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Corrosion Inhibition using Green Inhibitors: An Overview

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Abstract: Corrosion is an inevitable phenomenon causing economic, time and material losses. Iron and its alloys are the most metallic materials used in industry and life uses. H^+ ion and dissolved oxygen - called natural motors - near H_2S and CO_2 in Oil and gas industries seem to be the major aggressive agents, leading to a horrible cost attaining some billion dollars… Researchers throughout history acted so that metals are used as long as possible. In this review, we report the use of inhibitors as one of the best ways of protecting metals and alloys against corrosion. Inhibitors acted by adsorption to create a film facing the arrival of aggressive agents on the metal surface. The presence of heteroatoms such as P, S, N and O… as well as the cyclic rings and unsaturated bonds, interact chemically and/or physically to avoid metal destruction. The environmental toxicity of organic corrosion inhibitors has prompted the search. Last decades, plant products were inexpensive, readily available, and renewable to be part of the so-called "green inhibitors." Their use is thanks to their environment-friendly character environmentally friendly and ecologically acceptable. The richness in tannins, alkaloids, organic amino acids and organic dyes of the plant becomes the principal to explain their reactivity as good/excellent inhibitors of corrosion.

Introduction

Corrosion includes the spontaneous attack of a different metal by chemical products in an aggressive environment, such as seawater, alkaline media and acids. It is a constant and continual problem that is often difficult to eliminate. (Ahmed ES and Ganesh, 2022 ; Fouda et al., 2014). Metals and alloys return to their original state after corrosion; in other words, it's the reverse process of metallurgy. The energy stored during the refining process is converted to kinetic energy during the deterioration stage leading the metal (or alloy) to its native state (oxides, chloride…) (Sun et al., 2022). The Pourbaix diagram for iron summarizes some of the most important zones of coexistence of iron states in binary pH-potential scale (Pourbaix, 2021). Three zones are possible: Immunity zone, where iron is thermodynamically stable. The second one corresponds to the oxidation of iron to Fe^{2+} or Fe^{3+} (corrosion region) and the third part, called passivation, is explained by forming a layer of oxides (solids such as Fe_2O_3) on the metal surface. Figure 1 shows the Pourbaix diagram of iron superposed to the region known as the stability of H_2O in the grew region of E-pH. In higher potential, water reacts toward O_2 and at lower potential becomes H_2. The principal anodic reaction of corrosion is the oxidation of iron:

$$Fe(s) \longrightarrow Fe2+ + 2e-$$

This cannot occur without the reduction of an aggressive agent such as H+ or dissolved oxygen O2(dis) according to the following cathodic reactions:

$$2H+ + 2e- \longrightarrow H2$$

$$O2(dis) + 4H+ + 4e- \longrightarrow 2H2O$$

The phenomena are often encountered in aerated acid solutions used in industrial acid cleaning, acid pickling, acid descaling, and oil well acidizing… (Al Hamzi et al., 2013 ; El Ouafi et al., 2002).

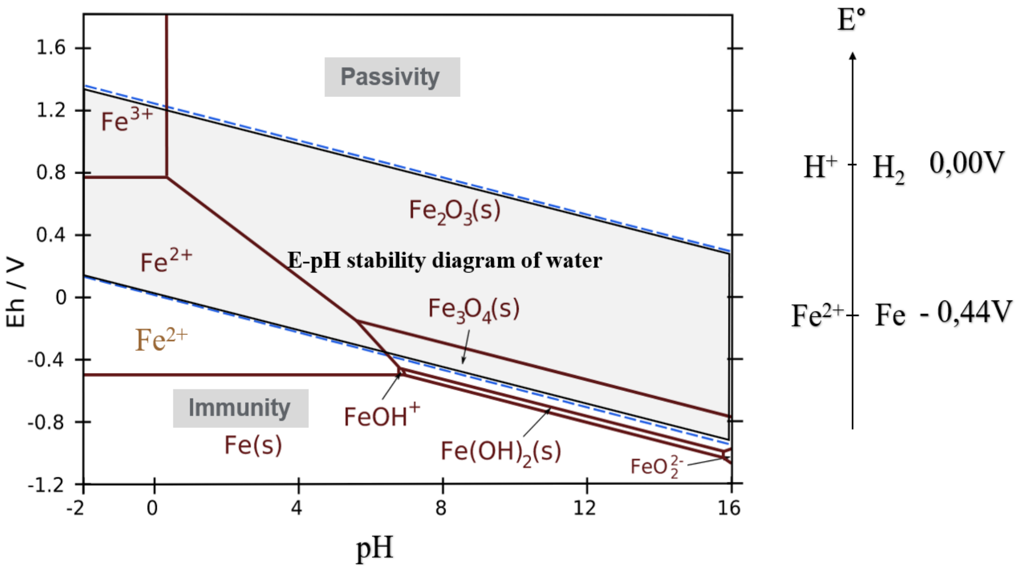

Figure 1. **E-pH diagram of iron (steel) and water**

Corrosion cannot be eliminated; it can be controlled and lessened to a great extent (Al Jahdaly et al., 2022 ; Bouklah et al., 2006). Corrosion is a natural process that is inevitable on the one hand and, on the other hand, has a financial burden on industry and amounts to trillions of dollars.

1. Corrosion Costs

To save a lot of money, industrial companies must commit to making maintenance or shutdown of production. Corrosion is thought to cost approx. 4% of GDP in industrialized countries (Koch, 2017 ; Javaherdashti, 2000). Uhlig proposed its method by defining corrosion cost as the total expenditure by manufacturing industries and corrosion-protection measures (Uhlig, 1950). Applying knowledge to draw the rational route of mechanisms can avoid many corrosion problems and greatly reduce the time and cost associated with corrosion maintenance and repair (ASM, 2000). In their recently published paper, Farh et al, 2023, state that the rehabilitation and repair of stray current corrosion damage to UK infrastructure cost approximately £550 million per year. Electrical insulation is required to give sufficient insulation between the tracks and the earth to avoid and control stray current corrosion. Rail fasteners can be effectively insulated from the tracks using plastic membrane, concrete track bed, PE jacket, waterproof coatings, grout slabs and wooden sleepers on high resilience neoprene pads. (Du et al., 2020 ; Riskin and Khentov, 2019)

(Mazumder, 2020), Table 1 presents the global economy into economic regions with similar economy categories collected from the World Bank data.

Table 1: Global cost (Billion US$) of corrosion by sectors and regions (Mazumder, 2020).

Economic Regions	sector					
	services	Industry	Agriculture	Total	Total GDP	%GDP
United States	146	303.2	2	4S1.3	16.270	2.7
Rest of the World	117.6	382.5	52.4	552.5	16.057	3.4
Russia	41.9	37.2	5.4	84.5	2.113	4
Arab World	92.6	34.2	13.3	140.1	2.789	5
India	32.3	20.3	17.7	70.3	1.670	4.2
European Region	297	401	3.5	701.5	18.331	3.8
Arab World	92.6	34.2	13.3	140.1	2.789	5
China	146.2	192.5	56.2	394.9	9.330	4.2
Four Asian Tigers Plus I Lacan	27.3	29.9	1.5	58,6	2.302	2.5
Japan	5.1	45.9	0.6	51.6	5.002	1
Global	906	1446.7	152.7	2505.4	74.314	3.4

2. General inhibitors

Among the easiest ways to protect corrosion is using inhibitors added at lower concentrations to block the corrosion process completely or partially by blocking the anodic and/or cathodic reactions. Inhibitors act via adsorption at the surface to create a layer facing the arrival of aggressive compounds.

Synthetic inhibitors have been widely used for many decades as azoles, pyridines, thiophenes, aldehydes, ketones, … (Njoku et al., 2021 ; Boulhaoua et al., 2021 ; Tourabi et al., 2013 ; Riskin and Khentov, 2019). Inhibitors decrease or retard the reactions at the metal/media interface. They reduce the corrosion rate by:

(i) adsorption of ions/molecules onto the metal surface,
(ii) decrease the reaction of the anodic and/or the cathodic,
(iii) decrease the rate of diffusion of the reagents towards the metal surface,
(iv) increase the surface electrical resistance of the metal.
(v) Inhibitors that are easy to apply and have the advantage of being applied in situ..

These actions at the metal surface occur via an active center capable of interacting physically or chemically with d-orbitals of metals.

3. Green Inhibitors.

The heteroatoms. O, N and S are organic inhibitors characterized by a basicity and electron density that are more important and act as corrosion inhibitors. O, N and S react as active centers of adsorption of the metal surface. Their effectiveness followed the following sequence O < N < S < P. generally, The chemicals which contain oxygen, nitrogen and sulfur used to decrease the corrosive effect of steel have been studied. Thus, the existing report shows that most organic inhibitors adsorb onto the metal surface by moving the water molecules to the surface and creating a compact barrier. The presence of even and p-electrons within the inhibitor molecules facilitates electron transfer from the inhibitors to the metal. A coordinated covalent bond involving electron transfer from the inhibitor to the metal surface can be formed. The intensity of the chemical bond depends on the density of the electrons, the functional group donor atom and the polarization of the group. When an H atom attached to the C-ring is substituted by a substitution group ($-NH_2$, $-NO_2$, -CHO, or -COOH), it increases inhibition. (Xhanari et al., 2017). The metal's electron density at the point of bonding changes, delaying the anodic or cathodic reactions. The electrons are consumed in the cathode and fed into the anode. So the corrosion is delayed. The increase in carbon chain length causes an increase in inhibition. This is attributed to the decrease in solubility in the aqueous solution with increasing hydrocarbon chain length. However, the hydrophilic functional group present in the substance favors the solubility of the inhibitors. The performance of an organic inhibitor depends on the physicochemical properties, chemical structure, p-orbital character, electron density at the donor atom, and the molecule's electronic structure.

4. Natural extract Inhibitors

Several categories of naturally occurring substances may be considered green corrosion inhibitors, particularly for steels in acid environments, including plant extracts, ionic fluids, amino acids, amino esters, peptides, drugs, polymers and rare earth elements. (Hammouti *et al.*, 1995; Aouniti, *et al.,* 2013b; Barouni, *et al.,* 2014; Abed *et al.,* 2004; Dohare, *et al.,* 2017). Plant extracts have been used extensively to provide corrosion inhibition for metal protection. The corrosion inhibition efficiency of these extracts is usually estimated by electrochemical tests, which include techniques such as potentiodynamic polarization, electrochemical impedance spectroscopy, and weight loss measurement. The inhibition efficacy of different concentrations of extracts is a useful indicator to gain a clear perspective to select an extract for a particular purpose.

4.1. Bgugaine isolated from Arisarum wlgareTarg.-Tozz

Bgugaine was isolated from Arisarum wlgareTarg.-Tozz. (Araceae), (Figure 2) which is widespread in Morocco and is named “irni (Belakhdar *et al.*, 1978; Benamed et al. 2009). This alkaloid fraction of the MeOH extract was successfully published since its efficiency reached 100% at 5 10^{-4}M (Hammouti *et al.* 1995). The electrochemical behavior of bgugaine Bg was studied by the polarization method as an iron corrosion inhibitor in the acidic medium of molar HCl. It was observed that its inhibition efficiency increased with concentration and reached a maximum value of 96% at 2 × 10-4 M. Polarization measurements also indicated that bgugain is a mixed type inhibitor. (Hammouti *et al.* 1997).

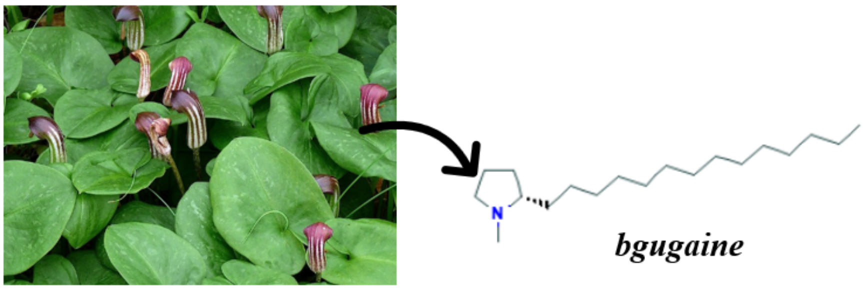

Figure 2. Arisarum wlgareTarg.-Tozz

4.2. Artemisia Herba Alba (AHA)

Artemisia herba-alba (AHA) extracts as green inhibitors were evaluated chiefly. The chemical variability of AHA from East Morocco was well documented (Paolini, et al., 2010). To this end, several works have shown that the aqueous extract of Artemisia Herba Alba (AHA) shows the corrosion behavior of the mild steel medium (MS) in HCl, H_2SO_4, which is a green inhibitor to decrease its corrosive action by exploiting a variety of techniques and methods namely: electrochemical techniques [electrochemical impedance spectroscopy (EIS), Weight loss and potentiodynamic polarization (PDP)], SEM-EDX, XPS, and theory calculations for the major molecules. (Bouyanzer *et al.*, 2004; Bouklah, *et al.,* 2006b; Bammou, *et al.,* 2011; Ouachikh *et al.,* 2009; Berrissoul, *et al.,* 2020a). Bouyanzer *et al.*, 2017, resumed using essential oil and extracts of Artemisia herba alba both in protecting metals against corrosion and other effects (antibacterial, antiviral, etc. Based on the richness of chemical compounds (1,6-Dimethylhepta-1,3,5-triene, Camphor, Thujone, Eucalyptol and Camphene...), Diass *et al.,* 2023 studied phytochemistry and determined the antioxidant activity. Theoretical docking study confirmed the superior activity of the following components: 1,6-dimethylhepta-1,3,5-triene, chrysanthone, eucalyptol, alpha-pinene against SARS-CoV-2 S-protein, as well as binding to SARS-CoV-2 S-protein 6CS2 in different ways and that the stable complex forming as a result of these interactions can inhibit the binding of CEA2 with the peak S-protein of SARS-CoV-2 (Figure 3).

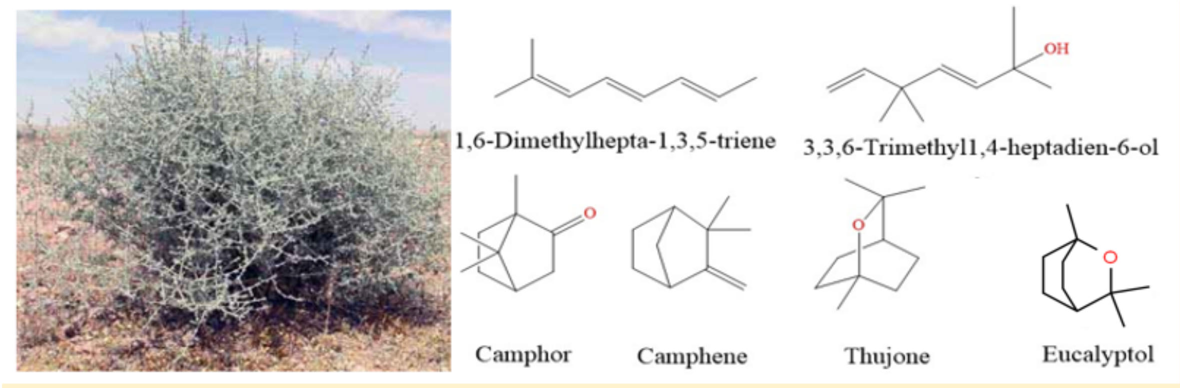

Figure 3. Artemisia Herba Alba (Diass *et al.,* 2023)

4.3. Lavandula species

According to Upson and Jury, 2002, eight native Moroccan species of *Lavandula L.* section Pterostoechas Ging., based on herbarium studies, were characterized by SEM photographs and field observations.

Due to its countless applications, steel is the most widely used metal for corrosion inhibition testing. This section delves into the most recent reports on plant extracts evaluated for use as corrosion inhibitors on steel. *Lavandula mairei Humbert* extracts (LM), The corrosion rate decreases using an ecological mild steel corrosion inhibitor (MS) in 1 M HCl, while the inhibition efficiency increases with increasing concentration of the pante extract (92% at 303 K) for a concentration of 0.4 g/L. The study is focused on weight loss and electrochemical measurements, scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS), (Berrissoul et al., 2020b). The other species, *Lavandula* dentata, officinalis, or stoechas exhibited good to excellent inhibitory effect on steel in an acidic medium (Bouammali *et al.*, 2013; Zerga *et al.*, 2009; Laqhaili *et al.*, 2013; El Ouadi *et al.*, 2014; Berrissoul *et al.*, 2020). The biological and pharmaceutical properties of the essential oils of *Lavandula* officinalis L. were largely discussed in the literature (Diass *et al.*, 2021). The characterization of lavender essential oil was investigated. The most important compounds are β-pinene (18.8%) and trans-pinocarveol (12.4%) (Figure 4) and the inhibitory efficiency of *Lavender LD* essential oil was estimated. The results showed that the inhibition efficiency increased with the inhibitor concentration at 1M in hydrochloric acid (HCl) up to 92% for a concentration of 5 mL/l oil (El Ouadi *et al.*). At the same time, what concerned the aqueous extract of the compound Quercetin-3-glucuronide was the major compound detected in this extract and it is shown high efficacy, as indicated previously (Berrissoul et al., 2020b).

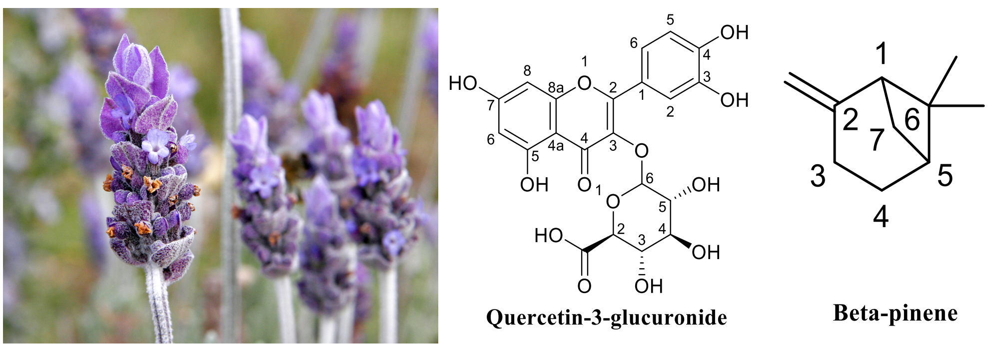

Figure 4. *Lavandula species*

4.4. Mentha Suaveolens

As discussed previously, all plant parts can be used to obtain extracts. For instance, (Hamdani et al., 2017; Salhi et al., 2017) coworkers studied the aqueous leaf extract of *Mentha suaveolens* (EAFMS). They have confirmed that the inhibitory effect of EAFMS aqueous extracts by gravimetric and electrochemical measurements has a powerful inhibitory action. The two aqueous extracts act as excellent inhibitors of mild steel corrosion in the two media, 1M HCl and 0.5M H_2SO_4, whose inhibitory efficiency increases with the concentration of the inhibitor with maximum efficiency for a concentration of 200 mg/L at 35°C of 89% of EAFMS in 1M HCl, respectively and 81% 88% in 1M HCl and 0.5M H_2SO_4. The presence of high amounts of Piperitenone oxide (44.3%) near Z-Piperitone oxide (19.1%), Terpinen-4-ol (3.8%), 1,2- Epoxymenthyl acetate (3,5%) and trans-hydrate Sabinene (3.1%) favored the *M. suaveolens* essential oil to play excellent protection for steel in sulfuric acid solution (Salhi et al., 2017) (Figure 5).

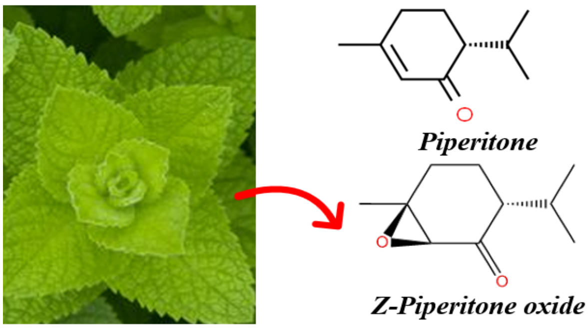

Figure 5. *Mentha Suaveolens*

4.5. Cistus monspeliensis

To achieve the description of corrosion inhibition provided by the techniques presented above, several authors, including (Mechbal *et al.,* 2021), wish to study the plant *Cistus monspeliensis* L (CM) as a green inhibitor of mild steel in hydrochloric acid solution. As previously mentioned, all plant parts can be used to obtain extracts (Figure 6). For example, the authors report that the electrochemical method has tested the CM aqueous extract. The obtained solutions show that the inhibition efficiency increases with the increase of the inhibitor dilution and the variation of the inhibition efficiency mainly depends on the mixture of bioactive compounds present in the CMAE (leaves, flowers and stems) at 0.2 g/l to reach 88%, 81% and 70%, respectively. The presence of aromatic rings and OH groups, ketones in the major compounds as gallic acid and quercetin increases the adsorption on metal surface (Mechbal *et al.,* 2021).

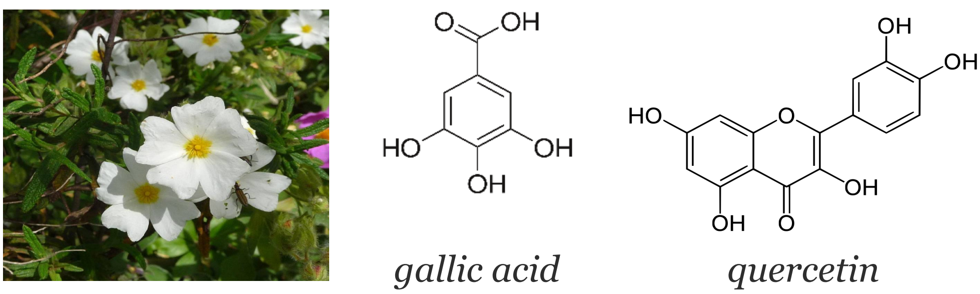

Figure 6. *Cistus monspeliensis*

4.6. Cannabis sativa L

Natural extracts have become one of the most eco-friendly sources of inhibitors because they provide high efficiency at a low cost. These inhibitors do not contain heavy metals. The effect of the corrosion inhibitor using Cannabis sativa L. seed oil grown in northern Morocco as a green inhibitor was also evaluated by (Damej et al., 2020). The authors showed that the efficacy of the corrosion inhibitor is 93,79% is increased with the concentration of the inhibitor with maximum efficiency for a concentration of 1g/L at 35°C. *Cannabis sativa L.* seed oil extract is an excellent protective effect of the green inhibitor. It is attributed to forming an inhibitor film on the surface of 60Cu-40Zn metallic brass in seawater. The use of the cannabis plant as a green inhibitor is not only focused on the seeds but also the leaves of this plant (Haldhar et al., 2021). used leave extract of the *Cannabis sativa* plant for the corrosion resistance of low carbon steel in the acidic medium at 0.5 M sulfuric acid. The latter has an exceptional inhibition efficiency of 97.31% at an inhibitor concentration of 200 mg/L utilizing the weight-loss method, Tafel and EIS. The authors also evaluated the phytochemical analysis of the *Cannabis sativa* plant. They found the following compounds Cannabinol, Cannabinoid and Cannabichromenic acid are the main compounds, which are generally responsible for this critical inhibition. Plant oils and extracts rich in active molecules have become a source of eco-friendly inhibitors that provide high corrosion inhibition performance at a low cost. So, (Damej et al., 2022), have worked on cannabis seed oils and reported that this oil is rich in bioactive compounds, with linoleic acid (51.3%), oleic acid (20.3%), and palmitic acid (7.9%) being the main compounds detected by gas chromatography (Haddou et al., 2022). Thus, based on weight loss and electrochemical results, an inhibition efficiency of more than 92% was achieved using 1 g/L of this oil in an acidic medium of 1 mol/L HCl (Figure 7).

Figure 7. *Cannabis sativa seed and linoleic acid*

Conclusion

It can be defined that green inhibitors are excellent inhibitors that protect the environment against corrosion for different metals due to their non-toxicity and biodegradability.

The dangerous effects of most synthetic organic inhibitors and restrictive environmental regulations are driving researchers to focus on the need to develop low-cost, non-toxic and eco-friendly natural products as effective corrosion inhibitors for many metals and steels in various aggressive environments.

The purpose of many researchers is to synthesize new formulations based on plant extracts and their use as green corrosion inhibitors. Plant extracts are considered a source of biosynthesis chemical compounds that simple procedures can extract at low cost, soluble and biodegradable in water, not environmentally and humanly harmful.

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Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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