An Overview of Silane-based Hybrid Sol-gel Coatings for Highly Efficient Metal Corrosion Protection

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Editor's note: This invited mini-review presents an overview of the corrosion protection potential of hybrid silane-based coatings applied through sol-gel fabrication on different metal substances. Sheydaei and Edraki focused on the environmental impact of such coatings and the importance of corrosion inhibitors on the final performance of silane-based corrosion protection systems for different application fields.

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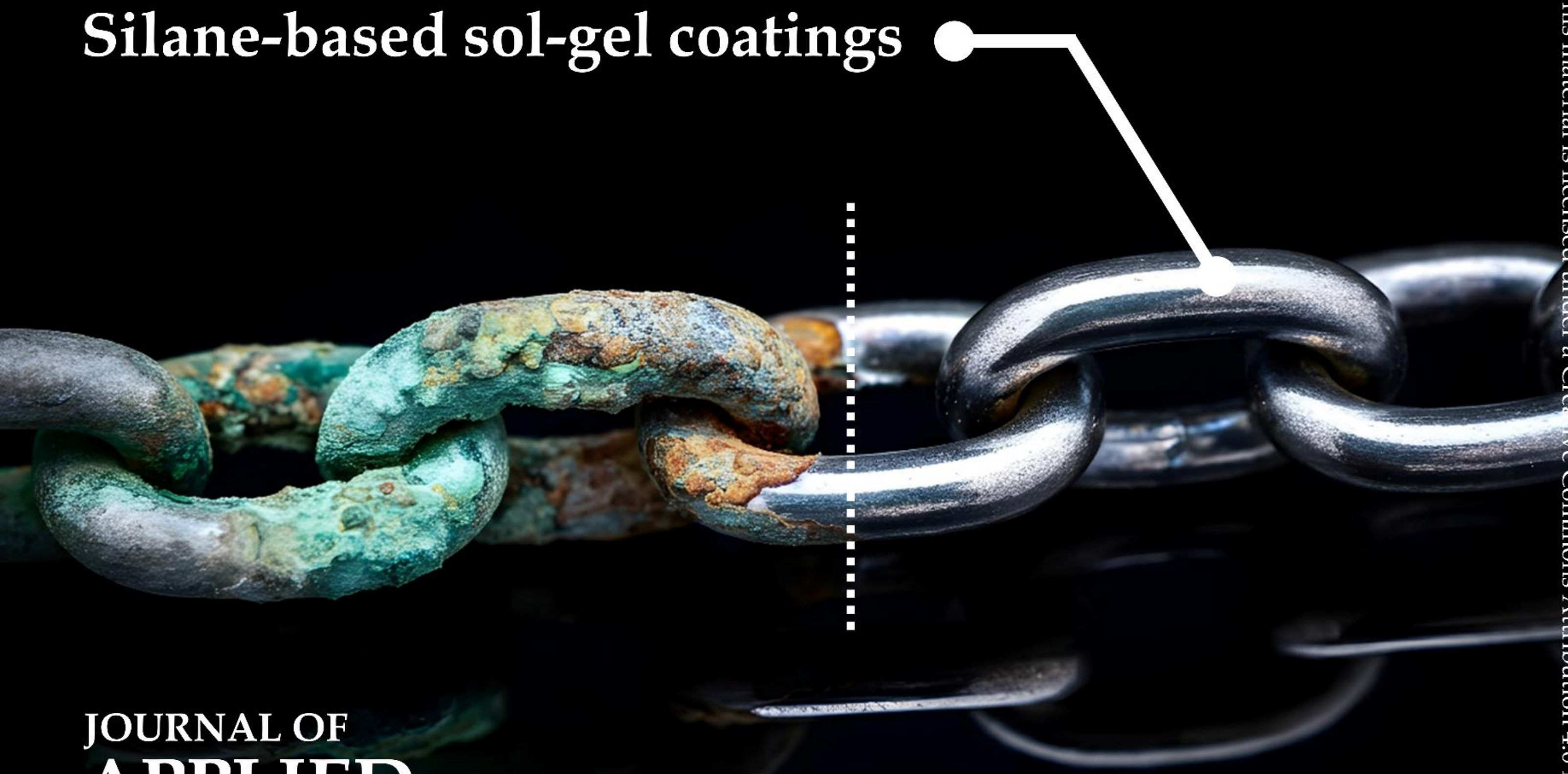

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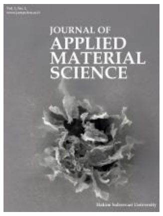

Invited Review

An Overview of Silane-based Hybrid Sol-gel Coatings for Highly Efficient Metal Corrosion Protection

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Abstract

The most important problem of metal structures is corrosion, which can lead to significant losses and is considered a serious threat to safety. Corrosion can be considered a slow but continuous phenomenon. Researchers are continually seeking low-cost, high-performance solutions. In the meantime, coatings have traditionally been the most popular method to mitigate corrosion. However, there is a growing focus on developing environmentally friendly protective coatings, as many conventional coatings can harm the environment after they deteriorate. Many coatings incorporate corrosion inhibitors, providing a synergistic effect that helps prevent corrosion. Unfortunately, many of these inhibitors contain toxic and harmful substances posing additional environmental risks. Therefore, selecting the right coating is crucial. Among the various types of coatings, silane-based sol-gel coatings emerge as suitable candidates. This paper, discusses silane-based sol-gel coatings, the sol-gel method, and the factors that influence the efficiency of, these products. Additionally, it explores the use of plant-extracted inhibitors in sol-gel coatings.

Keywords: Environmental pollution, Green chemistry, Green inhibitors.

Contents

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1. Introduction

Metal structures, despite their unique properties, have a big problem, which is a vulnerability in corrosive environments [1]. Corrosion can be considered a mysterious and quiet phenomenon, but it is very damaging because it can cause huge costs [2]. Although corrosion occurs in very aggressive environments such as the sea, it appears and spreads very quickly [3]. Corrosion has been a concern of researchers and manufacturers of metal instruments for many years, and many methods have been investigated to delay and slow corrosion. Corrosion, in addition to creating financial costs, also causes environmental pollution [4]. Researchers have found that one factor in the release of heavy metals into the environment is corrosion [5]. Many industries such as oil and gas and marine are constantly involved in the phenomenon of corrosion and suffer economic damages [6,7]. Corrosion can be considered an electrochemical and spontaneous process, although many factors contribute to its creation and development, such as temperature, humidity, pH, and chemicals [8]. In the industry, manufacturers are always looking for low-cost and high-performance solutions, although the new legislations require the use of methods and chemicals that are safe for the environment and humans [9]. Figure 1 shows the various methods used to prevent corrosion.

Corrosion inhibitors are very effective, but many of them are toxic, such as chromates, and their release into the environment can cause serious harm [10,11]. Therefore, researchers are investigating green corrosion inhibitors (such as plant extracts, pharmaceuticals, and natural gums) [12]. It can be said that coatings have the best effect in corrosion protection [13]. They are an outer layer that prevents the penetration of corrosive species [14]. Among the classifications of coatings, polymers are in a special place due to properties such as hydrophobicity and self-healing [15].

Among the coating production methods, the sol-gel method has received much attention in the past years,

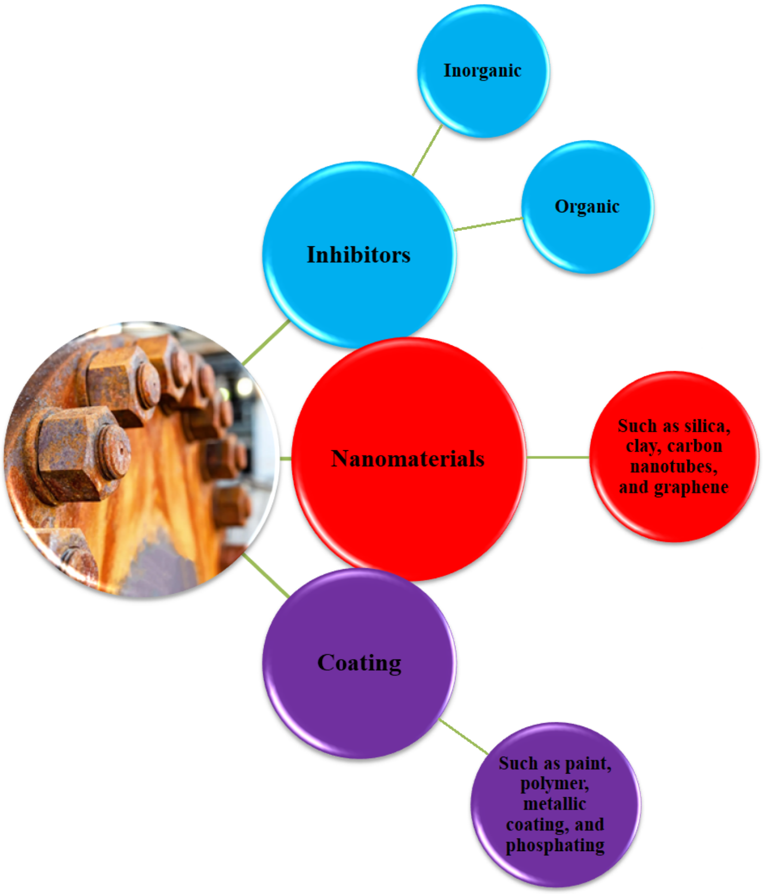

Figure 1. Different methods to prevent corrosion

especially the silane-based hybrid coatings that are prepared by the sol-gel method [16]. The sol-gel method is very economical, low toxicity, and produces a high-purity product [17]. Considering the environmental concerns caused by chromate-containing coatings, silane-based hybrid coatings can be considered a suitable alternative for them. Hybrid organic-inorganic materials synthesized by sol-gel method have been widely used since the last decade. It can be said that the sol-gel method consists of three stages of hydrolysis, polycondensation, and thermal densification and can be applied at ambient temperature and atmospheric pressure [18]. In this review, research on the use of silane coatings to reduce the corrosion of metal substrates has been investigated.

2. Background

The history of using coatings resistant to environmental factors goes back thousands of years [19]. The Egyptians developed wax-based weatherproof coatings that were applied in the molten state [20]. Natural bitumen was also used as a coating to protect the wooden body of ships [21]. Coatings are prepared to create a barrier and separate the object from the environment. There is no such thing as perfect (lifelong) coating, and no coating can provide complete protection [22]. In general, as the effective life and efficiency of the coating system increases, the costs of its implementation also increase. With the expansion of various types of surface coatings, organic-inorganic hybrid coatings that were prepared by the sol-gel process have received much attention (see Figure 2).

These coatings provide the possibility of achieving special properties by having organic and inorganic materials together. Hybrid organic-inorganic coatings that have inorganic particles on a nanoscale are proposed as nanocomposite coatings. Composites are the result of the physical mixing of two or more materials, in which two main phases are called the matrix phase and the dispersed phase. Usually, the purpose of preparing a composite is to create properties that the constituent phases alone do not have. Organic-inorganic hybrid nanocomposites prepared by the sol-gel method have uniform morphology and good

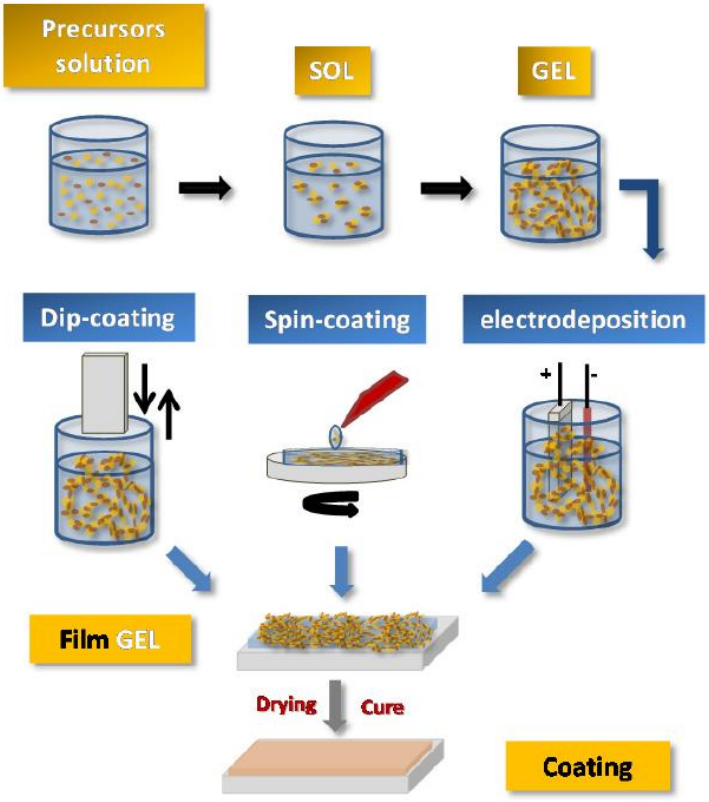

Figure 2. Example of processing routes to obtain sol-gel coatings for a surface. Reprinted with permission from [23]. 2016, MDPI.

properties and can be used in many applications such as anti-wear, anti-scratch, anti-corrosion, anti-fungal and bacterial coatings and even in the field of pharmaceuticals [24,25]. A group of sol-gel coatings is based on silanes, which, contrary to their high corrosion protection potential, are electrochemically inactive unless they have chemically active groups [26]. Therefore, these coatings do not provide active protection. The protection they provide is generally based on passive protection through the formation of a layer that adheres well to the surface. However, long-term exposure of these coatings to water/electrolyte eventually causes moisture to penetrate the metal/coating interface [27]. The protective properties of sol-gel coatings can be improved by using active species such as corrosion inhibitors that add an active protective mechanism to the coating (see Figure 3) [28]. Corrosion inhibitors can be added directly to the formulation or immobilized in places to reduce possible interactions with the matrix and control the release of the inhibitor [6-8].

A sol is a colloidal dispersion of nano-solid particles in a liquid medium, and a gel is defined as a colloidal system that is more like a solid than a sol [29]. Sol-gel chemistry is related to the process in which a gel is formed by the accumulation of sol [16]. As a result of hydrolysis reactions with the help of an acid or base catalyst, a sol is formed, and condensation reactions lead to the gelation of this sol [16,29]. In fact, as a result of condensation reactions, molecules with high molecular weight are produced, which are continuously joined together [30].

The effective factors in the sol-gel process are the molar ratio of reactants, solvent, temperature, and pH [16,17,29]. Some of the advantages of the sol-gel method include low process temperature (close to ambient temperature), ease of operation, high chemical diversity, the possibility of coating different geometric shapes, the strong interaction of the coating with the surface, simple process, low price, and chemical and physical stability [16,30,31]. Also, in the sol-gel process, it is possible to add special properties to the coating by adding additives, based on which sol coatings with properties such as UV resistance and anti-reflection are prepared [16,17,30]. Despite the mentioned advantages, this method also has disadvantages, such as that some raw materials used in this method are sensitive to environmental factors such as humidity [32]. Also, the possibility of the solvent remaining in the coating and creating cracks in the coating [16,32].

In the past years, special attention has been paid to the production of organic-inorganic hybrid coatings using the sol-gel method, which showed good corrosion protection properties [33]. For several reasons, organic-inorganic hybrid coatings synthesized by the sol-gel method are used more than pure organic or inorganic sol-gel coatings to protect the corrosion of metal surfaces, which are mentioned below [16,17,33]. The hybrid coating leads to the formation of a film without cracks better than organic or pure inorganic coatings, and for heat treatment, they usually require lower temperatures (usually 100 °C) than the pure organic or inorganic sol-gel coatings (600-800 °C) [34,35]. On the other hand, there is a possibility of compatibility with various added anti-corrosion agents such as inhibitors and pigments for organic-inorganic hybrid coatings [34].

Hybrid coatings are divided into two groups based on the interaction between organic and inorganic implementation [16,17]. The first group is coatings in which there is no chemical bond between organic and inorganic components and these components are only together by weak van der Waals or hydrogen interactions [34,35]. The hybrid coating in this group is formed by mixing organic and inorganic phases in the presence of a contacting organic polymer [35]. Therefore, these composites show little stability against moisture. In the second group, there are strong covalent bonds between organic and inorganic [34]. Regarding the

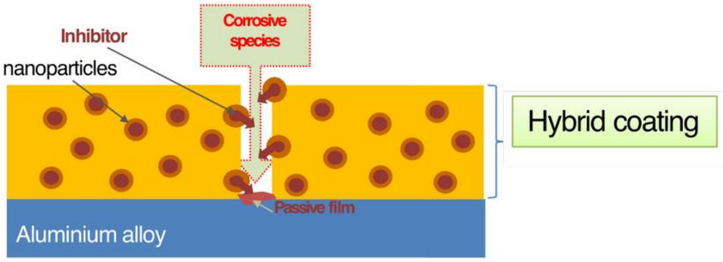

Figure 3. Generic scheme of the corrosion healing mechanism of a ca hybrid sol-gel coating containing inhibitory nanoparticles. Reprinted with permission from [23]. 2016, MDPI.

second group, we can mention organo alkoxysilane [16,17,35]. One of the advantages of these coatings is increasing the adhesion of the final coating to the metal surface and other coatings, increasing the mechanical resistance of the coating, and producing a crack-free and completely uniform coating [36]. In summary, it can be said that in the sol-gel process, the hydrolysis reaction is performed by increasing the water content and replacing the alkoxy groups (OR) with OH groups (see Figure 4 (a and b)). Next, successive condensation reactions between Si-OH and Si-OR groups occur, and Si-O-Si is produced with alcohol as well as water as by-products (see Figure 4c). In this section, the sol-gel process was briefly reviewed, and the studies conducted are discussed in the next section.

3. Using sol-gel coatings to prevent corrosion in metals

Corrosion prevention has always been a concern and various methods based on electrochemical principles have been explored for this task, although methods of separating metal from aggressive media have been a field of study and investigation in materials science for many years [23]. Corrosion and subsequent rate of reaction depend on two physical and chemical factors of the substrate, and also, the type of corrosion (such as pitting, filiform, and bimetallic) depends on the substrate and the aggressive media. Meanwhile, aggressive species such as chloride ions play an important role in determining the type of corrosion and its reaction rate [37]. One of the popular and developing methods of surface coating is the sol-gel method [38]. The most common sol-gel coating matrices are based on silane, some of their precursors are reported in Table 1.

Coatings based on silanes such as TEOS are very popular because they have high thermal stability and their reactivity is possible in a wide range of temperatures [38,39]. Also, this class of precursors provides good wear resistance, adhesion, and corrosion protection [39]. Rodríguez-Alonso et al. [40] investigated corrosion protection of AZ61 magnesium alloys by using sol-gel method (the precursors were TEOS and 3-

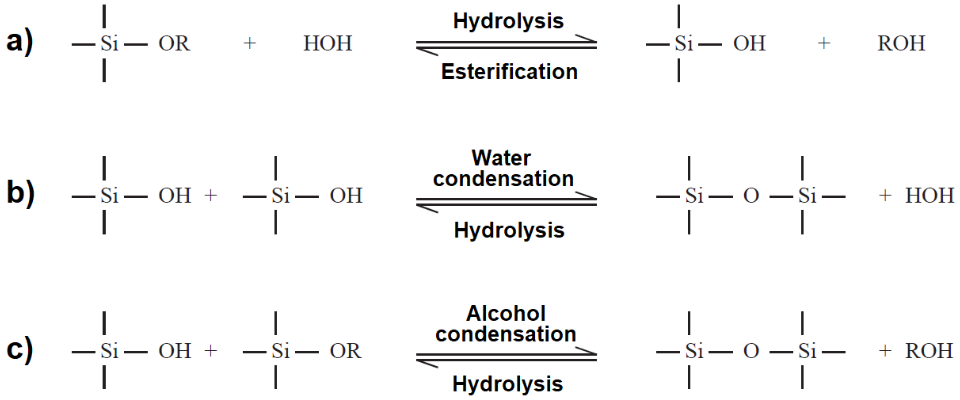

Figure 4. Hydrolysis and condensation reactions in sol-gel processes. Reprinted with permission from [37]. 2005, IPPI.

Table 1. Full names and abbreviations of some precursors of silane (data from [37,39])

Chemical Name	Abbreviation
Tetraethoxysilane	TEOS
3-Glycidoxypropyltrimethoxysilane	GPTMS
Methyltriethoxysilane	MTES
Glycidoxy propyl trimethoxy silane	GPS
Polymethyltrimethoxysilane	PMTMS
3-Aminopropyltrimethoxysilane	APS
Bis [3-(triethoxysilyl)propyl]tetrasulfide	BTESPT
Methylmethoxysilane	MTMS
3-Glycidoxypropyltrimethoxysilane	GPTMS
Methyltriethoxysilane	MTES

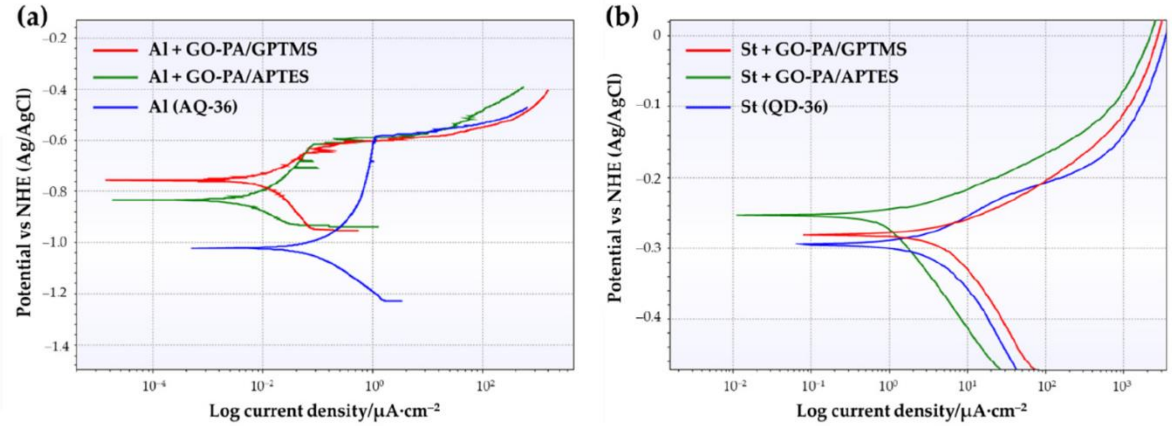

Figure 5. Polarization curves of aluminum (a) and steel (b). Reprinted with permission from [41]. 2022, MDPI.

(Trimethoxysilyl)propyl methacrylate) and inhibitors such as L-cysteine, dimethyl glyoxime, quinine, 2-Aminopyridine, graphene oxide sheet; and titanium dioxide nanoparticles. The results showed that these coatings have good adhesion and performance even without any pretreatment. The results showed that these coatings have good adhesion and performance even without any pretreatment. All inhibitors improved corrosion resistance except graphene. Sfameni et al. [41] prepared a coating using graphene oxide (GO) and phytic acid (PA), as well as GPTMS and APTES precursors. The performance of coatings on aluminum and steel layers was investigated. The results of the polarization (see Figure 5) and neutral salt spray test (see Figure 6) showed improved anti-corrosion performance.

Li et al. [42] created a coating on Q235 carbon steel substrate using MTMS, colloidal silicon dioxide nanoparticles, silica, and titania powders by sol-gel method. Electrochemical impedance spectroscopy results showed that this coating shows excellent anti-corrosion properties. Also, the authors clearly showed the process of the sol-gel process using Transmission Electron Microscopy (TEM) images (see Figure 7).

Inorganic thin films prepared using the sol-gel method are used in various fields. These layers contribute to low conductivity and high purity for excellent sensitivity. Finally, materials with high surface area and porosity increase the sensitivity in mechanisms dominated by surface phenomena [43].

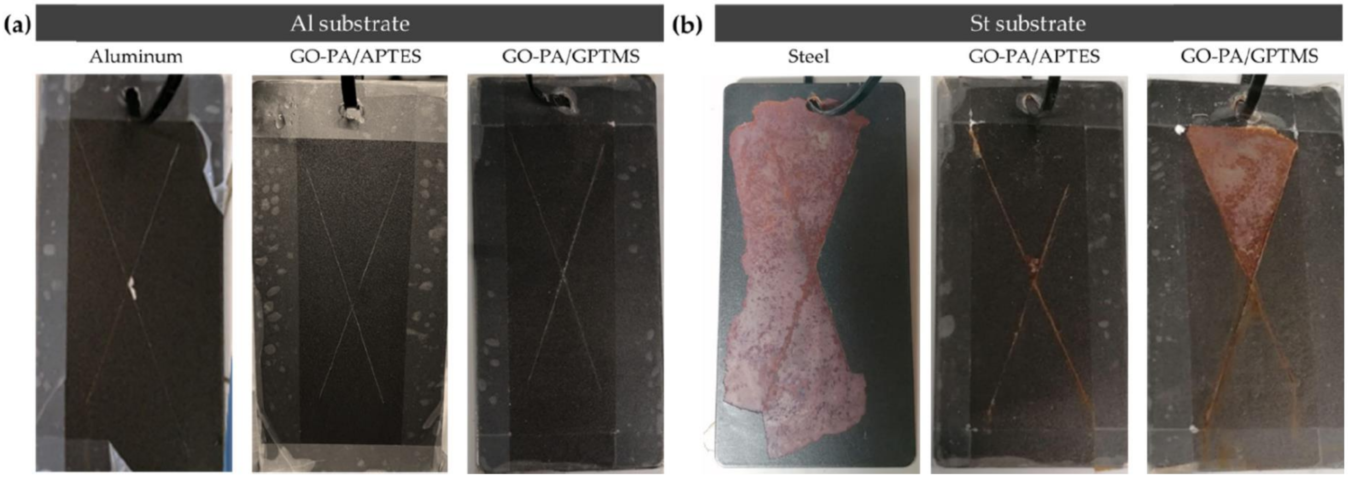

Figure 6. Visual images of coated substrates without and with nanofiller after salt spray test (aluminum: 900 h; coated aluminum 1300 h; steel: 50 h; coated steel: 400 h). Reprinted with permission from [41]. 2022, MDPI.

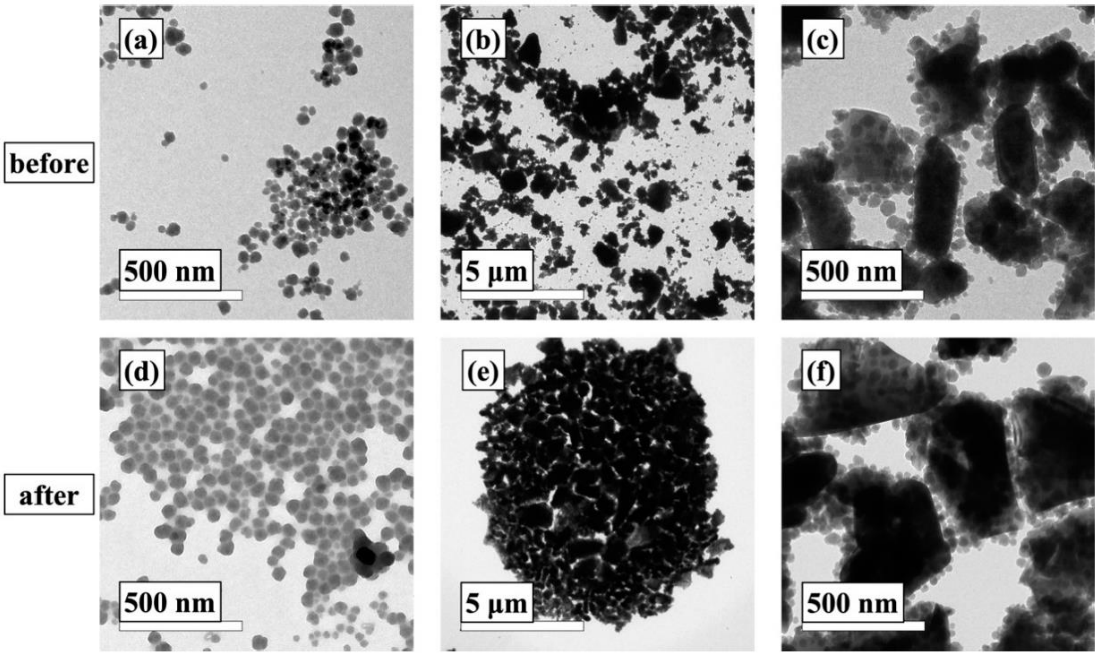

Figure 7. TEM images of colloidal silica nanoparticles (a,d) and silica nanoparticles/filler mixture before (b,c) and after (e,f) the sol-gel process. Reprinted with permission from [42]. 2019, MDPI.

Table 2. Performance results of some coatings in different corrosion environments

Coatings	Sol-Gel Solution Composition	Substrate	Corrosive medium	Effect	Ref.
Bare sol-gel coating	Mixing TEOS and GPTMS	AZ31	Sodium chloride (NaCl) (0.1 M)	The hybrid coatings reduced the corrosion current density by about three orders and the protection range increased up to 160 mV.	44
Sol-gel coating + corrosion inhibitor	Mixing MTES, GPTMS, and TEOS	AZ31	Simulated Body Fluid (SBF) solution	The Rf of silane coating modified with hydroxyapatite nanoparticles was more than 100 times higher than that without modification after being soaked for 4 days.	45
Composite sol-gel coating (PEO/sol-gel)	Mixing TEOS and MTES.	AZ31	Hanks' solution (pH = 7)	icorr PEO = 1.6 × 10^{-7} A/cm^2 icorr PEO/sol-gel = 2.50 × 10^{-8} A/cm^2	46
Composite sol-gel coating (PEO/sol-gel)	Mixing GPS and TEOS	AM6	NaCl (3.5 wt.%)	Rp PEO = 3.37 ×10^5 W cm^2 Rp PEO/sol-gel = 3.58 × 10^9 W cm^2	47
Composite sol-gel coating (N-GQDs/sol-gel)	PMTMS: (MTMS: ethanol: DI water = 3: 10: 20)	AZ91D	NaCl (3.5 wt.%)	Rct N-GQDs /sol-gel = 1.7 × 10^4 W cm^2	48
Composite sol-gel coating (sol-gel/GO/sol-gel)	Mixing APS/BTESPT	AZ31B	NaCl (3.5 wt.%)	icorr sol-gel/GO/sol-gel = 1.381 × 10^{-8} A/cm^2	49
Composite sol-gel coating (MAO/sol-gel)	PMTMS: (MTMS: ethanol: DI water = 3:10: 20)	AZ31	NaCl (3.5 wt.%)	icorr MAO/sol-gel = 2.86 × 10^{-8} A/cm^2 Rct MAO/sol-gel = 2.24 × 10^6 W cm^2	50
Composite sol-gel coating (CLP/sol-gel)	Mixing TEOS and MTES	AZ91	NaCl (3.5 wt.%)	Rtotal conversion coating = 0.681 kW cm^2 Rtotal conversion coating /sol-gel = 127.382 kW cm^2	51

Notes: Rf: the overall resistance of the coating response, icorr: Corrosion current density, Rp: Polarization resistance, Rct: The charge transfer resistance, Rtotal: values are calculated as the sum of all the faradaic resistance by using the fitted data, PEO: Plasma electrolytic oxidation, CLP: Clinoptilolite, MAO: Micro-arc oxidation, GQD: Graphene quantum dots.

In fact, the complexing and dispersing agents determine the size, shape, and porosity of the particles [23,43]. Also, the porous aerogels materials increase the conductivity and maintain the amorphous morphology [43]. One of the applications of sol-gel materials (such as olivine-type materials, nickel-based materials, manganese-based materials, carbon aerogels, and perovskites) is to produce supercapacitors [23,43]. The sol-gel process can be considered a flexible method to produce coatings because a denser layer with high adhesion can be created with this process, in this regard studies in this field are widely underway, and some of the results of these studies in Table 2 are reported.

Although sol-gel is considered a green method, some of the additives used in this method, especially corrosion inhibitors, are very toxic and harmful to health and the environment [8]. Hence, in the past years, researchers have investigated the use of green corrosion inhibitors (GCIs) in this method. Among the classifications of GCIs, plants and their extracts are of great interest [6,8]. Because plants are rich in bioactive compounds, many of them have antimicrobial properties in addition to anti-corrosion properties [8,52-55]. All parts of a plant can be used to prepare the inhibitor, but usually, the leaves are more important due to the abundance of phytochemicals [8]. In general, the reason for their popularity can be attributed to their cheap price, easy preparation method, and non-toxicity [8,52]. The inhibition mechanism and their inhibition efficiency (IE) depend on the active phytochemical compounds in them, and also the absorption mechanism is through three methods of chemisorption, physisorption, or a combination of the two (mixed) [8]. Table 3 reports the use of some plants as corrosion inhibitors in the sol-gel process.

Table 3. Use of plants in sol-gel hybrid coatings

Plants	Sol-Gel Solution Composition	Substrate	Corrosive medium	IE (%)	Ref.
Clitoria Ternatea	Mixing TEOS and GPTMS	MS	NaCl (3.5 wt.%)	86	18
Clitoria Ternatea	Mixing TEOS and GPTMS	Mild steel (MS)	HCl (0.5 M)	83.78	56
Caffeine extracted from tea leaves	Mixing TEOS and APTES	MS	NaCl (3.5 wt.%)	84.22	57
Ethanolic extract of Beta vulgaris	Mixing TEOS and APTES	MS	NaCl (3.5 wt.%)	83.03	58
Tea leaf aqueous extract	Mixing TEOS and APTES	MS	NaCl (3.5 wt.%)	85.66	59
Henna extract	Mixing TMSM and poly(methyl methacrylate)	Carbon steel (CS)	HCl (0.5 M)	95.6	60
Aqueous and ethanolic extracts of Mulberry	Mixing TEOS and GPTMS	Low-CS	NaCl (3.5 wt.%)	85.57	61
Henna extract	Polymerized vinyltrimethoxysilane	316L Stainless steel	SBF solution	92.53	62

4. Conclusions

In summary, silane-based sol-gel coatings were explored for their potential to protect metal structures, offering an alternative to toxic chromate-containing coatings. The sol-gel process has several advantages, including low processing temperatures, ease of operation, high chemical diversity, and the ability to coat various geometric shapes. Additionally, the use of plants as GCIs was reviewed. Given the acceptable performance of sol-gel coatings under different conditions and their environmental compatibility the adoption of these coatings is expected to increase in the future.

Conflict of Interest

The author declares no conflict of interest.

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