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CHEMISORPTION-TYPE AMIDE-BASED CORROSION INHIBITORS: A REVIEW[1]

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Corrosion, a pervasive phenomenon in both natural environments and industrial processes, poses significant safety hazards and incurs substantial economic losses. Consequently, the study of corrosion mechanisms and anti-corrosion strategies has become a focal point of research. The article discusses metal corrosion inhibitors based on organic amides The article examines the features of the chemical structure and the presumed mechanism of anticorrosive action on metal surfaces of corrosion inhibitors such as imidazoline, its quaternary ammonium salts, polyamide corrosion inhibitors, and compounds with heterocyclic nitrogen-containing structures. It has been shown, that a promising direction in the development of technology for obtaining metal corrosion inhibitors based on amides is their modification with naturally occurring products.

Keywords: *corrosion, corrosion inhibitor, amide, imidazoline, method of preparation, natural modifiers.*

Introduction. Metal materials are extensively utilized in the construction, petroleum, metallurgical, and aerospace industries as a result of their excellent physical-chemical properties and high ductility [1]. Metals are highly active, resulting in an oxide film on the metal surface under the action of an aerobic environment. Metal corrosion is the process by which the action of the surrounding media damages metal materials. Corrosion is a spontaneous chemical reaction that can be divided into chemical corrosion and electrochemical corrosion, according to the reaction mechanism. Whether chemical corrosion or electrochemical corrosion occurs, metallic materials and chemical substances in the environment, such as acids and alkalis, are subject to redox reactions. The oxidation-reduction reactions of metals become stronger over time, and eventually, metals lose their original capacity. Every year, about a quarter of the world's metal products are scrapped due to corrosion. The loss caused by metal corrosion accounts for 30% of its annual output. In industrialized countries, the loss caused by corrosion accounts for about one tenth of the national income, and the maintenance and replacement costs of equipment and pipelines are far more than the cost of materials [2]. Therefore, the study of corrosion and anti-corrosion has important economic significance.

The second important aspect of studying corrosion and corrosion resistance is based on safety. Corrosion has caused many serious casualties, and many accidents involving cars, ships, and airplanes are more or less related to corrosion.

The third area of the importance of studying corrosion and corrosion-proofing is the conservation of resources and the protection of the environment. Because of corrosion, a lot of precious earth resources are wasted. It has been estimated that the world's metal resources are becoming increasingly depleted, and even with an optimistic estimate of 10 times the current reserves plus 50% recycling, the years of maintenance will not be long. The waste of materials is also a waste of energy, because the extraction of metals from ore consumes a lot of energy [3].

Studying corrosion, mastering corrosion, and being expert in anti-corrosion can effectively reduce economic losses, powerfully prevent disasters, and save resources and energy further.

There are many kinds of corrosion inhibitors, which can be classified in different ways. From the perspective of composition and structure, corrosion inhibitors can be divided into organic and inorganic categories. Organic compounds used as inhibitors, occasionally, they act as cathodic, anodic, or together, as cathodic and anodic inhibitors; nevertheless, as a general rule, they act through a process of surface adsorption, designated as a film-forming process. Naturally, the occurrence of molecules exhibiting a strong affinity for metal surfaces compounds showing good inhibition efficiency and low environmental risk. These inhibitors build up a protective hydrophobic film adsorbed molecules on the metal surface, which provides a barrier to the dissolution of the metal in the electrolyte. They must be soluble or dispersible in the medium surrounding the metal [4].

Organic corrosion inhibitors can be classified into nitrogen-containing corrosion inhibitors, sulfur-containing corrosion inhibitors, oxygen-containing corrosion inhibitors, and other types. Nitrogen-containing organic corrosion inhibitors show excellent corrosion inhibition activity because there is an unshared electron pair on the nitrogen atom, so the research on nitrogen-containing organic corrosion inhibitors is in full swing.

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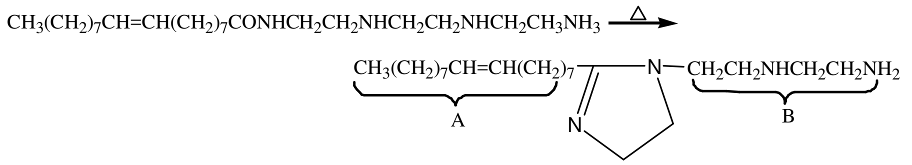
Main part. The use of corrosion inhibitors is an important means of corrosion protection. It plays an important role in the production, storage, and transportation of oil and gas. Amide-based corrosion inhibitors are widely used because of their superior performance. They can be classified into many types according to their molecular structure and active groups, and each type has unique structural characteristics and corrosion inhibition properties. Understanding these molecular structural characteristics is crucial for the rational design of high-performance corrosion inhibitors.

Imidazoline corrosion inhibitors. Imidazoline and its derivatives are the most widely used corrosion inhibitors in oil and gas fields, accounting for about 90% of the total corrosion inhibitors [5]. These kinds of corrosion inhibitors are characterized by non-irritating odor, low toxicity, good thermal stability, and easy biodegradation. They can effectively inhibit the overall corrosion and local corrosion of metal in the corrosion environment. The nitrogen atom in the imidazolin ring of its molecular structure, along with adjacent functional groups such as hydroxyl and amino-group, collectively form multiple adsorption centers, which enhance their binding capacity to the metal surface.

Imidazoline corrosion inhibitors are generally formed by the condensation of fatty carboxylic acid and polyamine. First, the fatty carboxylic acid reacts with one of the amino groups in the polyamine and forms a salt. Then, this salt will lose a molecule of water and form an amide when the temperature is above 100°C. At last, the other nitrogen atoms in the polyamine undergo nucleophilic addition reaction to the carbonyl group in the amide group when the temperature rises continuously, and generate nitrogen-containing heterocyclic compounds, that is, substituted imidazoline derivatives. Their structure is related to the structure of fatty groups in carboxylic acids and polyamines. The substituent groups play an important role in determining the corrosion inhibition effect of corrosion inhibitors [6]. Taking oleic acid and triethyltetramine as an example, the reaction process can be expressed as follows:

$$\underbrace{CH_3(CH_2)_7CH{=}CH(CH_2)_7}_{A}COOH + H_2NCH_2CH_2NH\overbrace{CH_2CH_2NHCH_2CH_2NH_2}^{B} \longrightarrow CH_3(CH_2)_7CH{=}CH(CH_2)_7COO\overset{+}{N}H_2CHCH_2NHCH_2CH_2NHCH_2CH_3NH_2 \quad (1)$$

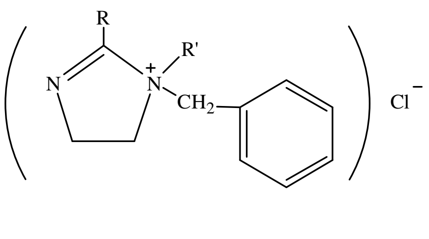
$$CH_3(CH_2)_7CH{=}CH(CH_2)_7COO\overset{+}{N}H_2CHCH_2NHCH_2CH_2NHCH_2CH_3NH_2 \xrightarrow{\triangle} CH_3(CH_2)_7CH{=}CH(CH_2)_7CONHCH_2CH_2NHCH_2CH_2NHCH_2CH_3NH_3 \quad (2)$$



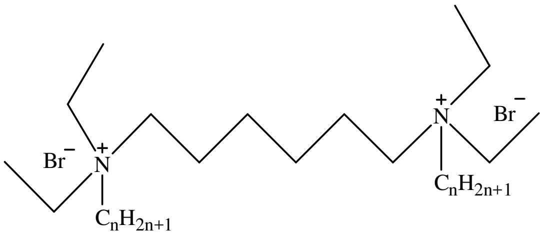
(3)

In the example reaction, the product obtained is a derivative of substituted imidazoline. The structure of the substituted imidazoline is influenced by group A in carboxylic acid and group B in polyamines. While group A does not participate in adsorption, its length and structure significantly influence the molecule's water solubility. Group B also contains a nitrogen atom, which not only has an unshared electron pair that can participate in adsorption but also serves as an active center for substitution or nucleophilic addition reactions. A functional group with strong adsorption activity can be introduced onto the nitrogen atom of group B, thereby forming a stronger and more stable adsorption effect with the metal surface.

Quaternary ammonium salt corrosion inhibitors. An imidazoline quaternary ammonium salt is formed by quaternization modification treatment through changing the valence of the nitrogen atom on the imidazoline ring to five valence. Imidazolinium quaternary ammonium salts exhibit superior corrosion inhibition performance compared to unmodified imidazoline due to the stronger adsorption of N+ ions on metal surfaces [6]. The quaternary ammonium cation can be strongly adsorbed by the negatively charged metal surface. They are arranged on the metal surface just like they are making the metal surface positively charged, and this makes it difficult for hydrogen ions to approach the metal surface, enhancing isolation, obstructing charge transfer, and decreasing cathodic reaction rate significantly [7]. The basic structure of imidazoline quaternary ammonium salt corrosion inhibitors is shown below [8]:



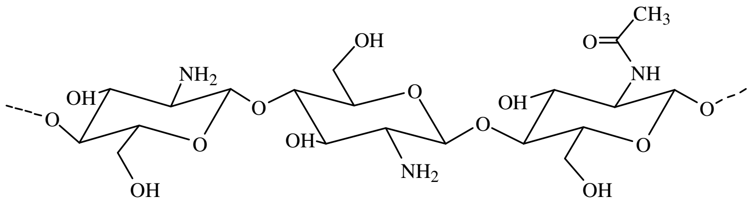
Quaternary ammonium corrosion inhibitors can also be composed of positively charged nitrogen atoms and amphiphilic structures without imidazoline:



They still exhibit relatively good corrosion inhibition effects. In addition, a new type of twin quaternary ammonium salt contains two hydrophilic head groups and two hydrophobic tail groups, connected by the head groups or the spacer groups connected to the head groups. The spacer may have different structures, rigid or flexible, hydrophobic or hydrophilic. The neutral charge of the molecule is maintained by the presence of organic or inorganic anions. Twin quaternary ammonium salts exhibit an exceptionally low critical micelle concentration (cmc), hundreds of times lower than that of their corresponding monomeric surfactants. Twin quaternary ammonium salts have a larger molecular area than monomeric analogues, which makes them more effective as corrosion inhibitors [9].

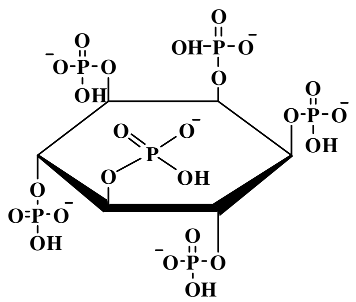
Natural product modified amide corrosion inhibitors. According to the chemical structure, the commonly used corrosion inhibitors include inorganic corrosion inhibitors and synthetic organic corrosion inhibitors. These two kinds of corrosion inhibitors are easy to synthesize and apply, and have high corrosion inhibition efficiency at low concentration [10]. But they are highly toxic, not biodegradable, and they will cause potential pollution to the environment. For example, phosphate corrosion inhibitors can lead to eutrophication of water bodies, and this disadvantage greatly limits their development and application prospects [11]. In recent years, global attention has increasingly focused on environmental protection and green development, and researchers have started to explore the anti-corrosion effects of plant and natural product extracts [12]. Plant and natural product corrosion inhibitors are naturally effective components extracted from natural products. Compared with traditional metal corrosion inhibitors, they have the characteristics of being easy to obtain, low cost, green, and environmentally friendly, and have a broad development prospect. Using common analytical methods such as high-performance liquid chromatography (HPLC), Fourier transform infrared spectroscopy (FTIR), gas chromatography-mass spectrometry (GC-MS), and ultraviolet spectroscopy (UV), it can be known that these natural products are rich in triterpenoid saponins, Gynostemma pentaphyllum alkaloids, flavonoids, and other bioactive components [13]. These compounds contain polar groups centered on nitrogen (N) and oxygen (O), such as -O-H, -C=O, -N-H, -C-O, benzene rings, and aromatic heterocycles, which possess corrosion-resistant functional groups [14]. For example, garlic contains allyl propyl disulphide, mustard seeds contain an alkaloid berberine, which has a long chain of aromatic rings and an N atom in the ring, and, at several places, carrot contains pyrrolidine and castor seed contains the alkaloid ricinine. Eucalyptus oil contains monomtrene-1,8-cineole. Lawsonia extract contains 2-hydroxy-1,4-naphthoquinone resin and tannin, coumarin, gallic acid, and sterols. Gum exudate contains hexuronic acid, neutral sugar residues, volatile monoterpenes, canaric and related triterpene acids, and reducing and nonreducing sugars. Garcinia kola seed contains primary and secondary amines, unsaturated fatty acids, and bioflavonoids. Calyx extract contains ascorbic acid, amino acids, flavonoids, pigments, and carotene. Many of these active ingredients have been used as corrosion inhibitors [15].

Chitosan is a linear polysaccharide of randomly distributed N-acetyl-D-glucosamine and D-glucosamine connected together by a β-1,4-glycosidic linkage:



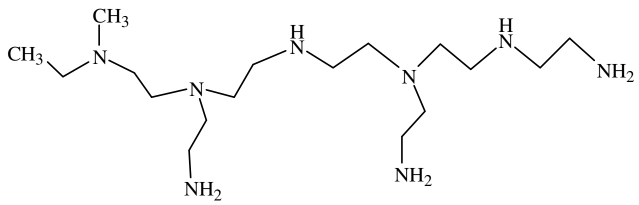
N-Acetyl-D-glucosamine and D-glucosamine are called acetylated and deacetylated units, respectively. Generally, the synthesis of chitosan is achieved by deacetylation of chitin. Chitin is primarily derived from the shells of crustaceans such as shrimp and crabs, the cuticle of insects, and the cell walls of fungi like mushrooms. The anticorrosive effect of chitosan for metals and alloys is widely reported. Because of their natural origin, chitosan-based corrosion inhibitors are considered friendly. Generally, chitosan modified with various organic compounds exhibits a higher corrosion inhibition efficiency than pure chitosan [16].

Phytic acid, which is the hexaphosphate of inositol, is a kind of organic phosphorus compound that exists naturally and widely in the seeds and rhizomes of higher plants:

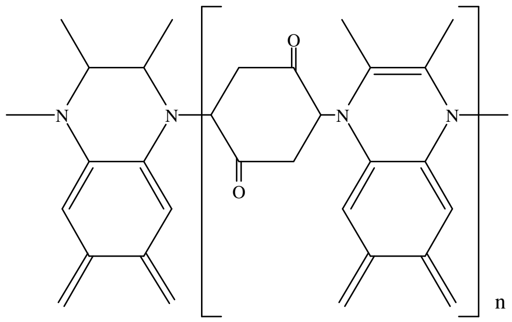


It has the characteristic of chelating metal ions, and it can be used as a corrosion inhibitor. Six phosphate anions linked to inositol can accept or donate up to 12 protons[17], so that it can effectively shield H+ from metal in the corrosion environment to achieve the effect of corrosion protection. The mixture of phytic acid 13～15g/L, sodium benzoate 11～13g/L, sodium gluconate 11～13g/L, and sodium citrate 3～5g/L was prepared and used as a corrosion inhibitor. This corrosion inhibition rate on carbon steel can reach 95%. [18]

Polymer corrosion inhibitors. Polyethyleneimine (PFI) is one of the earliest discovered organic polymers with notable corrosion inhibition properties. The number of methylene groups in its molecular structure influences the bond angles of the -C-N-C- bonds, thereby affecting its corrosion inhibition capability [19, 20]:

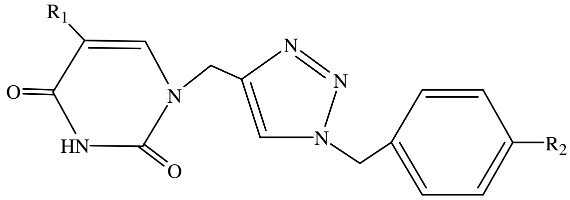


Polyvinylpyrrolidone (PVP) also exhibits excellent corrosion inhibition properties, effectively suppressing the corrosion of aluminum in hydrochloric acid, iron in sulfuric acid, and copper in nitric acid. Polyamine-based benzochinone polymers can be synthesized through the homopolymerization of diamines, triamine, and p-benzoquinone(PAQ). PAQ is a hybrid corrosion inhibitor that significantly reduces the surface penetration current of steel in acidic solutions. It can form a protective film on metal surfaces, with its effectiveness increasing over time. PAQ is suitable for industrial acid washing and secondary oil recovery processes [19]. The structure is as follows:



Other amide corrosion inhibitors. In recent years, many novel corrosion inhibitors have been designed and synthesized by researchers. These inhibitors exhibit excellent corrosion inhibition effects, but their structures are complex, and their synthesis routes are intricate.

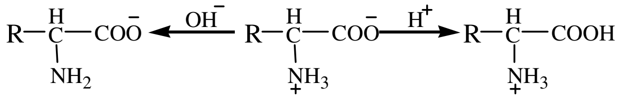
1,2,3-triazoles are present in a number of compounds with assorted biological activities such as anticancer, antibacterial, antifungal, anti-tubercular, and anti-HIV properties. In the last years, the corrosion inhibition of steel in acid solutions by nitrogen-containing heterocyclic compounds has been extensively studied. In this regard, 1,2,3-triazole derivatives are considered to be effective acidic corrosion inhibitors:



New mono-1,2,3-triazole pyrimidine nucleobases were synthesized by copper(I)-catalyzed 1,3-dipolar cycloaddition reactions between N-1-propargyluracil and thymine, sodium azide, and benzyl halides. This compound was investigated as a corrosion inhibitor for steel in 1 M HCl solution, using the electrochemical impedance spectroscopy (EIS) technique. The results indicate that these heterocyclic compounds are promising acidic corrosion inhibitors for steel [21].

Polyamides, which are based on tartaric acid as well as thiourea, are considered promising corrosion inhibitors, with efficiency up to 90.6 % at 250 wppm. Factors affecting the inhibition mechanism are: (i) polyamide with 3 categories of high electron density atoms which are active centers and they donates lone pairs of electrons, hence, polyamide molecules show easy adsorption on the metal surface; (ii) these polyamides show easy production in pure state; and (iii) the polymeric compounds with comparatively high molecular weight are more inhibition efficient than the monomeric compounds [22].

Amino acids have become the research object of metal corrosion because they are non-toxic, non-polluting, and strong corrosion inhibitors. There are approximately 200 types of amino acids in nature, all with the amino and carboxyl groups attached to the same carbon. In neutral solutions, amino acids exist as zwitterions. When the pH of the solution changes, the molecular form of amino acids undergoes corresponding transformations as follows [23]:



In neutral and acidic solutions, amino acid molecules adsorb onto the electrode surface via +NH_3 (in the cathodic region of a galvanic cell), slowing the cathodic reaction rate and consequently reducing the anodic reaction rate (metal dissolution). In alkaline solutions, amino acid molecules adsorb on the metal surface, suppressing corrosion active sites, decreasing exposed metal surface area, increasing the corrosion potential, and reducing the corrosion current density. Under normal circumstances, the adsorption of amino acid molecules on the metal surface is divided into two steps: first, the amino acid molecules replace the water molecules on the metal surface, and then the amino acid molecules combine with the metal ions formed on the surface to form a metal-corrosion inhibitor clathrate [24].

Conclusion. With the attention of human beings to the living environment, the harmlessness of production and life has become a hot topic. Therefore, the production process of corrosion inhibitors and the inhibitors themselves also need to be harmless. In other words, the corrosion inhibitor products themselves are low-toxic and harmless, unpolluting the environment. The production process should not use toxic raw materials and should not discharge by-products that pollute the environment. The whole process is environment friendly and is not harmful to people or living things. Making full use of natural products and rational utilization of industrial wastes is an important way. Another crucial point is to achieve an outstanding product with high effectiveness and considerable economic benefits. A promising direction for the development of technology for obtaining metal corrosion inhibitors based on amides is their modification with products of natural origin.

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ИНГИБИТОРЫ КОРРОЗИИ ХЕМОСОРБЦИОННОГО ТИПА НА ОСНОВЕ АМИДОВ (ОБЗОР)

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Коррозия, широко распространенное явление как в природных средах, так и в промышленных процессах, представляет значительную угрозу безопасности и влечет за собой существенные экономические потери. В связи с этим изучение механизмов коррозии и способов борьбы с ней является актуальной задачей. В статье приведен обзор современных ингибиторов коррозии металлов на основе органических амидов, таких как имидазолин, его четвертичные аммониевые соли, полиамидные ингибиторы коррозии и соединения с гетероциклическими азотсодержащими структурами. Рассмотрены особенности их химической структуры и предполагаемый механизм антикоррозионного действия на металлические поверхности. Показано, что перспективным направлением в разработке технологии получения ингибиторов коррозии металлов на основе амидов является их модифицирование продуктами природного происхождения.

Ключевые слова: *коррозия, ингибитор коррозии, амид, имидазолин, способ получения, природные модификаторы.*