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# INVESTIGATING THE PLANT EXTRACTS PERFORMANCE AS ALTERNATIVE CORROSION INHIBITORS AGAINST SALINE MEDIA IN PETROLIUM INDUSTRIES: A MINI REVIEW

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## ABSTRACT

Corrosion inhibitors are considered one of the best techniques used to control the corrosion problem, but on the other hand, they pose environmental and economic burdens that constantly drive the search for alternatives, especially considering the environmental awareness that makes environmental safety a priority in modern society. So, this paper aims to review some of the recent literature to investigate the ability of plant extracts to inhibit corrosion in the oil industry, which is controlled with massive amounts of chemical inhibitors annually. Since low-carbon steel is the leading metal in this industry, the study of its corrosion behavior under the influence of these green inhibitors was selected as the starting point. Studies conducted in similar corrosive environments (NaCl environments) were particularly reviewed for more accurate comparisons and conclusions. The research discussed the factors affecting the performance of the plant-based inhibitors, starting from the effect of the preparation stage until the application conditions (extraction technique, inhibitor concentration, temperature, exposure time, velocity, and pH), besides some basic related theories. It was found that many plant extracts showed acceptable performance under different conditions, but there were some gaps and challenges that needed to be highlighted by future studies.

**Keywords: Corrosion, low carbon steel, NaCl, green inhibitors**

## INTRODUCTION

One of the problems known to specialists is the corrosion that affects all industrial sectors and constitutes a potential threat and danger to safety, the economy, and the environment unless it is constantly controlled.

Corrosion in oil industries constitutes the most difficult challenge of the corrosion problem due to the harsh operating conditions and the environment with high concentrations of salts, sulfur, and acids, in addition to water, microorganisms, and other potential corrosion causes (Groysman, 2014; Montemor, 2016). Therefore, only the internal corrosion of oil pipelines is responsible for more than half of the total corrosion accidents and the loss of billions of dollars annually around the world, which prompts the expenditure of tons of corrosion inhibitors to control this problem to the maximum extent possible (Hamied et al., 2018; Montemor, 2016; Tamalmani & Husin, 2020).

Corrosion inhibitors are among the most efficient techniques used to control internal corrosion since the 1930's (Askari et al., 2021), but in fact they constitute a burden in a number of other important aspects, as most of them are carcinogens, toxic, and unsafe for the environment (Aliofkhazraei, 2014; Pinzón-Espinosa & Kanda, 2020; Poste et al., 2014; Vachtsevanos et al., 2020), in addition to their high costs, which make them economically undesirable solution (Montemor, 2016). These obstacles constitute a source of concern considering continuous industrial development, which is expected to lead to an increase in corrosion problems in the future. This is what prompted many researchers to search for alternative corrosion inhibitors.

Although green chemistry is a huge reservoir of many possible alternatives, plants are the most attractive due to their abundance and biological origin (Alrefaee et al., 2021; Zakeri et al., 2022), in addition to containing many polar and electron-rich functional groups with the ability to create physical or chemical interactions with the metallic surface, thus protecting it (Alrefaee et al., 2021).

A large number of literary articles are constantly being published in this regard and are usefully reviewed from time to time by researchers. (Yang, 2021) rehearsal the latest studies on plant inhibitors used in acidic environments and their role in sustainable society. (Salleh et al., 2021) conducted a review of the performance of plant inhibitors in relation to ferrous alloys and discussed the general principles in the methodologies of the latest research. (Zakeri et al., 2022) reviewed a collection of recent landmark research and discussed the advantages and disadvantages of plant extracts in corrosion inhibition. While we aim in this research paper to review the performance of plant extracts in corrosion inhibition in oil industries, which constitutes an environmental and economic obstacle that must be seriously looked at to be solved in alternative ways, So this paper deals with the impact of the most important factors on the performance of these inhibitors, starting with preparation and ending with application, as a way to employ research towards discovering the related gaps and challenges and covering the most prominent developments that pave the way to actual production of plant-based inhibitors.

**Salts In Crude Oil**

Crude oil contains varying proportions of sodium chloride, magnesium chloride, and calcium chloride salts depending on the type of crude, often at 75%, 15%, and 10%, respectively (Groysman, 2014; Rajendran & Santhana, 2014). Calcium and magnesium chlorides are hydrolyzed to hydrochloric acid (Equations 1 and 2) in the presence of water and at high temperatures (Groysman, 2014).

$$MgCl_2 + H_2O \longrightarrow Mg(OH)Cl + HCl \quad (1)$$

$$CaCl2 + H_2O \longrightarrow Ca(OH)Cl + HCl \quad (2)$$

pH values decrease to 2 or less (Groysman, 2014). So, this saline environment is considered one of the most aggressively corrosive media. While the presence of 3.5% NaCl or less contributes to reducing the solution's electrical resistance, thus increasing the corrosion current (Groysman, 2009).

## THE CHEMICL INHIBITORS

Corrosion inhibitors are substances that, when added in a small amount (ppm), are capable of reducing corrosiveness to the maximum extent possible (Aliofkhazraei, 2014; Grassino et al., 2021; Pourzarghan & Fazeli-Nasab, 2021; Roberge, 2019; Zakeri et al., 2022). They protect the metal surface by several mechanisms, depending on the type of inhibitor.

The chemical inhibitor is required to be highly efficient, available in components, effective in different conditions, stable at high temperatures, renewable, non-toxic, and biodegradable, and must not interact with components or tend to form a foam or emulsion (Dewangan et al., 2021; Shang & Zhu, 2021). Most chemical inhibitors, such as chromate, phosphate, lead, azole derivatives, and others (Akbarzadeh et al., 2020) are approved industrially under strict environmental rules due to their toxicity and carcinogenic nature (Aliofkhazraei, 2014; Pinzón-Espinosa & Kanda, 2020; Poste et al., 2014; Vachtsevanos et al., 2020). However, many of them are not completely subject to these rules (Aliofkhazraei, 2014). Inhibitors can be classified on different bases. The classification based on their work mechanisms may be the most comprehensive, as shown in Fig. 1.

### Adsorption of Corrosion Inhibitors

Although the inhibitors work according to various mechanisms that are not limited to adsorption, but it the basic principle for the work of most of them, especially if we talk about plant-based inhibitors, so it is necessary to closely identify how the inhibitors adsorbed on the surface and what is the most prominent theories that adopt the interpretation of this.

The inhibitor's molecules bind to the surface as a result of the interaction energy among them, which exceeds the water molecules-metal interaction energy; this enables them to occupy their places as shown by equation (3) below (Sastri, 2012; Sastri et al., 2007):

$$M(nH_2O)_{ads} + I \longrightarrow MI_{ads} + nH_2O \quad (3)$$

Where: M= metal, n= moles number, I= inhibitor.

Metal surface nature, its charge, and the inhibitor composition determine adsorption's type, which can be chemical, physical, or a combination between them (Laamari et al., 2011; Sastri, 2012; Sastri et al., 2007). Chemical adsorption occurs when there are heterogeneous atoms and electron-rich regions as a result of a chemical reaction between π-electrons or uncharged electrons and the metallic-surface atoms, while physical adsorption occurs because of the hydrostatic attraction of the metal surface to the polar molecules (Sastri, 2012; Sastri et al., 2007). Non-polar molecules act as a buffer for corrosive molecules, which means that a protective layer is formed (Aliofkhazraei, 2014; Sastri, 2012; Sastri et al., 2007). Fig. 2 illustrates how the metal can be isolated from the corrosive environment by the protective film adsorbed on a surface.

## Adsorption Isotherm

The adsorption isotherm enables us to identify the nature of the interaction between the adsorbed particle and the surface, predict what happens in the formed film, and deduce several important adsorption parameters. The adsorption isotherm is represented by graphic curves describing the relationship between the degree of adsorption and some influencing variables such as concentration, pressure, time, etc. Table 1 summarizes the most important theories adopted to study corrosion inhibitor adsorption and their equations (Ituen et al., 2017). The parameters in the indicated equations can be calculated according to the following relationships (Kumar et al., 2021):

$$\theta = \frac{EI}{100} \quad (4)$$

$K_{ads}$ relate to the adsorption Gibbs free energy as follows:

$$\Delta G_{ads} = -RT \ln(55.5K_{ads}) \quad (5)$$

ΔH can be determined by Van't Hoff relation and ΔS can be determined by the following thermodynamic low (Kumar et al., 2021):

$$\Delta G_{ads} = \Delta H_{ads} - T\,\Delta S_{ads} \quad (6)$$

Where: **C** = concentration, $\mathbf{K_{ads}}$ = equilibrium adsorption-desorption constant, **θ** = the coverage **α** = parameter of molecular interaction, **n** = empirical constant, **x** = a constant related the $H_2O$ molecules number (displaced by the inhibitor), **EI**= inhibitor efficiency, **R**= gas constant, **T** = temperature, $\mathbf{G_{ads}}$= adsorption Gibbs energy, $\mathbf{H_{ads}}$= adsorption enthalpy energy, $\mathbf{S_{ads}}$= adsorption entropy energy.

## Plant-Based Inhibitors

Plants are among the greenest sources that attract attention as alternative corrosion inhibitors. In addition to being available and environmentally friendly, most of them are rich in active groups such as polar components (S, O, N, P and Se atoms) (Zakeri et al., 2022), non-polar components (aliphatic chains, aromatic rings and heterocyclic, etc.) and many heterogeneous atoms, which all have a very good ability to adsorb on the metal-surface (Hossain et al., 2021). What makes them studyable inhibition sources in whether in the oil industries or other industries like concrete, vapor phase corrosion, cooling systems, electrical industries and many more (Payal & Jain, 2021). Many plant extracts have successfully inhibited corrosion with high efficiencies, as can be seen in Table 2, making them promise regenerative inhibitors.

## FACTORS AFFECTING INHIBITION EFFECIENCY

Like other industrial inhibitors, plant-inhibitors efficiency is affected by specific factors, starting from inhibitor preparation, and ending with different operating conditions, as shown in Fig. 3.

### Extraction Technique

Extraction techniques can be divided into two classes: conventional techniques (such as maceration, soxhlet extraction, and hydrodistillation) and modern techniques (UAE, PEF, EAE, MAE, PLE, and SFE) (Azmir et al., 2013), each with many advantages and disadvantages. In general, "modern extraction techniques" lead to a higher-quality product with less energy, less time, less solvent, and less waste, unlike other methods that require more energy consumption, longer time, and more solvent quantities to reach an equivalent quality (Gupta et al., 2012). (Liu et al., 2020) compared four extracts prepared with different techniques and found that the one prepared with the help of ultrasound for one hour gave four times better performance than the other three extracts prepared by soaking for 24 hours under different conditions. Table 3 presents a summary of the most important modern extraction methods, their principles, and their strengths that make them eligible to be exploited in this regard.

Besides the extraction technique itself, there are many factors related to it that decide the composition and the final properties of the product, namely the solvent used, solute-solvent ratio, extraction conditions, extraction time, and other possible influencers at this stage. In terms of solvents, water, ethanol, methanol, acetone, and sodium hydroxide are the most common used for extraction. The solubility of active groups varies in plant cells; some of them have a tendency to dissolve within the polar solvents, while some of them are vice versa, so it's required to carefully choose the suitable solvent. (Wang et al., 2008) compared the performance of ethanol, methanol, and acetone in the extraction of phenolic substances. The results showed that the largest percentage of phenols was obtained by ethanol, while the lowest percentage was obtained by acetone. It should be noted that mixing more than one substance as a solvent may lead to better results due to its synergistic effect (Gupta et al., 2012), while the use of a high-purity solvent may lead to denaturation of protein cells and thus the difficulty of extracting them (Medina-Torres et al., 2017). In addition, the extraction period plays a key role in determining the product quality; the long time may be positive or negative, depending on the target sample and the extraction technique used. For example, excessive exposure time to ultrasound waves can cause the decomposition of some of the active components in the extract, which decreases its efficiency. This, of course, highlights the importance of accurately identifying the nature of the target components.

### Concentration

The inhibitor's effectiveness increases as its concentration increases. The higher concentration means higher surface coverage (Verma et al., 2022). This result has been reached by many researchers, such as (Heakal et al., 2018), (Devikala, Kamaraj, Arthanareeswari, & Pavithra, 2019), and (Alibakhshi et al., 2019) who demonstrated corrosion inhibition efficiency increases of Persian licorice extracts, Asafoetida, and Centaurea cyanus aqueous with their concentrations (respectively). However, an excessive increase in some inhibitor concentrations does not only lead to a constant efficiency, but even a decrease in it. This was observed, for example, with the lignin extract concentration rising (Muzakir et al., 2019) and Elettaria cardamomum extract (Shyamvarnan et al., 2021). The stabilization of the inhibitor efficiency, despite the increase in its concentration, indicates that the reaction of the inhibitor on the metal surface has reached equilibrium. The decrease in efficiency with increased concentration is mostly related to the increase in protective film thickness with time, as more time allows the adsorption of more bio-layers on the metal surface, especially for the highly polar extracts that can form multiple layers during adsorption, which weakens the adhesion of the adsorbed layer to the metal surface and makes it more susceptible to desorption.

### Temperature

The high temperature is negatively affecting chemical corrosion inhibitors in general, as many of them fail at less than 200 $^{o}$C (Heidersbach, 2018). This raises some concern about the possibility of sustaining the performance of green inhibitors in oil industries with extremely aggressive environments. It was observed that the performance of Centaurea cyanus aqueous extract (Heakal et al., 2018) and lignin extract (Muzakir et al., 2019) decreased in inhibiting low-carbon steel corrosion with increasing temperature. This is as a result of their molecules' increased kinetic energy, which prevents them from adhering to the metallic surface, along with the temperature effect on the corrosion rate (Berrissoul et al., 2020; Verma et al., 2022). However, the film-forming inhibitor's behavior under temperature influence depends on how it is attached to the surface. As is known, chemical adsorption is stronger at high temperatures (Sastri, 2012; Sastri et al., 2007), which can increase the efficiency of chemically adsorbed inhibitors, while the opposite is true for physical adsorption. For instance, (Noor, 2007) and (Fouda et al., 2017) found that higher temperatures enhanced the adsorption of an aqueous extract of Fenugreek leaves on the mild steel surface and Salicornia behoove extract on the aluminum surface, respectively, and this was inferred from the significant increase in inhibition efficiency with temperature.

### Exposure Time

A sufficient exposure time is necessary to obtain the maximum efficiency from the inhibitor. Therefore, increasing exposure time is mostly considered to have a positive effect on the inhibition process (Ghaedi, 2021). This effect has been observed on a number of plant-based inhibitors, such as Persian licorice extract (Alibakhshi et al., 2019), Centaurea cyanus aqueous extract (Heakal et al., 2018), and Juglans regia shell (Haddadi et al., 2019). Longer exposure times often allow for the sedimentation of a larger number of active functional groups, which promote the adsorbed protective film. However, exceeding the critical time can cause a gradual efficiency decrease; for example, the results reached by (Shyamvarnan et al., 2021) demonstrated a reduction in Elettaria cardamomum extract performance because of the inhibitor molecules' looseness and weak nature of absorption on the surface. (Muzakir et al., 2019) also noticed a loss of more than 10% of the lignin extract inhibition efficiency after 30 days.

At the same time, some researchers reached different results. For example, (Bahlakeh et al., 2019) noticed a gradual decline in the eucalyptus extract's performance up to 24 hours, then it improved. But in fact, it is likely that the efficiency increase after this period is not related to the inhibitor behavior but rather to the development of a thin layer of corrosion products, shielding the metal from its corrosive surroundings.

### Flow Velocity

Velocity is one of the most severe negative factors affecting the performance of film-forming inhibitors through two main aspects: First, velocity increases the corrosion rate by increasing the oxygen in the medium. The relative movement of the corrosive solution leads to a decrease in the thickness of the hydrodynamic boundary layer and the diffusion layer through which oxygen diffuses to the metal surface; this means lower resistance to oxygen transport and thus a higher percentage of it, according to equations (7) and (8). Oxygen penetrates the protective layer and interferes with its components. So, most inhibitors can’t work efficiently at more than 0.5 ppm of oxygen (Heidersbach, 2018).

$$No_2 = K\ Co_2 \quad (7)$$

$$\text{Or } No_2 = (D/\delta)\ Co_2 \quad (8)$$

Where: $\mathbf{No_2}$ = moles of oxygen, **K** = mass transfer coefficient, $\mathbf{Co_2}$ = oxygen concentration, **D** = diffusivity of dissolved oxygen, and **δ** = diffusion layer thickness.

Secondly, the shear stress displaces the protective layer formed by the inhibitor on the metal surface (Heidersbach, 2018). One of the requirements for industrially approved chemical inhibitors today is that they are effective under the influence of 20 m/s (Saji & Umoren, 2020). This is what necessitates taking this factor into account when evaluating green inhibitors.

**pH**

The pH affects in a complex and variable manner, positively or negatively depending on the type and composition of the inhibitor. For example, each commercial inhibitor works within a certain pH range; exceeding it can cause the inhibitor to fail completely. For instance, sodium benzoate is ineffective when the pH drops below 5.5 (Saji & Umoren, 2020). This confirms the importance of investigating the performance of efficient plant-based inhibitors under the influence of different pH ranges.

Further, hydrogen ions have a complex effect on the rate of dissolution of the metals; the corrosion rate of low-carbon steel is often increased with pH decrease because of the increased metal dissolution through the interaction of ($H^+$) with Fe, as shown in the following equation (Africa, 2008):

$$Fe + H^+ = Fe^{2+} + 2e^- \qquad (9)$$

On the other hand, the high acidity impedes the formation of a negative passive layer of corrosion products, unlike what happens in the basic media as a layer of ferrous hydroxide $Fe(OH)_2$ is created, followed by a more stable layer of magnetite $Fe_3O_4$, preventing corrosion except in exposed places that could experience localized corrosion, as the following equations show (Kazemipour et al., 2020; Omran et al., 2022):

$$Fe^{+2} + 2\,H_2O \longrightarrow Fe\,(OH)_2 + 2H^+ \qquad (10)$$

$$3Fe\,(OH)_2 \longrightarrow Fe_3O_4 + 2\,H_2O + 2H^+ + 2e \qquad (12)$$

$$2H^+ + 2e \longrightarrow H_2 \qquad (13)$$

$$3Fe\,(OH)_2 \longrightarrow Fe_3O_4 + 2H_2O + H_2 \qquad (14)$$

## CONCLUSIONS

Many plant-based extracts show superior performance in inhibiting the corrosion of mild steel; this is a strong motivation for developing them to withstand the harshest possible level of operating conditions and thus consuming them as renewable green sources to inhibit corrosion in the oil industries. The most important influences are summarized as follows:

i. The extraction technique, with its various conditions, plays the first and most key role in determining the composition and properties of the resulting extract and its ability to withstand the influence of several factors later.

ii. The performance of most plant extracts is positively impacted by increasing their dose in the corrosive medium, considering the critical values of some types, which exceeding leads to a harmful increase in the thickness of the adsorbed protective layer.

iii. The chemical adsorption improved with increasing temperature, which gives the extracts that can be chemically adsorbed on the target metal surface the priority of being developed as corrosion inhibitors in oil industries characterized by high temperatures. Employing high temperatures to enhance the extract's performance is also an important way to avoid the effectiveness declining under velocity influence.

## KNOWLEDGE GAPS AND RECOMMINDATIONS

i. There is an urgent need to move away from conventional extraction methods and exploit the studies to expand on the modern techniques, which are most industrially suitable. It should also be expanded in the study of the influential factors during the extraction, without being limited to the study of operating conditions.

ii. Intensifying studies on diagnosing the contribution percentage of each functional group within the extract in the inhibition process can open broad horizons for developing stronger solvents that help in obtaining double-quality extracts.

iii. Most of the plant-based inhibitor are evaluated at a static system. This is a preliminary, not final, evaluation. Therefore, more attention should be paid to investigating the performance of these inhibitors under velocity influence as a prelude to applying them in operating units.

iv. It is important to study the effects of often-overlooked factors, including pH, oxygen, and higher temperatures, as this is a necessary means to discover more weaknesses and search for appropriate solutions.

v. Optimization of experimental conditions by adopting modern methodologies that consider the interaction of factors to simulate the actual corrosive environments and thus reach more accurate results.

vi. Consider the economic aspect to encourage investors to go towards producing green inhibitors.

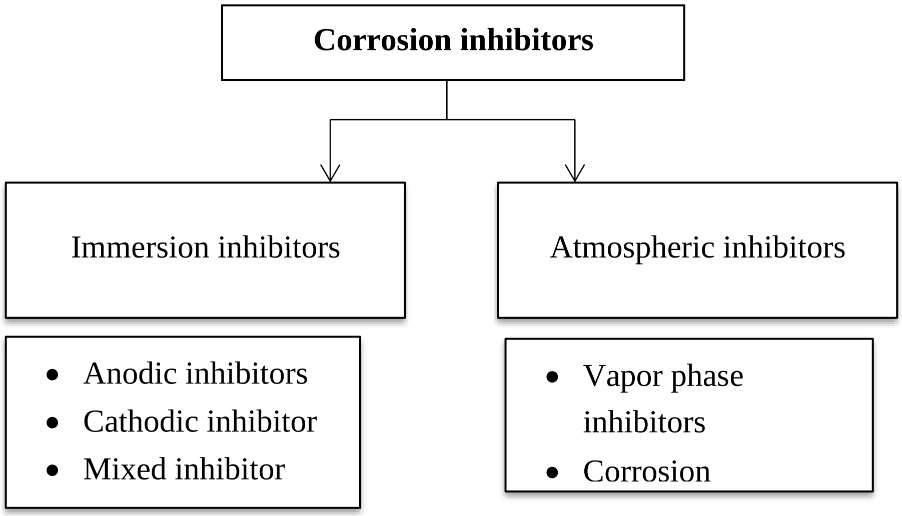


Fig. 1: Classification of corrosion inhibitors [reformed from source (Roberge, 2019)].

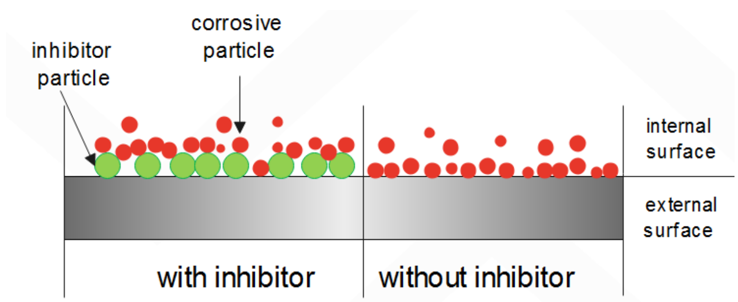


Fig. 2: Adsorption of corrosion inhibitor particles on the surface.

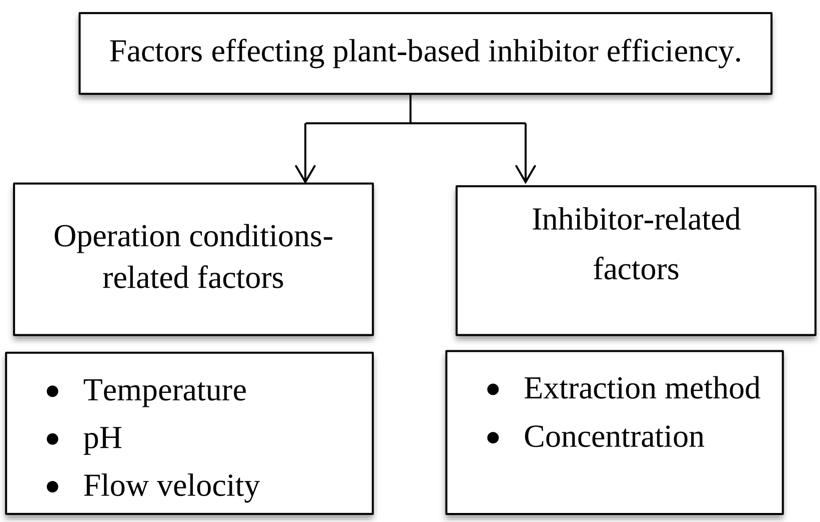


Fig. 3: Factors effecting plant-derived inhibitor performance.

Table 1. Adsorption isotherm models

| Model | Representative equation |
|---|---|
| Langmuir | $\frac{C}{\theta} = \frac{1}{Kads} + C$ |
| Temkin | $e^{-2\alpha\theta} = K_{ads}C$ |
| Freundlich | $\theta = K_{ads}C^{1/n}$ |
| Flory-Huggins | $\theta = \frac{KadsC}{X - KadsC}$ |

Table 2. Data on some plant-based-extracts used to inhibit low-carbon-steel corrosion in NaCl environment: plant, extraction method, extraction solvent and efficiency.

| Plant | Extraction technique | Solvent | Eff.% | Source |
|---|---|---|---|---|
| **Catharanthus roseus** | ultrasound-aided extraction | ethanol | 70 | (Palaniappan et al., 2020) |
| **Platanus acerifolia** | ultrasound-aided extraction | ethanol+ NaOH | > 99% | (Liu et al., 2020) |
| **aqueous Allium sativum** | solvent extraction | water | 95% | (Devikala, Kamaraj, Arthanareeswari, & Patel, 2019) |
| **Rice straw** | solvent extraction | ethanol/water | 92% | (Othman et al., 2019) |
| **Asafoetida** | solvent extraction | double distilled water | 90% | (Devikala, Kamaraj, Arthanareeswari, & Pavithra, 2019) |

| | | | | |
|---|---|---|---|---|
| **Elettaria cardamomum** | solvent extraction | ethanol | 73.5% | (Shyamvarnan et al., 2021) |
| **potato peels** | solvent extraction | ethanol | 70% | (Shekhar et al., 2021) |
| **Solanum melongena stem** | solvent extraction | deionized water | 75% | (Maurya et al., 2021) |
| **Lignin** | solvent extraction | NaOH | 99.83% | (Muzakir et al., 2019) |
| **Glycyrrhiza glabra** | solvent extraction | deionized water | 99.8% | (Parthipan et al., 2021) |

Table 3. Most prominent modern extraction methods: principle, strengths.

| Technique | Principle |
|---|---|
| **Ultrasound-aided extraction (UA. E)** | The principle of UA. E is acoustic cavitation, which eventually doubly enhances the arrival of the solvent to the functional groups by creating a large number of channels on the plant matrix surface. This leads to obtaining the largest amount of active ingredients in a short time (Azmir et al., 2013; Medina-Torres et al., 2017). |
| **Pulsed-electric field extraction (PF. E)** | In this technique, the plant cell walls are destroyed by an electrical voltage passing through them, which allows a significant increase in the permeability of the solvent through its (Azmir et al., 2013; Ranjha et al., 2021). |
| **Enzyme-aided extraction (EA. E)** | The addition of enzymes such as cellulose, pectinase, and α-amylase helps release hydrophobic bonded components that cannot be obtained by solvent alone. Enzymes break down cell walls and break down the structures of fat bodies and polysaccharides, which enhances the process (Azmir et al., 2013). |
| **Microwave assisted extraction (MA. E)** | In this technique, the solvent is heated when it meets the solute using microwave energy. This directly affects the polar materials, causing them to separate from their positions and enhancing their solubility (Alvi et al., 2022; Azmir et al., 2013; Boyapati et al., 2023; Llompart et al., 2019). |

| | |
|---|---|
| **Pressurized liquid extraction (PL. E)** | Is applying high pressure during a high-temperature extraction process to maintain the liquid property of a solvent when its boiling point is exceeded. High temperatures reduce viscosity, increase solubility, and increase mass transfer, which improves the extraction process with less time and less solvent (Alvarez-Rivera et al., 2020; Azmir et al., 2013; Mustafa & Turner, 2011). |
| **Supercritical fluid extraction (SF. E)** | A supercritical liquid combines the gas properties in terms of viscosity, diffusion, and surface tension with the liquid properties in terms of density and solubility, making it an excellent solvent capable of dissolving a high percentage of functional groups within a short period of time (Ahangari et al., 2021; Azmir et al., 2013; Ray et al., 2023). |

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