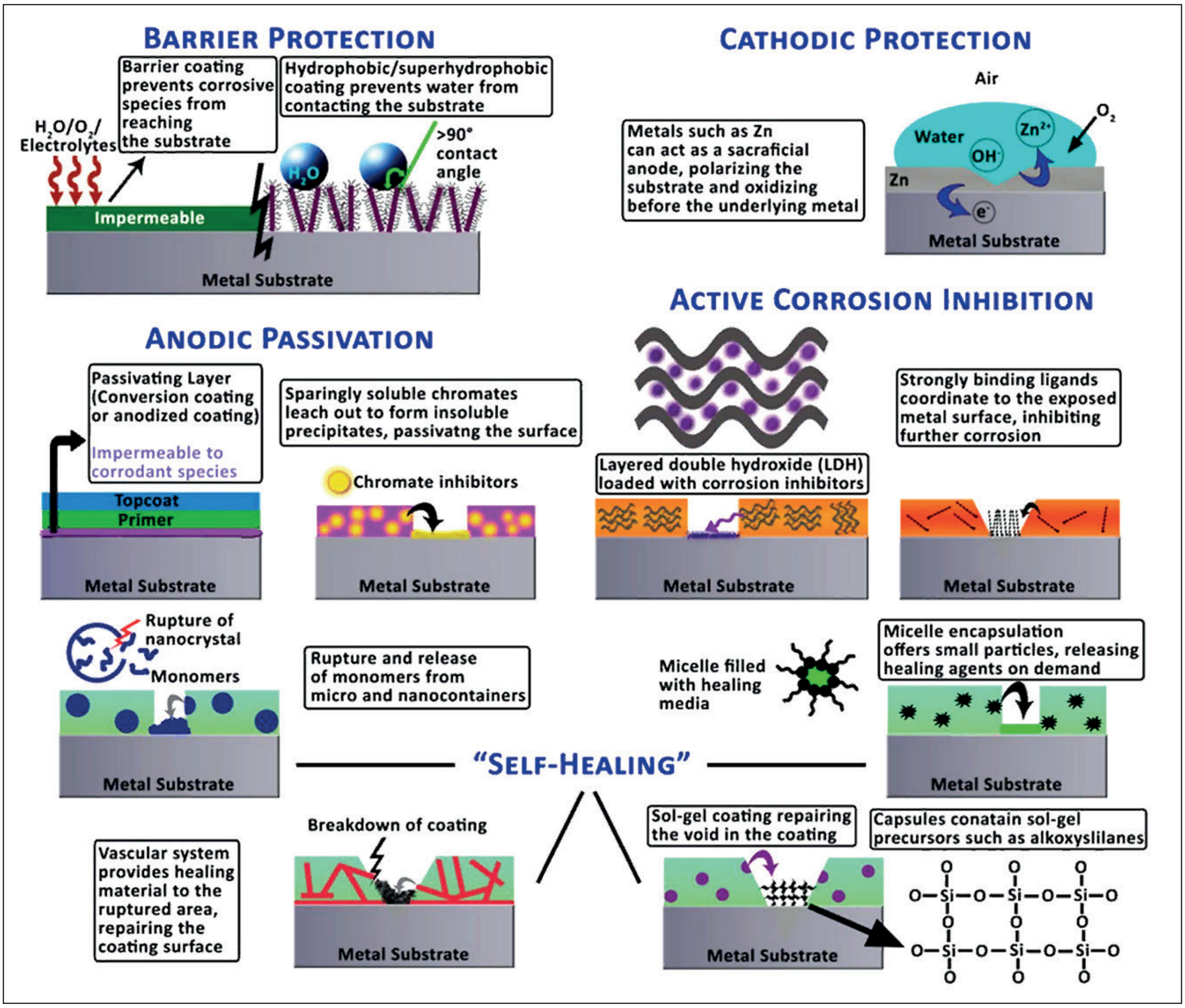

Fig. 3. **Corrosion protection methods**

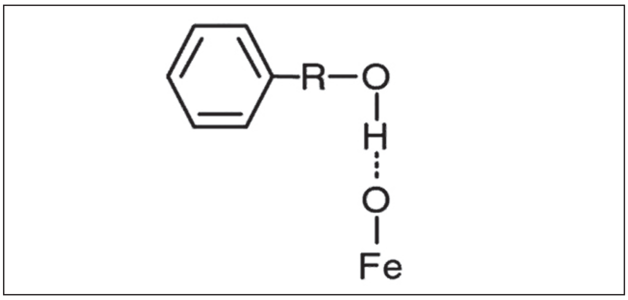

Fig. 4. **Formation of hydrogen bonds between the iron oxide layer on steel and epoxy resin**

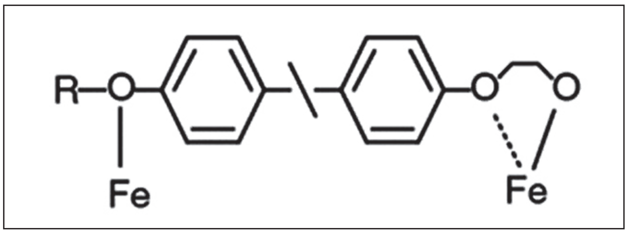

Fig. 5. **Adhesion between epoxy and steel**

through the oxygen atoms of the phenoxy and OH groups, while the epoxy resin molecule is oriented on the water surface in a certain way: the hydrophobic part of the epoxy resin molecule is directed in the direction opposite to the interface [90].

However, regardless of the mechanism of adhesion of the polymer coating to the metal, stronger adhesion can be achieved by pre-treatment using chromatization or phosphating methods. The conversion layer on the metal surface passivates the surface and acts as a barrier to aggressive substances [91]. Contaminants on metal surfaces (dust, grease, oxides, salts and remnants of old coatings) negatively affect the adhesion of coatings. This reduces

Table 2
Comparative characteristics of protective coatings

Coverage type	Areas of use	Advantages of coatings	Disadvantages of coatings
Barrier	Dive, marine, industry	– reduced permeability to liquids, ions, gases, – strength and abrasion resistance, – UV resistance, – used together with cathodic current protection or sacrificial anodes, – applied as a primer, intermediate or topcoat	– the possibility of incomplete removal of the solvent, – aluminum pigment reacts in acidic environments to release hydrogen, – in environments with high humidity, the likelihood of galvanic corrosion of coatings with aluminum-containing pigments applied to a zinc-containing primer, – in splash areas and atmospheric environments, defective coatings are not reliable enough, – the likelihood of rapid migration of aggressive particles to the substrate in case of incorrect orientation of lamellar pigments
Sacrificial organic	Splash zone, marine, industry	High anti-corrosion protective effect due to cathodic protection of the metal substrate	– high costs, – low adhesion and cohesion due to high metal content, – high zinc content coatings are not recommended for dipping or application in conjunction with impression current cathodic protection
Sacrificial inorganic	Splash zone, marine, industry	Zinc silicates are resistant to heat and immersion in water with chemical solutions	– zinc silicates require special conditions for curing and are characterized by low compatibility with other types of coatings, – for proper protection the need for electrical contact between metal particles
Inhibitory	industry	– the formation of a water-insoluble passivation layer that persists or remains insoluble in most environments, – lower costs compared to zinc pigmented coatings	– not applicable for submersible structures, – coatings must be semi-permeable to water to effectively inhibit the substrate, – risk of increased corrosion rate when adding insufficient anodic inhibitor

the mechanical and chemical adhesion of the coating to the base, and also increases the risk of destruction of the coating. Most water-soluble contaminants must be removed by treating with a detergent solution [92]. Next, abrasive blasting is used to prepare the surface and more thoroughly remove rust, mill scale and remnants of the previous coating before applying protective painting. In this case, the surface area of the base increases in order to improve adhesion [93]. The need to remove water-soluble contaminants is obvious, since they can dissolve when moisture penetrates into the coating and form swelling, delamination and accelerate surface corrosion processes [94–97].

Improving adhesion between a polymer coating and a metal surface can be achieved by using adhesion promoters – binding agents that form covalent bonds, providing stronger interfacial interaction between the substrate and the coating. Their role in enhancing the adhesion of coatings to metal surfaces has been thoroughly studied [97]. Adhesion promoters are basically short-chain organic molecules with different end groups: on the one hand, an organic functional group compatible with the coating material, on the other, an inorganic group compatible with the substrate. A kind of chemical bridge is formed between the base and the coating. Modified silanes are the most common commercial adhesion promoter between polymeric and inorganic materials [98]. They are introduced into the coating composition or applied directly to the substrate. Adhesion promoters are thought to migrate to the interface region and attach to the substrate or pigments before extensive curing occurs. Adhesion promoting molecules are known to be oriented perpendicular to the surface to which they are attached. Thus, the rough surface prevents the formation of an ordered adjacent layer. However, continuous thin layers are still capable of forming strong and durable adhesive joints [99].

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