Microstructure and Mechanical Properties of 6063 Aluminum Alloy Brazed Joints with AI-Si-Cu-Ni-RE Filler Metal

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A new low melting point filler metal, Al-Si-Cu-Ni-RE, was developed for the furnace brazing of aluminum alloy 6063. Flux-assisted brazing was conducted at 560 °C using the new filler metal and AlF₃-CsF-KF flux. Microstructure of the brazed joints were studied by means of SEM, TEM, and EDS. Shear strength and micro-Vickers hardness of joints had been tested. Results show that sound joints could be obtained with the filler metal and the flux. Microstructure characterization of the brazed joint shows dendritic CuAl₂ phase was distributed evenly and Si-phase was spheroidized and refined, which was embedded in CuAl₂ dendrites with modification of rare-earth element. Shear strength test results show that the joints with Al-Si-Cu-Ni-RE filler metal achieved average shear strength of 62.5 MPa, 14.5% more than the shear strength of brazed joints with Chinese HL401 filler metal. The micro-Vickers hardness of joint after T6 treatment is about 83 HV. The hardness of the joints after just brazing and after solution treatment was higher than the hardness of the base metal.

Keywords aluminum, brazing, joining, metallography, modification

1. Introduction

With advantages, such as high specific strength, low density, and superior corrosion resistance, aluminum and its alloys have been widely used in the aerospace, automobile, and construction industries (Ref 1). Brazing is used widely as an excellent assembly method for the bonding of aluminum components, especially in the manufacturing of radiators, condensers, and heat exchangers (Ref 2). Although a sound joint can be obtained with traditional filler metal (Al-12Si), its brazing temperature is too high due to its high melting point (577 °C). The high brazing temperature could degrade the mechanical properties or cause localized melting in some brazed joints.

It is well known that 6063 aluminum alloy is difficult to join, because the solidus temperature of the alloy is so low to be close to the liquidus temperature of commonly used Al-Si filler metal. Otherwise, for containing of magnesium, it is difficult to remove the 6063 aluminum alloy's surface oxide film away for conventional Nocolok flux. Therefore, to overcome this problem, recently many researchers have studied new filler metals. Suzuki introduced an Al-4.2Si-40Zn brazing alloy, and its melting point was 535 °C, but zinc is volatile and makes vacuum brazing process difficult (Ref 3). The brazing alloy Al-Ge-Si-Mg developed by Kayamoto et al., was used to

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produce a brazed joint in 6061 aluminum alloy. The tensile strength of which was close to that of base metal (Ref 4). However, high Ge content is costly. Humpston developed Al-5Si-20Cu-2Ni filler metal, and its melting temperature is 518-538 °C (Ref 5), because of the additions of Sr, Bi and Be, the filler metal is more suitable for vacuum brazing without flux assist.

Further studies of new low melting point filler metal for furnace brazing in a controlled atmosphere are needed. In this study, brazed joints of 6063 aluminum alloy were produced using the prepared brazing alloy Al-Si-Cu-Ni-RE and AlF₃-CsF-KF flux. The microstructure and mechanical properties of the brazed joints were studied. Mechanical properties were compared to alloy HL401, an important Al-Si-Cu aluminumbrazing alloy.

2. Experimental

Wetting experiments were performed to determine the stability of the flux, and the fluidity and wettability of the brazing alloy. In slow heating condition, many fluxes will decompose or react in a short time and lose their activity, and a lot of non-eutectic brazing alloys have poor fluidity and wettability, both which lead to failed brazing. For comparison, the melting of the Al-Si-Cu-Ni-RE rod $(0.2 \pm 0.02 \text{ g}, \text{ compo-}$ sition shown in Table 1) on 6063 aluminum alloy substrates (square plates, 40×40 mm, 2 mm height) (Ref 6), was accomplished by two heating regimes: (1) a slow heating condition comprised two rates (room temperature to 440 °C at 7.0 °C/min and 440 °C to 560 °C at 1.5 °C/min) and (2) a rapid rate of 18 °C/min. Temperature profiles for the slow and rapid heating are shown in Fig. 1. The experiments were conducted in N₂ flowing at 1-2 L/min in a resistance furnace. After the required time to reach a temperature of 560 °C, the

furnace was shut off and allowed to cool. The melting range of filler metal is from 515 to 533 °C. The composition of the 6063 aluminum alloy is shown in Table 2. The composition of AlF₃-CsF-KF flux was 8.8% CsF, 40.4% KF, and 50.8% AlF₃, all in wt.%.

The brazing process was carried out in the same condition as the wetting experiment with the rapid heating rate shown in Fig. 1. The brazed samples were treated with T4 and T6 temper condition, i.e., solution treated at 530 °C for 1 h, water quenched (T4), and then aged at 175 °C for 8 h (T6). The hardness of the joints was measured using micro-Vickers hardness tester under a vertical loading of 10 g. The shear strength test sample configuration was shown in Fig. 2.

Prior to the experiments, the 6063 substrates and the brazing alloy pieces were cleaned with acetone and ethanol to remove the oil and grease. An alkali solution and a nitric acid solution were used to remove the oxide film and then the samples were dried with an air gun.

The microstructure was observed with an XJG-05 optical microscopy (OM) and a JSM-6360LA scanning electron microscope (SEM) after metallographic preparation and etching in a mixture of 1 vol.% HF, 1.5 vol.% HCl, 2.5 vol.% HNO₃, and 95 vol.% H_2O at room temperature for 15 s. The phases of microstructure observed in SEM were identified with energy dispersive spectrometry (EDS).

For precipitated phase distribution determination, a transmission electron microscopy (TEM, JEOL JEM-2010) was used to investigate single particles. TEM was operated in bright-field mode using an acceleration voltage of 200 kV.

 Table 1
 Composition of brazing filler metal (wt.%)

Cu	Si	Ni	Re	Al	
25	8.5	1.5	0.2	Balance	



Fig. 1 Temperature profile in wetting and brazing experiments

Elemental analysis was performed with an EDS system (OXFORD-INCA) attached to the microscope.

3. Results and Discussion

3.1 Microstructure of Wetting Samples

The average wetting area of Al-Si-Cu-Ni-RE brazing alloy with rapid heating rate were more than 500 mm² and contact angles were less than 5°, which shows fine wetting of the brazing alloy. The macrograph of the wetting sample at the slow heating rate is shown in Fig. 3(a) and 3(b). The average wetting area of the brazing alloy is 121.8 mm² and the average contact angle is about 20°, which shows the brazing alloy's good wettability in the slow heating rate. In Fig. 3(b), there is a diffusion zone between brazing alloy and substrate. Figure 4 shows the interface microstructure of brazing alloy and substrate. As indicated in the result of EDS analysis, polygonal and strip silicon phases and dendritic CuAl₂ phase formed inside the brazing alloy and in the diffusion zone as shown in Fig. 4.

In wetting, once the brazing alloy is melted, diffusion of Si and CuAl₂ takes place from the brazing alloy to 6063 aluminum alloy substrate. In particular, the copper in brazing alloy diffuses very rapidly, for the irregular arrangement of atoms on the metal surface and the low diffusion activation energy on the substrate. Cu or CuAl₂ could diffuse into the substrate through the grain boundary at the Al/oxide film interface (Ref 7). Eutectic reaction of Cu atoms and Al atoms lower the surface free energy, and the surface tension was decreased, which could help in the removal of the oxide film and the spreading of filler metal. Silicon in brazing alloy also diffused into substrate metal and therefore polygonal and strip silicon particles were found at the interface of brazing alloy and substrate.



Fig. 2 Model of the shear strength test sample

Table 2	Composition	of 6063	aluminum	(wt.%)
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Mg	Si	Cu	Fe	Mn	Cr	Zn	Ti	Others	Al
0.6-0.9	0.3-0.6	0.10	0.15-0.35	0.15	0.05	0.15	0.10	0.15	Balance



Fig. 3 Macrograph of cross section from spreading sample in OM (a) and SEM (b) in slow heating condition



Fig. 4 Microstructure of the interface of brazing alloy and substrate alloy into substrate

3.2 Microstructure of Brazed Joint

Sound joints can be obtained for 6063 aluminum alloy using the developed Al-Si-Cu-Ni-RE filler metals at 560 °C. Figure 5 shows the brazing joint at low magnification and Fig. 6 shows



Fig. 5 SEM images of 6063 aluminum alloy joint with Al-Si-Cu-Ni-RE filler metal

the microstructure of the brazed joint in SEM. From Fig. 5, there was diffusion from brazing alloy to substrate. Figure 6 shows that the microstructure and EDS results of joint consisted of white dendritic phase and light gray bone-like phase and gray matrix. From EDS results, it is believed that the white phase was mainly CuAl₂ phase, the light gray and the gray phase were Si particle and α -Al, respectively. Primary phase of Si was not found and the Si particle was modified and spheroidized into bone-like and ball-like. The Si grains were uniformly distributed and the smooth grain boundaries were shown in Fig. 6(a). According to reinforcement theory, the fine and spheroidized Si particle would pretend dislocation movement and enhance material's strength. That was much related with the addition of Ni and RE in brazing alloy. RE elements could delay the nucleation process and lower the rates of the grain growth, and play a positive role on refining grain. Especially, the RE had the permanent modification to the shape of Si particle (Ref 8). The dendritic CuAl₂ was scattered and solid solution α (Al) had a larger proportion of total. This is benefit to the strength and ductility of the brazed joint and also counters balance for the brittleness from CuAl₂. In addition, addition of Ni elements in brazing alloy compensates for brittleness of CuAl₂.

In the diffusion process, once a "liquid-channel" is formed in the grain boundaries, Si will flow together with $CuAl_2$, which formed the microstructure shown in Fig. 6(a), where Si embedded in $CuAl_2$ (Ref 9).

In order to study ultra-fine precipitated phase and dispersed phase in the microstructure of brazed joint, joints were observed by TEM. Figure 7(a) and 7(b) show the microstructure of brazed joint using the bright-field image mode, and Fig. 7(e) shows the EDS result of intermetallic compound in Fig. 7(a). We found some strip black phase and very fine dispersed phase in Fig. 7(a) and 7(b). The main strip phase is intermetallic compound in which we found Fe atoms from EDS results. The composition of the intermetallic compound was about Al: 71.82%, Si: 6.29%, Fe: 19.69%, Cu: 1.94%, Ni: 0.27% (all in at.%), and deduced for the α (Al₁₂Fe₃Si) phase. In Fig. 7(a), there was the regular boundary intermetallic compound and in Fig. 7(b) there was smooth boundary intermetallic compound. In dispersed phase zone of Fig. 7(a)and 7(b), the size of dispersed precipitates was less than 10 nm, which is supposed to be unbalance θ'' (CuAl₂) phase after aging precipitation. As the brazing joints were taken out of furnace when furnace temperature was below 300 °C, and dry quenching and left as it is for a while, which has the efficacy of solution and aging treatment.



Fig. 6 Microstructure (a) and EDS results (b, c, d) of the 6063 aluminum alloy joint with Al-Si-Cu-Ni-RE filler metal

Compared with precipitated phase of brazing joint in Fig. 7(a) and 7(b), Fig. 7(c) and 7(d) show the morphologies of precipitated phase in aluminum matrix. The single needle-like phase in Fig. 7(c) could be β' -phase (Mg₂Si). There are no flaky β -phase in the TEM images and it indicates that the equilibrium phase has not enough time to form. Figure 7(d) shows the morphologies of precipitated phases along grain boundary.

3.3 Shear Strength and Hardness of Joints

Shear strength test results of brazed joints in the annealed condition are shown in Table 3. Compared to the brazed joint with Chinese brazing alloy HL401, the average shear strength of 6063 aluminum alloy joint without heat treatment with Al-Si-Cu-Ni-RE brazing alloy was 62.7 MPa, 16.4% more than the average strength of joints with HL401. It is worth mentioning that fracture occurs in the base metal of some of the tensile tests, and it also shows shear strength of brazed joint with Al-Si-Cu-Ni-RE brazing alloy approaches to that of the base metal. The advanced strength is related to addition of Ni and RE element. RE elements can change the mechanism from single transcrystalline fracture into mixture of transcrystalline fracture and intergranular fracture. Otherwise, Ni and RE element modified Si-phase and makes it refined and uniform, and it also makes CuAl₂ dendritic fine-grained and decentralized.

6063 aluminum alloy is usually used in solution heattreatment condition in practical applications. The hardness distribution of joints after solution treatment, aging treatment, respectively, was observed, compared with as-brazed joints, to evaluate the performance of brazing filler metal.

Figure 8 shows the micro-Vickers hardness of the as-brazed joints, the brazed + solution-treated joints, and the brazed + solution treated + aged joints. All the joints were brazed at 560 °C for about 10 min, the rapid heating rate shown in Fig. 1. The hardness of the joint, diffusion zone, and base metal for the brazed + solution-treated joints was higher than that for the as-brazed joints. This is due to the fact that (Cu,Si,Mg)-Al and (Si,Mg)-Al (for example, Mg₂Si, Al₂Cu, Al₈Mg₅, and Al₂CuMg) solid solutions were formed at the joint, diffusion zone, and the base metal, respectively. The hardness of the joints was increased by aging probably because of the precipitate hardening. Moreover, the hardness of the brazed joints for the as-brazed joints and the solution-treated joints was higher than that for the base metal for high content of Cu and Si in joint and diffusion zone, and the solid solution strengthening of the addition alloys. After aging, the hardness of the joints (83 HV) brazed at 560 °C was more than that of T6-treated 6063 base metal.

4. Conclusions

In the present work, a new Al-Si-Cu-Ni-RE filler metal was developed and the brazed joints of 6063 aluminum alloy with



Fig. 7 TEM images of coarse intermetallic phase (a, b) and precipitated phase (c, d) and the EDS result of intermetallic compound in Fig. 7a (e)

the filler metal were studied, including the microstructure and mechanical propertied of the brazed joints. The results can be summarized as follows:

- 1. Sound joints can be obtained for 6063 aluminum alloy using the Al-Si-Cu-Ni-RE filler metals at lower temperatures than the traditional Al-12Si filler metal.
- 2. Microstructure of the brazed joint consisted of α (Al)-CuAl₂ eutectic and Si-phase, dendritic CuAl₂ phase was distributed evenly and Si-phase was spheroidized and refined and embedded in CuAl₂ dendrites with modification of rare-earth element.
- 3. When the new filler metal is applied to brazing 6063 aluminum alloy at 560 °C for 10 min, the joints with

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Filler metals	Thickness, mm	Lap length, mm	Lap areas, mm ²	Tensile load, KN	Shearing strength, MPa	Average strength, MPa
HL401-1	2.71	14.34	38.86	2.10	54.04	54.69
HL401-2	2.80	13.97	39.12	2.46	62.89	
HL401-3	2.77	15.01	41.58	1.96	47.14	
Al-Si-Cu-Ni-RE-1	2.38	13.11	31.20	1.84	58.97	62.63
Al-Si-Cu-Ni-RE-2	2.60	13.65	35.49	2.56	72.13	
Al-Si-Cu-Ni-RE-3	2.34	13.70	32.06	1.82	56.77	



Fig. 8 Micro-Vickers hardness distribution of brazed joint

Al-Si-Cu-Ni-RE filler metal achieve average shear strength of 62.5 MPa, 14.5% more than the shear strength of brazed joint with HL401 Al-Si-Cu eutectic filler metal. The hardness of the brazed joint for the as-brazed joints and the solution-treated joints were higher than that for the base metal. The micro-Vickers hardness of the joints was significantly increased by aging at 175 °C. These values are quite close to those of base metal. We believe that a noble brazing of aluminum alloy 6061 using Al-Si-Cu-Ni-RE brazing alloy will be a very useful technique for the joining of 6063 aluminum alloy.

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References

- L.C. Tsao, T.C. Tsai, C.S. Wu, and T.H. Chuang, Brazeability of the 6061-T6 Aluminum Alloy with Al-Si-20Cu-Based Filler Metals, *J. Mater. Eng. Perform.*, 2001, 10(6), p 705–709
- J. Lacaze, S. Tierce, M.C. Lafont, Y. Thebault, N. Pebere, G. Mankowski, C. Blanc, H. Robidou, D. Vaumousse, and D. Daloz, Study of the Microstructure Resulting from Brazed Aluminum Materials Used in Heat Exchangers, *Mater. Sci. Eng. A*, 2005, 413–414, p 317–321
- K. Suzuki, M. Kagayama, and Y. Takeuchi, Eutectic Phase Equilibrium of Al-Si-Zn System and Its Applicability for Lower Temperature Brazing, *Jpn. Inst. Light Met.*, 1993, 43, p 533–538
- T. Kayamoto, J.H. Kim, S. Saito, and T. Onzawa, Brazing of Al-Mg Alloy and Al-Mg-Si Alloy with Al-Ge Based Filter Metals. *Proc. Workshop Jpn. Weld. Soc.*, Vol. 12, 1994, p 495–501
- D.M. Jocbson, G. Humpston, and S.P.S. Sangha, A New Low-Melting-Point Aluminum Braze, *Weld. Res. Suppl.*, 1966, p 243–250
- "Test Method of Spreadabillity and Clearance Filling Ability for Filler Meal," GB 11364-89, Metal Welding National Standards Committee, Chinese National Standard, 1989, p 168–170
- F. Gao et al., Solid State Si Diffusion and Joint Formation Involving Aluminum Brazing Sheet, *Mater. Sci. Eng.*, 2002, A337, p 228–235
- K. Noqita, S.D. McDonald, and A.K. Dahle, Eutectic Modification of Al-Si Alloys with Rare Earth Metals, *Mater. Trans.*, 2005, 45, p 323–326
- X.M. Qiu et al., Thermodynamical Analysis on Al Eutectic Reaction Brazing, *Chin. J. Nonferr. Met.*, 2001, 11(6), p 1017–1020