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Natural Corrosion Process and Inhibition by Plant Products at Various Metal in Different Media with Kinetic, Thermodynamic and Elctrochemical Parameters

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ABSTRACT

Metals and alloys are pre-eminent and important materials for the construction and fabrications of material. The destruction of metals is a usual and unavoidable process but my opting suitable measures it is controllable. Corrosion is impossible to eliminate but on implementing certain remedies we can control to some extent. Usage of the inhibitors for corrosion is one of the controlling measures used widely. Many corrosion inhibitors are banned due to their environmental hazards and toxicity they create. It leads to making use of naturally occurring extracts of plant materials. Natural corrosion inhibitors are nontoxic and environmentally friendly.

KEYWORDS: *corrosion, plant, metal, kinetic, thermodynamic, electrochemical, inhibition, media, products*

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INTRODUCTION

Traditionally, reduction of corrosion has been managed by various methods including cathodic protection, process control, reduction of the metal impurity content, and application of surface treatment techniques, as well as incorporation of suitable alloys. However, the use of corrosion inhibitors has proven to be the easiest and cheapest method for corrosion protection and prevention in acidic media. These inhibitors slow down the corrosion rate and thus prevent monetary losses due to metallic corrosion on industrial vessels, equipment, or surfaces. Inorganic and organic inhibitors are toxic and costly and thus recent focus has been turned to develop environmentally benign methods for corrosion retardation. Many researchers have recently focused on corrosion prevention methods using green inhibitors for mild steel in acidic solutions to mimic industrial processes. Mild steel, also known as plain-carbon steel, is now the most common form of steel because its price is relatively low, while it provides material properties that are acceptable for many applications [1]. However, the challenge is that it has low corrosion resistance especially in acidic environments [2]. Industrial processes such as acid cleaning, pickling, descaling, and drilling operations in oil and gas exploration use acidic solutions extensively and as such iron and steel vessels or surfaces used in these environments are prone to corrosion [3]. The use of many inorganic inhibitors, particularly those containing phosphate, chromate, and other heavy metals, is now being gradually restricted or banned by various environmental regulations because of their toxicity and difficulties faced in their disposal especially in the marine industry, where aquatic life is at threat [4]. Synthetic organic inhibitors have also been extensively applied but their use is now being marred by their toxicity and high cost of manufacturing. This has prompted researchers to explore other areas to produce eco-friendly, cheap, and biodegradable green corrosion inhibitors to replace inorganic and synthetic organic inhibitors. Natural products such as plant extract, amino acids, proteins, and biopolymers have been reported to be efficient corrosion inhibitors [5]. Plant extracts are viewed as rich source of naturally synthesized chemical compounds that can be extracted by simple procedures with low cost [6]. These natural extracts are analogous to the synthetic organic inhibitors and are being proven to work as much as their synthetic counterparts. Worldwide, corrosion causes the value of the gross domestic product to decrease in industrialized countries by 4.26% and causes significant losses to industries including infrastructure. As a result, corrosion prevention and research related to it are extremely important. Some researchers are working to develop plant-based natural corrosion inhibitors, and experimental and computational studies are being conducted widely to prevent corrosion through cheap and environmental friendly coatings.[7] A case study of Convolvulus microphyllus (C. microphyllus) extract was examined as eco-friendly for bio-corrosion inhibitor of mild steel in 0.5 M H2SO4 by using conventional weight loss, electrochemical polarization measurements, and electrochemical impedance spectroscopy (EIS) techniques. The compounds responsible for decreasing the rate of corrosion are kaempferol and phydroxycinnamic acid present in the extract. This inhibitor slows down the corrosion rate. Out of many observations, the best result 89.87% corrosion resistance efficiency was obtained at 600 mg/L of C. microphyllus as extract for mild steel in 0.5 M H2SO4 by applying electrochemical and weight loss measurements. The presence of a heteroatom in the main component of C. microphyllus as extract is believed to be an excellent inhibitor.[8]

Natural extracts have been widely used to protect metal materials from corrosion. The efficiency of these extracts as corrosion inhibitors is commonly evaluated through electrochemical tests, which include techniques such as potentiodynamic polarization, electrochemical impedance spectroscopy, and weight loss measurement. The inhibition efficiency of different extract concentrations is a valuable indicator to obtain a clear outlook to choose an extract for a particular purpose. A complementary vision of the effectiveness of green extracts to inhibit the corrosion of metals is obtained by means of surface characterizations; atomic force microscopy, scanning electron microscopy, and X-ray photoelectron spectroscopy analysis are experimental techniques widely used for this purpose. Moreover, theoretical studies are usually addressed to elucidate the nature of the corrosion inhibitor—metal surface interactions. In addition, calculations have been employed to predict how other organic substances behave on metal surfaces and to provide experimental work with fresh proposals. [9]

Inhibition of corrosion of mild steel in hydrochloric acid by natural plant leaves extract of of Bahunia Varigata (Aapta), Azadirachta Indica (Neem), Pongamia Pinnata (Karanj), Santalum Album (Sandal Wood) and Astonia Scholaris (Saptparni) has studied using weight loss measurement. It was, found that the leaves extract act as a good corrosion inhibitor for mild steel in all concentrations of the extract. The inhibition action depends on the concentration of the leaves extract in the acid solution. Results for weight loss measurements indicate that inhibition efficiency increase with increasing inhibitor concentration. Extract of natural plants is one of the most important metallic corrosion inhibitors. [10]They are readily available, nontoxic, environmentally friendly, biodegradable, highly efficient, and renewable. Several organic compounds with heteroatom such as N, O, S and P present in the plant extracts are, adsorbed directly onto the metals surface through polar atoms and thereby forming the protective layer. Plants represent a class of interesting source of compounds currently being, explored for use in metal corrosion protection in most systems, as possible replacement of toxic synthetic inhibitors.

Discussion

Aluminium, being a highly reactive metal, corrodes rapidly in acidic (pH < 6) and alkaline (pH > 12.5) media. Hence it has to be protected when it is likely to come in contact with such solutions, e.g., during cleaning or acid pickling. One method of protection is the addition of inhibitor to the corroding medium. In the present work, ethanol extract of *Azadirechta indica* and *Murraya koenigii* leaves have been investigated as corrosion inhibitor for aluminium in aqueous hydrochloric acid. The effect of inhibitor and acid concentration, exposure period, and temperature on the inhibitive action of the compound has been studied. Weight loss method has been used. The mechanism of the action of inhibitor has also been suggested. It was observed that the weight loss increases as the concentration of acid increases, and the same effect is observed with increase in temperature and time duration.[11]

In acid containing the inhibitor, it was observed that at low concentrations of both the plant accelerate the attack on aluminium in 0.5 M HCl. The acceleration decreases with increase in inhibitor concentration. Finally, it depends upon the inhibitor and its concentration, the process of inhibition sets in and then inhibition increases with further increase in inhibitor concentration.

The results show that the plant extract studied function as accelerators of corrosion at low concentrations but as inhibitors at high concentrations. Thus at low concentrations they may be useful for removal or recovery of aluminium from galvanized articles provided they do not attack the base metal like steel. But when inhibition of corrosion is desired, higher inhibitor concentrations are required.

The corrosion of Alluminium in plain hydrochloric acid, as well as inhibited, is found to increase with a rise in temperature. Thus in uninhibited 0.5 M HCl the loss in weight due to corrosion for an exposure period of 60 min increases from 736 mg/dm[2] at 35oC to 852, 922, and 958 mg/dm[2] at 45oC, 55oC and 65oC respectively.[12]

In inhibited 0.5 M HCl containing 1.30% of *Azadirechta indica*, it was observed that at 35oC and for an exposure period of 60 min *Azadirechta indica* confer 100.0% protection. As the temperature is increased, the extent of corrosion in inhibited acid also increases but the weight losses are much less than that in *Murraya koenigii* and in plain acid. As far as the inhibitor efficiency is concerned, it may be generalized that at 1.30% inhibitor concentration, the efficiency decreases with a rise in temperature, the effect being less pronounced in the case of Azadirechta indica.[13]

In *A. indica*, it is observed that the compound at 1.30% concentration show an efficiency 100.0% at 35°C. As the temperature is increased the efficiency decreases. In the case of A.indica, the efficiency decreases slightly and is found to be 72.96% at 65°C. It appears that in the case of Azadirechta indica, the adsorption is of physisorption type which decreases with a rise in temperature. The surface morphology of the Al samples in the absence and presence of *A.indica* and *M.koenigii* leaves extract was investigated after weight loss using SEM technique. The badly damaged surface obtained when the metal was kept immersed in 0. 5 M HCl for 60 min without inhibitor indicates significant corrosion. However, in presence of inhibitor the surface has remarkably Improve with respect to its smoothness indicating considerable reduction of corrosion rate. This improvement in surface morphology is due to the formation of a good protective film of inhibitor on aluminium surface which is responsible for inhibition of corrosion. The order of efficiency at 1.30% v/v inhibitor concentration in 0.5 M hydrochloric acid was found to be: *Azadirechta indica* (100.0%) *Murraya koenigii* (94.79%)[14]

The use of corrosion inhibitors is a cost-effective corrosion mitigation strategy for carbon steel. There is an increased focus on developing and using low-cost, biodegradable and environmentally friendly inhibitor formulations. Plant-based extracts have been evaluated in many studies using a multitude of electrochemical methods and characterisation techniques. Although plant extracts appear as promising alternatives for commercially synthesised inhibitor formulations, a significant amount of optimisation is required. The majority of the research on plant extracts does not elucidate the effect of other synergistic combinations in commercial inhibitor formulations. Therefore, further development of plant extracts as corrosion inhibitors is of significant interest. [15]Carbon steel is extensively used in the oil and gas industry, and due to its mechanical properties and low cost, is the preferred material for pipeline construction. Owing mainly to their much higher costs, it is not normally feasible to replace carbon steel with corrosion resistant alloys in large oil and gas trunklines. However, carbon steel is highly susceptible to corrosion. The presence of CO_2, H_2S and organic acids (acetic acid, propionic acid and formic acid) is one of the leading causes of corrosion in oil and gas production that can instigate localised or pitting corrosion[16]

Aluminum and its alloys are important industrial materials because of their high technological value and wide spectrum of industrial applications, including aerospace, household, marine applications etc. Aluminum is known for formability, electrical conductivity, mechanical strength, reflectivity and light weight. It is economical as compared to other materials of similar features and is abundantly available. In appearance it is a form of silvery tone to approximate gray. Their mechanical properties are due to the addition of alloying elements that, sometimes lead to a decrease in the corrosion resistance. However, localized pitting corrosion is most observed in the regions around the Fe-rich and Cu-rich intermetallic compounds. These intermetallic phases normally reveal distinctive cathodic activity and this turns aluminum susceptible to corrosive attack, displays the different fields influenced by corrosion.[17]

Figure 1 Effects of corrosion on various aspects.

A study carried out proposed that globally, corrosion causes a loss of about 3.4% of total GDP. Nevertheless, by implementation of corrosion prevention strategies in an appropriate manner, the metal degradation cost can be reduced up to 15-35%. Corrosion is a subject of great importance because of its economical and safety related concerns. Therefore engineers and scientists around the world are trying to address this issue. In the light of the aforementioned concerns, alluminum alloys demand protection in various process industries. Chromate conversion coatings (CCC) have been extensively used because of their excellent anti corrosion properties. Additionally, corrosion inhibitors have proved themselves effective in protecting the metals against corrosion with the clear advantages concerning cost, availability, ease of use and high protection efficiency. [18]

Corrosion inhibitor compounds normally contain N,S,O,P and other heteroatoms and electron-donating groups. They give out electrons and form a defensive film on the metal protecting the material from corrosion. Though organic inhibitors offer good protection efficiency against corrosion they have a complex process of synthesis and is not economically feasible. Moreover they are toxic to the environment. Therefore there is a necessity to substitute the toxic and harmful inhibitors with environmentally benign compounds like extracts from plants, biomass wastes and other natural products.

Results

The extraction methods can be classified into two categories: classical technique and modern technique. The former technique experiences several limitations, which involve the use of excess solvents, a long heating time and time-consuming. These could lead to the deterioration of the bioactive compounds. In most of the instances, use of these solvents was hazardous and poisonous to humans and the environment. Green house gases are released by these organic solvents, intimidating humans, agriculture and microorganisms. Furthermore, the excess solvent generates huge amount of waste by-products. Contrary to these hazardous techniques, eco-friendly approaches like ‘green processing’, ‘green solvents’, and ‘green product’ are encouraged.[19]

The most critical step in the analysis of plants is extraction. Generally, the extraction is a process of separation, where the actives are isolated from the plant. Various parts of the plant produce a variety phyto-chemicals due to the plant matrices. The appropriateness of an extraction method depends on the polar or non polar nature of the target compound, sample particle size and the presence of interfering substances. The extraction technique must be cautiously chosen according to the aim of the study. It has an impact on the purity, rate and yield and is subjective to the compound of interest and the degree of purity required. Various extraction methods and the solvents applied are illustrated in Table 1.

Table 1 Extraction methods employed for various plant products

Extraction method	Plant / Herb	Solvent
Reflux	*Andrographis paniculata*	50% ethanol (v/v)
Supercritical fluid extraction	P. lobatae root, P. massoniana needle and C. reticulata	supercritical CO2
solid-liquid extraction	*Andrographis paniculata*	Methanol/ethanol/acetone
reflux	cactus pear	45% ethanol
reflux	*Fagopyrum esculentum* Moench	50% (v/v) aqueous ethanol
Cold and hot extraction	Andrographis paniculata	Chloroform

Extraction techniques consist of solvent extraction, distillation method, pressing and sublimation as per the principle of extraction. Among these, most often used is solvent extraction method. The natural product extraction advances through the following stages: (1) the solvent penetratiom into the solid matrix; (2) dissolving of the solute in the solvent; (3) the solute diffuses out of the matrix; (4) the solute extracted is collected. Extraction is further facilitated by any of the factors enhancing diffusivity and solubility.

General Methods of Extraction of Medicinal Plants

Extraction of natural products demands various extraction methods. Few of them are displayed in Fig 2. Various extraction methods posses various technical features as well as drawbacks. It is implausible that an extraction technique alone can concurrently congregate all extraction requirements in terms of quality, operation, cost, consumption, and so on. Recently, extraction techniques are employed in combination to assimilate the advantages of different methods.

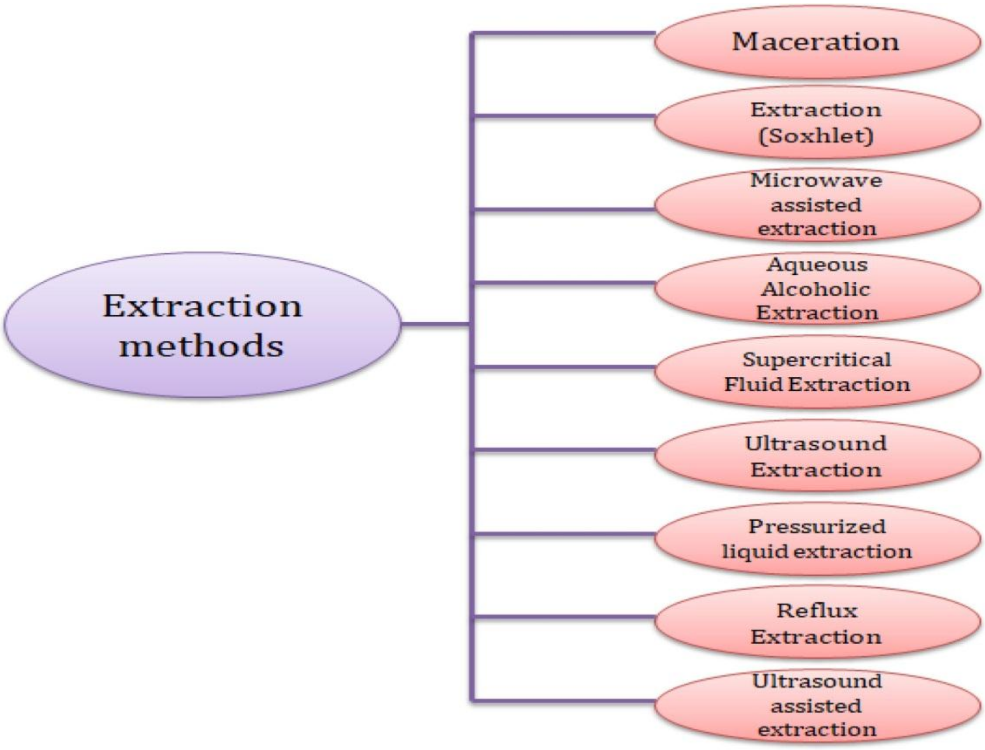

Figure 2 A brief summary of various extraction methods for natural products.

Solvents for extraction

The solvent utilized for medicinal plant extraction is also known as the menstruum. The selection of the solvent is based on the type of plant, part to be extracted, the availability of solvent and the nature of the bioactive compounds. Generally, extraction of polar compounds involves polar solvents, while nonpolar solvents are applied in extraction of nonpolar compounds. Factors to be considered in selecting solvents of extraction are depicted.

General Adsorption mechanism followed by plant extracts

Inhibitor molecules act by adsorbimng on an exposed metal/alloy surface as neutral molecules as a replacement for hydrogen ions adsorbed from the surface of the metal or by dislodgment of water molecules as given by the Eq. (8).

Inhibitor+nHads−−−−−−Inhibitorads+H2inhibitor +nHads−−−−Inhibitorads H2E4Inhibitor+nHads− −−−−−Inhibitorads+H2inhibitor+nHads−−−−Inhibitorads H2E4 **(8)**

The principal cause for the inhibition capability of a molecule is the presence of heteroatoms. These possess greater electron density and behave as active centres for physisorption or chemical adsorption. Other factors influencing the effectiveness of the inhibitor involve: the structure of functional groups; the presence of π bonds, non-bonding p-orbitals and the electronic properties. The inhibition efficiency of leaves extract as an is mostly related to its capability to get adsorbed on surface of the metal with the adherence

Additional explanation of adsorption mechanism from the study data necessitates assessment of the adsorption modes of the molecule (whether molecular or ionic). The prime adsorption mode will be a function of extract composition, chemical changes to the extract and the surface charge on metal.[18,19]

A negative surface charge will support the adsorption of cations whereas anion adsorption is preferred by a positive surface charge. Organic inhibitors often interact through two main mechanisms. Namely, chemical and physical adsorption. It has been recommended that physisorbed molecules attach to the surface at the cathodes and basically retard material dissolution by the cathodic reaction whereas chemisorbed molecules shield anodic areas and diminish the inherent reactivity at the sites where they are attached. It is a known fact that the values of (ΔG_{ads}) of the order of 20 kJ mol^{-}1 or lower point towards physisorption; those of order of 40 kJ mol^{-}1 or higher include charge sharing or transfer to the metal surface from the inhibitor molecules to form a coordinate bond.

Adsorption Isotherms

Isotherm equations were utilized to validate the inhibition mechanism that it is truly adsorption. In addition, to ascertain the closest equation that relates the dosage of inhibitors to the adsorbed concentration at saturation. The empirical equations such as hyperbolic, exponential, logarithmic and power are complicated to associate with the given adsorption mechanisms. There are numerous mathematical equations called adsorption models that estimate the adsorbate quantity in the absorbent at a constant temperature, such as Langmuir, Frumkin, Temkin, Flory–Huggins, Freundlich and Bockris–Swinkels. Mostly, the inhibitors follow Langmuir isotherm, some also obey Freundlich and Frumkin isotherms. The intention of the using adsorption isotherms is to illustrate the mechanism of interaction between the surface and the inhibitor.

Langmuir isotherms

There are few factors effecting the process of adsorption, but, at the fundamental level, the most extensively studied is temperature. The process of adsorption is generally studied at a given temperature and documented as an adsorption isotherm. The Langmuir isotherm is applicable to homogeneous adsorption where the adsorption of each adsorbate molecule on to the metal surface has equal sorption activation energy. Additionally, the saturation occurs when all of the surface active cites are fully occupied by the adsorbate molecules. The important features of the Langmuir model can be articulated in terms of separation factor or equilibrium parameter R_L that can be obtained from the relationship: $R_L=1/[(K_L*C_0)]$,

Where C_0 is the highest initial concentration (mg/l). The value of R_L indicates whether the type of isotherm is irreversible adsorption ($R_L=0$), favourable adsorption $0< R_L < 1$ unfavorable adsorption ($R_L>1$), or linear adsorption ($R_L=1$).

Freundlich isotherms

The Freundlich model describes the adsorption of metal ions on the heterogeneous surfaces. The Freundlich isotherm is not limited to monolayer coverage alone but also describe multilayer adsorption. The linear and non linear forms of the Freundlich model are given by the following equations (9) and (10).

qeq=KFCe1/nqeq=KFCe1/n **(9)**

log qe=logKF+1/nlogCelog qe=logKF+1/nlogCe **(10)**

where K_F is a constant related to the adsorption capacity, and 1/nF is an empirical parameter related to the adsorption intensity, which varies with the heterogeneity of the material.

Tremkin isotherms

The Temkin isotherm is a two-parameter isotherm. Temkin isotherm model considers the influence of indirect adsorbate/adsorbate interactions on the adsorption process; it is believed that the heat of adsorption of all molecules in the layer reduce linearly as a consequence of increased surface coverage. The linear form of Temkin isotherm model is given by the following Eq. (11).

qe=(Rt/b)lnKT+(Rt/b)lnCeqe=(Rt/b)lnKT+(Rt/b)ln Ce **(11)**

Where b is Temkin constant which is related to the heat of sorption (J mol^{-}1) and KT is Temkin isotherm constant (L/g), R is gas constant (8.31 Jmol^{-1} k^{-1}), T is absolute temperature, Ce is concentration of adsorbate at equilibrium (mg g^{-1}).

Conclusions

Merely a tiny portion of actives is included in the plant extracts, therefore, huge amount of plants is obligatory to accomplish the satisfactory inhibition ability resulting in the high cost. Additionally, the process of extraction is too complex to be appropriate for large-scale applications in the industries. One of the alternatives is to verify the essential active components that inhibits corrosion in the plant extracts. Preparation of plant extract plays a crucial role. Every extraction method possesses its own characteristics. Greenhouse gases are released by the organic solvents into the environment, threatening humans and nature. Furthermore, excess solvent usage leads to production of enormous waste by-products. More research is required in order to present efficient and ecologically friendly processes. Plant extracts have exceptional demands in quite a few fields including, cosmetic, pharmaceutical, neutraceutical etc. Consequently medicinal plants cannot be utilized for such applications unless they are copiously available. If corrosion inhibitors could be extracted from the plant waste, it will not only be economical but also be useful to refine the environment. Future studies may focus on the utilization of waste, dead and dried plants.

The varied set of studies discussed and summarized advocates the use of plant extracts as corrosion inhibitors for aluminum alloy. Intense research efforts has been put in to tackle the much problematic phenomenon of corrosion. Though plant extract offer several advantages, they suffer certain challenges. Recovery of phytochemicals requires efficient, cheap and environmental-friendly extraction techniques. Many extraction methods have been exploited ranging from conventional techniques to advanced extraction technologies. The extraction methods mostly depend on the choice of solvents and use of agitation and heat to enhance the solubility of the compounds and to improve the mass transfer to the extraction media from the plant matrix.[20]

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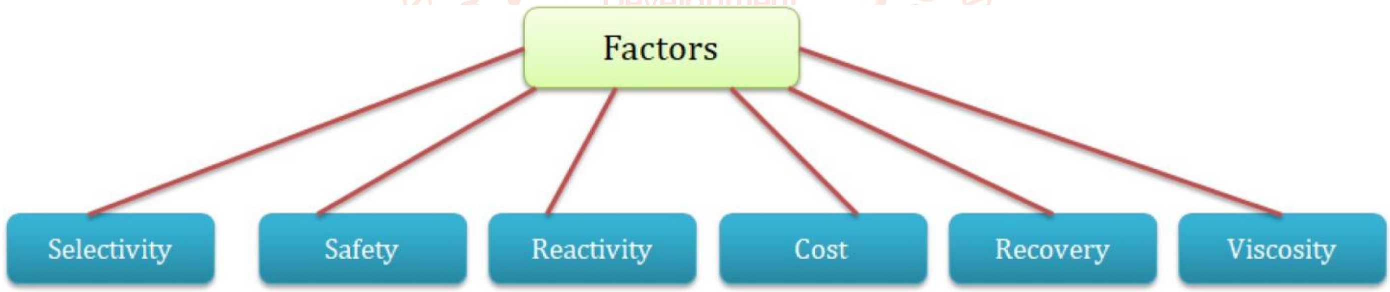

Figure 3 Schematic illustrations of various factors affecting the extraction process