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Green Corrosion Inhibitors

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Abstract

The corrosion inhibiting possibility examination of extracts obtained from fruits of rose hip and horse chestnut was investigated. Rose hip extracted at different temperatures exhibited different corrosion inhibiting effect. Rose hip obtained at 40^{o}C did not show any inhibiting effect in neutral pH medium, while in acidic media it expressed moderate protective impact (up to 55%) for steel and mild corrosion inhibiting impact for zinc (39%), while for aluminium and copper it did not have any inhibiting effect. Rose hip obtained at 25^{o}C exhibited significant inhibiting effect for steel (up to 62 %) and even better for zinc (up to 72%) in neutral media. For aluminium and copper neither of Rose hip extracts showed any inhibiting effect. The Horse chestnut extract exhibits moderate to significant inhibiting effect for steel (up to 55%) and zinc (up to 60%) in neutral media. In addition to this, because of the complex chemical composition of these fruit extracts, some further electrochemical investigation is suggested, aimed to determine a single component inhibitive effect. Firstly, as one of prevailing components, tannins are examined. Linear polarization resistance (LPR) and electrochemical impedance spectroscopy (EIS) were used as a testing techniques.

Key words: *Rose hip extracts, Horse chestnut extract, tannin, steel, zinc, aluminium.*

Introduction

It has been more than two decades since the first reported results on the investigation of corrosion inhibition effect of naturally occurring substances [1,2]. The interest increased over years, resulted in publishing investigations using very different substances which can be considered as green [3-10]. The rising consciousness of the necessity to protect the environment led to the interest in green technologies. Most of the highly effective corrosion inhibitors for steels and cast iron as key construction materials for industry and civil engineering are toxic and environmentally unacceptable. There is strong requirement for them to be replaced in near future. Sustainable and environmentally friendly approach for this replacement is to be based on the natural green inhibitors, able to substitute up to now used ones. Besides the request to be environmentally harmless, the green inhibitor should be effective, at least close to the already used. Finally, the price matters.

A number of highly effective corrosion inhibitors are withdrawn. Chromates and amines have been removed from the technically applicable inhibitors list due to their toxicity, the use of polyphosphates is still being reduced, while arsenic oxides are completely substituted [11]

Corrosion inhibitor is any chemical substance which, introduced to the corrosive environment (usually in small amounts), increases the resistance to corrosion of a metal [12]. Commonly used inhibitors were hydrazine, 2-mercaptobenzothiazole and sodium tripolyphosphate, up to now. According to Material Safety Data Sheet (MSDS), hydrazine is a highly reactive base and reducing agent. It is used for many industrial and military applications, even in household heating systems as a corrosion inhibitor. Besides corrosion inhibition, it is often necessary to obtain glossy surfaces in industrial facilities, which was associated with the application of toxic 2-mercaptobenzothiazole. It is identified also as a potential human carcinogen.

All of mentioned components and ingredients can find efficient substituents in natural products. It is known that citric acid vitamins, tannins, etc. can act in certain environment as efficient corrosion inhibitors of steels [3-9]. However, a broad research has been undertaken to examine various chemicals inhibitive effect on alluminium, copper, zinc and their alloys, as well as on other construction metallic materials [13-20].

These chemicals are typical constituents of most of herbal or fruits extracts and they have shown the corrosion inhibition effect in different experimental conditions. It has been a scientific quest for nontoxic, biodegradable i.e. environment friendly structure which can fulfill the aim at low concentration

Materials and Methods

Obtaining extracts from rose hip and horse chestnut fruits

The extraction of plant extracts is carried out by pressing, i.e. by grinding of plant fruits, followed by the dissolution phase with heating and simultaneous mixing, followed by extraction.

Horse Chestnut

The grinded fruit of chestnut is added to heated distilled water with a liquid/solid ratio of 20:1. To obtain the extract, a heating magnetic stirrer is used. The time to extraction depends on the temperature and concentration, as well as on the quality of the starting raw material. The extraction temperature was held below 60^{o}C, and the process of extraction lasted 3-5 h, depending on the lot.

After extraction phase, the obtained solution should be filtered. It is usual to obtain a heavy liquid, so the vacuum filtration technique had to be used. The obtained macerate was stored in a cool and dark place.

Rose Hip

The rose hip fruit was previously dried, at room temperature away from sunlight. The grinded fruit of dry rose hip is added to heated distilled water with a liquid/solid ratio of 20:1. In the preliminary testing phase, it was obvious that it is possible to obtain very different water extracts depending on the temperature and the rate of extraction. It is decided to obtain two different, low and high temperature extracts. The extraction process at 25 ˚C lasts for 24 h, while at the temperature at 40 ˚C it lasts for 5 h, with using a magnetic stirrer. After extraction phase, the obtained solution should be filtered. It is usual to obtain a heavy liquid, so the vacuum filtration is the best technique, as well. The obtained macerates were stored in a cool and dark place and marked as Rose Hip 25 and Rose Hip 40.

Electrochemical measurements

The electrochemical measurements were carried out in a standard three-electrode cell, the reference electrode was the saturated calomel electrode (SCE), the counter electrode was Pt-mesh, while the working electrodes were made of steel, copper, aluminum and zinc. Linear polarization resistance (LPR) and electrochemical impedance spectroscopy (EIS) measurements were carried out in solution made of 0,3 mol dm^{-3} NaCl and 0,1 mol dm^{-3} Na_2SO_4, under ambient conditions, without and with addition of fruit extracts. Concentration of extracts was in the range from 0,5-4 ml/130 ml of the cell. Measurements were performed by a GAMRY Reference 1010E Potentiostat /Galvanostat/ ZRA. Linear polarization resistance (Rp) was determined using Gamry software after measurement in the potentials range of -0.01 V – 0,01 V versus open circuit potential (Eoc). The active electrode surface (0.785 cm2) was wet polished with SiC papers (grit sizes 600-800-1000-2000) before each measurement. The polarization and EIS measurements were performed at different pH values of the electrolyte.

Results and discussion

In order to test the impact of these plant fruit extracts on different metals corrosion inhibiting possibility, the polarization resistance (Rp) was determined in acidic solution for pH=2 and pH=3 as well as in neutral pH=7 solution of 0,3 M NaCl + 0,1 M Na_2SO_4 for steel, copper, zinc and aluminium. Results are shown in Tables 1 for the cases where it has the inhibitive effect.
The corrosion inhibition efficiency, η_{inh} was calculated according to equation:

$$\eta_{inh} = (1 - R_{Me}/R_{inh}) * 100\%$$

Where R_{Me} and R_{inh} are determined values of polarization resistance (Rp) for metal without and with inhibitor, respectively.

Table 1. Determined values of corrosion inhibition efficiency for testing inhibitor: Rose Hip 40 ^oC

Metal	Concentration (ml/ml)	η_{inh}, pH7	η_{inh}, pH3	η_{inh}, pH2
Steel	0	0	0	0
	0,004	activating	24	28
	0,008	activating	30	39
	0,016	activating	53	43
	0,023	activating	54	48
	0,031	activating	48	55
Zinc	0	0	0	-
	0,004	20	neutral	-
	0,008	22	neutral	-
	0,016	39	neutral	-
	0,023	20	neutral	-
	0,031	activating	neutral	-

Rose hip 40 did not show any inhibiting impact at neutral pH, while in acidic media it expressed moderate protective impact (up to 55%) for steel and mild corrosion inhibiting impact for zinc (39%), while for aluminium and copper it did not have any inhibiting effect.
Rose hip 25 exibited significant inhibiting effect for steel (up to 62 %) and even better for zinc (up to 72%) in neutral media. The highest inhibiting effect was at medium concentration of inhibitor. As the inhibitor concentration increased, its protective effect decreased. For aluminium and copper it did not have any inhibiting effect. In the pH=3 solution, this extract showed no protective effect on any metal.
The Horse chestnut extract exhibits moderate to significant inhibiting effect for steel (up to 55%) and zinc (up to 60%) in neutral media. The highest inhibitive effect was for highest added inhibitor concentration. For aluminium and copper it did not have any inhibiting effect. In the pH=3 solution, this extract showed no protective effect on any metal.

Table 2. Determined values of corrosion inhibition efficiency for testing inhibitor Rose Hip 25 ^{o}C

Metal	Concentration (ml/ml)	η_{inh}, pH7	η_{inh}, pH3	η_{inh}, pH2
Steel	0	0	0	-
	0,004	51	neutral	-
	0,008	32	neutral	-
	0,016	62	neutral	-
	0,023	55	neutral	-
	0,031	54	neutral	-
Zinc	0	0	0	-
	0,004	67	neutral	-
	0,008	56	neutral	-
	0,016	72	neutral	-
	0,023	30	neutral	-
	0,031	12	neutral	-

Table 3. Determined values of corrosion inhibition efficiency for testing inhibitor Horse Chestnut: 60 ^{o}C

Metal	Concentration (ml/ml)	η_{inh}, pH7	η_{inh}, pH3	η_{inh}, pH2
Steel	0	0	0	-
	0,004	13	neutral	-
	0,008	23	neutral	-
	0,016	48	neutral	-
	0,023	43	neutral	-
	0,031	55	neutral	-
Zinc	0	0	0	-
	0,004	5	neutral	-
	0,008	4	neutral	-
	0,016	17	neutral	-
	0,023	21	neutral	-
	0,031	60	neutral	-

There are a lot of very different substances in such an extract, but the search for promising corrosion inhibitors is among those with lone electron pair. Analytical techniques such as gas chromatography, column chromatography, high-performance liquid chromatography (HPLC) and nuclear magnetic resonance (NMR) spectroscopy were used for the analysis and quantification of volatile compounds, for identification a single chemical compound in a mixture, or for separation and quantification each component i.e. for determining the content and purity of a sample as well as its molecular structure [21-23]. To illustrate the Rose hip and Horse chestnut extracts complexity, the containing substances are listed in Tables 4a and b.

Tables 4 a and b. Chemical composition of Horse chestnut and Rose hip extracts

Horse Chestnut sp.

Chemical composition	Content in 100 g
Water	60.21 g
Phenol, total	785.25 mgGAE/g
Simple phenols	37.82 mgGAE/g
Tannins	747.43 mgGAE/g
Flavonoids	47.0 mgGAE/g
Sugar, total	20.23 mg
Starch	27.83 mg
Minerals	
Calcium (Ca)	19 mg
Iron (Fe)	0.94 mg
Magnesium (Mg)	30 mg
Phosphor(P)	38 mg
Potassium (K)	484 mg
Sodium (Na)	2 mg
Zinc (Zn)	0.49 mg
Vitamins	
Vitamin A	1µg
Thiamin B1	0.144 mg
Riboflavin B2	0.016 mg
Niacin B3	1.102 mg
Pantothenic acid B6	0.352 mg
Folate B9	58 µg
Vitamin B12	0 µg
Vitamin C	40.2mg

This may lead to different mechanisms of corrosion inhibition, with inevitable cross-impact, even sometimes the opposite influence. Besides, some of the components exhibits corrosion activation effect, while some of them acts neutral.

According to the this analysis of Rose hip extracts, the highest content among listed substances in Rose hip extract belongs to vitamin C, indicating that it might be principal active component. Likewise, the analysis of Horse chestnut extracts shows that the highest content among listed substances in this extract belongs to tannins.

Tannins (or tannoids) are a class of astringent, polyphenolic biomolecules and the term is applied to any large polyphenolic compound containing sufficient hydroxyls and other suitable groups (such as carboxyls) to form strong complexes with various macromolecules. The tannin compounds can be find in many plant species and have been already tested on corrosion inhibitive properties. The two types of tannin is examined in this work, so called condensed tannin and tannin blank. The results are presented in Tables 5-8, only for inhibitin effects that can be taken into account.

Rose hip (Rosa Canina sp.)

Chemical composition	Content in 100 g
Water	58.66 g
Proteins	1.6 g
Lipids, total	0.34 g
Carbo hydrates	38.22 g
Sugar, total	2.58 g
Minerals	
Calcium (Ca)	169 mg
Iron (Fe)	1.06 mg
Magnesium (Mg)	69 mg
Phosphor(P)	61 mg
Potassium (K)	429 mg
Sodium (Na)	4 mg
Zinc (Zn)	0.25 mg
Copper (Cu)	0.113 mg
Manganese (Mn)	1.02 mg
Vitamins	
Vitamin C,	426 mg
Thiamin	0.016 mg
Riboflavin	0.166 mg
Niacin	1.3 mg
Pantothenic acid B6	0.8 mg
Vitamin A, RAE	217 µg
Carotene, beta	2350 µg
Carotene, alpha	31 µg
Cryptoxanthin, beta	483 µg
Vitamin A	4345 IU
Lycopene	6800 µg
Lutein + zeaxanthin	2001 µg
Vitamin E (alpha-tocopherol)	5.84 mg
Tocopherol, beta	0.05 mg
Tocopherol, gamma	1.34 mg
Tocopherol, delta	0.14 mg
Vitamin K (phylloquinone)	25.9 µg

Table 5 Determined values of corrosion inhibition efficiency of condensed tannin at pH=2 for steel

Concentration (ml/ml)	E_{kor} (mV)	R_p (Ω cm^2)	η (%)
0	-556	246	-
0,25	-552	314	21,7
0,50	-546	391	37,1
1,0	-541	444	45,7
1,5	-536	467	47,3
2,0	-532	490	49,8

Table 6 Determined values of corrosion inhibition efficiency of tannin blanc, pH=2 for steel

Concentration (ml/ml)	E_{kor} (mV)	R_p (Ω cm2)	η (%)
0	-560	210	-
0,25	-550	277	24,2
0,50	-546	331	36,6
1,0	-541	366	42,6
1,5	-540	436	51,8
2,0	-538	536	60,8

Table 7 Determined values of corrosion inhibition efficiency of condensed tannin, pH=7 for zinc

Concentration (ml/ml)	E_{kor} (mV)	R_p (Ω cm2)	η (%)
0	-1084	429	-
0,25	-1082	707	39,3
0,50	-1074	758	43,4
1,0	-1073	1007	57,4
1,5	-1081	858	50,0

Table 8 Determined values of corrosion inhibition efficiency of tannin blanc, pH=7 for copper

Concentration (ml/ml	E_{kor} (mV)	R_p (Ω cm2)	η (%)
0	-201	3338	-
0,5	-201	3970	15,9
1,0	-217	5750	42,0
2,0	-214	5991	44,3
3,0	-213	5816	42,6

For further information concerning the interaction nature between the inhibitor molecules and the metal surface, as well as among inhibitor molecules themselves, adsorption isotherm should be determined.

Conclusion

- Corrosion inhibiting impact of Rose hip 40:
    - in neutral pH not inhibiting impact at all,
    - in acidic pH moderate protective impact for steel and mild for Zn, but for Al and Cu not inhibiting effect.
- Corrosion inhibiting impact of Rose hip 25:
    - in neutral pH significant inhibiting effect for steel and even better for zinc in neutral media. The highest inhibiting effect at medium inhibitor concentration.
    - For Al and Cu do not have any inhibiting effect.
    - In acidic pH, this extract showed no protective effect on any metal.
- Corrosion inhibiting impact of Horse chestnut extract
    - in neutral pH exhibits moderate to significant inhibiting effect for steel and zinc. The highest inhibitive effect was for highest added inhibitor concentration.
    - For Al and Cu this extract do not have any inhibiting effect.
    - In acidic pH, this extract showed no protective effect on any metal.

- Corrosion inhibiting impact of tannin blank is moderate for Cu in neutral pH and significant for steel in acidic pH.
- Corrosion inhibiting impact of condensed tannin is moderate to significant for steel in acidic pH and for Zn in neutral pH.

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