

Sudan University of Science and Technology



College of Graduate studies



Corrosion of Carbon Steel and Chromium Steel in Different Aqueous Media

تاكل الفولاذ الكربوني وفولاذ الكروم في اوساط مائيه مختلفه

A Thesis Submitted for the Partial Fulfillment for the Requirements of M.Sc Degree in Chemistry

By

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استفتاح

بسم الله الرحمن الرحيم

قال تعالى :

(وهو الله لا اله الا هو له الحمد في الاولي والآخره وله الحكم واليه ترجعون)

سورة القصص الايه (70)

صدق الله العظيم

Dedication

To

My Parents,

Brothers and Sisters.

Abstract

This research was undertaken to examine the corrosion behavior of low carbon steel and chromium steel, in different aqueous medium, to test the corrosion behavior of it in: Sodium chloride, sodium sulfite, hydrochloric acid, sulfuric acid and nitric acid solutions, in concentration range from 0.1 to 0.4 mol | L in the same periods of time, for all seasons hence, to observe the temperature effect the corrosion rate.

Inhibitor effect was examined using thiourea in nitric acid and sodium chloride solutions, the obtained results showed that, the acids are more corroding than the salts, and the corrosion increased with increasing the concentration of acids, and the higher corrosion was founded in nitric acid solutions.

Thiourea in the solution decreased the corrosion rate and this decrease was found to be larger in sodium chloride solution in comparison with nitric acid solution.

المستخلص

في هذا البحث تمت دراسته تاكل الفولاذ الكربوني المنخفض والكروم في اوساط مائيه مختلفه لاختبار معدل التاكل في حمض الهيدروكلوريك, الكبريتيك والنتريك وملح كلوريد وكبريتت الصوديوم بتراكيز تتراوح بين 0.1 الى 0.4 مول / لتر لفترات زمنيه متساويه , في مدار فصول السنه لأن درجة الحراره لها تاثير في اختلاف التاكل. كما تم اختبار تثبيط التاكل بأستخدام مثبط الثيو يوريا بالنسبه لحمض النتريك وملح كلوريد الصوديوم, ووجد ان الاحماض تؤدي الي تاكل اعلي من الاملاح. وحمض النتريك كان اعلي تاكلا من الاحماض الاخري وجود مثبط الثيو يوريا قلل من التاكل بالمقارنه بدون مثبط. وكان هذا التخفيض في التاكل في محلول كلوريد الصويوم أعلى من حمض النتريك.

Acknowledgment

First of all thanks to Allah the Most Gracious and most Merciful for giving me the strength and good health to complete this research.

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Chapter One

1. Introduction and literature review

1.1- Definition of Corrosion

Corrosion may be defined as a destructive phenomenon, chemical or electrochemical, which can attack any metal or alloy through reaction by the surrounding environment and in extreme cases may cause structural failure. Corrosion can be also defined as the deterioration of material by reaction with its environment. The corrosion occurs because of the natural tendency for most metals to return to their natural states; e.g., iron in the presence of moist air will revert to its natural state, iron oxide. Metals can be corroded by the direct reaction of the metal to a chemical; e.g., zinc will react with dilute sulfuric acid, and magnesium will react with alcohols.

1.2- Importance of Corrosion Studies

The importance of corrosion studies is in two folds. The first is economic, including the reduction of material losses resulting from the wasting away or sudden failure of piping, tanks, metal components of machines, ships, hulls, marine, structures...etc. The second is conservation, applied primarily to metal resources, the world's supply of which is limited, and the wastage of which includes corresponding losses of energy and water resources accompanying the production and fabrication of metal structures [1]

1.3- Basic Causes of Corrosion

1.3.1- Conditions necessary for corrosion

For the purpose of this manual, electrochemical corrosion is the most important classification of corrosion. Four conditions must exist before electrochemical corrosion can proceed:

- 1- There must be something that corrodes (the metal anode).
- 2- There must be a cathode. [1]
- 3- There must be continuous conductive liquid path (electrolyte, usually condensate and salt or other contaminations).
- 4- There must be a conductor to carry the flow of electrons from the anode to the cathode. This conductor is usually in the form of metal-to-metal contact such as in bolted or riveted joints.

The elimination of any one of the four conditions will stop corrosion.

1.3.2- Effect of material selection

One of the fundamental factors in corrosion is the nature of the material. materials are usually selected primarily for structural efficiency, and corrosion resistance is often a secondary consideration in design.

1.3.3- Water intrusion

Water intrusion is the principal cause of corrosion problems encountered in the field use of equipment. Water can enter an enclosure by free entry, capillary action, or condensation. With these three modes of water entry acting and with the subsequent confinement of water, it is almost certain that any enclosure will be susceptible to water intrusion.

1.3.4- Environmental factors

At normal atmospheric temperatures the moisture in the air is enough to start corrosive action. Oxygen in water is essential for corrosion to at ambient temperatures. Other factors that affect the tendency of a metal to corrode are:

- 1- Acidity or alkalinity of the conductive medium (pH factor).
- 2- Stability of the corrosion products.
- 3- Biological organisms (particularly anaerobic bacteria).
- 4- Variation in composition of the corrosive medium.
- 5- Temperature.

The presence of salts and acids on metal surfaces greatly increases the electrical conductivity of any moisture present and accelerates corrosion. Moisture tends to collect on dirt particles.

1.4- Classification of Corrosion

All metallic materials consist of atoms having valence electrons which can be donated or shared. In a corrosive environment the components of the metallic material get ionized and the movement of the electrons sets up a galvanic or electrochemical cell which causes oxidation, reduction, dissolution or simple diffusion of elements.

The metallurgical approach of corrosion of metals is in terms of the nature of the alloying characteristics, the phases existing and their inter-diffusion under different environmental conditions. In fact, the process of corrosion is a complex phenomenon and it is difficult to predict the exclusive effect or the individual role involved by any one of the above mentioned processes.

Based on the above processes, corrosion can be classified in many ways as low temperature and high temperature corrosion, direct oxidation and electrochemical corrosion, etc. The preferred classification is:

Dry or chemical corrosion and wet or electrochemical corrosion. [2]

1- **Chemical corrosion:** In which the metal is converted into its oxide when the metal is exposed to a reactive gas or non-conducting liquids is.

2- **Electrochemical corrosion:** The formation of a hydrous oxide film occurs when the metal is immersed in a conducting liquid containing dissolved reactive substance. The reaction is considered to take place at the metal-solution interface, due to the heterogeneity on the metal surface, which creates local anodic and cathodic sites on the metal.

1.5- Factors Influence Corrosion

The nature and extent of corrosion depend on the metal and the environment. The important factors which may influence the corrosion process are:

- a) Nature of the metal, nature of the environment and the corrosion products.
- b) Temperature.
- c) Concentration of electrolyte.
- d) Electrode potential.
- f) Aeration.
- g) Agitation.
- h) Hydrogen over voltage and pH of the electrolyte.

1.6- Theories of Corrosion

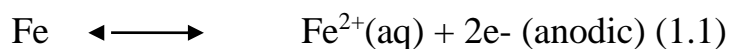
The corrosion of metals in aqueous solutions is an electrochemical process as established in the first half of the 19th century. Whitney [1] gave the most acceptable electrochemical theory. The other theories such as acid theory [2], chemical attack theory, colloidal theory [3, 4] and biological theory [5] were proved to form a part of electrochemical theory.

1.6.1- Electrochemical theory of corrosion

Electrochemistry is a branch of chemistry dealing with relationships between electricity and chemical reactions. It involves oxidation and reduction (redox) reactions. Corrosion is an example of a type of electrochemical reaction, since a substance -oxidizing agent- oxidizes a metal in its environment. In the natural environment, oxygen gas is a good oxidizing agent. Most metals have lower reduction potentials than O_2 ; therefore they are easily oxidized in the presence of oxygen. Metals such as gold, silver and platinum are not so easily oxidized and are sometimes referred to as noble metals. The reasons for the lack of oxidation in these noble metals are varied and sometimes complex.

One of the most familiar corrosion processes is the oxidation of iron (rusting). Iron metal is spontaneously oxidized in the presence of O_2 and an aqueous electrolyte solution. Physical strains (scratches, dents, bends, etc...) present on the iron are more easily oxidized than other areas. This directly relates to physics, i.e., the way electric fields are generated at the surface of the metal. Stronger fields are generated at the physically strained parts of the metal. The result is that these regions are anodic (oxidation occurs) and simultaneously different areas are cathodic regions at which a reduction reaction (usually of O_2) occurs.

The iron atom gives up two electrons to form the Fe^{2+} ion:



The electrons that are released flow through the iron metal to the cathodic region where they react with oxygen:

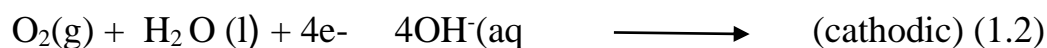
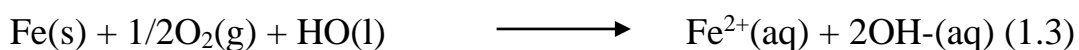
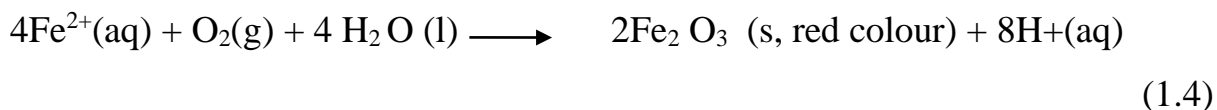


Fig. (1.1): The basic corrosion cell consists of an anode, a cathode, an electrolyte, and a metallic path for electron flow.

These two half reactions together give the overall reaction:



Common experience with this process (e.g., car fenders) tends to show that Fe^{2+} is eventually oxidized further to Fe^{3+} , in the compound iron (III) oxide (rust):



In the experimental setup, this last reaction does not occur. The $\text{Fe}^{2+} (\text{aq})$ reacts with $[\text{Fe}(\text{CN})_6]^{3-}$ in the gel, forming $\text{Fe}[\text{Fe}(\text{CN})_6]^-$, which is a vivid blue colour. The gel you use contains phenolphthalein, an indicator that is pink in basic solutions. Since the reduction of O_2 results in a basic solution, one will observe a pink color at the cathodic region. Thus, phenolphthalein and $[\text{Fe}(\text{CN})_6]^{3-}$ are used to show where the cathodic and anodic regions are.

Further complications arise if we have a system where two different metals are in contact with each other, both in the electrolyte solution. Consider iron metal in contact with another metal. When this situation is encountered, electric fields are set up due to the difference in the reduction potential of the two metals. These fields are stronger than any effect caused by physical strains. Thus, one metal will act entirely as the anode, while the other becomes the cathode. One can use standard reduction potentials to determine which metal will act as the anode/cathode.

When considering a bimetallic system, the metal with the more positive reduction potential will be reduced (cathode) and the metal with the more negative reduction potential will be oxidized (anode). For example, if copper and zinc were connected electrically and immersed in an electrolyte solution, we might expect that the copper would be the cathode and the zinc the anode, based on their relative reduction potentials. [6]

1.6.2- Effects of the anode and cathode areas on the corrosion process

Area Effects are important, especially in galvanic and localized corrosion. Consider the difference between a cell with a very large anode area compared to the cathode and one in which the cathode is significantly larger than the anode. Since metal is corroded at the anode only, the rate of corrosion will be proportional to the rate at which the anodic reaction proceeds. For the anodic reaction to proceed, however, there must be corresponding cathodic reactions. The cathode reaction therefore controls the rate of the overall reaction. With a large cathode and a small anode, there is more surface area on which cathodic reactions may proceed so the anodic reaction proceeds at a much faster rate than if the reverse (i.e. large anode, small cathode) is true. Where use of dissimilar metals is unavoidable, therefore, it is desirable to use the more noble (cathodic) metal in the smallest possible exposed area relative to the anode. [9]

1.7- Thermodynamic Principles of Corrosion

Corrosion resistance or chemical resistance depends on the following:

- 1- Thermodynamic principles.
- 2- Physical and chemical factors.
- 3- Metallurgical factors.
- 4- Electrochemical principles.

Thermodynamic and electrochemical principles play a major role in determining the corrosion behavior of materials.

Thermodynamic indicates the spontaneous direction of a chemical reaction. It is used to determine whether or not corrosion is theoretically possible.

Almost all metals and alloys used in service are actually in an unstable thermodynamic state. There is thus a fundamental thermodynamic tendency for them to return to a stable state through corrosion processes. Thermodynamics is

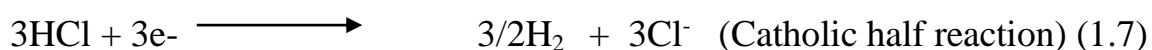
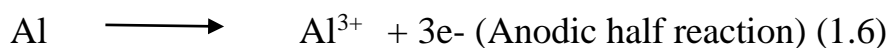
concerned with energy states. The original metallic ores are said to be in a state of low energy.

metals and alloys, transforming them to a higher energy state. They tend to revert to a lower (more stable) energy state by reacting with a corrosive environment. While thermodynamics can predict whether a corrosion reaction will take place, it does not provide an indication of the rate of corrosion reactions .

For a reaction to be occurred spontaneously there must be a free energy change, ΔG , and since the energy, in the case of spontaneous reaction, is given out, the sign is negative. The magnitude of the ΔG and its sign are of great importance, since it indicates whether or not the corrosion reaction will take place. For example the following reaction of aluminum in hydrochloric acid:



The reaction is thermodynamically favorable; i.e. free energy change ΔG must be negative. This reaction can be divided into anodic half and cathodic half reactions as follows:



These two half reactions have associated free energy change, thus, overall, the sum of the two free energies change must be negative, i.e.

$$\Delta G_{\text{anodic}} + \Delta G_{\text{cathodic}}$$

where:

ΔG_{anodic} is the free energy change for anodic iron dissolution.

$\Delta G_{\text{cathodic}}$ is the free energy change for cathodic hydrogen evolution.

The electrical potential, can be related to the free energy change by the following equation:

$$\Delta G = -n F E \quad (1.8)$$

where:

ΔG is the free energy change.

n is the number of electrons transferred in the half-cell reaction.

F is the charge transported by 1 mole of electrons and has the value of 96494 coulombs/mole.

E is the measured potential in volts (Vs. SHE).

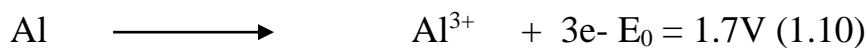
Similarly,

$$\Delta G^0 = -n F E_0 \quad (1.9)$$

where:

ΔG^0 is the standard free energy change.

n and F are defined as above. E^0 is the standard electrode potential and can be obtained from thermodynamic tables.



Under nonstandard conditions i.e., the respective ions not at unit activity, the electrode potential is related to the standard electrode potential by Nernst equation [8].

$$E = E_0 + (RT/nF) \ln (a_{\text{Ox}}/ a_{\text{Red}}) \quad (1.11)$$

As can be seen from this equation the activity of the oxidized species (a_{Ox}) increases as electrode potential (E) becomes more positive. In aqueous environments the thermodynamic consideration of electrochemical corrosion was the bases of the construction of (E)- pH diagram, which are also known as Pourbaix diagram [9]. Such diagrams are essentially isothermal phase diagrams, which represent metal-ion/ oxide equilibrium plotted with potential, and pH bas coordinates [10].

1.8- Types of Corrosion

1.8.1- General or uniform corrosion

Differences in electrical potential occur on the surface of a piece of metal due to small differences in chemical composition, phase differences, amount of cold

work, etc. These differences set up small corrosion cells each with an anode and cathode. Corrosion continues until the metal is consumed or the film of rust formed on the surface sets up a barrier to the electrolyte[15].

1.8.2- Pitting corrosion

Pitting corrosion is a complex but important problem that is at the root of many corrosion failures. It has been studied in detail for many years, yet crucial phenomena remain unclear. In pitting corrosion the surface of the metal is attacked in small-localized areas. Organisms in water or breaks in a passive film can initiate corrosion. In pitting corrosion very little metal is removed from the surface but the effect is marked.

In passivated metals or alloys that are exposed to solutions containing aggressive anions, primarily chloride, pitting corrosion results in local dissolution leading to the formation of cavities or (holes). The shape of the pits or cavities can vary from shallow to cylindrical holes and the cavity is approximately hemispherical [11]. The pit morphology depends on the metallurgy of the alloy and chemistry of the environment as well as the leading conditions. As observed first by Mc Adam in 1928, these pits may cause local increase in stress concentration and cracks may nucleate from them. Pitting corrosion occurs mostly in solutions containing halide or oxyhalides. Chlorides, bromides and hypochlorites are the most aggressive anions [12]. Fluoride, iodide and iodine containing anions were thought to be without pitting tendency. However, iodide ions were found to cause pitting corrosion in many cases [13]. Solutions of certain oxidizing cations produce the worst pitting attack. Ferric, cupric and mercuric halides are the most aggressive of all pitting reagents. The non-oxidizing metal halides such as Al, Ca and Na cause pitting to a lesser degree.

Electrochemical studies of pitting corrosion have found that there exist characteristic potentials. Stable pits form at potentials noble to the pitting potential, E_P , and will grow at potentials noble to the repassivation potential, E_R , which is lower than E_P . During upward scanning in a cyclic polarization experiment, a stable pit starts growing at E_P where the current increases sharply from the passive current level and, upon reversal of the scan direction, repassivates at E_R where the current drops back. It is generally considered that materials exhibiting higher values of E_P and E_R are more resistant to pitting corrosion, and cyclic polarization experiments are commonly used for this purpose.

Pitting is considered to be autocatalytic in nature; once a pit starts to grow, the conditions developed are such that further pit growth is promoted. The anodic and cathodic electrochemical reactions that comprise corrosion separate spatially during pitting. The local pit environment becomes depleted in cathodic reactant (e.g. oxygen), which shifts most of the cathodic reaction to the boldly exposed surface where this reactant is more plentiful. The pit environment becomes enriched in metal cations and an anionic species such as chloride, which electromigrates into the pit to maintain charge neutrality by balancing the charge associated with the cation concentration. The pH in the pit is lower owing to cation hydrolysis and the absence of a local cathodic reaction. The acidic chloride environment thus generated in pits is aggressive to most metals and tends to propagate the pit growth.

1.8.3- Stress corrosion cracking

Failure is due to the simultaneous influence of static tensile stresses and a corrosive environment and this is specific to a particular metal. The stresses may be internal such as those caused by cold work, welding, heat treatment or external forces caused by mechanical stresses set up by assembly practices. A

good example of this form of corrosion is 316 stainless steel in marine environments. 316 stainless steel was developed to withstand attacks in chloride environments, but if stressed the steel will fail by stress corrosion cracking.

1.8.4- Intergranular corrosion

Corrosion occurs at the grain boundaries due to a difference in potential between the anodic grain boundaries and the cathodic grains. "Sensitized" stainless steels, where carbides have been precipitated in the grain boundaries during improper heat treatment or in the heat-affected zone of a weld, are particularly susceptible to intergranular corrosion.

1.8.5- Corrosion fatigue

Failure under repeated cycling stresses in a corrosive environment[13]

1.8.6- Filiform corrosion

Filiform corrosion appears as a network of corrosion trails, of a wormlike structure, particularly beneath thin organic coatings. Salts containing chlorides, which have been left on the surface prior to coating, are suspected.

1.8.7- Crevice corrosion Crevice corrosion occurs when there is a difference in ion, or oxygen, concentration between the metal and its surroundings. Oxygen starvation in an electrolyte at the bottom of a sharp V-section will set up an anodic site in the metal that then corrodes rapidly.

1.8.8- Galvanic or bi-metallic corrosion

Galvanic corrosion takes place between two different metals, or coatings, which are joined together in the presence of an electrolyte. Each metal has a potential different from any other metal when placed in an electrolyte. A series can be built up of all the metals relative to each other.

1.8.9- Fretting corrosion

Fretting corrosion occurs when two or more parts rub against each other. The rubbing action removes the corrosion products and exposes new metal to the electrolyte.

1.8.10- Erosion corrosion

Erosion is the removal of metal by the movement of fluids against the surface. The combination of erosion and corrosion can provide a severe rate of corrosion.

1.8.11- Selective leaching or demetalification

Demetalification is the removal of one of the alloying elements in an alloy by the electrolyte. This results in a "spongy" metal. Typical example is the removal of zinc in chloride waters from brass.[15]

1.9- Cost of Corrosion

Corrosion is not only dangerous, but also costly, with annual damages in the billions of dollars! If this is difficult to believe, consider some of the direct and indirect effects of corrosion which contribute to these costs:

- 1- Replacement of corroded equipment.
- 2- Unscheduled plant shutdowns for replacement.
- 3- Process upsets resulting from corrosion.
- 4- Product contamination.
- 5- Product loss from a vessel that has corroded.
- 6- Otherwise unnecessary preventive maintenance.
- 7- Over design to allow for corrosion.
- 8- Inability to use otherwise desirable materials.

While the economic costs are frightening, we must consider them to be of secondary importance to the potential loss of life and damage to the environment problems, which can have widespread effects upon modern industrial businesses. It is essential, therefore, for operators of industrial process plants to have a program for controlling corrosion[16]

1.10- Methods of Corrosion Protection

1.10.1- Application of protective coatings

Metallic structures can be protected from corrosion in many ways. A common method involves the application of protective coatings made from paints, plastics or films of noble metals on the structure itself (e.g., the coating on tin cans). These coatings form an impervious barrier between the metal and the oxidant but are only effective when the coating completely covers the structure. Flaws in the coating have been found to produce accelerated corrosion of the metal.

1.10.2- Polarize or shift the potential of the metal

Another method of reducing corrosion is to polarize or shift the potential of the metal enabling it to act as a cathode (rather than an anode) in an electrochemical cell. One example is the galvanizing of steel with a coating of zinc. The iron and zinc then function as the electrodes of a cell. Zinc is the more readily oxidized metal of the pair and functions as the anode and corrodes, while the iron functions as the cathode. The zinc is used up and protection is effective as long as some zinc remains. Sacrificial anodes work on the same principal but use an external electrode made from a readily oxidized metal; this form of protection is commonly used on buried pipelines.

1.10.3- Cathodic protection

Cathodic protection using an impressed current derived from an external power supply is a related form of protection in which the metal is forced to be the cathode in an electrochemical cell. For example, most cars now use the negative terminal on their batteries as the ground. Besides being a convenient way to carry electricity, this process shifts the electrical potential of the chassis of the car, thereby reducing (somewhat) its tendency to rust.

1.10.4- Materials selection

Basic recommendations for eliminating corrosion are materials selection which are summarized as follows:

- 1- Use corrosion-resistant materials including plastics and non-metallics in severe environments.
- 2- Avoid dissimilar metal couples.
- 3- Avoid the use of back-to-back structural shapes, such as box beam sections and unistrut. When the exterior use of unistrut cannot be avoided, selection of appropriate material shall be considered, such as stainless steel. In addition, alternate structural shapes shall be considered such as C- or d-shaped channel, especially in highly corrosive areas.
- 4- Avoid the use of alloys susceptible to stress-corrosion.

1.10.5- Corrosion inhibitors

Corrosion inhibitors can be added to solutions in contact with metals (e.g. inhibitors are required in the antifreeze solution in automobile cooling systems). These compounds can prevent either the anode or the cathode reaction of corrosion cells; one way that they can do this is by forming insoluble films over the anode or cathode sites of the cell. Examples of anodic inhibitors are sodium

phosphate or sodium carbonate while zinc sulfate and calcium or magnesium salts act as cathodic inhibitors. New forms of paints are being developed which take advantage of similar properties. These paints promise to nearly eliminate corrosion in applications like painted car fenders.

1.11- Corrosion Inhibitors

1.11.1- Corrosion inhibition in acid solutions

Acid solutions are widely used in industry, where the most important fields of application are acid pickling, industrial acid cleaning and oil well acidizing. Because of general aggressiveness of acid solutions, the practice of inhibition is commonly used to reduce the corrosive attack on metallic materials.

The selection of appropriate inhibitors mainly depends on the type of acid, its concentration, temperature and velocity of flow, the presence of dissolved organic and/or inorganic substances, and on the type of metallic material exposed to the action of acidic solution.

Most of well-known acid inhibitors are organic compounds containing nitrogen, sulphate and oxygen .

1.11.2- Factors affecting inhibitors action in aggressive acid media

a- Interaction of the inhibitor with a metal surface

Physical (or electrostatic) adsorption and chemisorption are the principle types of interaction between an organic inhibitor and a metal surface

1.11.2.1- Physical adsorption

Physical adsorption is the result of electrostatic attractive forces between inhibiting ions or dipoles and the electrically charged surface of the metal. The surface charge of the metal is due to the electric field at the outer Helmholtz plane of the electrical double layer existing at the metal/solution interface. The

surface charge can be defined by the potential of the metal E_{corr} vs. its zero charge potential ($E_{\text{q}}=0$) when the difference $E_{\text{corr}} - E_{\text{q}} = \phi$ is negative charge, cation adsorption is favored. Adsorption of anions is favored when ϕ become positive. This behavior is related not only to positively or negatively charged compounds, but also to dipoles whose orientation is determined by the value of the ϕ potential.

According to Andropov [20], at equal value of ϕ for different metals, similar behavior of gives inhibiting species should be expected in the same environment.

In study the adsorption of ions at the metal/solution interface, it was first assumed that ions maintained their total charge during the adsorption. Giving rise in this way to a pure electrostatic bond. Lorenz [23-25] suggested that partial charge is present in the adsorption of ions; in this case a certain amount of covalent bond in the adsorption process must be considered. The partial charge concept studied by Shultz and Kappitz and others], they defined electrosorptionvalency as the coefficient for the potential dependence and charge flow of electro sorption process. The term electrosorptionvalency was chosen because of its analogy with the electrode reaction valiancy, which enters into Faraday's law as well as the Nernst equation. [24]

1.11.2.2-Chemisorptions

Another type of metal/inhibitor interaction is chemisorptions. This process involves charge sharing or charge transfer from the inhibitor molecule to the metal surface in order to form a coordinate type of bond.

The chemisorptions process takes place more slowly than electrostatic adsorption and with higher activation energy. It depends on the higher temperature; higher degree of inhibition should be expected at higher temperature.

Chemisorptions is specific for certain metals and is not completely reversible [28]. The bonding occurring with electron transfer clearly depends on the nature of the metal and the nature of organic inhibitor. In fact, electron transfer is typical for transition metals having vacant, low-energy electron orbitals.

The structure characteristics of the rest of the molecule influence the electron density on the heteroatom and as a consequence, the strength of the chemisorptions bond. Regular and systematic changes in the molecular structure, such as the introduction of substituent's in various positions on the aromatic and heterocyclic compounds, may influence the electron density and the ability of compounds to inhibit corrosion.[28]

Relationship between electronic structure and efficiency of various classes of inhibitors has been deduced from quantum mechanical calculations [30-33]. In this way, the electron density, the order of the bond along the bond line, the index of free valence and the charge of atoms in the free and adsorbed states have been calculated. Other structure parameters influencing the inhibiting efficiency may be mentioned. Thus, the projected molecular area [34], molecular weight [33] of various series of organic compounds has been correlated with variation in inhibiting efficiencies.

b- Interaction between adsorbed inhibitors

When the coverage of the metal surface by adsorbed inhibitor species, lateral interaction between inhibitor molecules may arise, influencing the inhibition efficiency. Attractive lateral interactions usually give rise to stronger adsorption and higher inhibition efficiency. This effect has been show in the case of compounds containing long hydrocarbon chains due to attractive Vander Waals forces In the presence of ions or molecules containing dipoles, repulsive interaction may occur, weakling the adsorption and diminishing the inhibition efficiency.

c- Interaction of the inhibitor with water molecules

Due to adsorption process of an inhibitor, water molecules are being removed from the metal surface. During the adsorption process of a molecule the interaction energy between molecule and water changes as it passes from the dissolved to adsorbed state. This change forms an important part of the free energy of adsorption, i.e., the higher the solvation energy of the adsorption species, the higher the free energy of adsorption. Increasing the size of the hydrocarbon portion of an organic molecule will increase the solvation energy of an inhibitor leading to decrease the solubility and increasing adsorbability .

d- Relationship between inhibitor reactivity and efficiency

The nature of the inhibitor initially present in acid solutions may change with the time and/or the electrode potential as a consequence of reduction reactions, polymerization reactions, or formation of surface products. The inhibition due to the reaction products is usually called secondary inhibition, whereas primary inhibition may be higher or lower than secondary inhibition, depending on the effectiveness of the reaction product [25].

1.11.3- Adsorption isotherms

The adsorption isotherms describe the relation ship between the coverage (θ) of an interface with adsorbed species and concentration of the species in solution. Various adsorption isotherms have been listed as follow .

a- The Henry isotherm: It has the following form:

$$\theta = K C \quad (1.12)$$

where:

K is the adsorption equilibrium constant.

C is the concentration of the adsorb ate in the bulk phase in moles / liter.

θ is the degree of surface coverage.

b- The Freundlich isotherm: It has the following form:

$$\theta = K C^n \quad (1.13)$$

where:

K and C are defined as in Henry isotherm.

n is a constant where, $0 < n < 1$.

c- The Langmuir isotherm: It has the following form:

$$\theta / (1 - \theta) = K C \quad (1.14)$$

d- The Volume isotherm: It has the following form:

$$K C = \theta / (1 - \theta) \exp(\theta / (1 - \theta)) \quad (1.15)$$

e- The Amagatisotherm: It has the following form:

$$K C^n = \theta / (1 - \theta) \exp(\theta / (1 - \theta)) \quad (1.16)$$

which has the same form as the Volmer isotherm but the concentration is raised to the power n where, $0 < n < 1$.

f- The Franking isotherm: It has the following form:

$$K C = \theta / (1 - \theta) \exp(-2 a \theta) \quad (1.17)$$

where:

a is a parameter characterizing the interaction between the adsorbed particles..

$a > 0$, Attraction while $a < 0$, Repulsion. 15

g- The Parsons isotherm: It has the following form:

$$K C = \theta / (1 - \theta) \exp((2 - \theta / (1 - \theta))^2 - 2 a \theta) \quad (1.18)$$

h- The Temkin isotherm: It has the following form:

$$\ln K C = a \theta \quad (1.19)$$

where all terms are defined as described above and θ varies between

$0.1 < \theta < 0.9$.

Interpretation of adsorption and performance of organic inhibitors can be evidenced by fitting the data to one of the adsorption isotherms.

1.12 - Carbon steel and Chromium steel:

1.12.1-Carbon steel:

The two types of carbon steel are plain carbon steel and alloy steel. Plain carbon steel is one which contains in addition to carbon and iron certain elements notably manganese, phosphorus, sulphur and silicon. In small quantities it may also contain traces of elements like copper, boron, cobalt and so on.

-low carbon steel

-medium carbon steel

High carbon steel

(Young, J.F. 1982 materials and processes)

Low carbon steel :

Low carbon steel by weight contains 0.05% to 0.30% carbon. Low carbon steel is made in the basic – oxygen and open –hearth furnaces. Low carbon steel is used for forge work, rivets, chains and machine parts that do not need great strength. It is also used for almost every product that was once made of wrought iron.

Some low carbon steel is cold – rolled between highly polished rollers under great pressure. This improves its tensile strength, and gives it a very smooth finish and exact size. (10)

1.12.2- Chromium Steel :

Chromium steel has long dominated the landscape of the wrought industry because of their high performance capabilities at modest cost level. Historically chromium steels have had difficulty penetrating the powder metallurgy market because of oxidation issues.

Wrought materials containing Cr have found widespread use in high performance components because of improvements in hardenability and mechanical properties at modest cost. The powder metallurgy (PM) industry has adopted Cr through a range of commercially available products because Cr tends to form stable oxides, PM alloys containing this element have traditionally been high temperature sintered above 1205 ° C to avoid the adverse effects of oxygen on mechanical properties. The recent instability of raw material prices has forced an increased desire to develop alloys that jointly utilize the benefits of Cr and can be effectively sintered at conventional temperatures using a 30 minute time-at-temperature cycle. [30]

1.13- Objectives of the Study:

The objectives of this study can be divided to:

- firstly the corrosion of carbon and chromium steel alloys studies in different aqueous solution includes: sodium chloride, sodium sulfite, hydrochloric acid, sulfuric acid and nitric acid solutions at different concentration.

- secondly the kinetic and inhibitor effect of this corrosion will be studied and finally evaluate and prediction of this behavior can be discussed

Chapter Two

2. Materials methods

Experimental

2.1. Chemicals

The following materials, chemicals, and apparatus were used for the research: 400 mL beakers; ; conc. nitric acid(HNO_3), hydrochloric acid (HCl), and sulfuric acid (H_2SO_4) sodium chloride (NaCl) -sodium sulfate(Na_2SO_3) distilled water; each of which were analytical grade type.

2.2- Samples:

Carbon steel and chromium steel obtained from Giad Company, the composition and the different constituents of each sample were examined using X-ray fluorescence (XRF)

2.2.1- Carbon steel:

Table 1 the composition of carbon steel samples showed by XRF

C%	Si%	Mn%	Cr%	Mo%	Ni%	Al%	Cu%	Ti%	V%	Nb%
0.416	0.11	0.631	0.030	0.034	0.068	0.076	0.052	0.004	0.010	0.008
0.421	0.105	0.606	0.027	0.034	0.103	0.072	0.053	0.004	0.013	0.010
0.418	0.108	0.619	0.028	0.034	0.095	0.074	0.052	0.004	0.012	0.009

Co%	W%	Pb%	Fe%
0.013	0.000	0.019	98.509
0.014	0.000	0.019	98.517
0.014	0.000	0.019	98.513

2.2.2- Chromium Steel:

Table2 the composition of chromium steel samples showed by XRF

C%	Si%	Mn%	Cr%	Mo%	Ni%	Cu%	V%	Co%	W%	Fe%
1.316	0.355	0.491	4,306	4.535	0.247	>0.100	2.782	0.197	4.731	80.981
1.230	0.386	0.475	4,610	4.610	0.205	>0.100	2.050	0.184	5.772	80.598
1.273	0.371	0.483	4.672	4.572	0.226	>0.100	2.416	0.191	5.252	80.80

2.3-Corrosion Method:

The metal specimens were first of all pretreated, polished, and cleaning with tissue paper. They were washed first with tap water, then distilled water, and finally washed with acetone in order to degrease them. The specimens were then completely air dried, and weighed on an analytical balance. The weights of the specimens were noted, and they were dipped into test solutions of HCl, HNO₃, and NaCl and H₂SO₄ in concentration range from 0.1 to 0.4 M 7 days, the specimens were removed from the test solutions and first washed with tap water. This was followed by washing in distilled water, cleaning with fine cloth, washing again with distilled water, and further washing with acetone. The specimens were finally weighed on an analytical balance, and the differences in weights at each interval were noted. The weight loss was calculated using the formula: $W.L = (W_b - W_a)$

where:

W_b and W_a are the weights before and after immersion in corroding solution.

2.4-Kinetic Study of corrosion:

The initial weight of the specimens and weight of specimens at various instants of time in all concentrations of the three acid media and tow salt were used in calculating the specific reaction rate values, using first order rate expression; $k = (2.303/t) \text{ Log } [(initial\ weight\ of\ specimen)/(weight\ at\ time,\ t)]$. The half-life time values were then calculated using the formula $t_{1/2} = 0.693/k$ (20).

2.5-Inhibitor Method :

the same method as corrosion method was used in the corrosion inhibitor test using thiourea as inhibitor and nitric acid and sodium chloride solution of concentration 0.4 M for 7 days as corrosion solution test, the specimens (carbon steel and chromium steel) were removed from the test solution , washed with tap water , distilled water and cleaned with fine cloth, washed again with distilled water and acetone and finally weighed

Chapter Three

3-Results and Discussions:

. k increases as acid concentration increases. There is then a corresponding decrease in the half-life time values, thus confirming the first order kinetics for the corrosion, it has been confirmed that HNO₃ corrodes these metals the most because it has the highest k values, and the lowest half-life times at all concentrations. The specific reaction rates follow this order for the three acids used: HNO₃ > H₂SO₄ > HCl. Inhibitor effect

The calculate weight loss of carbon steel and chromium steel in different concentration of sodium chloride plotted in figures 1 and 2 It could be observed that corrosion of carbon steel and chromium steel increase with increase the concentration of salt, but the carbon steel is more corroded than chromium steel in 0.4M

3. 1 - Sodium Chloride:

3.1.1-Carbon Steel:

Table 3 show the value of K increase as the concentration of sodium chloride increase and the half time decrease

Table 3

Concentration	Weight loss (g)	K (min ⁻¹)	Half life (min)	Corration rate g cm ⁻² min ⁻¹ (
0.1	9.9	1.63×10^{-7}	0.423×10^7	7.2×10^6
0.2	11.9	1.98×10^{-7}	$0,35 \times 10^7$	7.5×10^6
0.3	21.9	3.83×10^{-7}	0.18×10^7	0.13×10^6
0.4	51.9	10.78×10^{-7}	0.06×10^7	0.34×10^6

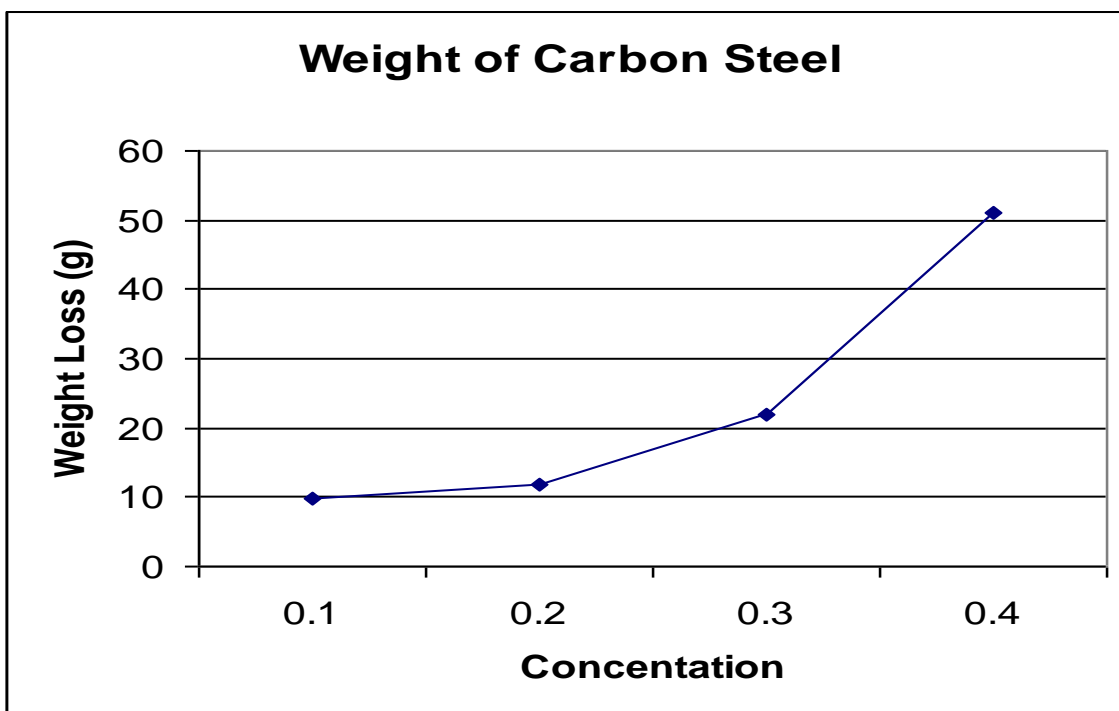


Figure1 weight loss of carbon steel in NaCl

3.1.2-Chromium steel:

Also in the table 4 the value of K increasing as the sodium chloride concentration increasing but the value of k in 0.4 M is lowest comparing the value of K in table 3 (carbon steel).

Table 4

concentration	Weight loss (g)	K (min ⁻¹)	Half life (min)	Corrosion rate g cm ⁻² min ⁻¹ (
0.1	7.3	1.19×10^{-7}	0.58×10^7	4.6×10^6
0.2	12.51	2.09×10^{-7}	0.33×10^7	8.81×10^6
0.3	16.16	2.75×10^{-7}	0.25×10^7	0.118×10^6
0.4	19.81	3.43×10^{-7}	0.20×10^7	0.49×10^6

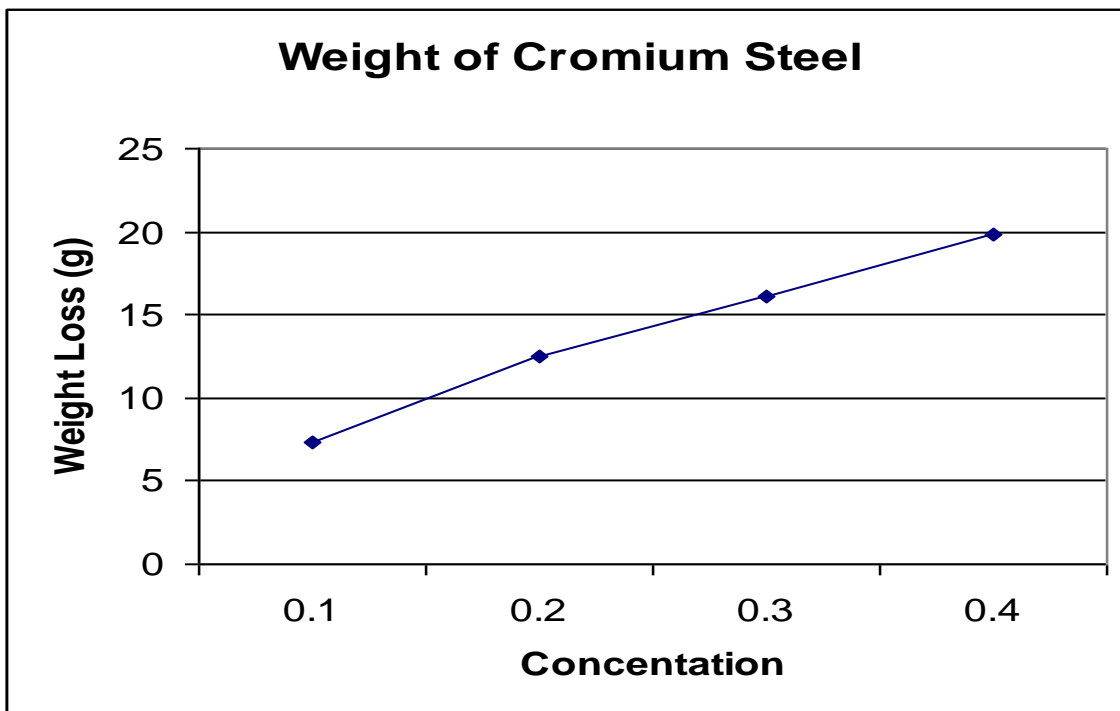


Figure 2 weight loss of chromium steel in NaCl

3. 2- Sodium Sulfito:

Figure 3 -4 represent weight loss plots of carbon steel and chromium steel in sodium sulfate in different concentration of sodium sulfate it could be observed the corrosion of this samples in sodium sulfate is a few than sodium chloride and the chromium steel is a few than carbon steel in sodium sulfate.

3.2.1- Carbon steel:

It could be observed that the specific reaction rate, k increases as the sodium sulfate increases There is then a corresponding decrease in the half-life time values,

Table 5 carbon steel in sodium sulfite

Concentration	Weight loss (g)	K (min ⁻¹)	Half life (min)	Corrosion rate g cm ⁻² min ⁻¹ (
0.1	1.04	0.165×10^{-7}	4.2×10^7	0.76×10^6
0.2	9.38	1.54×10^{-7}	0.45×10^7	5.94×10^6
0.3	18.25	3.13×10^{-7}	0.22×10^7	11.5×10^6
0.4	48.44	9.83×10^{-7}	0.07×10^7	32.9×10^6

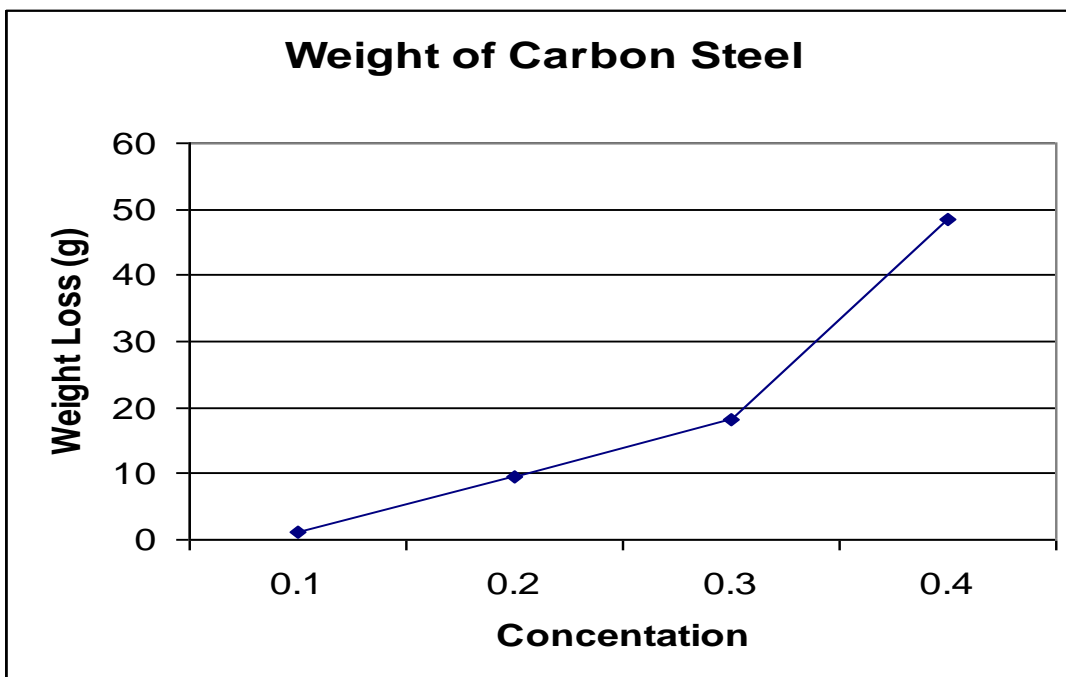


Figure3 weight loss of carbon steel in Na₂SO₃

3.2.2- Chromium Steel:

Table 5 Chromium steel in Sodium Sulfit

Concentration	Weight loss (g)	K(min-1)	Half life (min)	Corrosion rate g cm-2 min-1)
0.1	5.73	0.93×10^{-7}	0.74×10^7	3.6×10^6
0.2	11.47	1.91×10^{-7}	0.36×10^7	8.08×10^6
0.3	13.55	2.28×10^{-7}	0.30×10^7	9.91×10^6
0.4	18.77	3.23×10^{-7}	0.21×10^7	18.31×10^6

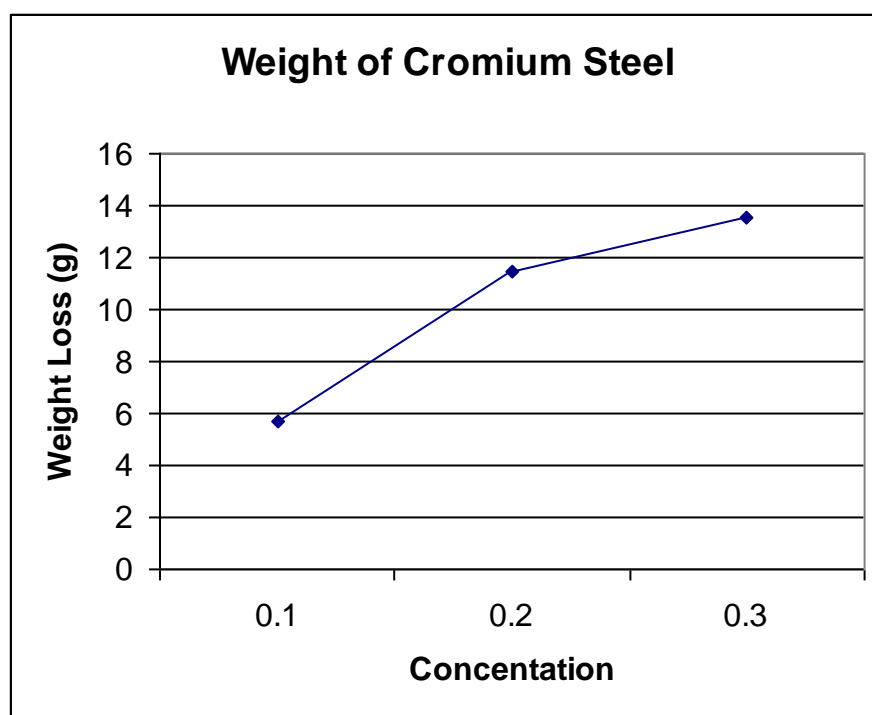
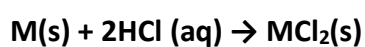
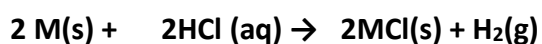


Figure4 weight loss of chromium steel in Na_2SO_3

3.3- Hydrochloric Acid:

The calculated weight loss of carbon steel and chromium steel in four different concentrations of HCl are plotted in figures 5 and 6 . It could be observed that corrosion of corrosion increase with increase concentration of HCl. Carbon steel and chromium steel reacts as shown by the reaction equations below:



In 0.4 M chromium steel gave high corrosion than carbon steel although in the same time immersion in solution.

3. 3.1- Carbon steel:

K increase in the table7, 8 as the concentration of hydrochloric acid increase and half-life decrease

Table 7

Concentration	Weight loss (g)	K(min ⁻¹)	Half life (min)	Corrosion rate g cm ⁻² min ⁻¹) (
0.1	19.81	1.72×10^{-7}	0.40×10^7	7.62×10^6
0.2	10.41	3.43×10^{-7}	0.20×10^7	0.12×10^6
0.3	21.37	3.73×10^{-7}	0.18×10^7	1.35×10^6
0.4	38.58	7.38×10^{-7}	0.09×10^7	2.62×10^4

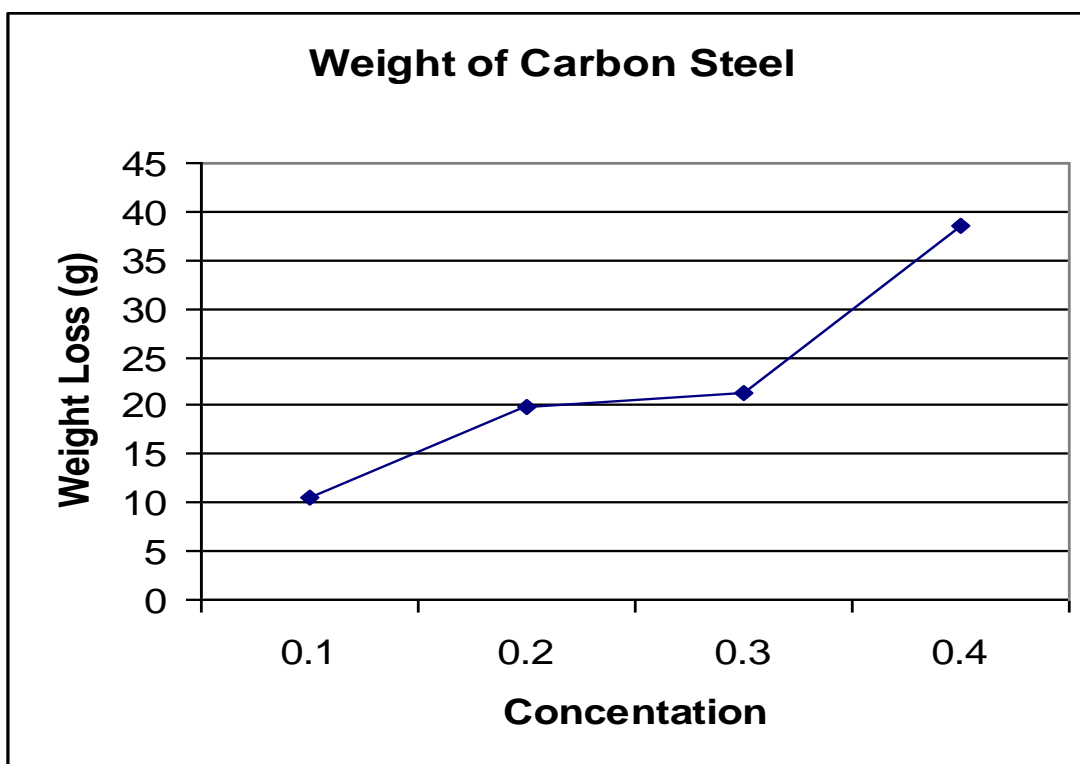


figure 5 weight loss of carbon steel in HCl

3. 3.2- Chromium Steel in Hydrochloric acid

Table 8

Concentration	Weight loss (g)	K (min ⁻¹)	Half life (min)	Corrosion rate g cm ⁻² min ⁻¹ ()
0.1	54.50	4.34×10^{-7}	0.15×10^7	1.55×10^5
0.2	30.24	5.52×10^{-7}	0.12×10^7	2.13×10^5
0.3	35.45	6.66×10^{-7}	0.10×10^7	2.5×10^{-5}
0.4	43.27	8.51×10^{-7}	0.08×10^7	4.22×10^5

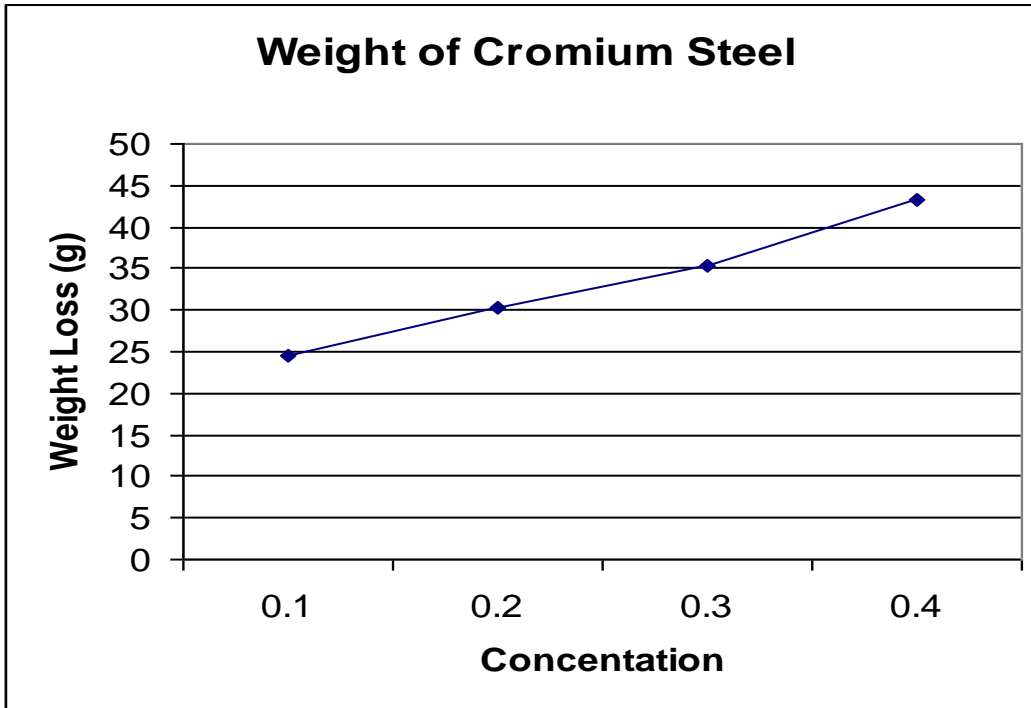
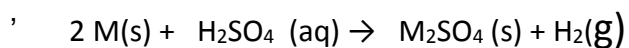


Figure 6 weight loss of chromium steel in HCl

3.4- Sulfuric Acid:

figures 7 and 8 represent weight loss of carbon steel and chromium steel in four different concentrations of H₂SO₄ are plotted in. It could be observed that corrosion of corrosion increase with increase concentration of H₂SO₄ increase. Carbon steel and chromium steel reacts as shown by the reaction equations below:



3.4.1- Carbon steel:

The tables 9, 10 show the specific reaction of carbon steel and chromium steel in sulfuric acid, the value of K increase as the sulfuric acid increase and the half

life decrease and the values of K 0.4 M in (carbon steel and chromium steel) are the same .

Table 9 carbon steel in sulfuric acid

Concentration	Weight loss (g)	K (min ⁻¹)	Half-life (min)	Corrosion Rate (g cm ⁻² min ⁻¹)
0.1	43.27	8.51×10^{-7}	0.08×10^7	3.16×10^{-5}
0.2	47.97	9.71×10^{-7}	0.07×10^7	3.04×10^{-5}
0.3	78.21	20.0×10^{-7}	0.03×10^7	4.96×10^{-5}
0.4	113.15	54.4×10^{-7}	0.012×10^7	4.22×10^{-5}

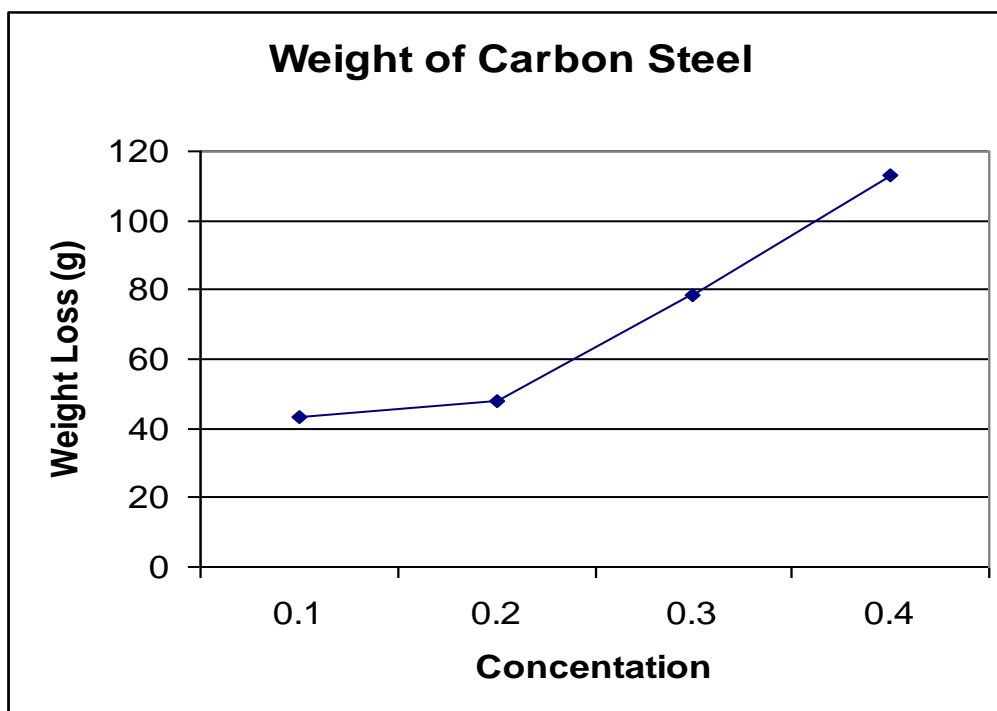


figure 7 weight loss of carbon steel in H₂SO₄

3.4.2- Chromium Steel:

Table 10 shows the value of K, half life and corrosion rate of chromium steel in sulfuric acid

Concentration	Weight loss (g)	K(min-1)	Half life (min)	Corrosion Rate g cm-2 min-1) (
0.1	10.45	1.73×10^{-7}	0.40×10^7	6.94×10^4
0.2	41.91	8.17×10^{-7}	0.084×10^7	2.95×10^5
0.3	62.75	14.08×10^{-7}	0.049×10^7	4.59×10^5
0.4	113.15	54.4×10^{-7}	0.012×10^7	1.10×10^4

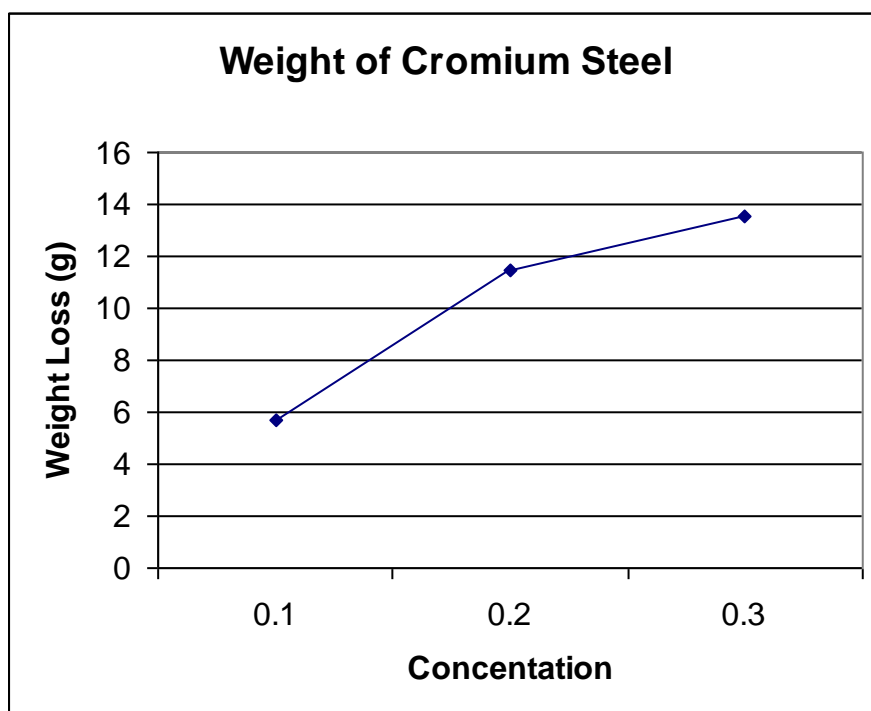
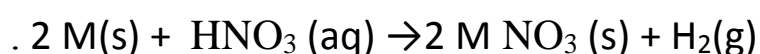


Figure8 weight of Chromium Steel in H₂SO₄

3.5-Nitric Acid :

Weight plots for carbon steel and chromium steel corrosion are plotted in figures 9 -10 .carbon steel and chromium steel corrosion in this medium followed increase corrodibility with increase in time and acid concentration HNO₃

Carbon steel and chromium steel reacts as shown by the reaction equations below:



3.5.1- Carbon Steel:

The specific reaction in the tables 11 ,12 increase as the concentration of nitric acid increase and the half life decrease , the chromium steel gives the highest value comparing the carbon steel and the different medium (sodium chloride , sodium sulfate , hydrochloric acid and sulfuric acid).

Table 11

Concentration	Weight loss(g)	K (min ⁻¹)	Half life (min)	Corrosion Rate (g cm ⁻² min ⁻¹)
0.1	32.32	5.9×10^{-7}	0.117×10^7	2.36×10^5
0.2	47.45	9.57×10^{-7}	0.072×10^7	3.0×10^5
0.3	79.25	20.55×10^{-7}	0.033×10^7	5.02×10^5
0.4	99.07	33.23×10^{-7}	0.020×10^7	6.73×10^5

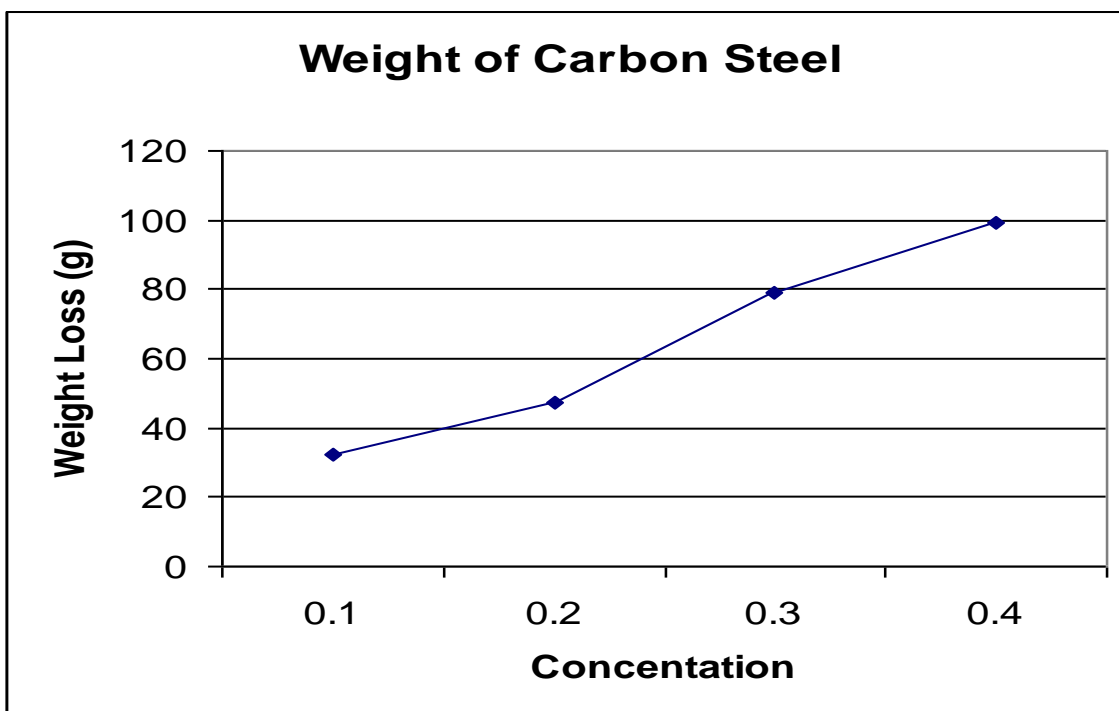


figure 9 weight loss of carbon steel in HNO₃

3.5.2- Chromium Steel:

Table 12 Chromium steel in nitric acid

Concentration	Weight loss (g)	K (min ⁻¹)	Half life (min)	Corrosion Rate g cm ⁻² min ⁻¹ (
0.1	48.79	9.43×10^{-7}	0.069×10^7	3.09×10^5
0.2	53.70	11.29×10^{-7}	0.061×10^7	3.78×10^5
0.3	73.0	17.83×10^{-7}	0.038×10^7	5.34×10^5
0.4	147.56	74.12×10^{-7}	0.004×10^7	1.43×10^4

Figures (1-10) represent weight loss plots (carbon steel and chromium steel) in different media:

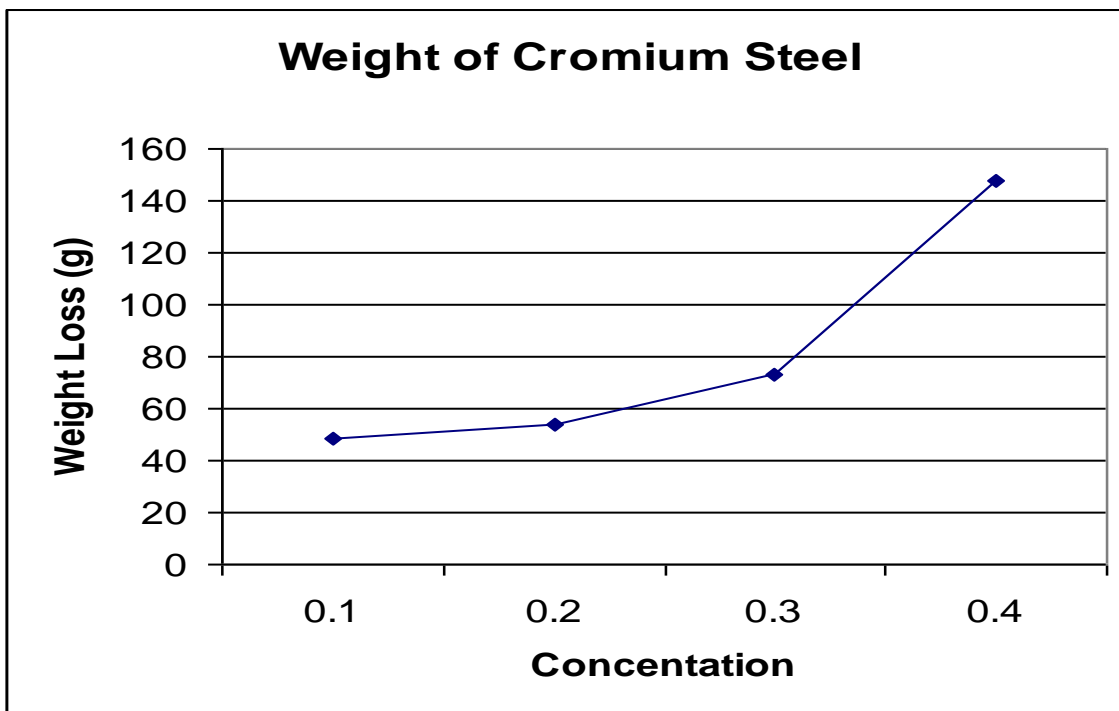


figure 10 weight loss of chromium steel in HNO₃

3.6-Inhibitor

Figures 11-12 represent weight loss plots of galvanized thiourea in NaCl, and HNO₃. The illustrations show that Increase in concentration increases weight loss specimen. The weight loss also increases proportionally with increase in immersion time the acid solutions. HNO₃ has in 0.4 M is more corrosion in carbon steel than chromium steel, and chromium steel is more corrosion in 0.4M NaCl than carbon steel.

3.6.1- Nitric acid:

Figure 11 , 12 and Table13 , 14 illustrated the effect of inhibitor on the corrosion of carbon steel and chromium steel in Nitric acid and sodium chloride in 0.4 M and the values of K and half life are less than without inhibitor . and the values of K in chromium steel in sodium chloride and nitric acide are less than carbon steel

Table 13 carbon and chromium steel in nitric acid

	Concentration(M)	Weight loss(g)	K (min ⁻¹)	Half life (min)	Corrosion rate (g cm ⁻² min ⁻¹)
Carbon steel	0.4	4.69	0.75×10^{-7}	0.92×10^7	0.31×10^{-5}
Chromium steel	0.4	4.27	0.68×10^{-7}	1.01×10^7	0.411×10^{-5}

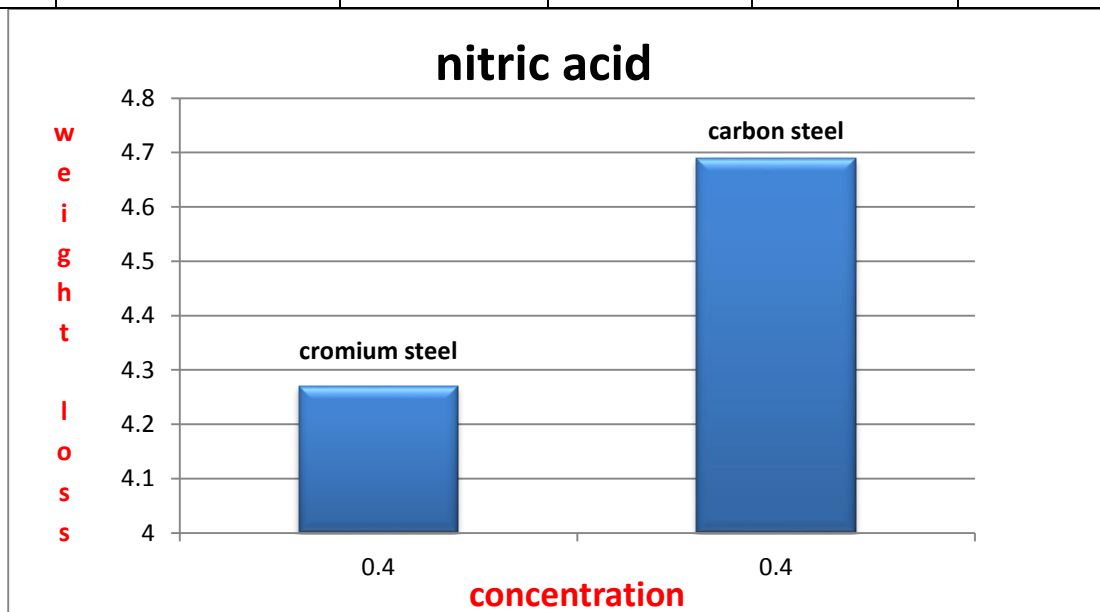


Figure 11 weight loss of carbon steel and chromium in HNO₃

3.6.2- Sodium Chloride:

Table 14 carbon steel and chromium steel in sodium chloride

	Concentration(M)	Weight loss(g)	K (min ⁻¹)	Half life (min)	Corrosion rate (g cm ⁻² min ⁻¹)
Carbon steel	0.4	2.24	0.35×10^{-7}	1.98×10^7	0.16×10^{-5}
Chromium steel	0.4	2.08	0.33×10^{-7}	2.08×10^7	0.13×10^{-5}

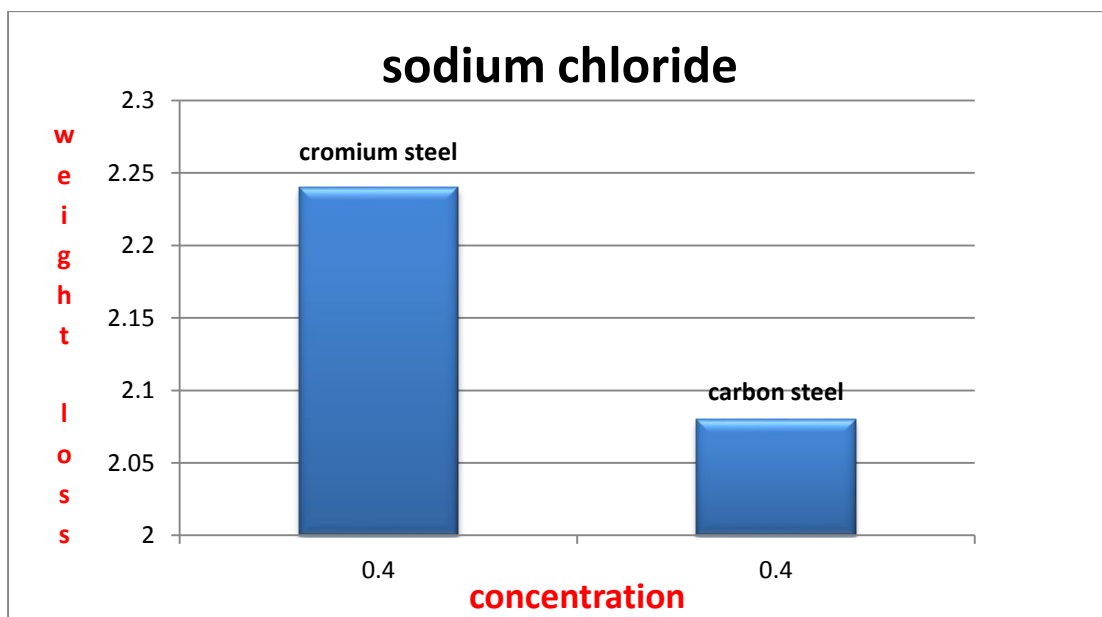


Figure 12 weight loss of carbon steel and chromium steel in NaCl

Conclusion

The corrosion of carbon steel and chromium steel have been studied in different media by weight loss method four different concentration 0.1,0.2 ,0.3 and 0.4 M in one year I made this experiments in different season of year as you know that the different temperature effected the corrosion (increase the temperature increase the corrosion) . also used inhibitor thiourea to study the inhibition of corrosion.

The corrosion of acids are more corrosive than the other salts.

High corrosion of carbon steel and chromium steel is acid solution .

The nitric acid is higher than hydrochloric acid and sulfuric acid.

The illustration shows that increases in concentration of this media increases the weight loss of specimen , and the weight loss increase proportional to the increase in immersion time.

The thiourea decreases the corrosion of carbon steel and chromium steel.

Rate of corrosion reaction (specific reaction rate) of metals increases with a first order degree as acid concentration increases. The half life on the other hand decreases with an increase in concentration.

Recmondations

- To increase the studys and reserachs in this subject on the another type of carbon steel.
- Apply another technical (method)
- Transfer the result of this expirment to industrios.

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