

A Review on Effect of Alloying Element on Aluminium Anode

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Abstract

Aluminium is most widely used as a structural material world wide because of its unique properties and ability to alloy with other elements. These elements improve desirable mechanical properties and remarkably affect the chemical properties by changing the microstructure of Aluminium from homogeneous solid solution to complex structure with multiple intermetallic phases. This will change the corrosion resistance of pure aluminium as it belongs to a group of passive metals. Aluminium is the most preferable metal for sacrificial anode for cathodic protection techniques because of its lightweight, high anode efficiency and current capacity. Pure and unalloyed Aluminium is not suitable for a sacrificial anode due to the development of the protective layer on its surface this will restrict direct contact of Aluminium with the Environment. The Reactivity of Aluminium can be changed from passive to active by adding other elements. Chemical composition is the primary parameter that governs the passivity and corrosion resistance of metals. This article deals with the overview of the role of alloy chemistry and important aspects like a breakdown of passive film and formation of an intermetallic compound that will affect the behaviour of Sacrificial Aluminium Anode.

Keywords: Aluminium alloy, Sacrificial anode, Passivity, Electrochemical Behaviour.

1. Introduction

Metal degradation or corrosion is one of the far reaching challenges in the industrial world. It is nothing but the reaction of metal with its service Environment and this will lead to the deterioration of valuable service properties of the metal. Several techniques have been developed to prevent corrosion like inhibitors, design modification, coating and anodic-cathodic protection from which Cathodic protection (C.P.) being deployed by major industries.[1-3] Cathodic Protection is nothing but reduce or restrict the corrosion by making the metallic structure as a cathode in corrosion cell. There are two cathodic protection methods that impressed current method and sacrificial anode method respectively. Out of these two methods sacrificial anode method is most probably used for preventing oil pipelines, marine structure and some domestic structures. This is most widely accepted due to several benefits like ease of installation, no need for an external power source, and also suitable for localized protection. Main objective of this method is that the metallic structure to be protected is coupled with more active metal (anodic metal) and makes it cathode. Hence, all the corrosion concentrated at active metals called a sacrificial anode. This is brought off by altering the electrode potential of the metallic structure so that it can be placed in the immunity region, therefore further corrosion is not allowed [2,4].

Nowadays, Magnesium (Mg), Zinc (Zn) and Aluminium (Al) are most preferable sacrificial metals for cathodic protection. From which, Aluminium is acquire appreciable properties as the sacrificial metal like light in weight and density, ease of availability, large electrochemical equivalent, thermal and electrical conductivity, high current capacity, and reasonable cost.[5]

2. Passivity of Aluminium

Passivity is nothing but loss of chemical reactivity. Some active metal in a certain environment reacts with the environment and forms a layer of corrosive product on its exposed surface; this product may be oxide, sulphide, and hydroxide; this layer acts as a barrier for the direct contact of metal to the environment further reduces the reactivity of metals. In such a way Aluminium is also covered with an oxide layer when aluminium comes in contact with atmospheric oxygen, whether in solid or liquid form.[6]

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The existence of this film was first reported by Joseph W Richards in 1896. There is just about the immediate formation of an oxide film having the general formula $Al_2O_3.nH_2O$ as given by the reaction a, b and c respectively, which is few nanometres thick and this film isolates the aluminium from direct contact with the environment. [7, 8]

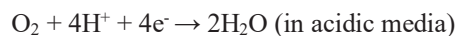
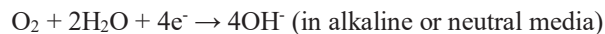
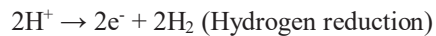
Table1. Comparison between sacrificial anode materials [3]

Properties	Aluminium	Zinc	Magnesium
Density (gm/cc)	2.70	7.13	1.74
Open circuit potential (SHE)	-1.66	-0.76	-2.38
Anode capacity (amp.hr/kg)	2500	780	1251
Consumption rate (kg/amp.yr)	3.4	11.5	7
Environment	Seawater Freshwater Backish water High and low resistive medium	Low resistive medium	High resistive medium Subsoil Freshwater

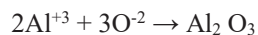
[a] Oxidation:



[b] Reduction:



[c] Passivation:



This passivating behaviour of aluminium is subject to the stability of oxide film and this is express by their Pourbaix diagrams which give idea about the thermodynamic oxidizing and reducing power of the major stable oxides, hydroxides, and oxyhydroxides of a various chemical compound. If we talk about aluminium oxide then it is a very stable oxide and also Rpb (Pilling-Bedworth ratio) for this oxide is 1.28 which concludes that this oxide film is denser than metal. This property of oxide film is enough for protecting the aluminium from further corrosion that's why aluminium has great atmospheric corrosion resistance. The parameter that affects the development of oxide is oxygen pressure, temperature, the surface of the metal and chemical substances present in the metal.[9,10]

3 Depassivation of Aluminium

Pure and unalloyed aluminium cannot be used as the sacrificial anode because of the passive behaviour of aluminium as we discussed in the above paragraph. The breakdown of this passive film (Al_2O_3) improves the performance of the sacrificial aluminium anode. [1,2] Breaking or weakening of the passive film moves the aluminium surface from the naturally passive state to the active state. Methods for this adjustment of aluminium surface by applying cathodic current, or by addition of alloying elements at a low quantity having a suitable negative potential which encourages depassivation and shifts the operating potential of the aluminium to the more

electronegative direction. [11-12]

In 1966, Reading and Newport carried out study to understand event of the addition of different metals on the performance of aluminium sacrificial anodes in seawater and found that many metals which improve the efficiency of the anode by forming intermetallic and other second phase particle depends on solubility limit of the solvent metal in Aluminium matrix (Table 2) and make more active aluminium electrode potentials. Generally, alloying elements are added into Aluminium are term as depassivators and modifiers. [8,13]

Table 2 Solubility of the elements in aluminium [14]

Alloying element	Temp (°C)	Solid solubility(wt%)	Liquid solubility(wt%)	Type of system
Zn	380	82.8	95.0	Eutectic
Mg	450	14.9	35.0	-
Cu	550	5.67	33.15	Peritectic
Si	580	1.65	12.16	-
Sn	230	<0.01	99.5	Eutectic
Ti	665	1.0	0.15	Peritectic
Zr	660	0.28	0.11	Peritectic
Pb	660	0.15	1.52	-
Ga	30	20	98.9	-
Bi	660	0.87	<0.1	Monotectic

Addition of alloy metal and impurities present in aluminium can pay-off degradation in passivating properties and it is basically governed by the nature and distribution of intermetallics compounds, the shape and particle size of the intermetallics. Beginning with the physical metallurgy of Al alloys clearly shows that all the alloying elements have negligible solubility at room temp and the addition of elements beyond this limit will give rise to the formation of another solid solution or compound. These compounds may be binary, ternary or even quaternary depending on the chemistry of the developed alloy. However, an alloying element can also form compounds with or without aluminium. This is possible in the system of Mg and Zn they will produce Mg_2Si and $MgZn_2$ in the 6XXX and 7XXX series respectively. These intermetallics compounds form in the liquid state by eutectic and peritectic reaction or in the solid state by precipitation process during the cooling or heat-treatment process.[15-16]

These intermetallics compounds take part in the galvanic corrosion process of the alloy, which may be anodic or cathodic to the aluminium matrix depending on their open circuit potential as listed in table 3. Therefore, this brings out development of galvanic corrosion cells between intermetallics and aluminium matrix and result is dealloying or localized corrosion. The Driving force for localized corrosion is the potential difference between intermetallics and aluminium matrix. These intermetallics lead to generalized surface corrosion like pitting; this will be initiated by an oxygen-reduction process on cathodic intermetallics. Pitting is usually starting when the oxide layer is breaking and it is the origin of localized corrosion.[16-17]

The intermetallics may be Age-hardening precipitates, in the ageing process, the usual sequence is the formation of supersaturated solid solution (SSSS)-GP zones-metastable phases- stable phases. In GP zones [Guinier-preston zone], decomposition of SSSS of Al is producing precipitates (ppts) that may be fine or coarse. fine ppts distributed throughout the matrix and coarse ppts concentrated on grain boundaries, and intergranular corrosion and stress corrosion will be initiated. The coarsening of ppts restricts the growth of the protective oxide layer and breaking in the oxide layer initiates corrosion. As suggested by several theories, at the breaking point where is no oxide layer is present from that area pitting corrosion is started in presence of chlorides which leads to Al dissolution [17-18]

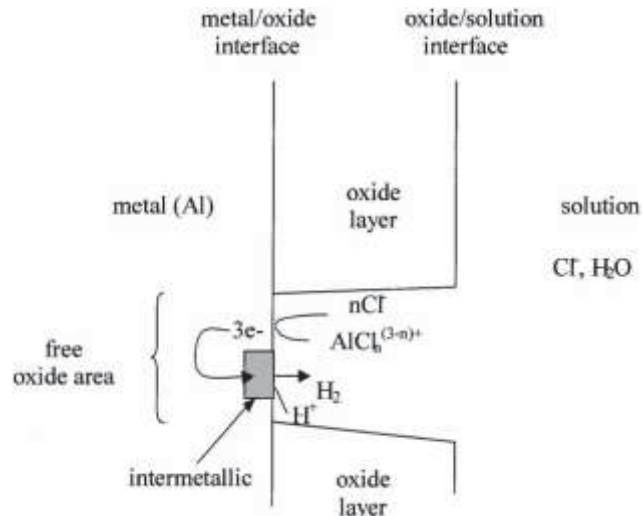


Fig 1 Formation of pit in oxide free area [7]

Table 3 open circuit potential of intermetallic compounds (NaCl, H₂O₂ solution, ASTM G 69)[16]

Position	Intermetallic Compound	Open Circuit Potential (mV SCE)
Anodic	Cu	-110
	Si	-170
	Al ₃ Fe	-470
	Al ₂ Cu	-640
	1050A	-750
Cathodic	Al ₆ Mn	-760
	MgZn ₂	-960
	Al ₃ Mg ₂	-1150
	Mg ₂ Si	-1200

4. Effect of Chemical Composition

Various research studies suggest that alloying addition has the strongest effect on the aluminium anode. Here, we summarized some of their research papers mainly focusing on the effect of alloying elements addition for improving dissolution of alpha α aluminium matrix in the binary compound of Al-X (X-Zn, Mg, Fe, Sn, Ti etc) and Ternary compound of Al-X-Y (X, Y is Zn, Mg, Fe, Sn, Ti etc).

4.1 Effect of Zinc

Zinc is a major alloy in the 7XXX series, which is heat treatable and has the highest strength from the other Aluminium alloy. [19] Zn has the greatest solid solubility in the aluminium matrix which is 66.4 atomic weight percentage and Zn makeup a eutectic type of system with aluminium, in which liquid aluminium give α solid solution of Aluminum and Aluminum rich intermetallic phase [20].

As suggested by the reference paper [2], pure aluminium has an anode efficiency of 21.23% which can be increased by increasing zinc content from 1-8% and maximum anode efficiency is available at 6% Zinc which is 86.59%. This paper established that at this amount of Zinc, the formation of β phase will start which is confirmed from the Al-Zn phase diagram. This phase takes part in the breakdown of an aluminium oxide film by forming a galvanic cell within the α - Al matrix. This detailed phenomenon is studied in this paper. This β phase is more negative to the α - Al matrix so it will act as cathode and the α - Al matrix will act as anode and the dissolution of the α - Al matrix will increase. Thus, anode efficiency is increasing with an increase in Zn content.

As given information by the reference paper [21], in this case also the addition of Zn in pure Al will improve anodic oxidation of developed alloy and it is confirmed by developing Evans diagram for that alloy that shows shifting of potential in anodic direction.

4.2 Effect of Tin

Sn has less solid solubility in an aluminium matrix which is less than 0.002 atomic weight percentage and Sn forms a eutectic type of system with aluminium. Sn has maximum solid solubility of 0.10% between the melting point of Aluminium (660°C) and eutectic temp (228.3°C) is confirmed by the binary diagram of Al-Sn. [20].

In the reference paper [1] information has been provided regarding Sn addition in different proportions 0.01%, 0.05% and 0.1% respectively in Al-Zn-Mg alloy. Paper opines that anode efficiency increases with increasing Sn content and maximum efficiency available at 0.1 Sn which is 98.65% and it will also increase with a time exposure. This paper points out that due to the addition of tin in different proportions in the Al matrix, Sn globules will form in the matrix which will act as a nucleation site for the corrosion of the matrix and self corrosion of anode will improve progressively.

4.3 Effect of Titanium

Ti has solid solubility in the aluminium matrix which is 0.57 atomic weight percentage which is less than 1% and Ti forms a peritectic type of system with aluminium and forms an intermetallic phase with Aluminium. Maximum solubility occurs at peritectic temp which is around 665° C [20].

This reference paper [22] clearly states that titanium acts as a grain refiner which affects the distribution of alloying elements within the matrix. This paper also communicates that Ti will form the second phase precipitates $TiAl_3$, as the Ti content increases the number of precipitates increases and Ti also helps in the uniform distribution of Zinc & Indium through the Al matrix. Uniform distribution of alloying elements avoids segregation at the grain boundary and which will reduce intergranular corrosion. Also, corrosion uniformity & self corrosion due to the formation of second phase particles will be improved by Ti addition.

4.4 Effect of Titanium and Magnesium

Mg is a major alloying element in the 5XXX series[19] Mg has maximum solid solubility in an aluminium matrix which is 16.26 atomic weight percentage around 450° C and Ti have solid solubility in an aluminium matrix is 0.57 atomic weight percentage around 665° C [20].

A detailed study of this reference paper [23] clearly state that the addition of Ti and Mg in Al-Zn-In Alloy will give rise to the formation of $TiAl_3$ and $MgZn_2$ particles in Al matrix which are excellent nucleation site for the galvanic corrosion of anode. In this situation, the second phase particles of Mg and Zn act as anode and the passive film acts as a cathode. Due to this dissolution of the second phase particle in an aluminium matrix is observed. Now, these particles are more negative compared to Al-matrix so the micro galvanic cell is formed within the matrix in which particles act as cathode and dissolution of the Al matrix will start. This will increase self corrosion of anode. Ti also acts as grain refiner in the Al matrix so the addition of Ti also increases uniformity of corrosion and it will be beneficial to improve self corrosion.

4.5 Effect of Titanium and Zirconium

Ti and Zr have solid solubility in an aluminium matrix which is 0.57 and 0.085 atomic weight percentage respectively which is less than 1% and form a peritectic type of system with aluminium and form intermetallic phase with Aluminium. Maximum solubility occurs at peritectic temp which is around 665° [20].

As suggested by the reference paper [24], Ti and Zr both act as grain refiner in Al-matrix. A detailed study of Titanium and Zirconium addition in Al-Zn-In alloy establishes that during solidification of alloy Zn is segregated at grain boundary which reduces the uniformity of corrosion and sometimes it will form interdendritic structure within a matrix and reduce self corrosion. Grain refiner reduces this effect and makes the alloying element uniformly

distributed through the Al matrix. Ti will form the $TiAl_3$ phase which produces a good nucleation site for corrosion and the dissolution of Al-matrix will start. Due to the addition of grain refiner more finer & uniform structure will be produced which will promote more uniform dissolution and anode performance.

4.6 Effect of Silicon

Silicon is the primary element in the 6XXX series and they are heat-treatable alloys. [19] Si has maximum solid solubility in an aluminium matrix which is 1.59 atomic weight percentage around $1080^\circ C$ [20].

As mentioned in the reference paper [15] after adding Si in Al-matrix it will form an equiaxed fine-grain during solidification which will reduce grain boundary segregation of Zn and improve corrosion uniformity. In presence of Mg, it will form Mg_2Si particles and these particles uniformly distributed at the grain boundary. These second phase particles of Mg and Si will act as a cathode and make an aluminium matrix anode. Silicon improves hardness, wear resistance and castability.

4.7 Effect of Lanthanum

Lanthanum has maximum solid solubility in an aluminium matrix which is 0.05 weight percentage at room temp and beyond that limit will give rise to form the intermetallic compound. As suggested by the reference paper [25] addition of La in Al-matrix will give rise to the development of Al-Zn-La phases i.e Al_2LaZn_2 . This particle segregates at the grain boundary which inhibits grain growth so fine grain and a uniform structure will be formed after the addition of lanthanum. Second phase particles $MgZn_2$ also form within the matrix. As discussed in earlier cases these phases are more negative to Al-matrix; this will provide the formation of galvanic cells within the matrix in which particles act as a cathode and matrix will act as an anode. Thus, the formation of these particles will improve the dissolution of the Al matrix.

4.8 Effect of Gallium and Bismuth

Ga has solid solubility in an aluminium matrix which is 8.82 atomic weight percentage around $30^\circ C$ and Bi have solid solubility in an aluminium matrix is 0.01 atomic weight percentage around $660^\circ C$ which is less than 1% and form a monotectic type of system with the aluminium and form the intermetallic phase with an aluminium [20].

As discussed in the reference paper [26] during solidification Al-Zn-Sn, aluminium crystallises first because it has a high melting point compared to Zn, Sn & Bi and these segregate at the grain boundary. Ga and Bi reduce the number of precipitates at grain boundary along with a decrease in width of diffuse boundaries which in turn improving the uniform dissolution of alloying element in an aluminium matrix, distribution of alloying element which forms heterogeneous phases that will improve dissolutions of the matrix. Addition of gallium and bismuth form microstructure with small grains.

4.9 Effect of Titanium and Strontium

Ti and Sr both have solid solubility less than 1% in Aluminium at peritectic Temp[20]. As proposed by reference Paper [27] Ti will act as a grain refiner and Sr can modify the Aluminium structure. Here, they have added Titanium varies from 0.03-0.1 wt% and Sr varies from 0.01-0.05 wt% in Al-5Zn-0.02In alloy.

Experimental studies confirm that Ti will affect the current capacity of anode increasing by 12% at 0.03wt% Ti and also anode efficiency will improve from 77% to 87%. Generally, indium in the Al-Zn system tends to form an interdendritic structure or grain boundary which is reduced by the addition of titanium and also form $TiAl_3$ particle and the greater amount of formation of small grain. These changes will lead to improved performance of the alloy. Strontium acts as modifiers and improves current capacity by 7% at 0.01 wt% and improves efficiency from 77% to 83%; this is achieved due to the formation of intermetallics at the grain boundary and interdendritic places due to high Sr content

4.10 Effect of Silver

Silver improves the corrosion resistance of Aluminium and will promote stability of passive layers. As discussed in the reference paper [28], adding Ag in ranges 1.5 & 2.5 will improve hardness and strength of the alloy but decrease the current capacity of alloy. The corrosion potential of developed alloy occurs simultaneously with the pitting potential and above that potential passive layer is stable. So, from that, we conclude that Silver will shift the potential of Aluminium in a noble direction.

4.11 Effect of Selenium

Information obtained by the reference paper [29], studied the effect of Se in Al - 5% Zn alloy and kept Sn & Bi 0.10 wt%. They added Se 0.01%, 0.05%, 0.10% and 0.50% respectively and got the best result at 0.5% Se. That is 90% Anode Efficiency this is achieved due to the formation of Se inclusion will give metallographical improvement and form uniformly grain. Efficiency is improved from 71.% to 90% as Selenium increases.[29]

4.12 Effect of Gallium and Zinc.

As discussed in section 4.1 and 4.8 Zn and Ga have formed peritectic and monotectic systems with Aluminium respectively. Reference paper [30] proposed improvement in anode efficiency. They studied the effect of Gn and Also the synergistic effect of Zn and Ga adding 0-1wt% and Zn 0-4 wt%. Here, both the elements decrease corrosion potential as Ga increases from 2.6wt% shift corrosion potential in a negative direction up to 0.5 volts and Zn will reduce 0.1-0.3 volt.

4.13 Effect of Tin and Magnesium

As discussed in section 4.2 and 4.4 Sn and Mg have formed a peritectic system. The reference paper [32] will give an idea about the effect of Sn and Mg in Al-5%Zn adding Sn vary from 0.1-1.0 wt% and Mg vary from 0.5-2.0 wt%. Both the elements will shift potential in the negative direction. This is because Sn distributes its particles locally on grain boundaries which affect the performance by increasing Sn up to 1.0% progressively shifted potential from -0.92 to -1.1 volt. Here, intergranular corrosion of alloy will give rise to the breakdown of a passive layer as a discontinued passive layer at the same time formation of the pit will be started. Mg will act as a grain refiner which reduces grain size from 150-200 micrometre to 70-100 m.

4.14 Effect of Copper

Copper is a major alloying element in 2XXX series Alloy. These are the heat treatable and natural aging alloy and have good strength. [32] Cu has 5.67% solubility at 550° C in Aluminium and form peritectic type of system with Aluminium.[14]

As mention in reference [16], Copper can affect the quality of oxide film, it will reduced density of oxide film and improve rate of amorphous alumina formation this will weaken the oxide film and leading to the breakdown of film and as we discussed section 3 pit formation is start at oxide free region. This will decrease corrosion resistance of developed alloy.

5. CONCLUSION

From the all above review we conclude that chemistry will play the most versatile role in corrosion morphology. The addition of the alloying element will change the chemical property of the sacrificial anode by forming another intermetallic, second phase particle and precipitates. The distribution of this new phase in the microstructure will alter the electrochemical and chemical properties of the sacrificial anode. All the alloying elements like Mg, Zn, Sn, Ga, Ti will improve electrochemical properties.

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