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Žbulj, Katarina; Bilić, Gordana; Hrnčević, Lidia; Simon, Katarina

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The Potential of Using Plant Extracts as Green Corrosion Inhibitors in the Petroleum Industry

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Katarina Žbulj¹; Gordana Bilić¹; Lidia Hrnčević¹; Katarina Simon¹

¹ University of Zagreb, Faculty of Mining, Geology and Petroleum Engineering, Pierottijeva 6, 10000 Zagreb

Abstract

In this paper, preliminary studies of ten (10) different plant extracts as potential corrosion inhibitors of carbon steel were examined. For each extract, the concentration range in which they show anti-corrosion action was first determined, and then the most effective concentration was determined for each extract. The tests were performed in a brine solution saturated with CO₂ at room temperature. The aim of this study was to isolate extracts with high effectiveness and subsequent electrochemical and surface methods to determine the mechanism of inhibitory action. For this purpose, potentiodynamic polarization was performed with Tafel extrapolation. Among all the tested extracts, lady's mantle (92.17%) and dandelion root (95.07%) stood out with their inhibitor efficiencies (IE). Both tested extracts showed the behaviour of a mixed corrosion inhibitor with a dominant influence on the anode process.

Keywords:

CO₂ corrosion; petroleum industry; carbon steel; plant extracts; green corrosion inhibitors

1. Introduction

From its very beginning, the petroleum industry has been facing the problem of corrosion since most of the equipment and pipelines in the petroleum industry are made of carbon steel. Through the process of oil and gas production, gathering, treatment and transportation, most of the process equipment and pipelines are made of carbon steel. Hydrocarbon production, depending on the type of the reservoir, also implies the production of a certain amount of brine and other impurities (dissolved gasses, sand, additives applied during production, etc.). With the production progress, i.e. the maturing of the production field, the share of the produced brine increases. As mentioned, the produced fluid can also contain, among other things, carbon dioxide (CO₂) and hydrogen sulphide (H₂S), which, dissolved in water (brine) cause corrosion and damage to the equipment.

In general, carbon steel is susceptible to corrosion. The steel most sensitive to corrosion is low carbon steel, which is mostly used in the petroleum industry. Low carbon steel is the carbon steel that contains less than 0.8% of carbon (Byars, 1999). Corrosion can be defined as the destruction of a material caused by an aggressive environment to which that material is exposed (Sastri, 2011). The consequences of corrosion in the petroleum industry could affect the environment (potential cause of a fluid spill) and also, due to equipment damage, but environ-

mental impacts also have a large economic impact. In 2014, in the IMPACT report (International Measures of Prevention, Application, and Economics of Corrosion Technologies Study) estimations of the global cost caused by corrosion were given. It was reported that the global costs due to corrosion are around 2.5×10^{12} USD, which is about 3.4% of global Gross Domestic Product (GDP) (Koch et al., 2016).

In systems where water is present, electrochemical corrosion will occur. As already mentioned, flowlines are the most sensitive part of an oil and gas transportation system. Fluid, transported in flowlines, is not yet treated, it contains brine and impurities, such as carbon dioxide. Flowlines, gathering pipelines and waterlines are mostly made of low carbon steel, and that makes them susceptible to CO₂ corrosion.

According to the data from CONCAWE (CONserva-tion of Clean Air and Water in Europe) (Cech et al., 2021), during a five-year period (2015–2019), there were nine spill incidents caused by corrosion. Corrosion is the cause of the largest volume of spill, and has thus the biggest negative impact, with 233 m³ of average spillage in comparison with mechanical failure (193 m³ of average spillage) and operational causes (17 m³ of average spillage).

One of the ways to control the corrosion problem is with the application of corrosion inhibitors. Since there are some limitations in the usage of conventional corrosion inhibitors due to their toxicity, plant extracts, among other things, have been studied as so-called *green corrosion inhibitors*. These green corrosion inhibitors could

Corresponding author: Gordana Bilić
gordana.bilic@rgn.unizg.hr

Table 1: Plant extracts as green corrosion inhibitors tested in different media on different types of metals

Metal	Corrosion medium	Plant extract as corrosion inhibitor	IE (%)	Reference
Mild steel	HCl	<i>Artemisia pallens</i> extract	93 – 98	Garai et al., 2012
Carbon steel	HCl	<i>Osmanthus fragrans</i> extract leaf	90	Li et al., 2012
Carbon steel	HCl	Grape pomace extract	97	da Rocha et al., 2012
Mild steel	HCl	<i>Lavandula dentata</i> extract	95	Bouammali et al., 2013
Mild steel	H ₂ SO ₄	<i>Argemone mexicana</i> leaf extract	80 – 92.5	Ji et al., 2013
Copper	HCl	Mangrove tannin	87.6	Shah et al., 2013
Copper	H ₂ SO ₄	<i>Myrtus communis</i>	> 85	Bozorg et al., 2014
Mild steel	H ₂ SO ₄	Marigold flower extract	98.07	Mourya et al., 2014
Carbon steel	HCl	<i>Opuntia ficus indica</i> extract	91	Flores-De los Rios et al., 2015
Carbon steel	H ₂ SO ₄ HCl	Rice husk extract	94.24	Olawale et al., 2017
Carbon steel	carbon dioxide saturated chloride carbonate solution	Olive leaf extract	> 90	Pustaj, 2017
Mild steel	sodium chloride solution	<i>Persian Liquorice</i> extract	98.8	Alibakhshi et al., 2018
Mild steel	HCl	Coffee husk extract	93.9	Cordeiro et al., 2018
Low carbon steel	HCl	<i>Prosopis juliflora</i> plant extract	91.5	Fouda et al., 2018
Low carbon steel	HCl	<i>Xanthium strumarium</i> leaf extract	94.82	Khadom et al., 2018
X70 steel	HCl	<i>Ginkgo</i> leaf extract	92.5	Qiang et al., 2018
Mild steel	H ₂ SO ₄	Saraca ashoka extract	95.48	Saxena et al., 2018a
Mild steel	H ₂ SO ₄	<i>Sida cordifolia</i> extract	99	Saxena et al., 2018b
Stainless steel	HCl	<i>Taraxacum officinale</i> leaf extract	91.5	Ugi et al., 2018
Copper	HCl	Alchemilla Vulgaris extract	> 95	Ahmed i Zhang, 2019
Mild steel	3.5% NaCl	<i>Allium sativum</i> (garlic) extract	92	Devikala et al., 2019a
Mild steel	3.5% NaCl	Asafoetida extract	90	Devikala et al., 2019b
Mild steel	HCl	<i>Citrullus lanatus</i> fruit extract	91	Dehghani et al., 2019
Mild steel	HCl	Carrot Peel extract	88.08	Saeed et al., 2020
Carbon steel	seawater	<i>Taraxacum officinale</i> extract	> 88.2	Deyab i Guibal, 2020
Mild steel	HCl	<i>Paederia foetida</i> leaf extract	73.77	Hossain et al., 2021

have a great potential with their application in the petroleum industry.

Due to the maturity of the production fields and the obsolescence of the infrastructure, the corrosion problem is one of the major issues in the Croatian petroleum industry. The parts of the oil and gas gathering and transportation system that are most exposed to corrosive fluid are the flowlines, because the produced fluid, which is transported via flowlines, is still untreated, and the pipelines for extracted brine transport. Regarding the hydrocarbon gathering system pipeline leaks in Croatia, the data have been analysed for the period between 2010 and 2019. For the analysis, only fluid leaks with spill volume equal to or larger than 1 m³ were considered. The analysis has shown that corrosion was the main

cause of pipeline leaks with an approximate share of 71% in total number of leaks. The analysed data indicated the highest frequency of leaks occurred in flowlines (INA d.d., 2021). The corrosion protection of these pipelines is usually done with the application of conventional organic corrosion inhibitors, which, due to their toxicity, have environmental limitations.

Alternatively, so-called green inhibitors, that can be, compared to conventional inhibitors, equally effective and are biodegradable and affordable, are being tested (Sastri, 2011; Pustaj, 2017; Goni i Mazumder, 2019). Due to their non-toxicity and biodegradability, the green corrosion inhibitors are environmentally acceptable (Montemor, 2016; Shehata et al., 2018; Popoola, 2019). Currently, the most researched green corrosion

Table 2: Chemical composition of the steel sample (wt.%)

C	Si	Mn	P	S	Cr	Mo	Ni	Cu	Fe
0.32	0.25	1.38	0.016	0.009	0.24	<0.01	0.02	0.01	balance

inhibitors are plant extracts. In **Table 1**, some of the plant extracts already researched as green corrosion inhibitors are listed. As can be seen from **Table 1**, most research was conducted in an acidic corrosion medium, while the experiments in this paper refer to a nearly neutral medium with the presence of CO₂.

In order to examine their effectiveness as corrosion inhibitors, extracts of some wild plants that can be found in Croatia were tested. For the overview of wild self-seeded plants, Grlic's encyclopaedia was used (Grlić, 1990). In the research, ten commercially available plant extracts were selected as potential carbon steel corrosion inhibitors in a simulated brine solution saturated with CO₂. The extracts were selected based on their chemical composition, more specifically the active components they contain (flavonoids, saponins, polyphenols), which represent potential adsorption centres in adsorption on a metal surface. In this paper, the preliminary results of the laboratory research conducted as part of a doctoral thesis are given. The aim of the conducted laboratory research was to select certain plants based on their corrosion inhibition efficiency and to examine the possibility of their application as green corrosion inhibitors in the petroleum industry. Further research on the subject will include comprehensive laboratory research, which should include experiments in dynamic conditions, using specific electrochemical and surface methods, and biodegradability testing for determining the behaviour of the chosen plant extracts as corrosion inhibitors.

2. Materials and methods

The carbon steel sample used in the corrosion experiments is a steel sample that flowlines and brine pipelines are composed of. In **Table 2**, the chemical composition of the steel sample is shown. The chemical composition of the steel sample was determined by a GDS 850 A, LECO spectrometer using the optical emission spectrometry method.

The brine used in the research was prepared as a solution of sodium chloride (NaCl), sodium bicarbonate (NaHCO₃) and calcium carbonate (CaCO₃), which are all the degree of purity of Pro Analysis by Sigma Aldrich. For each electrochemical measurement conducted, one litre of fresh solution was prepared, with or without the addition of different concentrations of plant extracts as corrosion inhibitors. The chemical composition of the simulated brine solution is given in **Table 3**.

For preliminary research, ten plant extracts were chosen:

- *Punica granatum* (pomegranate) (seeds, fruit, peel),
- *Melissa officinalis* (lemon balm) leaves,

Table 3: Chemical composition of the simulated brine solution

Chemical compound	γ (g/L)
NaCl	30.0
NaHCO ₃	0.1
CaCO ₃	0.1

- *Vitis vinifera*, *Vitis* (grape pomace),
- *Taraxacum officinale* (dandelion) root,
- *Passiflora incarnata* L. (passionflower) (leaves and flowers),
- *Arctium lappa* L. (burdock) root,
- *Glycyrrhiza glabra* L. (liquorice) root,
- *Crataegus oxyacantha* (hawthorn) (leaves and flowers),
- *Sempervivum tectorum* L. (houseleek),
- *Alchemilla vulgaris* (lady's mantle) leaves.

The plant extracts were added to the brine solution in various concentrations, varying from 100 mg/L to even 20 000 mg/L, based on the available literature on the previously conducted research. Before every measurement, the prepared solution was saturated with carbon dioxide for 45 minutes, and continuously stirred at 300 rpm during electrochemical measurement. Measurements were conducted at room temperature. The experiments were conducted by using three-electrode corro-

**Figure 1:** Three electrode corrosion cell

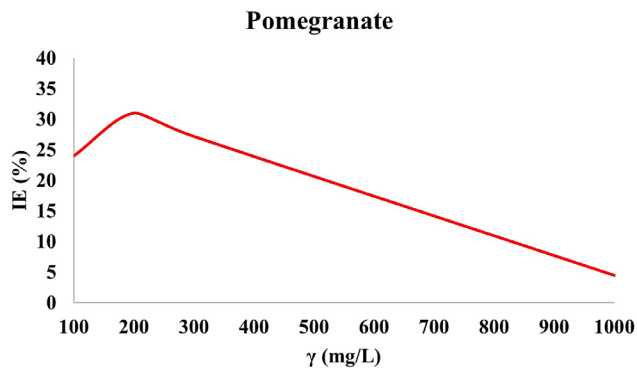


Figure 2: Inhibitor efficiency of pomegranate

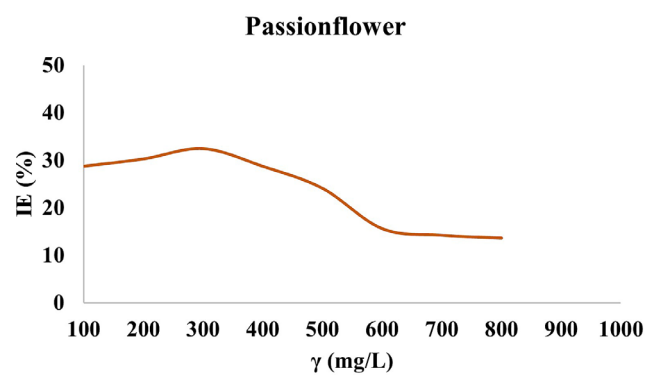


Figure 6: Inhibitor efficiency of passionflower

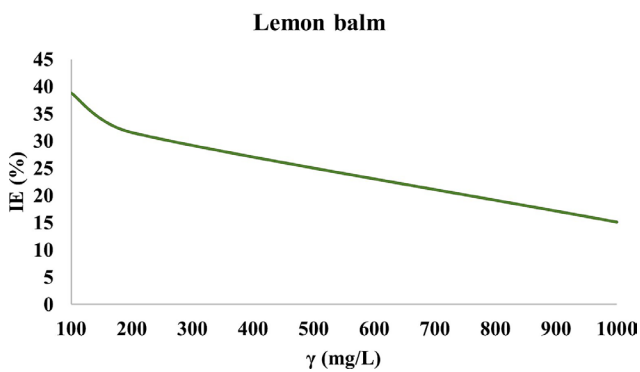


Figure 3: Inhibitor efficiency of lemon balm

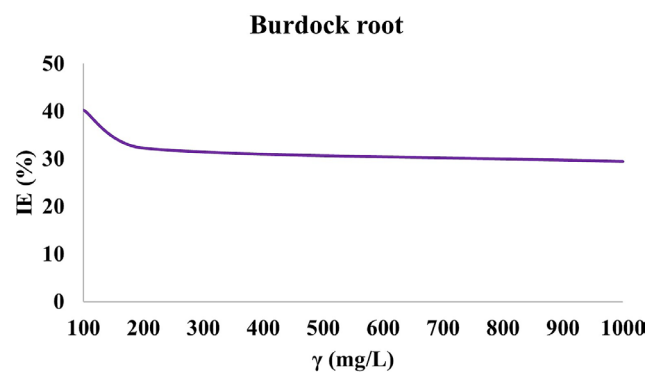


Figure 7: Inhibitor efficiency of burdock root

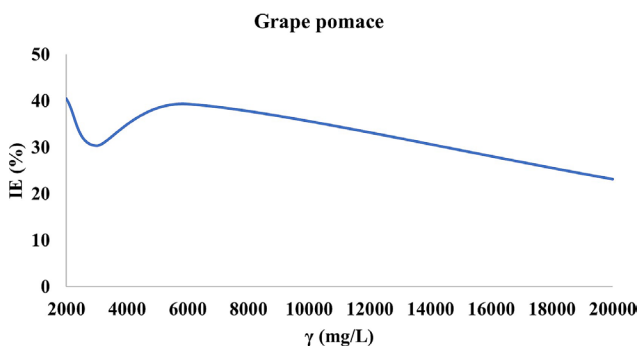


Figure 4: Inhibitor efficiency of grape pomace

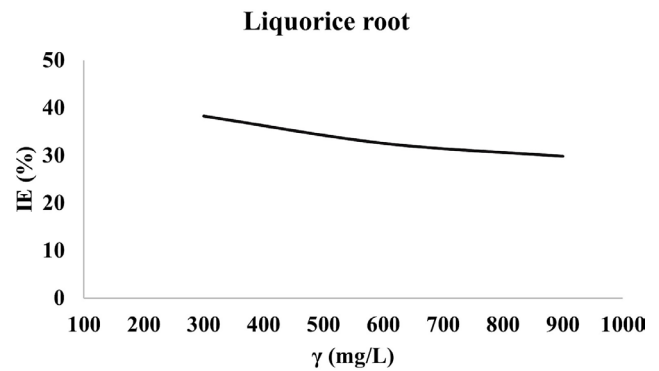


Figure 8: Inhibitor efficiency of liquorice root

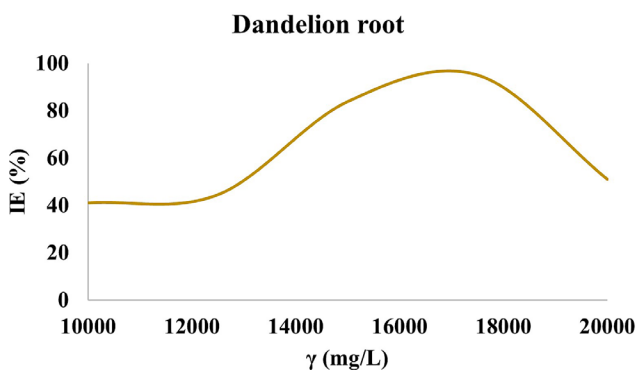


Figure 5: Inhibitor efficiency of dandelion root

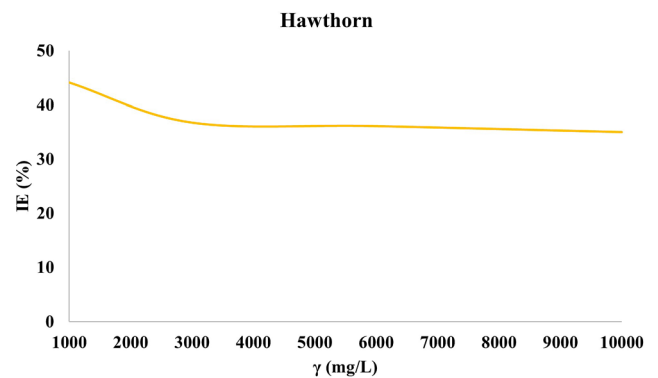


Figure 9: Inhibitor efficiency of hawthorn

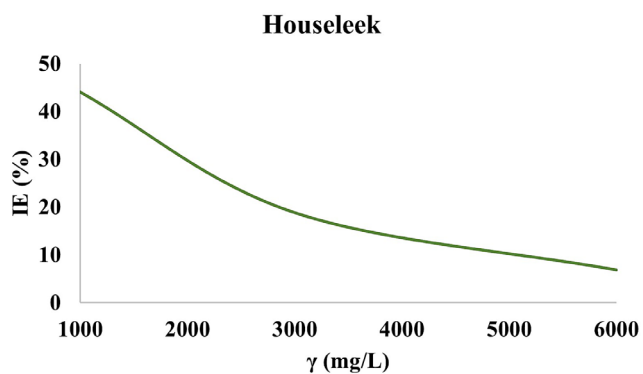


Figure 10: Inhibitor efficiency of houseleek

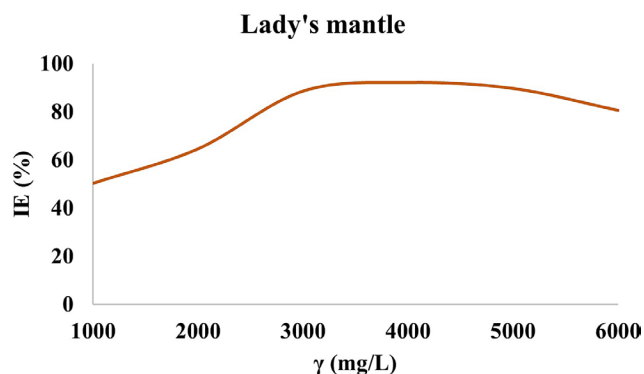
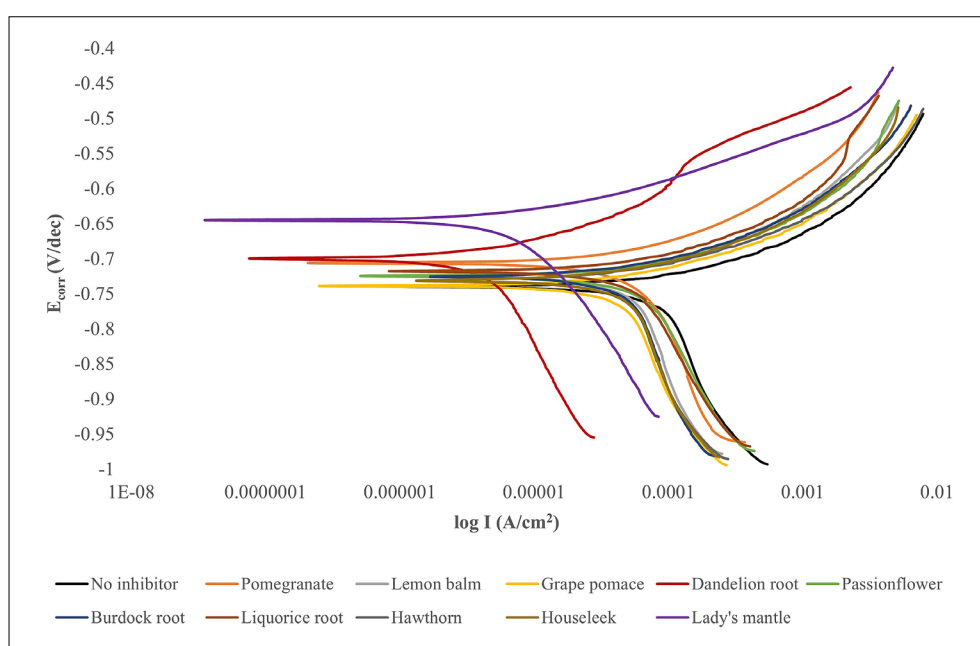


Figure 11: Inhibitor efficiency of lady's mantle

Figure 12: Tafel polarization curves for steel sample in brine solution saturated with CO₂, without and with the most efficient concentration of different plant extractsTable 4: Electrochemical parameters for steel sample in brine solution saturated with CO₂, with and without the most efficient concentration of different plant extracts

	γ (mg/L)	E_{kor} (mV)	j_{kor} ($\mu\text{A}/\text{cm}^2$)	$-\beta_k$ (V/dec)	β_a (V/dec)	v_{kor} (mm/y)
No inhibitor	-	-741	97.627	0.517	0.065	1.136
Pomegranate	200	-708	67.351	0.413	0.093	0.784
Lemon balm	100	-725	59.702	0.469	0.068	0.695
Grape pomace	1000	-738	50.880	0.459	0.060	0.592
Dandelion root	17500	-699	4.809	0.324	0.062	0.056
Passionflower	300	-726	65.918	0.365	0.063	0.767
Burdock root	100	-726	58.372	0.535	0.068	0.679
Liquorice root	300	-720	60.312	0.348	0.071	0.702
Hawthorn	1000	-732	54.540	0.494	0.061	0.635
Houseleek	1000	-732	54.581	0.419	0.067	0.635
Lady's mantle	4000	-663	7.690	0.172	0.037	0.09

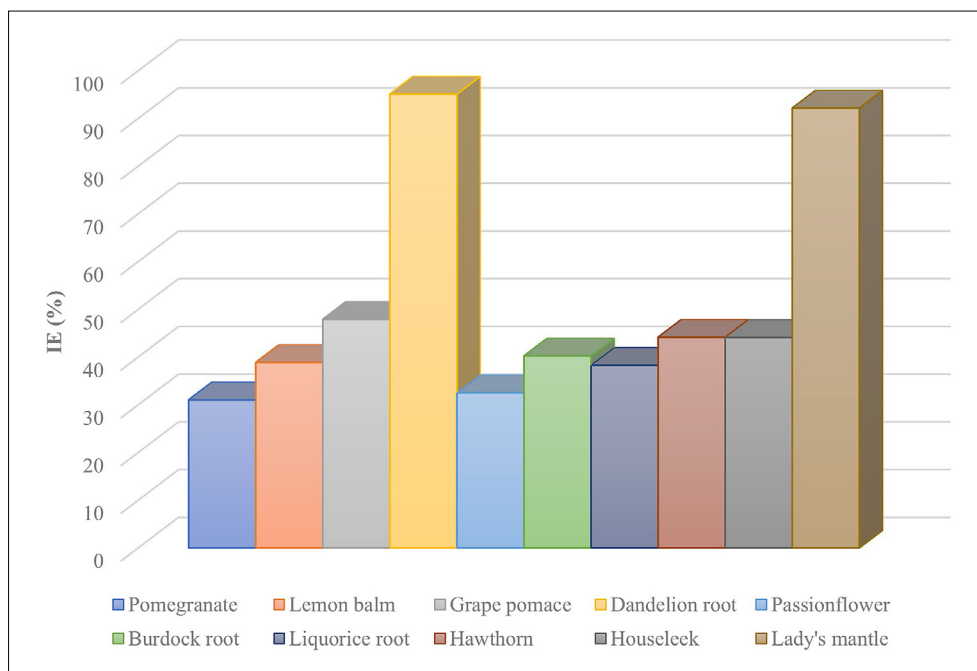


Figure 13: Highest corrosion inhibitor efficiency of each plant extract

Table 5: Highest corrosion inhibitor efficiency of each plant extract with corresponding concentration

Plant extract	γ (mg/L)	Highest IE (%)
Pomegranate	200	31
Lemon balm	100	38.84
Grape pomace	1000	47.88
Dandelion root	17500	95.07
Passionflower	300	32.47
Burdock root	100	40.2
Liquorice	300	38.22
Hawthorn	1000	44.13
Houseleek	1000	44.09
Lady's mantle	4000	92.12

sion cells (shown in **Figure 1**). The carbon steel sample was used as the working electrode, with 1 cm² area exposed to the brine solution. Before every measurement, the carbon steel sample was sanded with 360, 600 and 1200 grit paper, washed with distilled water and degreased with 96% ethanol. Two graphite rods were used as the counter electrode and a saturated calomel electrode as the reference electrode.

All measurements were performed using an SP1 potentiostat and SmartManger software with IVMAN programme for analysing data. Prior to polarization measurement, the working electrode was stabilised at open circuit potential for 1 hour. For Tafel polarization curves, the corrosion potential was set to ± 250 mV versus open circuit potential with a scan rate of 0.166 mV/s. For determining the corrosion rate and inhibition efficiency of various concentrations of plant extracts, polarization

measurements (with Tafel extrapolation) were performed. Each measurement was repeated three times for reproducibility.

3. Results and discussion

In order to examine the inhibitory activity and determine the concentrations of the extracts at which they show the highest effectiveness, potentiodynamic polarization by Tafel extrapolation was performed. Polarization curves were recorded for all the mentioned extracts in a certain range of concentrations. However, due to the large number of tested extracts, as well as better visibility of the results, all scanned polarization curves are not shown, but rather the graphical dependence of inhibitor efficiency (IE) on extract concentration (γ) (see **Figure 2 to Figure 11**). Inhibitor efficiency can be calculated as shown in **Equation 1** (Sastri, 2011):

$$IE (\%) = \frac{v_{kor}^o - v_{kor}^{inh}}{v_{kor}^o} \times 100\% \quad (1)$$

Where:

IE – inhibitor efficiency (%),

v_{kor}^o – corrosion rate in solution without inhibitor (mm/year),

v_{kor}^{inh} – corrosion rate in solution with inhibitor (mm/year).

As can be seen from **Figures 2 to 11**, for most of the tested plant extracts, the corrosion inhibitor efficiency was around 40%, but lady's mantle and dandelion root stood out with an inhibitor efficiency of more than 90%. Tafel polarization curves for the most efficient concentration of each plant extract in comparison with the brine

solution without an inhibitor is shown in **Figure 12**. Electrochemical parameters determined from Tafel extrapolation are listed in **Table 4**.

It can be seen from **Table 4** that with each tested plant extract, the corrosion potential (E_{kor}) has moved towards positive values, which could indicate the formation of a protective film on the steel surface. The change in corrosion potential was below 85 mV. Also, for all plant extracts, there is a decrease in corrosion current density (j_{kor}) which indicates lower corrosion rates. The presence of plant extract in the simulated brine solution, in the case of all plant extracts, except burdock root extract, results in a decrease of the value of the cathodic Tafel slope (β_k), which points to a diminishing effect of diffusion on the cathodic process (**Equation 2**):



Regarding anodic Tafel slopes (β_a), there is a small change in values in brine solutions containing plant extracts. This could be a sign of inhibitor blocking active sites preventing a reaction to occur. Taking everything stated into consideration, it can be concluded that the researched plant extracts are behaving as mixed-type corrosion inhibitors for low carbon steel in a brine solution saturated with CO_2 , thus slowing down both corrosion reactions with a main influence on the anode process (**Equation 3**):



The highest corrosion inhibition efficiency of each plant extract is shown in **Figure 13**. A list of the plant extracts with corresponding concentrations at which the highest efficiency is achieved is given in **Table 5**.

According to **Figure 13** and **Table 5**, two plant extracts stood out – dandelion root (inhibitor efficiency of 95.07% at a concentration of 17 500 mg/L) and lady's mantle (inhibitor efficiency of 92.12% at a concentration of 4000 mg/L). To get a better insight in their behaviour and adsorption on a metal surface, further electrochemical and surface analysis research will be conducted.

5. Conclusions

In this paper, a review of previously conducted laboratory research of green inhibitors' efficiency is shown, as well as the results of the laboratory research of some plant extracts that can be found in Croatia. The plant extracts were studied as potential inhibitors of carbon steel corrosion in a simulated brine solution saturated with CO_2 . The aim of this study was to isolate extracts of high inhibitor efficiency (more than 90%), which, if their effectiveness was confirmed by subsequent tests, could be successfully applied in the protection of carbon steel in hydrocarbon production and transportation systems in Croatia. The study showed a significant decrease of cor-

rosion rate in the cases of the dandelion root extract (0,056 mm/y) and the lady's mantle extract (0.09 mm/y)) compared to the uninhibited system (1.136 mm/y). Potentiodynamic polarization with Tafel extrapolation determined inhibitor efficiencies of 92.17% for the lady's mantle extract at a concentration of 4000 mg/L, and 95.07% for the dandelion root extract at a concentration of 17500 mg/L. Both tested extracts showed a small change in corrosion potential compared to the uninhibited system (less than 85 mV), which points out to their behaviour as mixed-type inhibitors, as retarding both anodic and cathodic corrosion reactions.

The mentioned extracts, that showed the best inhibitor efficiency (the dandelion root extract and the lady's mantle extract), will be further tested by electrochemical impedance spectroscopy (EIS), and surface methods (FTIR - Fourier-transform infrared spectroscopy and SEM - Scanning electron microscopy) to determine their mechanism of action under the test conditions. The same measurements will be conducted in dynamic conditions. To examine the effect of the chosen plant extracts on the environment, biodegradability and toxicity tests will be performed.

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SAŽETAK

Mogućnost primjene biljnih ekstrakata kao zelenih inhibitora korozije u naftnoj industriji

U ovome radu provedena su preliminarna ispitivanja deset različitih biljnih ekstrakata kao potencijalnih korozivskih inhibitora ugljičnoga čelika. Za svaki ekstrakt najprije je određen raspon koncentracija u kojemu pokazuje antikorozijsko djelovanje, a potom je za svaki ekstrakt određena najdjelotvornija koncentracija. Ispitivanja su provedena u sintetskoj slojnoj vodi zasićenoj s CO₂ pri sobnoj temperaturi. Cilj rada bio je izdvojiti ekstrakte s visokim djelotvornostima te naknadnim elektrokemijskim i površinskim metodama utvrditi mehanizam inhibitorškoga djelovanja. U tu svrhu provedena je potenciodinamička polarizacija s Tafelovom ekstrapolacijom. Od svih ispitanih ekstrakata sa svojim djelotvornostima izdvajaju se vrkuta (92,17 %) i korijen maslačka (95,07 %). Oba ispitana ekstrakta pokazala su ponašanje mješovitoga korozivskog inhibitora s dominantnim utjecajem na anodni proces.

Ključne riječi:

CO₂ korozija, naftna industrija, ugljični čelik, biljni ekstrakti, zeleni inhibitori korozije

Author's contribution

This paper is a part of the PhD research of the author **Katarina Žbulj** (PhD student, graduate engineer of petroleum engineering) who initialized the idea, lead the laboratory research, and participated in the interpretation of the laboratory results. The supervisor of the PhD thesis, **Gordana Bilić** (PhD, Assistant Professor) participated in the determination of research methods, supervised laboratory research, participated in the interpretation of the results, in the discussion of the results and made a critical revision of the paper. The supervisor of the PhD thesis, **Lidija Hrnčević** (PhD, Associate Professor) participated in the interpretation of the results, in the discussion of the results and made a critical revision of the paper. **Katarina Simon** (PhD, Full Professor) participated in the interpretation of the results, in the discussion of the results and made a critical revision of the paper. All the authors participated in writing the paper, as well as defining future research.