CHAPTER II

LITERATURE SURVEY
CHAPTER 2

LITERATURE SURVEY

This Chapter is divided into three Sections. The work reported in literature on the corrosion of aluminium in hydrochloric acid is reported in Section I. The inhibitors previously reported for the corrosion of aluminium in hydrochloric acid are narrated in Section II. If the substances investigated by us as corrosion inhibitors for aluminium in hydrochloric acid have been used as the inhibitors for corrosion of metals in diverse aggressive media, these uses are reported in Section III.
SECTION I
CORROSION OF ALUMINIUM IN HYDROCHLORIC ACID

The corrosion of aluminium in hydrochloric acid does not start immediately because of the slow attack on the oxide film. Oxides of the type $\text{M}_2\text{O}_3$ always display a sluggish reaction with acids. A long period of induction—which must elapse before the attack becomes vigorous—seems to be connected with the gradual destruction of the skin through dissolution or undermining.

Müller and Low\(^1\) investigated the corrosion rates of aluminium of different grades of purity in hydrochloric acid. They observed that 99.998% pure aluminium was attacked only by 4.0N hydrochloric acid. The dissolution process is characterised by an incubation period during which the metal does not dissolve, an induction period during which the corrosion velocity increases rapidly and the reaction period during which the reaction velocity becomes constant or falls off. With an increase in acid concentration, the incubation and induction periods become shorter.

Straumanis\(^2\) observed that if 99.998% aluminium is first etched in concentrated hydrochloric acid and then introduced into more dilute acid, it is immediately attacked as the induction period is eliminated by the prior dissolution of the oxide film, although the final corrosion rate is not greatly altered.
Gonet observed that increase in acid concentration, temperature or stirring decreases the induction period. The induction period is shorter for an abraded surface than for a polished one.

Aronson and Yoffe reported that the corrosion of aluminium in hydrochloric acid increases with acid concentration to a maximum up to 9.0N and then decreases rapidly. The corrosion of aluminium in 12.0N HCl is about the same as that in 1.0N HCl. The decrease in rate of corrosion is explained on the basis of the formation of a protective chloride layer.

Müller and Löw reported that the presence of 0.5% impurities reduce the corrosion resistance by 1/7000.

Glistenko studied the corrosion of aluminium in 1.0N hydrochloric acid in the presence of several inorganic compounds and reported that addition of potassium dichromate, silver nitrate, ammonium hydroxide, potassium permanganate, magnesium sulphate, ammonium nitrate, ferric chloride and mercuric chloride accelerate the corrosion of aluminium. An addition of hydrogen peroxide in hydrochloric acid increases the corrosion of aluminium.

Nishimura observed the corrosion of aluminium in dilute hydrochloric acid solutions by loss in weight method as well as by gas evolution method. An addition of 0.5% manganese to aluminium-manganese alloy is beneficial to its corrosion
resistance in hydrochloric acid whereas addition of antimony or zinc is not beneficial. Aluminium alloys containing 1.5% manganese and 0.3% chromium were not attacked easily, their corrosion resistance being analogous to that of pure aluminium.

Kuwahara\textsuperscript{7} studied the corrosion of aluminium-silicon alloys in 10.0% hydrochloric acid at 20°C and observed that aluminium containing 3.0% silicon is corroded to a greater extent than that containing 0.26% silicon.

Belyaev\textsuperscript{8} reported that the corrosion of commercial aluminium in 5.0% hydrochloric acid is accelerated by increasing amounts of impurities, especially iron, small amount of titanium do not affect the dissolution rate (less than 0.2%). 0.5% beryllium affects the corrosion only slightly, cobalt and rhenium accelerate corrosion. However, niobium decreases the dissolution rate.

Arora\textsuperscript{9} et al. observed that intergranular corrosion of aluminium in 16.0% hydrochloric acid becomes more rapid with increase in copper content, as little as, 21 p.p.m. copper gives a significant effect. The effect of copper is greater than that of any other residual element. This might be due to an autocatalytic acceleration produced by copper.

Tavadze\textsuperscript{10} et al. reported that aluminium-titanium alloys completely resist hydrochloric acid in all concentrations at
Inui et al. stated that corrosion of aluminium in dilute hydrochloric acid (less than 1.0N) is first order reaction in the beginning but gradually changes to second order. The dissolution in less than 0.5N aqueous hydrochloric acid is 1/2 order but in presence of atmospheric oxygen it is of a first order.
REFERENCES


SECTION II

INHIBITION OF THE CORROSION OF ALUMINIUM AND ALUMINIUM BASE ALLOYS IN HYDROCHLORIC ACID

Sievertz and Lueg reported that alkaloids retard the corrosion of aluminium in hydrochloric acid.

Sievertz and Lueg reported that at 0.0025M concentration, strychnine affords 95% protection to aluminium in 1N HCl. Strychnine affords 66-94% protection to aluminium zinc alloy in 0.5N hydrochloric acid for 1-6 hours. It acts by influencing cathodic reaction. It affords more than 95% protection to aluminium copper alloy (91% Al) in 1N HCl for 1-3 hours.

Quinine hydrochloride was found to be a satisfactory inhibitor for aluminium-copper alloy (91% Al) in 1N HCl, the efficiency being > 95%. It acts by influencing the cathodic reaction. Quinine hydrochloride affords 70-90% protection to aluminium-zinc alloy in 0.5N HCl.

It is possible to afford complete protection to aluminium in hydrochloric acid by impressing low cathodic currents in presence of quinine hydrochloride.

Lichtenberg reported the use of nicotine sulfate for the
prevention of corrosion of aluminium in hydrochloric acid.

In 1N HCl, for 1-6 hours, Nicotinic acid affords 84% protection to SIB-HH alloy (99.5% Al)\(^{10, 11}\), 81-86% protection to aluminium manganese alloy\(^{12}\) (92% Al).

Nicotinic acid protects aluminium-zinc alloy (86% Al) to an extent of 78-96% in 0.5 NHCl\(^{49, 2}\) and to an extent of 88-98% in 1N HCl\(^3, 6\). Aircraft aluminium-copper alloy (91% Al) is protected in N HCl to an extent of 95-99%\(^5, 3, 6, 7\).

Nicotinic acid is an excellent inhibitor for aluminium\(^{13-19, 19A}\).

Nicotinic acid functions by influencing the cathodic reaction\(^{20}\).

Complete protection can be afforded to 99.5% aluminium in 1.25N HCl by a combination of 2 gms/liter nicotinic acid and 0.024 amp/cm\(^2\) cathodic current\(^{20}\).

Jenckel and Waltman\(^{21}\) reported that at 0.003M concentration, acridine and \(\beta\) phenyl acridine afford more than 97% protection to aluminium in 1N HCl. Aronson\(^{22}\) and Yoffe studied acridine bases as corrosion inhibitors for aluminium in hydrochloric acid and observed that while acridine protects aluminium to an extent of 92%, substitution by sulfur increases, inhibitor activity.

Brooks\(^{25}\) also reported acridine derivatives as corrosion inhibitors for aluminium in HCl.

The efficiency of acridine in 1N HCl for 1 to 6 hours is 96% for SIB-HH alloy (99.5% Al)\(^{10, 11}\), 93% for commercial aluminium
(92% Al)\(^{10}\) and 97-99% for aircraft aluminium copper alloy (91% Al)\(^{10,5,3,6}\) and 97-98% for aircraft zinc alloy\(^{3,6}\).

Acridine leaves an adherent, brown deposit\(^{10}\). For aluminium manganese alloy (92% Al), the efficiency was more than 97% in 1N HCl for 2-6 hours\(^{12}\). The corresponding value for aluminium zinc alloy in 0.5N HCl (86% Al) being 94-99%\(^{4,3,7}\).

Acridine is an excellent inhibitor for aluminium \(^{23,14,16,18,19}\).

Jenckel\(^{24}\) and Braucker reported that \(\beta\) napthoquinoline inhibits the corrosion of aluminium in 3-6N hydrochloric acid at 25-50\(^{\circ}\)C by decreasing the surface tension of the medium and by increasing the effective potential. Jenckel\(^{21}\) and Woltman reported that 0.003M \(\beta\) napthoquinoline and 2-phenylquinoline afford more than 97% protection to aluminium in 1N HCl. Brooks\(^{25}\) reported that quinoline derivatives retard the corrosion of aluminium in hydrochloric acid.

Geller\(^{26}\) studied amines as inhibitors for the corrosion of aluminium in hydrochloric acid solutions and observed that methyl, dimethyl, ethyl, diethyl, triethyl, propyl, butyl and amyl amines afford temporary protection to aluminium in 1 to 5% HCl to an extent of 90% or more. The protection is roughly proportional to nitrogen concentration and to chain length of the hydrocarbon radical. The efficiency diminishes with increase in time and temperature. Colgate\(^{27}\) reported that 1% concentration of methyl, ethyl, propyl, and butyl amines are effective inhibitors for corrosion of aluminium in HCl, but the protection decreases as temperature rises.
Complete protection can be afforded to 99.5% aluminium in 1.25N HCl by 2.5 gms/liter methylamine\textsuperscript{20} and 0.064 amp/cm\textsuperscript{2} cathodic current or 2.5 gms/liter ethylamine\textsuperscript{20} and 0.032 amp/cm\textsuperscript{2} cathodic current\textsuperscript{6}.

Butylamine satisfactorily retards the corrosion of aluminium-manganese alloy in 1-2N HCl at 32\textdegree{}C\textsuperscript{28}, the efficiency being 16-75\%. For pure aluminium, the efficiency of butylamine ranges from 63 to 68\%, the optimum inhibitor concentration being 0.5 gms/liter\textsuperscript{30}. The efficiency of butylamine is less than that of dibutylamine and tributylamine\textsuperscript{29, 31, 32}.

The efficiency of dibutylamine in hydrochloric acid ranges from 40-90\%\textsuperscript{50}. For pure aluminium in 1.25N HCl, the optimum inhibitor concentration is 0.5 gms/liter, the efficiency being from 80 to 87\%\textsuperscript{30, 32}.

Tri-n-butylamine was studied as an inhibitor for the corrosion of pure aluminium in HCl\textsuperscript{33}. It shows a significant inhibitor action up to 3 hours\textsuperscript{34, 30}. Inhibitor efficiency increases with increase in inhibitor concentration up to 0.5 gms/liter and then becomes constant\textsuperscript{32}.

The order of efficiency of butylamines is tri-n-butylamine $>$ di-n-butylamine $>$ n-butylamine. All these substances exercise the inhibitive action by influencing the cathodic reaction.

Brook\textsuperscript{25} reported hexamine as an inhibitor of the corrosion of
aluminium in hydrochloric acid. Sheth and Ramachar studied hexamine as an inhibitor of corrosion of 99.5% aluminium in HCl and observed that it acts by influencing the cathodic reaction.

Lichtenberg reported that colloids protect aluminium in hydrochloric acid.

Dextrine protects aluminium manganese alloy (92% Al) to an extent of 73-84% in 1N HCl for 3-4 hours. For SIB-HH alloy (99.5% Al) the efficiency of dextrine ranged between 58 and 92%. With time, efficiency decreased for 2 hours and was constant thereafter. For aluminium zinc alloy, its efficiency is only 40% in 1N HCl, for 1 hour. Dextrine is a good inhibitor for aluminium in HCl.

It is possible to obtain complete protection by a combination of low impressed cathodic current and inhibitor. Thus 99.5% aluminium is completely protected in 1N hydrochloric acid for 2 hours by 2 gms/liter dextrine and 0.032 amp/cm² cathodic current.

De Angelis reported that tannic acid or colophony inhibit the corrosion of 99.0% aluminium in 1N hydrochloric acid. At 25°C, 50°C, 75°C for 1 to 10 hours, both substances afford protection to an extent of 99%. In a subsequent communication he observed that tannic acid exercises its inhibitor action by influencing both cathodic and anodic reactions, because it gets adsorbed on the metal surface generally.
Tannic acid protects aluminium manganese alloy in 0.5-4.0N HCl at 32°C to an extent of 80-86%. It also inhibits the corrosion of 99.5% aluminium in 0.5-1.5N HCl \( ^{11, 23, 14, 19} \).

De Angelis\(^ {40} \) studied Phenol, Pyrocatechol, resorcinol and hydroquinone as inhibitors of the corrosion of 99% Al in 1N hydrochloric acid. Efficiencies of the inhibitors at lower concentrations are in the order, resorcinol > hydroquinone > pyrocatechol > phenol whereas at higher solutions the order is changed to hydroquinone > phenol > pyrocatechol > resorcinol. He also tested pyrogallol, phloroglucinol, \( \alpha \) napthol and \( \beta \) napthol as corrosion inhibitors for aluminium in 1.0N hydrochloric acid.

Jablczynski\(^ {51} \) and Pierzchalski reported that 0.005N thiourea protects aluminium in 1.4N hydrochloric acid to an extent of 75%. The efficiency of thiourea in 1N HCl for 1-6 hours is 94% for commercial aluminium (91% Al)\(^ {10} \), 50-31% for SIB-HH alloy (99.5% Al)\(^ {10, 11} \), more than 94% for aircraft copper alloy (86% Al)\(^ {10, 5, 3, 6} \) and more than 99% for aircraft zinc alloy (91% Al)\(^ {10, 5, 3, 6} \), and 94% for aluminium manganese alloy (92% Al)\(^ {12} \). It was indicated that thiourea acts by influencing the cathodic reaction\(^ {41, 42} \). Satisfactory protection could be afforded to pure aluminium by a combination of an improved cathodic current of 0.1 amp/sq. dm and 1.5 g/liter thiourea\(^ {20} \). Thiourea is an excellent inhibitor for aluminium\(^ {13, 23, 14, 16, 6, 18, 19, 37} \).
Diphenylthiourea is a moderate inhibitor for the corrosion of aluminium-manganese alloy in HCl. It acts by influencing the cathodic reaction.

Acridine, thiourea and nicotinic acid were studied as inhibitors of corrosion of commercial aluminium (92% Al), (1) SIB-HH alloy (99.5% Al), (2) Aircraft zinc alloy (86% Al), (3) Aircraft copper alloy (91% Al), and (4) in 1N HCl at 32°C.

For all the inhibitors, the order of efficiency was

4 > 3 > 1 > 2.

For all the alloys, the order of efficiency was acridine > thiourea > nicotinic acid > dextrine.

For aircraft aluminium zinc alloy (80% Al) and aircraft aluminium-copper alloy (91% Al) and pure aluminium the order of efficiency in HCl solutions is acridine > thiourea > nicotinic acid > dextrine.

Titova reported that CHM, (found as a byproduct of the oil industry) containing quinoline and pyridine, naphthalene and phenol inhibit the corrosion of aluminium in hydrochloric acid. It is proposed that such organic compounds, approach the charged surfaces of the metal, attract electrons, and combine with the metal through the primary valencies of the nitrogen.

Reuther patented the use of dioxodisiloxane or trialkoxyoxysilanes as inhibitors for the corrosion of aluminium in hydrochloric acid.
Desai and Desai\textsuperscript{45} reported that furfuraldehyde is an excellent inhibitor of the corrosion of aluminium 2S and 3S in hydrochloric acid solutions. It is effective even at very low concentrations. With an increase in the concentration of furfuraldehyde, the efficiency rises up to a particular concentration and then decreases due to the cathodic depolarisation at higher concentrations of aldehyde. It functions by polarising the cathodic areas. The efficiency of furfuraldehyde increases with the increase in acid concentration.

Lichtenberg\textsuperscript{9} reported the use of dibenzyl sulfide, resin and phenol formaldehyde condensation products as inhibitors for corrosion of aluminium in hydrochloric acid and observed that phenol formaldehyde condensation products afford 90\% protection to aluminium in 2N hydrochloric acid.

Jablczynski and Pierzchalski\textsuperscript{51} reported that hydrogen sulfide and carbondisulfide strongly accelerate the corrosion of aluminium in 1.4N hydrochloric acid whereas iodine and thio-cyanic acid retard it to an extent of 60\% and 47.6\% respectively.

Balezin\textsuperscript{46} and Dobychina reported that PB-5, PB-8 and anabasine oxalate are excellent inhibitors for the corrosion of aluminium alloys in hydrochloric acid.

Horner\textsuperscript{47} studied dibenzylsulfoxide, dibenzylsulfide, triphenyl-arsinoxide, benzyl isoquinolinium bromide, tribenzylsulfonium
hydrogensulfate, diphenyl-dibenzyl-phosphonium chloride, diphenylidithiophosphonic acid, thiourea, o-tolythiourea, mercapto benzthiazole, alizarin, phenanthridine, acridine, as inhibitors for the corrosion of aluminium in 1N HCl. Dibenzylsulfide affords practically complete protection (98%). The performance of dibenzylsulfoxide (92%), triphenylarsin oxide (87%), tribenzylsulfonium hydrazine sulfate (87%) and Acridine (86%) is also satisfactory.

Substituted ethynyl cyclohexyl compounds where the substituent is -OH,-OCOCH₃,-OCH₂-CHOHCH₂ or -NH₂ have been patented as corrosion inhibitors for aluminium in hydrochloric acid.
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SECTION III

LITERATURE SURVEY OF SUBSTANCES STUDIED
BY US AS CORROSION INHIBITORS FOR OTHER
METALS AND ALLOYS

I. ALCOHOLS

Cyclohexanol has not been reported in literature as corrosion inhibitor so far.

Methyl cyclohexanol retards the corrosion of metals in petroleum residues\(^1\) and has been used as a constituent of anticorrosion oil\(^2\).

II. KETONES

2-butanone has been used to retard the corrosion of copper\(^3\) and 63/37 brass\(^75\) in potassium persulphate solutions.

2,4 pentanedione has been investigated as an inhibitor of corrosion of iron, zinc and cadmium in hydrochloric acid\(^4\) and the corrosion of aluminium\(^5\), and copper in sodium hydroxide solutions\(^76\). It has also been used to retard the corrosion of copper\(^3\) and brass\(^75\) in sodium hydroxide and potassium persulphate solutions. 2,5 hexanedione has been used to retard the corrosion of aluminium in chlorinated hydrocarbons by Minford\(^6\).

Cyclohexanone has been used to retard the corrosion of steel in sulphuric acid at 70-100\(^\circ\)C.\(^7\), the corrosion of copper and brass in sodium hydroxide solutions\(^76\), and potassium persulphate solutions\(^77, 75, 3\), and the corrosion of 63/37 brass in alkali\(^79, 75, 3\).
III. AMINES

Geller\textsuperscript{8} reported that several aliphatic amines inhibit the corrosion of aluminium in 1.4N hydrochloric acid but the protection afforded is temporary.

Methylamine, dimethylamine and trimethylamine retard the corrosion of tins by canned foods\textsuperscript{9}.

Morpholine, cyclohexylamine and ethylenediamine are reported to be suitable inhibitors for steam corrosion\textsuperscript{10}.

Mann\textsuperscript{10A} et al. studied aliphatic and aromatic amines as inhibitors for the corrosion of iron by acids. The salts formed by aromatic amines with corroding acids form blanket ing layers of positive ions adsorbed on the cathodic areas of the metal surface on the Nitrogen atom. The inhibitor structure determines the closeness of packing of the ions in the covering layer and the character of the film as to its penetrability. Later, Mann and Chiao\textsuperscript{10B}, correlated the molecular structure of inhibitors with the efficiency.

Antonucci\textsuperscript{11} investigated a number of amines as inhibitors for the corrosion of iron in 1.0N sulphuric acid by loss in weight method. He reported that the inhibitive power depends on the stereochemical properties of the amine, i.e., the area of the metal covered and protected by a single adsorbed molecule. At 0.25% Nitrogen, the order of efficiency was found to be trimethylamine = diethylaniline $>$ triethylamine.
triethylamine $>_{\text{m}}$ methylaniline $>_{\text{e}}$ ethylaniline $>_{\text{d}}$ dimethylaniline $>_{\text{d}}$ dimethylamine $>_{\text{de}}$ diethylamine $>_{\text{e}}$ ethylamine $>_{\text{m}}$ methylamine.

Nathan$^{12}$ studied retardation of the corrosion of iron powder and strips by 1.0N hydrochloric acid by aliphatic and aromatic amines, viz. methylaniline, ethylaniline, dimethylaniline, diethylaniline, ethylamine, diethylamine, triethylamine. The effectiveness of amines increases with increase in molecular weight in the homologous series. Steric factors which are more favourable to adsorption of the amine on metal solution interface increase its inhibitor efficiency.

Diethylaniline, dimethylaniline, ethylaniline, diethylamine, were observed as corrosion inhibitors for iron in sulphuric acid by Ride, who reported that the amines are adsorbed in a single and fixed mode of molecular orientation$^{13}$.

Aniline, methylaniline, ethylaniline, dimethylaniline, diethylaniline, methylamine and ethylamine were investigated as corrosion inhibitors for pure iron in 1.0N hydrochloric acid by Kaesche and Hackerman who observed that these substances act as inhibitors by mixed control$^{14}$.

Morpholine and ethylenediamine have been suggested as inhibitors for steel by dimethyl sulfoxide$^{15}$.

Ethylamine, diethylamine, triethylamine, ethylene diamine,
methylamine, were evaluated as corrosion inhibitors for metals in aqueous non acid solutions. The amines which were effective in acidic media were ineffective in non acid solutions. For soluble amines of low molecular weight, high pH was necessary in dilute solutions.

Dimethylamine, diethylamine and ethylenediamine have been investigated as volatile corrosion inhibitors for steel.

Antropov and Banerjee studied diethylaniline, ethylaniline, dimethylaniline, diethylamine, as corrosion inhibitors and calculated the inhibitor coefficients from electrocapillary measurements.

Trimethylamine and triethylamine have been suggested as corrosion inhibitors.

Diethylamine and dimethylamine retard the corrosion of steel by combustion products of sulphur containing fuels.

Aniline, o-chloroaniline, m-chloroaniline, o-toluidine and m-toluidine have been studied as inhibitors for acid corrosion of iron.

Zwierzykowska used aniline, diethylaniline, ortho and para toluidine to retard the corrosion of zinc and cadmium in 1.0N hydrochloric acid and correlated the inhibitor action with electrocapillary curves.
Dimethylaniline and ethylenediamine have been investigated as corrosion inhibitors by R. Jenny. Diethylamine and triethylamine have been patented as inhibitors for the corrosion of oil well casing and tubing. Dimethylaniline has been used to prevent corrosion of steel equipment during rectification of hydrocarbons. Diethylamine has been investigated as inhibitor for corrosion of lead and tin in nitric acid, as inhibitor of corrosion of automobile engines to protect equipment of oil wells.

Ethylenediamine has also been used as a vapourphase inhibitor.

Cyclohexylamine is used to retard the corrosion of condensers and boiler corrosion, as volatile inhibitor. It also prevents the corrosion of carbon steel in carbon tetrachloride.

Morpholine retards the corrosion of metals in sulphurous media, the corrosion of condensers and boiler corrosion as inhibitor, as inhibitor for the corrosion of copper in petroleum, as vapourphase inhibitor.

Aniline has been used to retard the corrosion of iron in acids, the corrosion of copper in benzene, in sulphuric acid, the corrosion of copper, iron and nickel in acids, the corrosion of copper and brass in nitric acid, as a vapourphase inhibitor. Its adsorption on mercury has also
been studied\textsuperscript{69-71}. o-anisidine has been studied as an inhibitor for the corrosion of copper\textsuperscript{72} and brass in nitric acid. Dimethylaniline has been patented as corrosion inhibitor\textsuperscript{73}, diethylaniline retards the acid corrosion of steel\textsuperscript{74}. 
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