Dendritic Boundary Corrosion of AA2198 Weld Using Fiber Laser Welding with Al–Cu Filler Wire

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Abstract

The microstructures and corrosion behaviors of AA2198–T851 alloy and weld were analyzed under corrosive conditions. Weld was formed using an innovative fiber laser welding process with AA2319 Al–Cu filler wire. The metallurgical morphology and distribution of the chemical compositions were determined using imaging techniques such as optical micrograph, scanning electron micrograph, high-resolution transmission electron microscopy, energy-dispersive X-ray spectrometry and X-ray diffraction. Corrosion was evaluated using an immersion test and electrochemical impedance spectroscopy in 3.5% NaCl solution at room temperature. Results indicate that the parent alloy suffered from pitting corrosion during the initial 4-h immersion which was caused by the inhomogeneous distribution of its chemical components and the different intermetallics formed during the rolling process. The weld experienced dendritic boundary corrosion under the same conditions due to the addition of the Al–Cu filler and rapid solidification during laser welding, which led to the precipitates Cu enrichment along the grain boundary. When a welding joint was immersed in the solution for 5 days, a big crack was observed across the center of the weld. In comparison, there was good corrosion resistance in the heat-affected zone with a compact protective film.

Keywords 2198 Aluminum alloy · Fiber laser welding · Dendritic boundary corrosion · Al–Cu filler wire

1 Introduction

The third-generation Al–Li alloy, 2198, has been developed from Al–Cu–Mg alloy by adding a small proportion of the light metal Li and decreasing the Cu content. The addition of 1% Li to the Al alloy reduces the alloy density by 3% and increases the elastic modulus by 6% [1, 2], while significantly improving the fatigue intensity. In addition to Li, AA2198 is introduced to lower the Cu content of the 2195 Al–Li alloy from about 4 to 3% to reduce weight and significantly increase elongation. The combination of these properties and the resultant 10% lower density and 10% higher modulus than conventional aluminum alloys make the AA2198 Al–Cu–Li alloys to be

more attractive for applications in aircraft and other aerospace structures.

In addition to low density and high intensity in 2198 aluminum alloys, cost savings of up to 50% may be possible with alternate design strategies using integral and welded structures to replace build-up structures. Compared to other grades of age-hardenable aluminum alloy, AA2198 has better weldability, but it also suffers from lower welded joint strength. Laser welding, as a novel fabrication method, can decrease heat input, and it involves rapid solidification of the alloy with a very small microstructure (< 10 μm diameter) with matching filler can be attained, which is a very favorable mechanical welding property for joints. Rapid solidification during laser welding also induces the inhomogeneous distribution of microstructures and creates unstable phases, so the corrosion resistance of a joint may be deteriorate due to microsegregation, unfavorable phase content, the presence of microporosities and cracks and also vaporization. The presence of filler wire induces the microstructural and compositional differences between the weld and base metal, and also induces the Galvanic cell in the corrosive liquid. Our study found that

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the joint strength of welded AA2198 with Al–Si filler wire only reaches about 65% of the strength of the parent alloy [3]. There is uniform corrosion of the weld due to the addition of Si, which facilitates Cu scattering [4]. In order to further increase the joint strength for providing a greater filler wire choice for industrial welding applications, we tried to use the Al–Cu filler wire. According to Al–Cu and Al–Si binary alloy phase diagrams [5], the solid solution with more Cu (5.7%) than Si (1.65%) in α -Al can expect to be beneficial to improve the joint strength by the solid solution strengthening effect. In addition, the higher Cu content in Al–Cu filler than the parent alloy will also promote the compatibility between the parent metal and the filler wire and therefore will help to reduce defects, such as cracks and pores. Our previous study found that the tensile strength of welding joints in AA2198 with Al–Cu filler wire was about 57% as strong as the parent alloy [6]. This study investigates the corrosion behavior of AA2198 with Al–Cu filler wire using high-power fiber laser welding.

2 Experimental

A 2-mm-thick 2198–T851 Al–Cu–Li alloy sheet and AA2319 Al–Cu alloy filler wire with a diameter of 1.2 mm were used in this study. Their chemical compositions are shown in Table 1.

Prior to welding, the surfaces of the AA2198–T851 plate were polished by abrasive paper and degreased by acetone. Welding experiments were carried out with a YLS-6000 Fiber Laser (IPG Photonics) operating at 3 kW, as shown in Fig. 1. Laser conditions (power and speed) were optimized depending on the focal lens used. The welding speed and filler wire input speed were 3 and 2 m/min, respectively. The diameter of the laser beam spot was focused at 1 mm. An argon shielding gas with flux of 10 L/min was used to protect the heated surface from oxidation.

The microstructures of the weld and parent alloy were studied by an Olympus optical micrograph, a Hitachi S3400N scanning electron micrograph equipped with a Bruker 5010 energy-dispersive X-ray spectrometer (EDX) and a JEOL-2010 high-resolution transmission electron microscope (HRTEM). For the HRTEM studies, plates were mechanically thinned to 0.2–0.3 mm by manually polishing both surfaces of the disks with 240- to 600-grit abrasive papers. The 3-mm-thick disks were cut and then

Fig. 1 Schematic illustration of the laser welding experimental setup

electropolished in a 30% HNO_3 plus 70% methanol solution at -20 to -30 °C and at 20 V in a Model 110 Twin-Jet Electropolisher. The procedure produced sufficiently large, thin regions to facilitate HRTEM studies. The phase compositions of the parent alloy, weld and HAZ were characterized by X-ray diffraction (CuK_α radiation). Because the weld and HAZ regions of the joints are very narrow, it is very careful for the specimen preparation. Before experiment, they were machined by electrical discharge machining for the specimen size as large as possible. And then their cross sections were tightly glued on a metal plate together using wax. Subsequently, mechanical grinding was applied to the opposite side until the only weld and HAZ regions were left. X-ray spectra were collected in the 2θ range from 20° to 85° with a scan rate of $0.5^\circ \text{ min}^{-1}$.

Prior to preparing the corrosion specimens, square specimens ($1.5 \text{ cm} \times 1.5 \text{ cm}$) were obtained from a welded joint. For the electrochemical tests, these specimens were embedded in Epofix resin, with an electric Cu wire contact fixed on the inner surface so that, after preparation, only their outer surface ($20 \text{ mm} \times 20 \text{ mm}$) was exposed to the aggressive solutions. This surface was mechanically polished with 2000-grit SiC paper, washed with distilled water, degreased with acetone and dried at room temperature for 24 h. The aggressive medium was 3.5% NaCl solution. Electrochemical corrosion was tested using a VMP3 analyzer with a saturated calomel electrode as the reference electrode and Pt electrode as the counter electrode.

Table 1 Chemical compositions of test materials (mass fraction %)

Alloy	Cu	Li	Mg	Mn	Zr	Ti	Fe	Si	Zn	Ag	Ni	Al
AA2198	3.24	0.99	0.33	< 1	0.11	0.03	0.04	0.03	0.01	0.29	< 1	Bal.
AA2319	6.0	–	0.21	0.29	0.19	0.06	0.3	0.2	0.1	–	–	Bal.

3 Results and Discussion

3.1 Microstructural Analysis

Figure 2 shows optical micrographs of the AA2198 joint laser welded with Al–Cu filler wire. The parent alloy in Fig. 2a shows fibrous grain structure with an average thickness of about 30 μm and length of several hundred microns. AA2198 is a typical age-hardening aluminum alloy. Precipitation hardening is the main strengthening mechanism. This occurs through the formation of θ' (Al_2Cu), δ' (Al_3Li), T_1 (Al_2CuLi) or T_2 (Al_6CuLi_3) and S (Al_2CuMg) precipitates dispersed in the alloy [1, 7–9]. The weld shown in Fig. 2b was found to have a dendritic structure with narrow dendritic arm spacing due to rapid solidification. From the weld center to the fusion line, a columnar grain zone with a width of about 50 μm is shown in Fig. 2c. The growth direction of the columnar grain is along the heat flow direction. Between the columnar grain zone and the fusion line, an equiaxed fine grain zone with a width of about 15 μm was also identified because of the large supercooling there. Along the fusion line and close to the base metal, there was a wide heat-affected zone (HAZ) decorated with plenty of tiny precipitates.

In order to further identify the phase distribution, X-ray diffraction was conducted as shown in Fig. 3. In the weld, θ' , δ (AlLi) and T_2 (Al_6CuLi_3) were the main precipitates, whereas, in parent alloy and HAZ, besides θ' , δ and T_2 , a small amount of other precipitates including $\text{Al}_3\text{Li}_2\text{Si}_4$, Al_8Mn_5 and AlLiSi were also found. A small quantity of the precipitates in the weld also implies the lower hardness and tensile strength than those of the base metal [6]. HRTEM of the parent alloy (Fig. 4a) shows the densely distributed dark structure presenting an aciculate and granular shape with tens to hundreds of nanometers in diameter. Meanwhile, a plenty of dislocations were found due to the extensive plastic deformation and flow of materials during rolling. The selected area electron diffraction shows that the light area is α -Al with [001] direction (Fig. 4b). The dark structure is identified as a

Fig. 3 XRD patterns of AA2198–T851 plate, weld and HAZ using fiber laser welding with Al–Cu filler wire

hexagonal structure with [011] direction (Fig. 4c). The lattice constants of a and c were 4.97 Å and 9.35 Å, respectively, which corresponds to T_1 precipitate. Around the main diffraction spot, plenty of vaguely visible satellite spots were also found. The literature has reported that the addition of Cu to Al–Li alloy promoted the precipitation of T_1 and θ' phases [10, 11]. Cavaliere et al. [12] also found the δ' phase in 2198–T851 aluminum alloy. In our study, precipitates of T_2 and Al_8Mn_5 are also found besides of T_1 , θ' and δ' phases. The formation of the T_1 , T_2 , θ' and δ' phases in aluminum alloys greatly increases their mechanical properties [2]. Meanwhile, these phases also accelerate the occurrence of corrosion [11]. This also implies the complicated corrosion mechanism.

High strength is achieved in aircraft aluminum alloys with the use of Cu as an alloying element; therefore, Al–Cu filler is usually used to increase the strength of a welding joint. However, the distribution of Cu will directly affect the corrosion properties of the joint. Figure 5 shows the EDX face scanning mapping of Al and Cu in AA2198 weld with Al–Cu filler wire using fiber laser welding. The results indicate that the distribution of Cu in the weld was

Fig. 2 Optical micrographs of **a** AA2198 parent alloy, **b** weld with AA2319 Al–Cu filler, **c** interface between weld and parent alloy

Fig. 4 TEM result of AA2198–T851 plate material showing **a** bright field structure; **b** SAED spot in bright area; **c** SAED spot in dark area

Fig. 5 EDS mapping of AA2198 weld, **a** SEM image, **b** element distribution, **c** Cu mapping, **d** Al mapping

inhomogeneous. From Table 2, we can clearly see that the light zone (see square 1) consists of plenty of Cu (about 13% Cu), whereas the dark zone (square 2) is mainly composed of Al, only about 0.7% Cu. From these results, it is clear that plenty of Cu was distributed on the dendritic boundary. Rapid solidification during laser welding caused the strengthening phases containing Cu, such as T_1 , T_2 and

θ' , to precipitate, particularly along the dendritic boundaries. Kim and Lee [13] also demonstrated the directionality of the strengthening effect associated with the T_1 phase, and finally, it affects the tensile properties.

Figure 6a shows a representative transmission electron microscope image of a weld. From the bright field image, there were not observed precipitates in the substrate of

Table 2 Element contents detected by EDS

	Al	Cu	Mg	Fe	Zn
Light zone	84.76	13.57	1.26	0.21	0.20
Dark zone	97.96	0.67	1.10	0.13	0.14

weld. This result is consistent with that in the XRD. The majority of chemical elements in the grain boundary (Fig. 6b) and intracrystalline are (Fig. 6c) Al, Cu, Mg, and Fe. As shown in Fig. 6b, the high value of Cu spectrum in the grain boundary implied high Cu content. From the selected area electron diffraction (inset of Fig. 6a), the θ' phase with $[\bar{1}12]$ direction was identified. The precipitates with high Cu content distributed along the dendritic boundary in the weld were responsible for the high electrode potential there.

3.2 Corrosion Behavior of AA2198 Joint After Laser Welding

Figure 7 shows the macroscopic corrosion morphology of an AA2198 joint after immersion in 3.5% NaCl solution for 5 days. The base metal (Fig. 7a) contains big pits with wide diameter. In comparison, HAZ presented good corrosion resistance with a compact protective film. Weld shows big corrosive cracks locally that passed through the whole weld (Fig. 7b).

Different corrosion characteristics in the base metal, weld and HAZ are associated with the microstructure and

chemical distribution. Figure 8 shows the microscopic corrosion morphologies of different regions in the AA2198 joint. From these figures, distinctive pit morphologies could be seen. After immersion in 3.5% NaCl solution for 4 h (Fig. 8a), the surface of the AA2198 base metal showed plenty of pits. This is corrosion along the dendritic boundaries of the weld (Fig. 8b). Serious corrosive attack occurred at the dendritic boundaries with dissolution phenomenon occurring in places (inset) when the immersion time was increased to 5 days (Fig. 8b). In HAZ, small, densely distributed pits were found in immersion for 4 h compared with the parent metal (Fig. 8c).

Interestingly, the morphology of pits in the eroded surface of the parent alloy was very different from the fibrous grain structure of the parent alloy. Two kinds of pit were identified based on their Cu content. One (labeled 1) had a sandwich structure with bright center and black outer; the other (labeled 2) showed typical pitting corrosion. According to the EDX results, the sandwich's bright center was enriched with Cu. The pits labeled as 2 were identified as the phase with low Cu. But, Li was undetected. This implies that θ' , δ and T_2 precipitates in the base metal were responsible for the pits initiation. In comparison, weld showed corrosion at the dendritic boundaries when it was immersed in 3.5% NaCl solution for 4 h. Rapid solidification during laser welding enriched the eutectic structure θ' phase and other precipitates along the dendritic boundaries. Cu was therefore particularly deleterious to the corrosion behavior of Al alloys owing to the Galvanic couples setup between Cu-rich intermetallic particles and the

Fig. 6 **a** TEM image of the weld with AA2319 Al–Cu filler wire, **b** element spectrum in the grain boundary, **c** element spectrum in intracrystalline

Fig. 7 **a** Macrostructure of AA2198 joint after immersion in 3.5 wt% NaCl solution for 5 days, **b** enlarged view in the site A

Fig. 8 SEM images showing corrosion morphology of joint in 3.5 wt% NaCl solution: **a** parent alloy for 4 h, **b** weld for 4 h, the inset for 5 days, **c** HAZ for 4 h

adjacent Cu-depleted matrix. Enriched Cu particles at the dendritic boundaries acted as the cathode compared with the Al base when immersed in the electrolyte, leading to the preferential creation of Galvanic cells along the dendritic boundaries. Increasing the immersion time to 4 days, there was obvious corrosion at the weld's dendritic boundaries (Fig. 8b, inset) and a macroscopic crack was observed in the surface (Fig. 7). Meanwhile, cathodic reactivity was found in a form of grooves around the dendritic boundary. HAZ showed different corrosion morphology. During laser welding, HAZ experienced solid-state phase transformation, leading to the formation of plenty of the precipitates. In the initial immersion for 4 h, a lot of small pits were created due to the Galvanic cell between the precipitates and the Al base.

Figure 9 compares the impedance responses of the parent alloy with weld. The data reveal that the impedance diagram of the parent alloy consisted of a large capacitive loop at high frequencies and a small inductive one at low frequencies during the initial 2-h immersion. When the immersion time increased to more than 2 h, only the capacitive loop was observed. The weld had a large capacitive loop at both high and low frequencies. The

bigger diameter of the capacitive loop in weld reflected the higher impedance value.

The inductive loop in the parent alloy may be attributed to the relaxation process of the oxide presenting on the electrode's surface, which was obtained by the adsorption of the species such as Cl_{ads}^- and H_{ads}^+ [14]. This was consistent with the proposed mechanism of chemisorption of Cl^- ions onto the oxide surface forming an oxide-chloride complex [15]. This complex exhibits fast dissolution in the presence of H^+ . Similar findings were reported by Lenderink et al. [16] during the corrosion of pure Al in acidic solutions containing chloride ions. Some authors [17, 18] also think that the inductive loop exists in the early stage of pitting corrosion, consistent with the results of this experiment. With corrosion proceeding, the oxide film is considered to be the parallel circuit of a resistor due to ionic conduction in the oxide film and a capacitor due to its dielectric properties. The impedance response presents the capacitive loop both in high and low frequencies. The electrode impedance in this case was determined by the metal-oxide interface, the oxide film and the oxide-solution interface. At this time, a high-frequency and low-frequency capacitive loop could be assigned to the relaxation

Fig. 9 **a** EIS of AA2198 parent alloy and the weld after immersing in 3.5 wt% NaCl solution, **b** the partial magnification picture in **a**

process due to the natural oxide present on the surface of the aluminum alloy samples and its dielectric properties.

4 Conclusions

Aluminum alloy is a hard-to-weld metal material using laser welding due to the high reflectivity of aluminum to laser and low joint strength. Light alloy with the addition of a small proportion of Li can improve the alloy's strength. Studying the corrosion behavior of an AA2198–T851 joint welded with Al–Cu filler wire using fiber laser welding, the following conclusions can be drawn:

1. The AA2198 parent alloy is composed of large elongated structures with fine T_1 , T_2 , θ' and δ' precipitates. In weld using fiber laser welding with an Al–Cu filler, very fine dendritic structures with narrow dendrite spaces were observed. The addition of Al–Cu filler induced the inhomogeneous distribution of Cu due to rapid solidification during laser welding. Cu was mainly distributed along the grain boundaries. HAZ had plenty of tiny precipitates.
2. The AA2198 parent alloy showed pitting corrosion behavior in 3.5% NaCl solution. Two kinds of pits were identified depending on the chemical species in the precipitates. In the precipitates containing Cu, such as T_1 and θ' phases, electrode transfer happened during corrosion; therefore, sandwich corrosion morphology was observed. In the precipitates not containing Cu, there was pitting corrosion. Weld showed dendritic boundary corrosion behavior, and when immersed in solution for a long time, some big cracks appeared. HAZ presented favorable corrosion resistance.
3. Electrochemical impedance spectroscopy measurements showed that parent alloy had an inductive effect in initial corrosion in 3.5% NaCl solution due to the

pitting effect and weld had a large capacitive loop at both high and low frequencies.

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