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Cellulose and its Derivatives: Towards Green Inhibitors of Metal Corrosion

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Abstract

Corrosion is a natural process motivated by energy considerations that create a critical problem that affects the industry, while inhibition is a defense against corrosive attacks on metal. To remove the oxide from the metal surface, mineral acids like HCl and H₂SO₄are mostcommonlyused in pickling baths. As a result, many studies have been done to create inhibitors that will lessen the effects of corrosion. Recently researchers have focused on discovering green inhibitors as replacements for conventional inhibitors to avoid toxicity. Among other options, biopolymers and carbohydrate polymers can effectively mitigate the corrosion process of alloys/metals. This review will describe the meaning of corrosion, followed by a recent survey on cellulose, its derivatives, and different techniques for evaluating corrosion inhibitors. Then, we describe an overview of applications as corrosion inhibitors for metal corrosion. Also, we conclude with perspectives on recommendations and challenges for future developments.

Keywords; Cellulose, Cellulose Derivatives, Green Environment, Corrosion Inhibition.

1. Introduction

Corrosionis the process through which materials (often metals) gradually deteriorate due chemical to or electrochemical reactions with their surroundings, which can occur at any time or point[1].Metal corrosion happens during the extraction of petroleum, the manufacturing of chemicals, farming, and drilling, and it is a significant public health risk[2].Corrosion results from the oxidation/ reduction reactions of metal with its environment; the oxidation reaction is the reaction that produces electrons, while the

reductionreaction is between two elements that bind the electrons.For example, copper dissociated in HCl by forming cuprous chloride on the copper surface, then cuprous chloride turns into copper chloride complex (soluble), and this complex further oxidizes to cupric ions [3].

$$Cu \rightarrow Cu^{+} + e^{-}$$

$$Cu^{+} + Cl^{-} \rightarrow CuCl$$

$$CuCl + Cl^{-} \rightarrow CuCl_{2}$$

$$CuCl_{2}^{-}(surfase) \rightarrow CuCl_{2}^{-}(solution)$$

$$CuCl_{2}(surfase) \rightarrow Cu^{+}_{2} + 2Cl^{-} + e^{-}$$

In general, metals play a significant role in business on a global scale because of their numerous qualities. But they undergo natural corrosion, which changes them into more stable oxides [4]. There are different methods for the protecting of metals from the corrosion such as using inhibitor [5], coating [6], alloying [7], etc. Due to its simplicity of use and efficiency, even in very small amounts, in a highly corrosive environment, the employment of inhibitors has been recognized as a practical technique to suppress corrosion[8].By passivating the metal's surface through creatinga shield of oxide against the corrosive environment, inhibitors can lessen corrosion. Additionally, inhibitors can engage with surfaces chemically, physically, or by a combination of these interactions to block the active sites and lower the rates of anodic, cathodic, or both types of reactions. In the physical interactions, the inhibitor binds to the surface with electrostatic forces and behaves loosely as temperature rises. But in the chemical interactions, charge-sharing and

transfer processes can be carried out between the inhibitor and the surface, so they are more powerful than physical interactions[9, 10].For example, chromates improve the substrate's ability to resist corrosion, but their use has been constrained due to their toxicity and the associated risk to human health[11]. The most well-known inhibitors heterocyclic are organic compounds containing heteroatoms like O, S, N, P, and/or delocalized electrons [12, 13]. The reaction hub for starting the adsorption process is typically considered the polar functional group. It has been found that organic molecules with nitrogen atoms are effective metal inhibitors in various hostile media[14].

Polymers are favored as inhibitors over simple organic compounds because they have numerous reaction centers that aid in forming complexes with metal ions[15]. In general, the presence of heteroatoms and cyclic rings is connected to the inhibitory capability of polymers. Sadly, synthetic polymers are frequently nonrenewable and nonbiodegradable. Therefore, the chosen molecule's availability. toxicity, and environmental friendliness are vital factors when estimating the expenses of the industrial and large-scale use of inhibitors. As a result, research on inhibitors has concentrated on using biopolymers. Biopolymers derived from plants have been developed as eco-friendly inhibitors due to their renewable, biodegradable, affordable, properties. and nontoxic However. biopolymers are rarely employed directly because of their high impurity content, the vast volume required for addition, the

20].Biopolymerscontain

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irrelatively unstable performance, and the ease of microbial reproduction in their presence. Biopolymers can be altered to get around these drawbacks, though. The most prevalent biopolymer, cellulose, is abundant in nature and can be used as an inhibitor[16, 17].

On the Market, different corrosion inhibitors are available, such as polymer stainless steel/copper corrosion inhibitor WDZ-02, carbon steel copper corrosion inhibitor BWF-XZ530, organic corrosion inhibitor LS-01, and organic carbon steel copper. However, unlike conventional corrosion inhibitors, which can be used in industrial production, there are no naturally occurring corrosion inhibitors. In most factories, harmful corrosion inhibitors are preferred inhibitors. However, natural in the laboratory, some natural inhibitors show a higher corrosion inhibition efficiency of over 97% compared to commercial corrosion inhibitors. But these findings have yet to be independently validated in the industry. This could result from sophisticated extraction methods, poor solubility, challenging material raw acquisition, and sizable а amount. Undoubtedly, cellulose may not perform higher corrosion inhibition in industrial applications than in the laboratory, which is still faced with significant challenges[18].

Therefore, in recent years scientific research hasmotivated eco-friendly inhibitors, called green inhibitorsin line with greenchemistry like, biopolymers.Biopolymers are typically regarded as rich sources of green corrosion inhibitors due to their moderate performance, low cost, low toxicity, and environmental friendliness [19, π bonds, and aromatic heterocycles, which are the main active parts for good corrosion inhibition. These atoms can act as adsorption centers (donating electrons) and are linked to the metal surface (accepting electrons) creating a bond and suppressing the corrosion[21, 22].Researchers also used biopolymers as a novel means of increasing the corrosion inhibition of metals, whether as corrosion inhibitors [23]or coatings [24], by utilizing nanotechnology.Due to the heteroatoms in the biopolymer's large molecules, they can exhibit adsorption capability, hence hindering the corrosion process [25]. For example, Jmiai et al. employed sodium alginate and chitosan to inhibit copper's corrosion[26].A combination of cellulose and ligninwas employed as a corrosion inhibitor for the steel surface and followed the Langmuir isotherm with 92% efficiency[21]. In another study, locust bean gum had 89.8% efficiency as a corrosion inhibitor for the O235 steel surface in an acidic solution [26]. Among biopolymers, polysaccharides and their derivatives include many active adsorption sites, which improve their ability to stop corrosion, they have garnered much interest. In addition, they contain many adsorption centers that can strongly bond with metal ions achieving high inhibition efficiency. But the results suggest that their inhibition efficiency on metals could not be better than expected [4, 27, 28]. Cellulose is more prevalent in plants and is a common polysaccharide. The two most significant characteristicshigh vield and accessibilitymust satisfy the requirements for industrial corrosion inhibitors.

Researchers have also discovered that cellulose exhibits a good inhibitory effect on a variety of metals in corrosive conditions [29]. So, it isconducive to a green environment for corrosion inhibitors. Therefore, recent studies on using cellulose and its derivatives as metalcorrosion inhibitors are comprehensively reviewed here.

2. Cellulose

It is a carbohydrate biopolymer in plant tissues, and in comparison with other biopolymers, it exists in the highest amount. It is composed of glucose units, and each unit contains three -OH groups. The cellulose molecule has a linear structure and a 3000-5000 glucose unit linked by β -1, 4glycosidic link and is a semi-crystalline material[30]. The degree of cellulose depends its crystallinity on origin. pretreatment, and extraction method. For example, the crystallinity ranges from 40 to 60% for plant-based and wood-based cellulose. while bacteria and tunicin cellulose range from 80 to 100%crystallinity. In addition, crystalline cellulose has different allomorphs, such as I, II, III, and IV. Cellulose I is natural and can be derived from plants, trees, bacteria, algae, and tunicates. It forms sheets stacked together by Van der Waals and hydrogen bond interactions and is composed of triclinic (I_{α}) and monoclinic (I_{β}) metastable structures. The $I\alpha/I\beta$ ratio depends on the source of cellulose. I_{α} can be converted into I_{β} through hydrothermal treatment in an alkaline solution at 260 °C. Cellulose I can be converted into II by regeneration (dissolution recrystallization) and or

mercerization (aqueous sodium hydroxide treatment), during which the parallel chain changes into a more stable antiparallel chain arrangement. Cellulose I or II can be converted into cellulose III through liquid ammonia treatment. Finally, cellulose IV can be obtained from cellulose III by thermal treatments [31, 32]. All conversions are summarized in **Figure 1**.Also, cellulose is classified based on its source, as wood-based, plant-based, bacteria-based, algae-based, and tunicate-based cellulose[33].

Cellulose can undergo a wide range of chemical modifications at the OH groups and the C atoms[34]. Various cellulose derivatives have been developed and utilized as corrosion inhibitors to upgrade the value or expand the versatility of cellulose by using chemical treatment or functionalization. The functionalization pattern along the polymer chain and the type and degree of substitution determines the cellulose derivatives properties. The most reactive groups on the cellulose chain are the hydroxyl groups which are relatively poor nucleophiles that lead to the for harsh requirement reactions[35].Cellulose derivatives, based on the response of the hydroxyl groups, include etherification and esterification (Figure 3), varying in solubility, water interaction, moisture sorption, surface activity, and chemical structure. The etherification of cellulose can be whole or in part by using different reagents, epoxides. e.g., halogenated carboxylic acids. and halogenoalkanes. Furthermore, the solubility of cellulose ethers is affected by the pH of the solution as in acidic, it dissolves very

alkalinesolutions, slowly, while in it dissolves rapidly. addition. In water solubility depends on the degree of substitution, water-soluble and most substances have 0.4–2 a degree of substitution[32].

The isolated cellulose can be classified into fibers, filaments, crystals, and micro/nanofibrils (**Figure 2**).

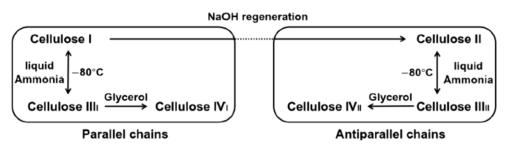


Figure 1. Phase transition between various crystalline allomorphs of cellulose (cellulose I, II, III, and IV)[31].

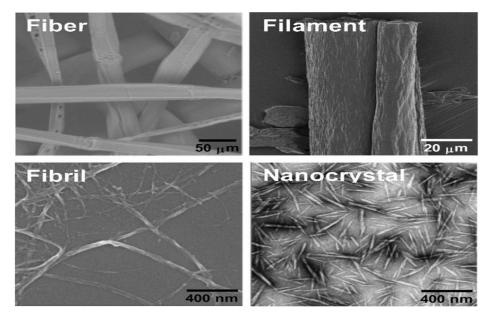


Figure2. Cellulosic particles in different forms: fiber, filament, micro/nanofibril, and nanocrystal [31].

$H = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0$			
Etherification		Esterification	
Cellulose Ethers	R Groups	Cellulose Ester	R Groups
Methylcellulose	H, CH ₃	Cellulose nitrate	H, NO ₂
Ethylcellulose	H, CH ₂ CH ₃	Cellulose sulfate	H, SO ₃ Na
Hydroxypropyl cellulose	H, [CH ₂ CH(CH ₃)O]H	Cellulose acetate	H, I
Hydroxylethyl methylcellulose	H, CH ₃ [CH ₂ CH ₂ O] _n H	Cellulose acetate trimelliate H,	H, I, II
Carboxymethyl cellulose	H, CH ₂ COONa	Cellulose acetate phthalate	H, I, III
Sulfoethyl cellulose	H, CH ₃ CH ₂ SO ₃ Na	Cellulose hydroxypropylmethylphthalate	H, CH ₃ , CH ₂ CH(OH)CH ₃ , III, IV
Cyanoethyl cellulose	H, CH ₃ CH ₂ -C≡N	Cellulose hydroxypropylmethylphthalate acetate succinate	H, CH ₃ , CH ₂ CH(OH)CH ₃ , III, V
Trimethylsilyl cellulose	H, Si(CH ₃) ₃		
CH3			
I	ло 11 111	IV V	

Figure3. Etherification and esterification of cellulose.

3. Corrosion Inhibitor

Corrosion inhibitor is a chemical substance; generally, liquid or gas is used to protect the substance and reduces the corrosion rate when added to the environment. The classification of corrosion inhibitors is summarized in Figure4[36].

Many distinct corrosion inhibitors exist, including organic, inorganic, and hybrid

(organic/inorganic) compounds. In addition, based on electrode process, there are three types of inhibitors; anodic, cathodic, and mixed (Figure 4). i) the anodic inhibitor, e.g., chromates, molybdates, nitrates, and tungstate acts by forming a protective oxide film on the surface of the metal, causing an anodic shift that forces the metallic surface into the passivation region. This reduces the corrosion potential of the

material. ii) the cathodic inhibitor. e.g., sulfite and bisulfite ions, slows down the cathodic reaction limiting the diffusion of reducing species to the metal surface. iii) the mixed inhibitor, e.g., silicates and phosphates, forms a film that reduces anodic and cathodic reactions[37].

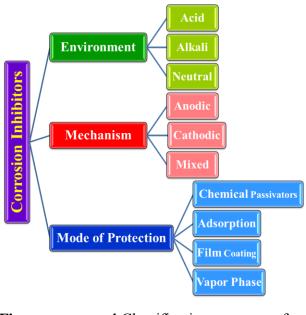


Figure4.Classificationofcorrosioninhibitors.

4. Evaluating Corrosion Inhibitors Techniques

This sectionsummarizesthe different techniques for evaluating corrosion inhibitors: weight loss, hydrogen evolution open reaction, circuit potential(OCP), potentiodynamic polarization, and electrochemical impedance spectroscopy.

4.1.Weight loss measurement[38].

It is a gravimetric experiment performed in a glass cell with a thermostatically cooling condenser, and the specimen should be rectangular. The specimen is cleaned and weighed before and after immersing in the test solution without and with the corrosion inhibitor material. After specific times the sample is removed, rinsed with water and acetone, and dried. The inhibition efficiency (IE %), corrosion rate (*CR*), percentage of protective effectiveness, and surface coverage (θ) can obtain using the following equations:

$$IE \% = \frac{W0 - W}{W0} X 100$$
$$CR = \frac{W0 - W (mg)}{area (cm2) - time (h)}$$
$$E_{WL} = \frac{CRuninhibt - CRinhibt}{CRuninhibt} X 100$$
$$\theta = E_{WL} / 100$$

where *WO* and *W* are the weight of the sample in the absence and presence of inhibitor, respectively.CR_{uninhibit} and CR_{inhibit} are the corrosion rates in the absence and presence of inhibitor, respectively.

4.2. Hydrogen evolution reaction[39].

The volume of evolved gas from a corrosion reaction can be measured by apparatus shown in **Figure 5**. The suggested mechanisms of the hydrogen evolution reaction can be discussed by proposing three different steps of the cathodic reaction, as follows:

i) Volmer reaction, a primary discharge step.

 $M + H_3O^+ + e \leftrightarrow MH_{ad} + H_2O$

H₂O or H₃O is released onto an electrode's surface, producing hydrogen atoms in an acidic solution and generating an adsorbed hydrogen atom (MH_{ad}).

ii) Heyrowsky reaction, an electrochemicaldesorption step.

 $MH_{ad} + H_3O^+ + e \leftrightarrow M + H_2 + H_2O$

The hydrogen evolution reaction occurs on a metal surface by transferring one electron to a hydronium ion.

iii) Tafel reaction, a recombination step.

 $MH_{ad} + MH_{ad} {\leftrightarrow} 2M + H_2$

After that, a pure chemical reaction occurs.

Despite the hydrogen evolution reaction mechanism having three states, no one occurs as a single step, but a combination must be. If the Volmer reaction is slow, the Heyrowsky or Tafel reaction must be faster. If the Volmer reaction is fast, the Heyrowsky and/or Tafelreactionsmust be slow.

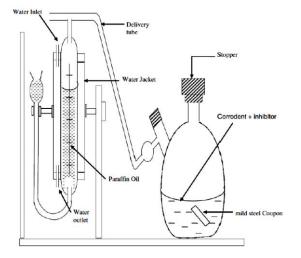


Figure 5. Measurement of evolved hydrogen gas[39].

4.3.Electrochemical Studies[40]

Before the electrochemical measurements, the sample should be cleaned. The platinum,

saturated calomel, and sample are used as auxiliary, reference, and working electrodes. The reference electrode tip is positioned close to the working electrode to minimize the ohmic potential drop.

4.4.OCP and Potentiodynamic Polarization Measurements

OCP is the difference between the electrical potential of two conductors in a specific electrolyte with zero current flow between them. Over time, the OCP informs the system's steady state, and transitions between different states, such as passive and active behavior, occur. Also, it can provide the type of inhibitor, anodic, cathodic, or mixed.

Before measurement, the electrodes are subject to cathodic pretreatment at -1300 mV to remove the native oxides on thesurface and allow them to stabilize for 1 h until a constant potential, which refers to the OCP. After this, the potentiodynamic polarization can be measured at a scan rate of 0.1 mV s⁻¹. The polarization curves for test specimens in the test solution with and without various concentrations of inhibitors will be recorded.

4.5.Electrochemical Impedance Studies

The electrochemical impedance studies can be carried out in the same setup used for potentiodynamic polarization studies by applying the ac perturbation signal of 10 mV within the 100 kHz to 1 Hz frequency range at OCP.

5. Cellulose in Corrosion Inhibitor Applications

Multiple hydrogen bonds and van der Waals forces hold the linear long-chain macromolecules together, preventing them from dissolving cellulose in water[41]. On the other hand. O atoms in cellulose can coordinate with metal atoms to avoid touchingof metal with corrosive media. However, the poor solubility of cellulose in water negatively impacts its usage as a corrosion inhibitor. So, there are limited data on cellulose's ability to suppress metal corrosion. Consequently, researchers are worried about functionalized cellulose to create active groups that increase its solubility and limit corrosion[42].A comparative corrosion inhibition study of and microcrystalline nano cellulose(MCC)on the SS316 alloy during the acid pickling process was investigated. It was found that nano and (MCC) acted as

mixed-inhibitor types with 93.1% and 96.3% as maximum protection capacities, respectively. Also, Density Functional Theory(DFT)calculation displayed that both interact with the metal interface through donor-acceptor attractions. A Monte Carlo simulation study revealed they adsorb via their electron-rich sites[43]. To increase the adhesion of the inhibitor onto a metal surface, colloidal lignin particles were mixed with TEMPO-oxidized cellulose nanofibers to inhibit the corrosion of hot-dip galvanized steel. Scanning electron microscopy showed the coalescence of colloidal lignin particles occurs during the drying process, forming a barrier layer on the steel (Figure 6). The cellulose-lignin layer reduced the electrolyte penetration (NaCl) to the metal-coating interface[44].

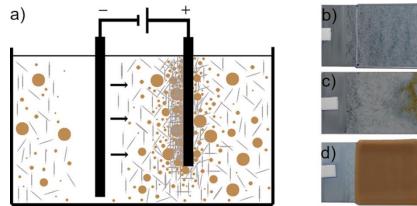


Figure 6. (a) Electrophoretic co-deposition of negatively charged colloidal lignin (brown spheres) and TEMPO-oxidized cellulose (gray lines) from aqueous dispersions onto a positively charged hot-dip galvanized steel surface, (b) appearance of the coating deposited from TEMPO-oxidized cellulose suspensions, (c) substrate deposited from colloidal lignin dispersion, and (d) coating obtained from co-dispersion of TEMPO-oxidized cellulose and colloidal lignin[44].

By functionalizing cellulose, some watersoluble groups, such as carboxymethyl, hydroxyethyl, methyl, hydroxypropyl, and hydroxyethyl methylcellulose, can be connected to cellulose, improving its watersolubility and corrosion inhibition performance[32].Also, cellulose can be synergized with surfactants, hexamethylenetetramine, halogen ions. methionine, metal nanoions, and other synergists to increase its stability and corrosion inhibition performance. It has been widely documented that hydroxypropyl methylcellulose(HPMC), carboxymethyl cellulose(CMC), and ethyl hydroxyethyl cellulose (EHEC) are effective corrosion inhibitors various corrosive in environments^[23].

Comparing chitosan and CMC as a single corrosion inhibitor with commercial inhibitor (NALCO Product, US), they have been found to inhibit API 5L X60 pipeline steel in а CO₂-saturated saline solution[45]. These substances often create very thin, long-lasting adsorbed films on metal surfaces, which slow anodic, cathodic, corrosion both or reactions[46]. Unfortunately, these inhibitors do not exhibit antibacterial behavior, and the dosage is very high. Gan and coworkers prepared functional cellulose derivatives by adding ester, amide, and borate groups to enhance cellulose's inhibitory and antibacterial effectiveness. For example, it was found that a borated aminated cellulose citrate with N and O acted as polar atoms reaction centers for adsorption and inhibited the inhibition effects in a simulated cooling water system on A3 steel corrosion. Also, it was found from polarization studies that it controlled the anodic process and operated as a mixed-type inhibitor [47]. Since the corrosion of steel involves the dissolution of iron (anodic) and theoxygen reduction reaction (cathodic) as follows:

 $O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$

Borated aminated cellulose citrate has many reaction centers, includingO and N atoms. The unshared electron pairs of these atoms have enabled interactions with the vacant dorbitals of iron atoms on the steel surface, forming chelates. Increasing the alkyl groups in cellulose derivatives, enhancing the electronic effect, and increasing the electron cloud density of N atoms stabilize the coordination bonds. This process isolated the metal surface from water and dissolved oxygen by forming an adsorption layer that inhibited corrosion on the metal surface (**Figure 7**).

The effect of metal oxide nanoparticles on the inhibition of corrosion efficiency of primary aminated cellulose on C-steel in acidic solutions was investigated. It was found that with/ without metal oxide nanoparticles exhibited superior inhibitive action. From PDP, these inhibitors act as inhibitors, mixed-type whereas the thermodynamic parameters are confirmed as chemisorption that follows the Langmuir isotherm model. The efficiency followed the order of aminated cellulose/NiONPs (98.6%) cellulose/Fe₃O₄NPs >aminated (96.1%)cellulose/CuONPs >aminated (93.2%) >aminated cellulose (88.1%)[48].

As a green corrosion inhibitor, CMCwas investigated, but it is insoluble in water; its salt Na-CMC is soluble in water. Therefore, the inorganic sodium ion is connected to cellulose, improving the water solubility and corrosion inhibition efficiency. CMCNahas been investigated on copper and examined by electrochemical impedance spectroscopy

 $Fe \rightarrow Fe^{2+} + 2e^{-}$

and polarization measurements. The results showed that the best inhibition efficiency could reach 83.34% when the concentration of CMCNais 5 mg/L at 20 °C [49].By studying the quantum chemical parameters of CMCNa, the more donating electrons trend to the metal is associated with the high energy of highest occupied molecular orbital (HOMO)of the inhibitor. While the lower the lowest unoccupied molecular orbital (LUMO) energy, the easier acceptance of electrons from metal. A low gap energy value means better inhibition efficiencies [50, 51]. The LUMO in the carboxyl functional group and the Mulliken charge of

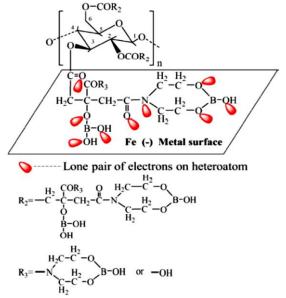


Figure 7.Corrosion inhibitionmechanism of borated aminated cellulose citrate in the simulated cooling water system[47].

oxygen atoms in the carboxyl group are -0.473 and -0.451, respectively. So, CMCNacould be adsorbed on copper by the carboxyl group. In another study, a CMC /Ag nanocomposite as anticorrosion for St37 specimen in 15% H₂SO₄medium with 93.94% efficiency wasafforded by 1000 ppm CMC /AgNPs[23].Manimaran et al. evaluated the corrosion inhibition efficiency of CMC with and without Zn^{2+} on carbon steel. They found that a composite of 250 ppm of CMC and 50 ppm of Zn^{2+} offers 98% inhibition efficiency to carbon steel immersed in groundwater [52].They suggested the following mechanism:

$$Zn^{2+}-CMC + Fe^{2+} \rightarrow Fe^{2+}-CMC + Zn^{2+}$$

 $Zn^{2+} + 2OH^{-} \rightarrow Zn(OH)_{2}\downarrow$

Protective film consists of: Fe²⁺–CMC complex and Zn(OH)₂

At anode: $Fe \rightarrow Fe^{2+} + 2e^{-}$

At cathode: $H_2O + \frac{1}{2}O_2 + 2e^- \rightarrow 2OH$

 Fe^{2+} -CMC \rightarrow CMC complex

The halide ions (Cl⁻, Br⁻, and I⁻) can affect the CMC corrosion inhibition by studying their effect on steel corrosion in H₂SO₄ using hydrogen evolution and weight loss methods. The study confirmed inhibitors' physical adsorption on the steel surface in the presence and absence of halide ions. Furthermore, adding Cl- antagonizes the inhibition process, while I- has a synergistic effect. Figure 8shows the weight loss and volume of evolved H2gas during steel corrosion in H₂SO₄ in the presence and absence of halide ions with CMC. It confirmed halide ions' antagonized and impacts on the corrosion synergistic inhibition efficiency of CMC[53].

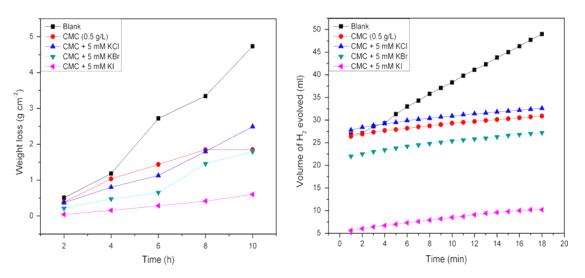


Figure 8. Variation of weight loss and volume of H_2 evolved against time for steel corrosion in H_2SO_4 in the presence and absence of CMC and its halide mixtures[53].

Soviziet.al. examined inhibition the efficiency of CMC alone and in combination with iodide ions toward the corrosion of aluminum in a 2M H₂SO₄ solution. They found that CMC inhibited aluminum corrosion and the presence of iodide ions stabilizes the adsorption of CMC molecules on the surfaces of aluminum and enhances inhibition efficiency of CMC the [54]. Arukalam et al. found that EHECis an effective inhibitor of mild steel in an acidic solution. Its inhibition efficiency was increased by adding KI due to the synergistic effect. The inhibiting action is due to the adsorption of molecular and protonated species on the anodic and cathodic sites forming a chemisorbed film on the steel surface. The Polarization studies confirmed that EHEC with/without KI was a mixed-type inhibitor with a predominant cathodic effect [55, 56].

The addition of surfactants to the natural inhibitor can increase its inhibition

efficiency. Mobin et al. studied the effect of adding different surfactants, such as sodium dodecyl sulfate, triton X 100, and cetyl pyridinium chloride, on the inhibition effect of hydroxyethyl cellulose (HEC) on the corrosion of carbon steel in HCl solution. It was found that the addition of surfactants synergistically increased the HECinhibition efficiency, and the triton X 100 was more pronounced than other surfactants. The adsorption of HECon a carbon steel surface is physical adsorption, while in the presence surfactant is mixed adsorption. of Furthermore, HECis a mixed-type inhibitor, predominantly anodic [57].

Incorporating nanometals, such as copper, iron, and nickel, enhances CMC's inhibition efficiency. El-Lateef et.al. found that the protection capacities for carbon steel of CMC, CMC/Fe, CMC/Cu, and CMC/NiNPsincreased from 25.3, 33.3, 39.7, and 43.5% at 50 ppm to 76.6, 94.9, 96.2, and 98.4% at 400 ppm, respectively. Suggesting

that these composites have potential applications in solar-driven water evaporation [58].Figure9 shows the potentiodynamic-polarization graphs for carbon steel in 2N HCl with various doses of composite inhibitors. It was clear that the addition of CMC or CMC/NPs composites led to a shifting in anodic and cathodic current densities to lower values.

Incorporating only 1% of polyurethane prepolymer into HEC (**Figure 10**) enhanced its inhibition efficiency even at high temperatures and in an aggressive acidic solution. Furthermore, this modified HEC functioned as a mixed-type inhibitor with 93% maximum efficiency at 80 °C[59].

Additionally, N atoms with functional groups are essential for inhibiting corrosion that can be adsorbed on the metal surface.

Therefore, adding polar groups with N atoms to modified cellulose can enhance the corrosion inhibition effect further. For by treating HEC with 2example, chloroethyl diethylamine hydrochloride, aminoated HEC was created, and in 1MHCl solution, it behaved as an effective inhibitor for mild steel. At 900 mg/L, aminoated hydroxyethyl cellulose(AHEC) had a 93 % corrosion inhibition efficiency at room temperature. The studies revealed that it acts as a mixed inhibitor type, and the adsorption mechanism is physical [12]. The steel corrosion by AHECis due to its adsorption at the metal surface, which depends on the electrochemical potential at the interface. The corrosion in an acid medium involves the dissolution of Fe (anodic) and hydrogen evolution (cathodic) (Figure 11).

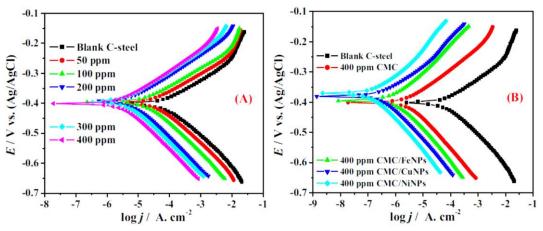


Figure9.Potentiodynamic-polarization curves of C-Steel in 2N HCl solutions without and with various concentrations of CMC (A) and CMC/NPs (B) at 50 °C.

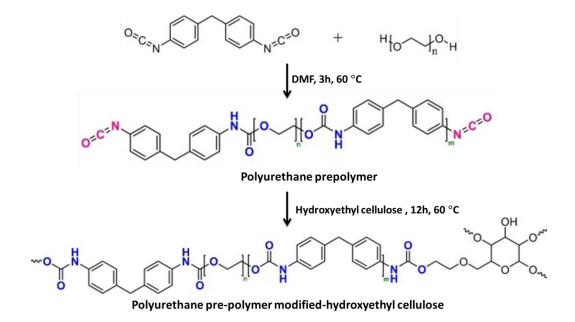
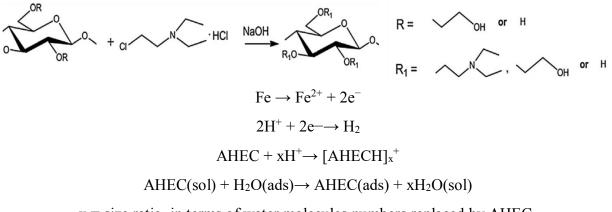


Figure 10.Synthesis of polyurethane prepolymer-modified HEC[59].



x = size ratio, in terms of water molecules numbers replaced by AHEC.

Figure 11. Synthesis of AHEC and its inhibition of Fe corrision.

In an acidic medium, the AHECacts as protonated species and neutral molecules, and the steel surface is positively charged. While the degree of hydration of Cl-ions is small, leading to the adsorption of positive AHECon the metal surface by the electrostatic interaction with chloride ions. Also, the neutral AHECmay adsorb on the metal surface by displacement of already adsorbed water molecules (**Figure11**). The interaction between the unshared electron pairs of N and O of AHECand vacant dorbital of Fe on the steel surface leads to the isolation of metal from aggressive solution, inhibiting the surface from corrosion[12].

another study. the anti-corrosive In performance of different cellulose-niacin composites for copper in NaCl solution has been evaluated. It is found that the efficiencies of ethvl cellulose-niacin. microcrystalline cellulose-niacin, and CMCniacin composites were 94.7%, 33.2 and 83.4 %, respectively, as corrosion inhibitors for copper in NaCl solution. Furthermore, the potentiodynamic polarization measurements confirmed that these composites act mixed-type as а inhibitors[60].

The corrosion inhibition of HEC on aluminum in HC1 and H₂SO₄ was investigated, also. The potentiodynamic polarization results confirmed the mixedtype inhibitor in two solutions with the predominant anodic effect. and the adsorption onto the aluminum surface followed the Langmuir isotherm. Furthermore, the inhibition efficiency was increased with an increase in inhibitor concentration, but rising temperature alters the sustainability [61].In another study, adding potassium iodide to hydroxypropyl cellulose increased its inhibitive effectiveness in dissolving steel in 0.5M H₂SO₄. The potentiodynamic polarization showed that hydroxypropyl cellulose alone controlled the cathodic reaction, while the presence of potassium iodide controlled the anodic processes. The inhibitive efficiency of hydroxypropyl cellulose can be attributed to the electrostatic interaction between its functional groups such as OH groups, propyl groups, and the aromatic structure and surface charge of steel in addition to the cover of a large metal surface area by complex formation [62].

Methyl hydroxyethyl cellulose was used as an inhibitor for the corrosion of copper in an HCl solution. It was found that the inhibition efficiency incremented with the concentration and immersion time increments, mainly physisorption[3].

Cellulose acetate (CA) can be considered an anticorrosion agent due to its costeasyof manufacture, and effectiveness. environment. In addition, it inhibited the corrosion of aluminum metal in the HCl solution with inhibition efficiencies of 55.71 and 51.87 % in 2M and 3M HCl solutions, respectively[63].CAfilms doped with amoxicillin high showed corrosion resistance on aluminum alloy. In addition, under anodic polarization, the doped film with amoxicillin demonstrated the effect of amoxicillin as a corrosion inhibitor[64].Its anti-corrosion effect can be attributed to its molecular structure and ability to form complexes. The ligands can react with the aluminumand copper ions of the alloy at forming positions Ι and II. metal complexes(Figure 12)[65]. The electrospun of CAnanofibers incorporated with biogenic silver nanoparticles was used to inhibit the corrosion of copper in artificial seawater and Escherichia coli inoculated solutions. It was found that this composite showed efficient anticorrosive properties in corrosive and biocorrosive marine solutions[66].

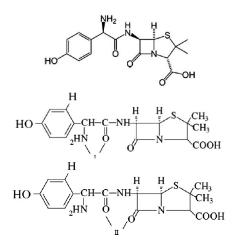


Figure 12.Structure of the amoxycillin and its anticorrosion mechanism on aluminum alloy.

6. Conclusion and Future Aspects

Recently, cellulose has been studied as a natural alternative corrosion inhibitor and shows great prospects as a green corrosion inhibitor, and its efficiency depends on modification methods, compounding agents, and dosage. This review summarized cellulose and its derivatives with further emphasis on their versatility for applications in inhibiting metal corrosion. Recent advances show cellulosic materials have inherent characteristics that can be tailored for corrosion inhibitor applications. In particular, CMC and HEC are used since they are ideal as versatile materials for corrosion inhibitor applications. However, no studies have been made on cellulose; there are still some limitations, such as effective corrosion inhibitors. In addition, their mechanism on the metal surface is required to be explored thoroughly, as well as the synergistic action mechanism. However, the efficiency of cellulose natural is significantly low, and it can be improved by chemical modification and complex formulation. Therefore, a significant way to enhance the inhibition efficiency and solubility of cellulose chemical is modification. Accordingly, an appropriate hydrophilic group should be introduced into cellulose to improve its water solubility and corrosion inhibition.

Abbreviations

Open circuit potential (OCP); Inhibition efficiency (IE), Corrosion rate (CR); Microcrystalline cellulose (MCC); (2,2,6,6-Tetramethylpiperidin-1-yl)oxyl (TEMPO); Nanoparticles (NPs); Highest occupied molecular orbital (HOMO); Lowest unoccupied molecular orbital (LUMO); Cellulose acetate (CA); Hydroxypropyl methylcellulose (HPMC); Ethyl hydroxyethyl cellulose (EHEC); Carboxymethyl cellulose (CMC); Hydroxypropyl cellulose (HEC); Aminoated hydroxyethyl cellulose (AHEC); Density Functional Theory (DFT).

Disclosure statement

No potential conflict of interest was reported by the author.

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Conflicts of interest

There are no conflicts to declare.

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