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Influence of Various Additional Elements in Al Based Filler Alloys for Automotive and Brazing Industry

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Abstract

Aluminium and its alloys are widely used in brazing various components in automotive industries due to their properties like lightweight, excellent ductility, malleability and formability, high oxidation and corrosion resistance, and high electrical and thermal conductivity. However, high machinability and strength of aluminium alloys are a serious concern during casting operations. The generation of porosity caused by dissolved gases and modifiers affects seriously the strength and quality of cast product. Brazing of Al and its alloys requires careful monitoring of temperature since theses alloys are brazed at around the melting temperature in most of the aluminium alloys. Therefore, the development of low temperature brazing filler alloys as well as superior strength Al alloys for various engineering applications is always in demand. In various heat exchangers and automotive applications, poor strength of Al alloys is due to the inherent porosities and casting defects. The unstable mechanical properties is therefore needed to be controlled by adding various additive elements in the aluminium and its alloys, by a change in the heat treatment procedure or by modifying the microstructure. In this regard, this article reports the effect of various elements added in aluminium alloys to improve microstructure, brazeability, machinability, castability as well as to stabilize the mechanical properties.

Key Words : Al alloy, Brazing, Microstructure, Strength, Automotive, Temperature.

1. Introduction

Aluminium is the most abundant metal, and third most common element comprising 8% of the earth's crust.[1)] Aluminium alloys have been widely used in automobiles and aerospace due to their high strength to weight ratio. A decrease in weight of these alloys is beneficial in various applications as it improves the fuel efficiency and carbon footprint, lowers power consumption and economical. Aluminium alloys received great attention in last few decades for joining and brazing various components of automotive such as heat exchanger, boiling and cooling devices, etc. because of their outstanding properties like corrosion and oxidation resistance, high strength, machinability and workability, improved damping capacity in various applications over the base alloys.[2)] The excellent combination of a wide range of different properties of aluminium alloys makes it most widely used metal after steel and cast iron.[3)] The combination of various properties depends mainly on the presence additional elements in the aluminium alloy matrix, cold working, and heat treatment procedures.[1,2)] There have been an enormous amount of investigations to improve the properties of Al alloys by various alloying additions, impurity additions, and/or modifiers, etc. The specific intermetallic compounds (IMC) particles arising from the additional elements are crucial in determining the mechanical properties and may impose damage due to poor distribution of IMCs.[4,5)] A poor distribution of IMCs inside the aluminium matrix may cause failure arising from the pitting and inter-granular type corrosion.[6)] The common alloying elements in aluminium are copper, silicon, magnesium, manganese, and zinc.[1)] Aluminium alloys have generally a shiny appearance in dry conditions due to the formation of aluminium oxide layer over its surface which also protects it from environmental corrosion. Al-Cu alloys

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Table 1 Designation of aluminium alloys and their applications[9)]

Alloy		Major alloying element	Applications
1XXX	→	No major alloying element	Electrical and chemical industries
2XXX	→	Cu	Aerospace applications
3XXX	→	Mn	Architectural applications
4XXX	→	Si	Welding and brazing, Heat exchangers
5XXX	→	Mg	Boat hulls, Marine industries
6XXX	→	Mg and Si	Architectural extrusions
7XXX	→	Zn	Aircraft components
8XXX	→	Other elements (Fe, Ni, Ti etc.)	
9XXX	→	Not assigned	

are prime choice as an aerospace material. Due to such reasons, these alloys were subject of several academic as well as industrial research in the past few decades.[1)] Therefore, the alloying elements should be selected on the basis of their effectiveness and suitability according to a particular application. Nowadays the Al-Si alloys found many industrial applications, particularly in brazing of automotive and heat exchanger parts.[7,8)] The alloying elements may be of various types, such as major and minor alloying elements, modifying agents, and impurity elements. It is also noteworthy point that the impurity elements in some alloys could be major elements in others.[1-3)] In addition, there may be certain microstructure modifying agents, flux, and grain refiners as well. In this paper the influences of different alloying elements, such as major such as (Cu, Si, Mg, Mg and Si), minor elements (Ni, Sn), modifiers (Ti and B, Sr, Na, Ca, Bi, P, S, Sb; Mn and Cr), impurity elements (Fe, Zn, Be), rare earth elements etc. on microstructures and brazing properties of Al alloys are reviewed.

2. Designation of Al alloys

The aluminium alloys have been designated according to the various additional elements system as shown in Table 1.[9)]

3. Major alloying elements

3.1 Copper

The formation of CuAl2 in age haredenable alloys like Al 2XXX alloy series is given as follows:

The supersaturated solution of Al if quenched in water or rapidly cooled and left for aging, it will pass through the formation of precipitates from GP Zones to Θ phase ($CuAl_2$). The strength increases in due course of hardening by the formation of GP zones. However, the formation of

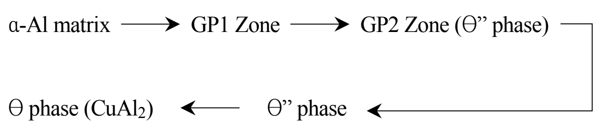

Fig. 1 Transformation sequence of GP zones in Al-Cu alloys[10)]

$CuAl_2$ also the corrosion resistance of the aluminium alloy and has been reported to increase the corrosion rate.[11)] The addition of copper in Al-Si-Mg alloy has been shown to improve the mechanical properties due to the precipitation of Cu-Al IMC. The best combination of mechanical properties is obtained with 1.5% Cu in Al-Si-Mg alloy after solidification.[12)] Zeren et. al. studied the effect of copper in Al-Si-Cu-Mg alloy and found an increase in tensile strength from 152 to 402 MPa with copper content from 1 to 6% due to the modification of eutectic silicon.[13)] The eutectic modification is beneficial for a brazed joint in various engineering applications. It has been reported that an excessive formation of the $CuAl_2$ IMC could lead to the formation of cracks and pores in the matrix that weaken the joints and accelerate the corrosion process.[6,11)] This can be harmful for various engineering applications in welding and joining with aluminium based brazing fillers.

3.2 Silicon

Al-Si alloys can be divided into three major categories: (1) Hypoeutectic containing less than 12 wt % Si, (2) Eutectic alloys containing 12-13 wt % Si, and (3) Hypereutectic alloys containing 14-25 wt % Si alloy. The strength of Al-Si alloy decreases with the formation of coarse polyhedral shaped particles.[14)] In brazing applications, Al–Si alloys with silicon in range, from 7-13% are useful in joining applications due to their excellent fluidity, higher strength and higher corrosion resistance to the joint.[15, 16)] These Al-Si brazing fillers have melting temperatures in the range of 575–610 ℃ according to the Al-Si phase diagram.[17)] The brazing temperature is very high relative to the melting point of other aluminum alloys having a solidus near 590 ℃. Thus, the development of low-melting-point filler metals is in great demand in the aluminum industry. Various low melting aluminium based fillers are developed in the past to reduce the brazing temperature in the past few years with the addition of different elements like Cu, Zn, Ni, etc. in Al-Si alloy.[18-20)] For example, an eutectic Al-6.5Si-42Zn and Al-6.5Si-42Zn-0.5Sr filler metals have been reported with a melting point around 520 ℃.[18)] The addition of zinc depresses the melting point of Al-Si alloy appreciably. However, zinc vaporizes very quickly and the process is not desirable for vacuum brazing operations. The vaporization of zinc may leave behind with cracks and weak joints after brazing. Niu et. al. designed a low melting point (solidus ~505 ℃) alloy of Al-Si-Ge-Zn with an improved brazeability and joint strength.[19] However, Ge is 400 times more expensive than Al, and is not economically feasible.

3.3 Magnesium

Magnesium is also an important in aluminium alloys which provides substantial strengthening and facilitates work-hardening characteristics in various operations and improves the brazeability. Magnesium alloys are considered as possible replacements for aluminum, or even steel, mainly due to their superior ductility, toughness, and better workability. Therefore Al-Mg alloy are generally used in plates, or extrusion type applications.[1] The precipitation sequence of Mg as an equilibrium β-phase (Mg_5Al_8) which increases the strength is given by Fig. 2.

It is also noteworthy point that for a durable aluminum heat exchangers, use of higher levels of magnesium in the core material is required in order to increase the strength of its components. However, there is a serious issue with the addition of Mg in Al alloys in especially vacuum brazing. Generally, alloys with higher magnesium content ~ 1 - 2 % or more are not easily brazeable due to increased oxidation layer is difficult to avoid by flux. The preferred concentration of Mg for brazing of the Al-Mg alloys (5XXX series) is up to 0.3 wt% with the standard brazing flux.[22] In the case of vacuum brazing, due to a vacuum of between 10^{-4} and 10^{-6} Torr, aluminium surface does not oxidize due to the high vacuum and reaction of Mg with traces of Mg inside the furnace to flush out trace oxygen additionally. As a consequence, brazeability will increase. This is known as getter effect in Mg containing brazing operations. The magnesium reacts with the traces of the oxygen or moisture according to the following reactions:[22]

$$Mg + H_2O \longrightarrow MgO + H_2 \quad (1)$$

$$2Mg + O_2 \longrightarrow 2MgO \quad (2)$$

$$Mg + CO_2 \longrightarrow MgO + CO \quad (3)$$

3.4 Mg and Si

The Al-Mg-Si (6XXX series) is tremendously used in the automotive industry due to their ability to absorb a greater amount of energy during collisions. The Al-Mg-Si alloys have a relatively higher tensile strength in the range of 220-390 MPa, with tensile strain remain in between 17-12% decreases with an increase in strength.[23] The ratio of the Mg and Si is crucial in Al 6XXX series and is responsible for the mechanical properties of the alloy. The well optimum behavior of alloys can be obtained at a Mg/Si ratio close to 1.74. The alloy with a higher concentration of Si that this ratio is generally known as excess Si alloy.[24] However, the ratio of 1.74 is difficult to achieve under ordinary operating conditions, therefore most of these alloys have excess in magnesium or silicon.[24,25] An excess in Mg amount produces superior corrosion resistance, however a lowering in strength and formability is detected, which may not be desired in various joint applications.[26-27] Excess in silicon provides higher strength without any appreciable loss of formability and brazeability, though it induces a tendency towards corrosion failure. Therefore, for brazing joint operations the Mg content should be wisely decided according to joint properties before practical application.

α-Al matrix ⟶ GP Zones ⟶ β' phase ⟶ β-phase (Mg_5Al_8)

Fig. 2 Precipitation sequence in Al-Mg alloy[21]

4. Minor alloying elements

4.1 Nickel

Presence of Ni in aluminium alloys generally provides additional strengthening by forming various nickel aluminide precipitates. The addition of Ni is kept minimum, however, to compromise with the cost and performance.[28] The nickel alloying with Al 2XXX alloy series mainly results in the formation of Cu-Ni aluminides which are atable at high temperatures and thus provide additional strengthening mechanism. However, the addition of Ni is ineffective at higher Cu concentrations. The amount of intermetallic Al_3Ni also increases with the nickel content and increases the hardness, compression and bending resistances, however it reduces the ductility.[28, 29]

4.2 Tin

Tin (Sn) is considered a soft metal used in aluminum castings for antifriction properties in bearing alloys. Presence of Sn in aluminium provides different lubricating properties to contact surfaces and increases the elongation as a result of softening.[30] This property is sometimes exploited in brazing filler development. It increases the recovery rate of Si in purification by Si-Al alloys.[31] In brazing applications, tin improves the additional brazeability by increasing the fluidity of the braze alloy.[32] However, a high content of Sn in the Al-Si alloy may weaken the brazed joints and increase corrosion. Sn may also act as Fe modifier producing a large rosette shaped Fe containing that act as nucleation sites for crack initiation. However, trace additions of Sn (e.g. 0.05%) are known to stimulate age hardening in the Al-2XXX series alloy which may be partly beneficial for increasing the brazed joint strength.[30]

5. Microstructure modifying agents

5.1 Titanium and Boron

Titanium is used as a grain refiner in common Al-Si alloys in minute concentrations, i.e., 0.12-0.15%.[33] It has been also reported that the refining action of Ti is also accelerated if it is coupled with Boron. Commonly, the mother alloys of Al-wt5% Ti-1wt% B are manufactured to serve as a regular grain refining objective in these Al 4XXX alloys. Titanium interacts with boron to form borides and additional aluminides like TiB2 and TiAl3 for better strengthening.[34] The grain refiners in brazing fillers play an important role as they improve the quality of brazed joint and produce minimum cracks and pores which may arise due to the non-uniform microstructures in Al-Si alloys.

5.2 Strontium

Strontium is an important modifier in almost all the aluminium based castings, especially in eutectic or hypoeutectic Al-Si alloys (Al 4XXX series). It serves as a modifying agent for silicon morphology from needle, blocky or irregular shape to round, or spheroidal one. There are various reports on the modification action of Sr in Al-Si alloys.[35-37] Wang et al also [ref] reported the optimum amount of Sr in the range 0.01% and 0.015%, while at a higher Sr level, i.e., 0.015%-0.02%, coarsening of Si particles is observed.[38] In the alloys with elements Fe and Cu, (Al2XXX, Al8XXX), the addition of strontium has been also shown to refine the Fe and Cu bearing phases in the castings.[39] However, at higher Sr levels, the coarsening of silicon particles is not understood well in literature. The recent model known as twin-plane reentrant edge (TPRE) model proposed by Major et al. to explain the coarsening at higher Sr contents in Al-Si alloys. They proposed the growth of flake-like silicon particles in an unmodified Al-Si alloy by repeated nucleation of fresh layers on the re-entrant edges and grooves created by the intersection of {1 1 1} twins at the solidification interface. This TPRE growth gets suppressed at higher concentration of Sr concentration.[38-39] Therefore, the strontium levels in the filler alloys for brazing application should be kept in the optimum range to achieve a proper Si modification and microstructural refinement.

5.3 Sodium, Calcium, Bismuth, Phosphorus, Sulphur and Antimony

The other group of the modifiers for eutectic silicon sodium and calcium used mostly in the form of flux or salts. To achieve this, a conducting electrolyte of sodium, calcium or combination of both, and halogens like F or Cl may be used in conjugation. Sarda et al found that the Na modified alloy has a more regular silicon morphology compared to Sr which also improved the mechanical properties.[37] It is also reported that the addition of Ca with Sr accelerates the modifying effect of Sr (where the amount of Ca lies in range, from 0 and 130 ppm and Sr from 0 and 312 ppm).[35] Addition of phosphorus is also used extensively to hypoeutectic Al-Si alloys in minute concentration from 0.001 to 0.02 depending upon various reports to improve the mechanical properties.[40] However, Al-P particles act as Si nucleation sites, reducing the modifying effect of Sr or Na. Higher levels of phosphorus can cause deleterious properties and can be detrimental to fluidity.[40] Other elements such as, bismuth and antimony, i,e Bi (<0.5wt %) and Sb (< 0.05wt %) to Al-Si alloys have also been shown to improve the strength as well as corrosion performance of the aluminium alloys.[41] It has been shown that embrittlement in Al-Mg-Si alloy (Al 6XXX series) can be significantly controlled by addition of Bi in 0.5 wt% concentration, and around 1wt% in Al-Si alloys.[41,42] Khan and his co-workers have reported that antimony behaves as a refiner rather than a modifier in the solidifying eutectic. He found that with 0.2 wt% Sb in Al-Si alloy, the interflake spacing of the eutectic Si gets reduced rather than flake refinement.[43-44] Recently, Onyia and his co-workers have studied the sulfur addition in cast Al-Si alloy. They found that sulfur also affects the modification of the Si particles in Al-Si alloys with sulfur, similar to the sodium or strontium modified silicon. The sulfur concentration increase decreases the sharpness of Si particles. However, sodium or strontium modified alloy produces more refined eutectic morphology and enhanced mechanical properties.[45-46]

5.4 Manganese and Chromium

It has been reported that Mn when used alone or in combination with chromium (Cr) in Al-Si alloy may change the morphology of the iron-rich Al_5FeSi phase with appearance of Chinese script/acicular to a more cubic $Al_{15}(MnFe)_3Si_2$, (and $Al_{13}(Fe,Cr)_4Si_4$ with Cr) and improve the ductility. The β-AlFe compounds are formed when Mn/Fe ratio is less than 0.5. Corrosion resistance is also improved by the addition of manganese and chromium.[47-50]

6. Effect of impurity elements

6.1 Zinc

Zinc is the only component of the Al 7XXX series alloys, which otherwise is sometimes an acceptable impurity in various casting alloys. Zhu and his co-workers studied the effect of the Zn concentration on tensile and electrochemical properties of Al 3003 alloy. They observed that an addition of about 1.5 wt% Zn enhances the UTS of the alloy.[51] Additions of Zn to the Al-Mg alloys (Al 5XXX series) in levels of 1− 2wt% have been shown beneficial to improve stress corrosion cracking resistance due to the formation of a stable ternary Al-Mg-Zn, the pie- phase.[52] In brazing applications of Al-Si (Al 4XXX series), addition of Zn reduces the melting point of the alloy appreciably to produce a low melting point filler alloy for brazing aluminium components. For example, an eutectic Al-6.5Si-42Zn and Al-6.5Si-42Zn-0.5Sr filler metals have been proposed to braze the aluminium joints with a melting point around 520 ℃, compared to the eutectic melting point of 577 ℃ of Al-Si alloy.[18] The addition of zinc also decreases the corrosion resistance of various aluminium alloys. However, Zn is prohibited very often due to its higher vapor pressure, which vaporizes early in due course of brazing operations, and may result in the formation of porosity and cracks in the joints which is not always desirable in vacuum operations.

6.2 Iron

Iron is the common element impurity in aluminium alloys and may be difficult to remove and affects the ductility and castability, especially in Al-Si (Al 4XXX series) based casting alloys. An increase in the amount of iron in Al-Si alloys may form the β-FeSi5Al IMC, which causes poor mechanical properties of the Al-Si alloys. It decreases the alloy ductility due to its weak bonding to the matrix and brittle thin platelets type form, and also results in excessive shrinkage casting defects.[39-40, 53] It is also noteworthy that, presence of Fe improves the high temperature strength and hot tearing resistance of aluminium alloys. Fe at low concentrations may act as a promoter for grain refinement, which can be a beneficial effect in brazing operations in Al–Si alloys at low concentrations although the effect diminishes shortly.[54] However, considering the damaging effects of iron, its reduction or control in Al–Si alloys are always suggested.

6.3 Beryllium

Wang and his co-workers examined the influence of Be in Al–7Si–0.4Mg–0.2Ti–xFe–xBe cast alloy and found that Be modifies the iron-rich compounds from needle or plate shapes to Chinese scripts and modifies the mechanical properties due to the Fe phase shape.[55-56] Murali et al and his co-workers checked the influence of iron and beryllium additions in squeeze-cast Al-7Si-0.3Mg alloy, and reported that trace additions of Be may completely neutralize the detrimental effect of iron, and improve the fracture toughness and strength of the alloy.[56]

6.4 Rare earth elements

Rare earth elements are known as vitamins for the metals and alloys in controlling the microstructural properties. Previous studies show that they are potential candidates for the improvement of the mechanical properties of aluminum alloys. For example, the addition of Yttrium and scandium has been shown to increase the recrystallization temperature of the aluminium alloy.[57-58] The high strength of aluminum alloy is related to the presence and size of Al_2Y phase. Scandium is an effective grain refiner component preventing recrystallization owing to the presence of Al_3Sc-phase particles in an aluminum matrix.[59] The addition of Ce to Al-Cu-Mg-Ag has been shown to enhance the thermal stability, though it raised the service temperature. The addition of La (0.05-0.1 wt%) in the Al-Si-Cu-Mg alloy reduced the mutual poisoning effect of the B and Sr by forming the LaB6 rather than SrB6.[60-62] It has been also demonstrated that addition of Nd also increases the strength of the Al-Cu alloy due to the uniform distribution of AlCuNd at high temperature.[61] In few recent reports, Ytterbium (Yb) has been added in aluminium alloys and has been shown to improve the mechanical properties of the Al-Cu-Mg-Ag alloy and Al-Zn-Mg-Cu-Zr alloy.[62-63]

7. Conclusion

Alloying elements in the aluminium play a vital role in deciding the final microstructure. A modification of microstructure drastically modifies the strength, ductility, castability, machinability, brazing properties as well as the energy efficiency in various engineering applications. The addition of Si increases the fluidity and thus improves the castability. However, the formation of big Si needles in hypereutectic alloys is not desired which weakens the strength of the alloy. Copper addition also improves the strength drastically by generating $CuAl_2$ intermetallic compounds as well as the formation of GP zones after heat treatment. Huge generation of $CuAl_2$ is also a problem in brazing and structural applications. Magnesium in aluminium alloys provides substantial strengthening and improvement of work hardening characteristics of aluminium alloy. It can impart good corrosion resistance and weldability or extremely high strength. However, a higher magnesium content (> 1 - 2 %) may impose a serious problem in brazing aluminium alloys due to increased oxide layer formation which cannot effectively be removed by fluxes. Titanium as a minor alloying element increase the strength if added in optimum quantities. Nickel enhances the elevated temperature strength and hardness while Sn reduces the friction characteristics of the joint. Addition of modifiers such as Ca, Na, Sr, P, S, and Sb also improves the morphology of the silicon and distributes the $CuAl_2$ IMC in the alloy. Addition of Mn and Cr improves the corrosion resistance and improves the morphology of iron complexes in the presence of Fe in the alloy. The presence of Be neutralizes the damaging effects of impurities like Fe, and also improves the strength. Addition of Zn depresses the melting point of the alloy, however in particular applications such as heat exchangers it is not desirable due to its highly vaporizing nature, and after effects in the form of cracks in the joints during casting and brazing operations. Rare earth metals have also been shown to improve the microstructure and eutectic modification to a great extent, if added in their optimum amounts

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