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Vacuum Brazing Tests on Powder Aluminum Brazing Filler Metals†

Tadashi TAKEMOTO*, Masami MIZUTANI**, Tatsuyuki UJIE***, Akiei TANAKA*** and Akira MATSUNAWA****

Abstract

Vacuum brazing tests on powder aluminum brazing filler metals were conducted using various atomized Al-Si-Mg powders to investigate their applicability for the vacuum process. The effect of atomizing condition, powder size and filler metal composition has been studied to obtain an optimum powder for the vacuum process. After brazing a part of each filler metal used made no contribution to the fillet formation and coalesced on the base metal. This indicated non-wetting tendencies among filler metal powders. Brazability was evaluated by the percentage of the filler metal remaining as fillet by measuring the weight change before and after brazing. After brazing, voids were found in the formed fillet. Although, completely sound fillets were not obtained, fine powders and air atomized powders gave better brazability than coarse powders and those atomized in argon gas atmosphere. The difference in vaporization characteristics of magnesium in the filler metals seemed to be responsible for void formation and poor brazability of the powder filler metals. Mixing of the filler metals with Al-Mg powder was found to be effective for the improvement of the fillet formation, however, completely sound fillets were not obtained. It is believed that both the control of magnesium vaporization rate and improvement in the quality of the vacuum are important to obtain sound fillets.

KEY WORDS: (Powder Brazing Filler Metal) (Vacuum Brazing) (Atomized Powder) (Aluminum Alloy) (Magnesium)

1. Introduction

In aluminum brazing, the use of a brazing sheet with clad filler metal on core material is popular[1)-3)], because it gives excellent results in furnace processes. Accordingly, the use of wire, rod and preformed filler metal is a minor process However, the use of preformed or powder filler metal is sometimes convenient in certain design circumstances[4)]. In the manufacture of large scale heat exchangers, loss of silicon by diffusion into the core material[5)] under the prolonged preheating conditions reduces the amount of molten filler metal available for fillet formation and leads to reduced strength of the brazements. To prevent silicon loss, the use of pre-set type filler metals is effective, however, these filler metals usually give poor brazability because of the stable surface oxide films on aluminum. Therefore, the use of powder filler metals with large specific surface areas could introduce difficulties for sound fillet formation. The brazability of powder aluminum filler metal, especially in vacuum condition, has not been systematically investigated. A variety of powder alloys could be produced by the atomizing method[6)7)], therefore, vacuum brazing tests on aluminum powder brazing filler metals were carried out to investigate the brazability of powder filler metals, the effect of atomizing conditions, filler metal compositions, powder sizes and shape.

2. Materials and Experimental

Many test methods and evaluation factors have been proposed for investigating the brazability of aluminum[8,9)]. In the present study, the amount of filler metal that contributed to the formation of a fillet was measured, i.e., the percentage of the weight of filler metal remaining as a fillet. **Figure 1** shows the experimental methods for the brazing tests. Stainless steel pipe with

† Received on Nobember 24, 1995
* Associate Professor
** Technical Assistant
*** Toyo Aluminum K.K.(Gamou-gun, Shiga, Japan)
**** Professor

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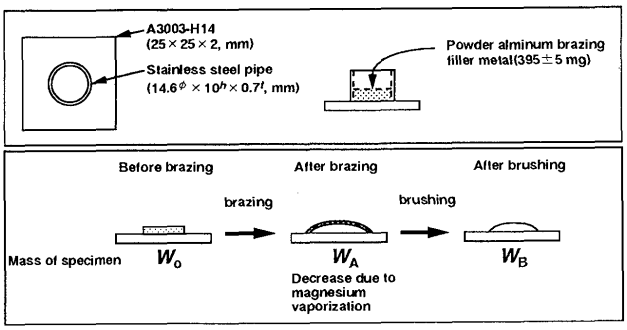

Fig. 1 Shape and size of specimen for brazing test (a) and measurement of fillet formability and vaporized magnesium by weighing (b).

an inner diameter of 14.6 mm, thickness of 0.7 mm, and 10 mm in height, was located on A3003 base metal of 25×25×2 mm and wound together with a fine stainless steel wire. A certain amount of powder filler metal, 395±5 mg, was put inside the stainless steel pipe. Organic binders and solvents were also put on the powder filler metal to prevent scattering during the evacuation process. It was confirmed the organics were decomposed during brazing heat cycle and had no detrimental effect on brazability. Brazability was evaluated by the amount of residual filler metal. Some of the filler metal was easily removed from the base metal by brushing with a stainless steel wire brush, indicating that the powder did not melt and coalesce with other powders. The percentage or residual filler metal is defined as fillet formability, F, by the following equation. The fillet formability in the present work is similar to coalescence efficiency.

$$F = 100\ (W_0\text{-}W_B)/W_0$$

F: Fillet formability

W_B: Weight of specimen after removal of filler metal by brushing

The magnesium vaporization rate, V_{Mg}, was also measured to study the role of magnesium in vacuum brazing process using powder filler metals. The magnesium vaporization rate is defined by the following equation and expressed as V_{Mg}.

$$V_{Mg} = 100\ (W_0\text{-}W_A)/(\text{Mg content in } W_0)$$

V_{Mg}: Vaporized magnesium (Magnesium vaporization rate), %

W_0: Mass of specimen before brazing

W_A: Mass of specimen after brazing

The powder filler metals used were produced by an atomizing method in three different atomizing atmospheres. The atomizing gas and atmosphere used were argon and air respectively. The composition and size of powder filler metal used is listed in **Table 1**, where the atomizing gas and atmosphere are also indicated. The powders were sieved according to the sizes

Table 1 Chemical composition, atomizing conditions, mesh size and nominal diameter of powder aluminum brazing filler metals.

Powder number	Nominal composition	Element Si (mass%)	Element Mg (mass%)	Atomizing condition: Atomizing gas	Atomizing condition: Atomizing atmosphere	Powder mesh size	Nominal powder diameter (μm)
No. 1	Al-10Si-1.5Mg	9.58	1.45	Air	Air	−100/+325	45～149
No. 2						−325	<45
No. 3		11.34	1.53	Ar	Air	−100/+325	45～149
No. 4						−250	<63
No. 5						−325	<45
No. 6		9.91	1.43	Ar	Ar	−100/+250	63～149
No. 7	Al-10Si	10.03	0.00	Air	Air	−325	<45
No. 8	Al-10Si-2.5Mg	9.99	2.82			−325	<45
No. 9	Al-10Si-4.5Mg	10.91	4.42			−100/+325	45～149
No. 10						−325	<45
No. 11	Al-10Si-9Mg	10.23	9.03			−100/+325	45～149
No. 12						−325	<45
No. 13	Al-20Si-1.5Mg	19.72	1.63	Air	Air	−325	<45
No. 14	Al-30Si-1.5Mg	28.87	1.46			−325	<45

shown. The effect of atomizing condition, filler metal composition and size were investigated. Brazing tests were conducted mainly in vacuum at a level of 2.67×10^{-3} Pa, and the tests in high purity nitrogen gas atmospheres were also conducted for comparison. The standard brazing temperature and time were 873K and 0.3ks respectively.

Magnesium distribution after brazing was measured by EPMA (Electron Probe Micro Analyzer) analysis.

3. Experimental Results and Discussion

3.1 Powder atomizing atmosphere

Figure 2 shows the fillet formability of Al-10Si-1.5Mg powders. A perfect formability of 100% could not be obtained with all powders, the maximum value in the vacuum process was about 75% for No.2 powder, and the minimum was 10% for No. 6 powder. The longer heating time at 873K and also the tests at higher temperature of 883K had no influence on fillet formability. An improvement in brazability could not be achieved by the high temperature long time brazing. Brazing in nitrogen gas atmosphere gave similar results to the vacuum process except that the brazability was relatively improved with No. 4 powder. From Fig. 2, the descending order of fillet formability is as follows.

No. 2 > No. 5 ≒ No. 4 > No. 3 ≒ No. 1 > No. 6

As regards powder size and atomizing process, the fine powders showed better results than the coarse ones,

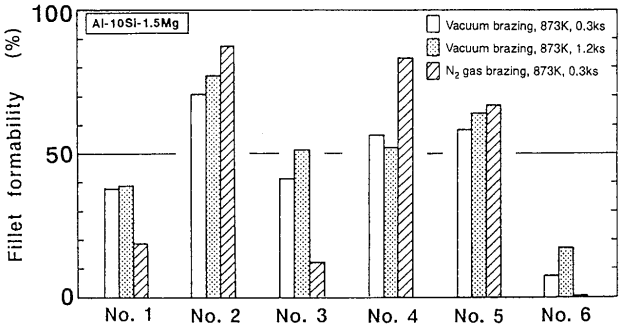

Fig. 2 Fillet formability of various powder filler metals after brazing in vacuum and nitrogen gas atmosphere at 873K.

especially the powders of -325 mesh (under 45μm) and -250 mesh (under 65μm) without removal of the fine particles. The use of an air atomizing atmosphere and also as atomizing gas is also preferable to obtain better fillet efficiency. The worst result was found in powder atomized in an argon atmosphere by argon atomizing gas.

Figure 3 shows the SEM (Scanning Electron Micrograph) of Al-10Si-1.5Mg powders produced in various atomizing conditions. Air atomized powders (Air → Air) show irregular shapes, whereas the argon gas atomized powders (Ar → Ar) have spherical shapes. The powders atomized by argon gas in air atmosphere (Ar → Air) show irregular shape but partially contain spheres,

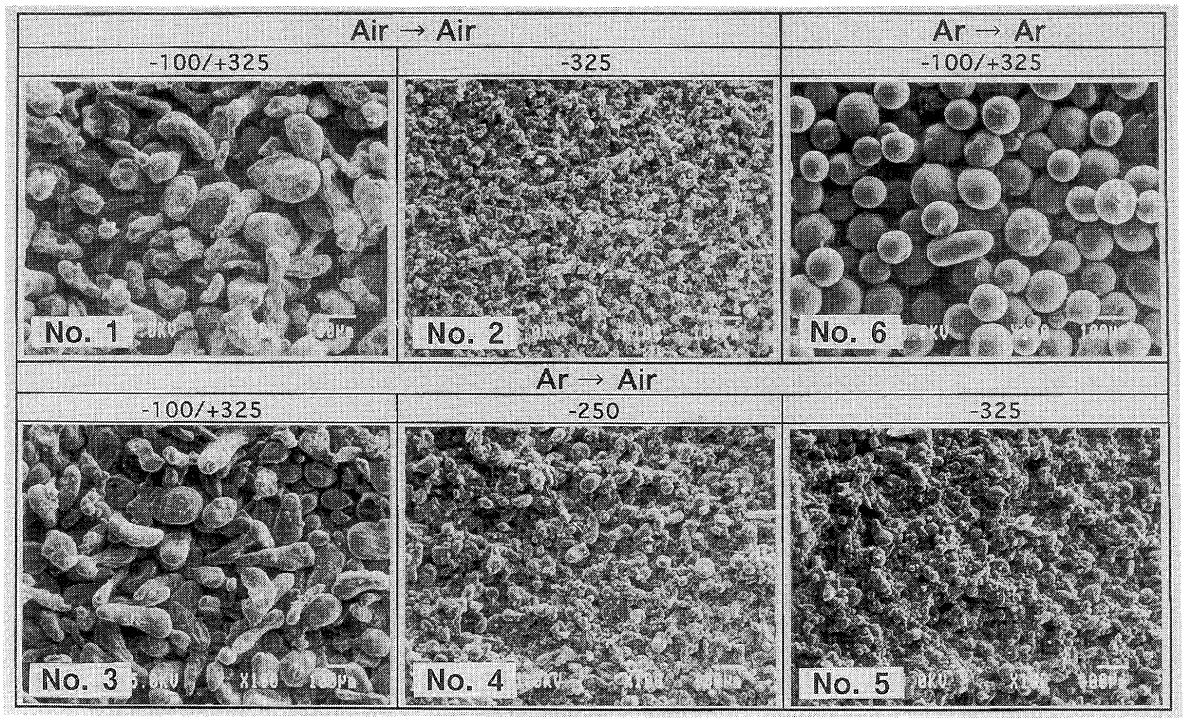

Fig. 3 Scanning electron micrographs of aluminum powder brazing filler metals.

and the percent of spheres becomes higher in finer powders. The oxygen content of No. 4 powder atomized into air with -250 mesh was 0.25 mass%, and in No. 6 powder produced in argon gas atmosphere it was 0.1 mass%. The oxide film thickness for each powder calculated from the oxygen content and specific surface area was almost the same, 5 and 4.7 nm respectively.

The reason that the fine and irregular shape powders showed better results in brazing than coarse and spherical powders could be explained as follows. The break down of surface oxide is the key to gather the molten filler metal particle and this coalescence leads the excellent brazability. Fine particles are more closely stacked with a higher packing density than coarse particles. The contact probability, the number of contact points and/or area ratio, for fine particles is larger than for coarse ones. Consequently, fine powders can more easily coalesce after break down of surface oxide under molten condition.

The molten liquid attempt to become spherical to minimize the surface area due to its surface tension, therefore, in irregular shape powders the surface oxide film will be easily broken after melting, because the powders behave to be spherical, whereas the film on spherical powder still remains unbroken, **Fig. 4**. This difference in break down probability of surface oxide film between irregular and spherical shape powders leads to different brazability, higher coalescence probability in irregular powders and gives better brazability.

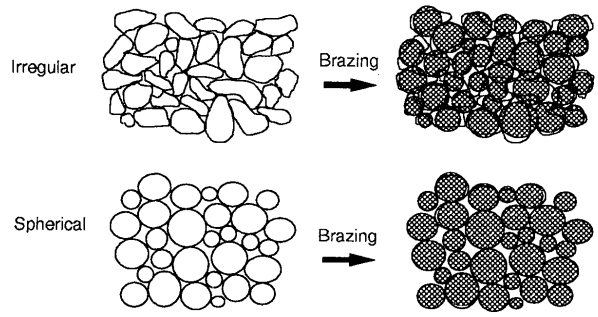

Fig. 4 Self break down of surface oxide film during melting, each particle is apt to be a sphere due to surface tension.

3.2 Magnesium content

The above results revealed that the air atomized irregular powder was adequate for the vacuum process under similar oxide thicknesses. Accordingly, the effect of powder composition on brazability was investigated using air atomized powders. **Figure 5** indicates the effect of the magnesium content in Al-10Si-Mg powder filler metal on fillet formability. Fillet formability decreased with the increase in magnesium content, and the powders with finer powders (-325 mesh) always had higher fillet formability than powders excluding fine powders less than 45μm (-100/+325) irrespective of magnesium content. At 9% magnesium, a fillet could hardly be formed.

Vaporized magnesium also decreased with increasing magnesium content in the powder filler metal, showing the same tendency to fillet formability. The results on powders of -325 mesh are indicated in **Figure 6**. The maximum vaporization rate was about 70% at 1.5%Mg powder, the rate gradually decreased to 20% at 9%Mg. The total amount of vaporized magnesium increased with magnesium content even the decrease in vaporized percentage.

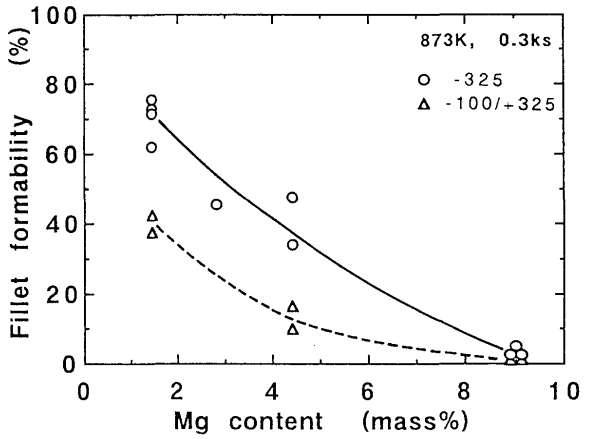

Fig. 5 Effect of magnesium content on fillet formability of Al-10Si-Mg powder filler metals after heating at 873K for 0.3ks.

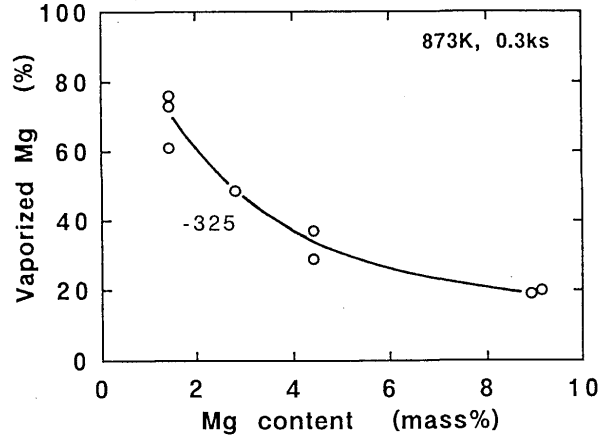

Fig. 6 Percentage of vaporized magnesium from Al-10Si-Mg powder filler metals after heating at 873K for 0.3ks.

3.3 Silicon content

The effect of silicon content was investigated on air atomized fine powders of -325 mesh, **Fig. 7**. The fillet

formability was almost the same up to 20%Mg, the values are around 70%, but further increases in silicon content decreased the fillet formability.

Figure 8 shows the relation between vaporized magnesium and silicon content. The vaporization rate of magnesium is known to be enhanced by the appearance of liquid phase[10)11)], therefore, the decrease in magnesium vaporization with silicon content must be related to the decrease in the liquid phase at brazing temperatures. The liquid phase percent at 873K calculated from Al-Si binary phase diagram[12)] is also indicated in Fig. 7 by a dotted line. Filler metals with less than 14%Si completely melt at the brazing temperature of 873K, in alloys exceeding 14%Si the liquid phase gradually decreases with increasing silicon content according to the increase in the liquidus. This trend corresponds to the decrease in vaporized magnesium. The main reason for the decrease in vaporized magnesium with increasing silicon content could be attributable to the increase in liquidus temperature of filler metals which lowered the amount of molten phase of filler metal.

Figure 9 shows the plots of fillet formability vs. vaporized magnesium on powders under -325 mesh (less than 45μm). The formability increased with an increase in the vaporized magnesium, and it seems to reach 100% by extrapolation, therefore, it seems to be essential to obtain a 100% vaporization rate for the improvement of the vacuum brazability of powder filler metal.

3.4 Powder mixing

Figure 10 shows the effect on fillet formability of the addition of Al-Mg powder to No. 2(Al-10Si-1.5Mg) or No. 7(Al-10Si) powders. Magnesium content on horizontal axis is the total content in the mixed powder. Mixing was found to be effective in improving the formability in both powders, but the improvement could not be obtained with magnesium contents more than 10%. The use of No. 7 powder without magnesium gave

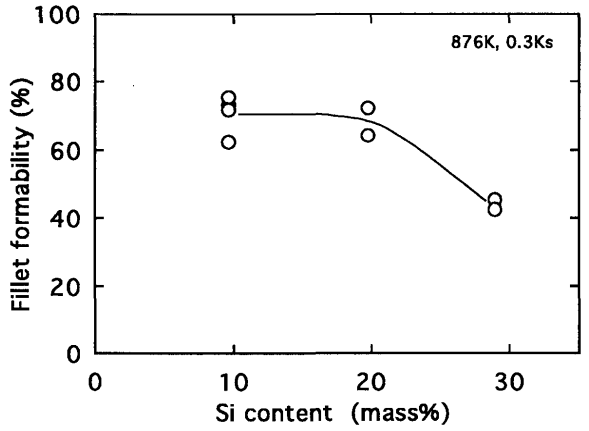

Fig. 7 Effect of silicon content on fillet formability of Al-Si-1.5Mg powder filler metal after heating at 873K for 0.3ks.

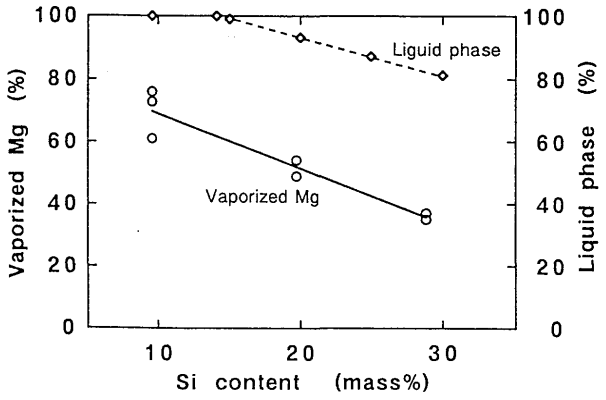

Fig. 8 Effect of silicon content in Al-Si-1.5Mg powder filler metal on percentage of vaporized magnesium and liquid phase at 873K.

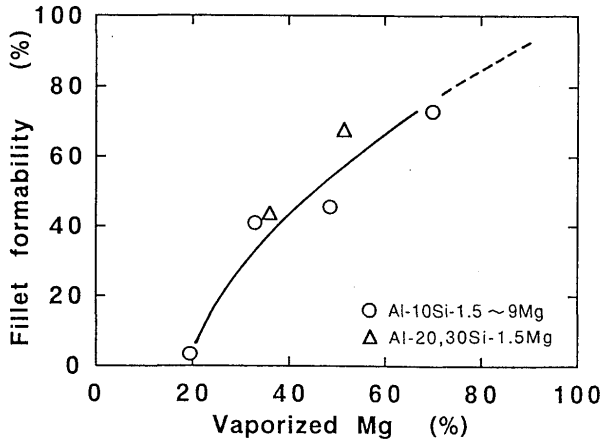

Fig. 9 Plots between fillet formability and vaporized magnesium obtained from Al-Si-Mg filler metals.

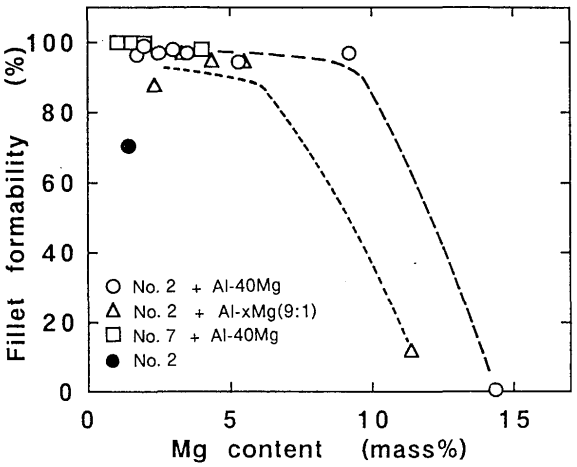

Fig. 10 Effect of magnesium content on fillet formability of mixed powder filler metals

the best result, both fillet formability and appearance, even at low magnesium contents (mark □), **Fig. 11**. The addition of Al-40Mg (mark ○) showed better results than Al-xMg(x=10, 20, 30) powders (mark △).

Although 100% fillet formability was obtained by mixing Al-Mg powders, sound fillets were not obtained, because defects similar to voids were found. **Figure 12** shows the cross section of a specimen using No. 7 powder + Al-40Mg with a mixing ratio of 9:1. The smooth and bright surface was obtained after brazing, however, many voids and non-gathered powders were included at the center of the specimen.

Figure 13 is the relation between fillet formability and vaporized magnesium. The figure shows fillet formability suddenly dropping when the vaporized magnesium becomes lower than a certain value, about 75% for No. 2 + Al-xMg mixed powder, about 65% for No. 7 + Al-40Mg powder and No.2 + Al-40Mg powder. In mixed powder filler metals, vaporized magnesium did not reach 100%, even with magnesium contents less than 9% where fillet formability of 100% was achieved. The complete magnesium vaporization is not necessary to achieve 100% fillet formability, because 100% fillet formability was obtained under the condition of only 70% magnesium vaporization. It is clear that a vaporized magnesium percent more than 65% is necessary to achieve 100% fillet formability. To obtain this high vaporized magnesium percent, the magnesium content should be restricted to less than 10% from Figs. 11 and 12.

3.5 SEM observation

Figure 14 shows the SEM of a cross section of the fillet on No. 7 + Al-40Mg powder. Many voids can be seen, even in the case of 100% fillet formability. The surface of the voids consisted of magnesium and oxygen, suggesting the formation of MgO, by the getter action of magnesium[13)14)]. A number of powder filler metal layers were stacked on the base metal, therefore, vaporized magnesium from the bottom of the stacked layer was trapped by the powders on upper layers, and became oxidized. MgO is a stable oxide, more stable than Al_2O_3 below 1500K[15)], and the formation of MgO remarkably reduces the brazability[16)]. The creation of fresh surface by active vaporization of magnesium is a key to obtaining excellent brazability. By the hot observation in scanning electron microscope, it is found that the exudation of molten filler metal through the broken oxide film is important to secure excellent brazability[17)].

Figure 15 shows the magnesium content in filler metal after heating of No. 2 powder and sheet filler of thickness 1 mm at 873K for 0s or 0.3ks . Magnesium

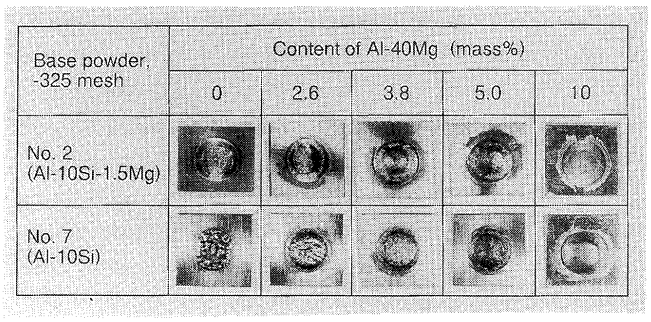

Fig. 11 Effect of powder mixing of Al-40Mg powder to Al-10Si-(Mg) powder filler metals.

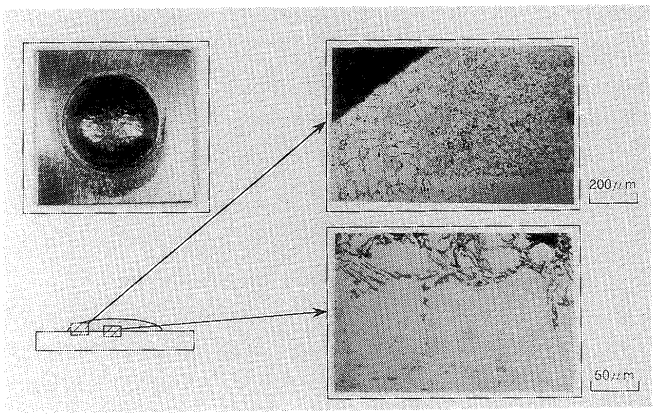

Fig. 12 Cross section of specimen brazed with No. 10 + Al-40Mg mixed powder filler metals.

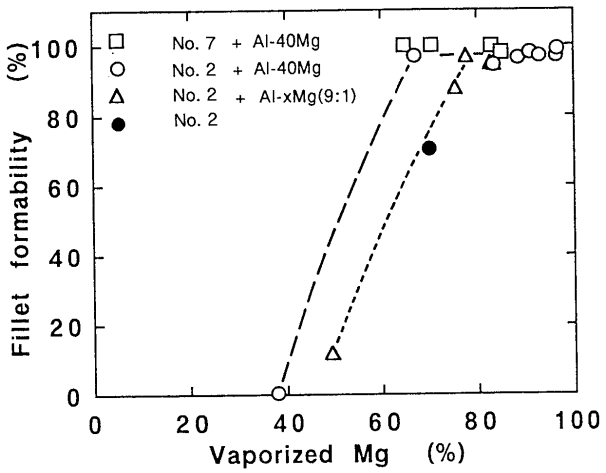

Fig. 13 Plots of fillet formability and vaporized magnesium obtained from mixed powder filler metals.

content is plotted against the analyzed position. In Fig. 14, "Surface" and "Middle" corresponds the depths of 20 and 400μm from the filler metal surface, whereas "Bottom" corresponds the position from 80μm apart from the filler/base metal interface. In all positions, magnesium contents decreased with longer holding time,

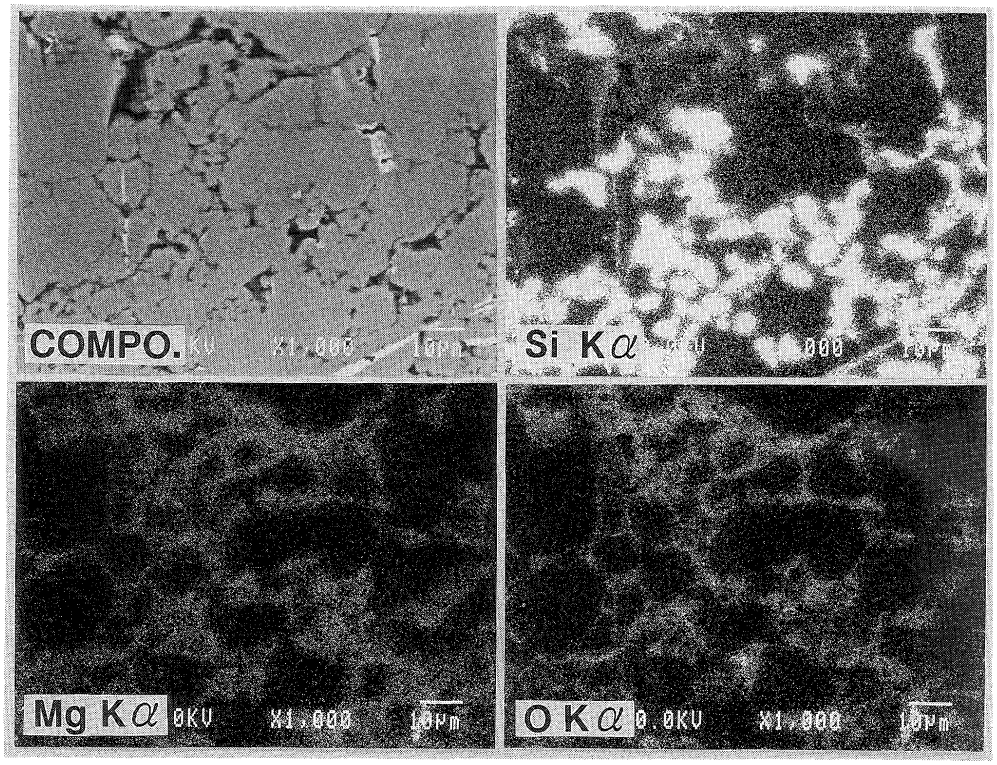

Fig. 14 Scanning electron micrographs of mixed powder filler metal heated at 873K for 0.3ks, mixture of No. 42(Al-10Si-1.5Mg) and Al-40Mg, 9:1

however, the behavior of magnesium is different. At "Surface", sheet filler contains larger amounts of magnesium than the powder just after reaching at 873K, whereas almost all of the magnesium had vaporized in powder filler metal. On the contrary, magnesium content is higher in powder filler metals than in sheet at "Middle" and "Bottom", the magnesium distribution in sheet filler is almost flat in both heating time. In powder filler metals, the existence of non-coalesced particles hinders the diffusion of magnesium, vaporized magnesium from a particle oxidizes and remains at the surface of each particle as indicated in Fig. 13, therefore, the supply of magnesium to the surface from the inside is delayed. On the other hand, in sheet filler, all the filler forms a continuous liquid, therefore, the supply of magnesium from inside is quite easy. Accordingly, a homogeneous magnesium distribution and a high magnesium vaporization rate is achieved.

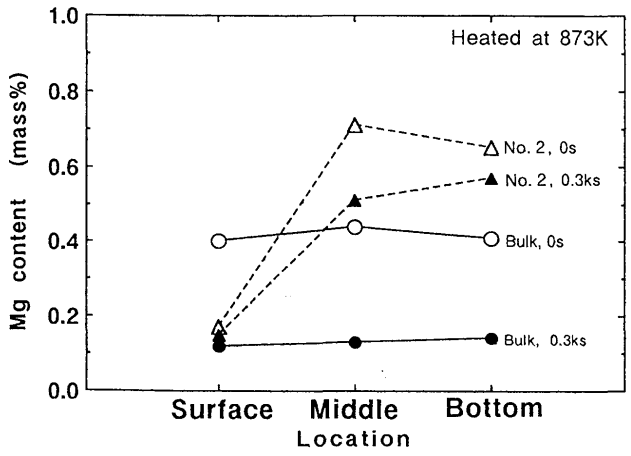

Fig. 15 Magnesium content of powder filler metal at different positions on base metal after heating at 873K.

4. Future Work

To obtain a perfect fillet by vacuum brazing with powder filler metal, the evaporation of vaporized magnesium without retention on powder particles is important. The present work proposes the following approaches to improve the brazability of aluminum powder filler metals.

1) Powder supply

Thin layers of filler metal to prevent retention of vaporized magnesium among particles.

2) Powder production method or process control

The control of magnesium vaporization from the surface of powder by controlling oxide thickness and characteristics. The establishment of a method to obtain the most appropriate values of magnesium content, surface oxide thickness, powder size, heating rate, and

vacuum level to secure the necessary amount of magnesium at the melting stage of filler metal.

3) Vacuum brazing condition

The prevention of oxidation of vaporized magnesium by reducing the moisture and oxygen levels in vacuum. The setting of getter magnesium or the use of ultra-high vacuum.

Coating of powder might be effective to control above mentioned factors.

5. Conclusion

Vacuum brazing tests on powder aluminum filler metals was conducted by measuring fillet formability, the residual percent of applied filler metal as deposit after braze heat cycle. The test revealed the difficulties of brazing with aluminum powder, however, guide lines to improve brazability were derived.

(1) The reason for the difficulties in obtaining sound fillets is thought to be due to the accumulation of vaporized magnesium on powder surfaces as oxide, prohibiting the coalescence of molten powder particles.

(2) The brazability is dependent on the powder shape and size, finer particles with irregular shapes are found to be preferable. The reason is the higher stacking density of fine powder and self break down of surface oxide films during melting of irregular particles, leading to the higher possibility of coalescence, which enhances the fillet formability.

(3) The increase of magnesium and silicon content in filler metal lowers the brazability.

(4) The fillet formability is improved by the enhancement of magnesium vaporization rate, but sound fillets could not be obtained even under 100% vaporization.

(5) Mixing of powder by adding Al-40Mg to Al-10Si(-Mg) is effective in improving both fillet formability and magnesium vaporization.

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