Original Article

Influence of Cu/Li ratio on the microstructure evolution of bobbin-tool friction stir welded Al–Cu–Li alloys

Jannik Entringer a,*, Martin Reimann a, Andrew Norman b, Jorge F. dos Santos a

a Helmholtz-Zentrum Geesthacht, Institute of Materials Research, Materials Mechanics, Solid State Joining Processes Department, Max-Planck-Str. 1, 21502 Geesthacht, Germany
b ESA, European Space Agency, Materials Technology Section, Noordwijk, The Netherlands

A R T I C L E   I N F O

Article history:
Received 15 May 2018
Accepted 16 January 2019
Available online 15 March 2019

Keywords:
Microstructure
Precipitation sequence
Bobbin tool friction stir welding
Aluminum
AA 2196
AA 2060

A B S T R A C T

Two modern aluminum lithium alloys were welded by semi-stationary bobbin tool friction stir welding. The influence of the Cu/Li ratio on precipitation phenomena under process heat impact was investigated by comparing the response of low Cu/Li alloy 2196-T8 and high Cu/Li alloy 2060-T8. Identical process parameters with a weld pitch of one rotation per mm were used to conduct flawless weldments. The thermal history and microstructural features were studied and correlated to the resulting mechanical properties of the welds. Analysis of microstructure using differential scanning calorimetry and high energy X-ray diffraction technique showed significant differences in the precipitation sequence of the base metal and in the welded samples of the two alloys of interest. A low Cu/Li ratio led to a higher softening resulting in a reduction of 43% of base metal yield strength while the high Cu/Li ratio alloy AA 2060 could demonstrate more thermal stability (38% reduction). Severe dissolution of the T1 precipitate and presence of equilibrium phases were confirmed for the stirred zone of both alloys. The heat affected zone suffered dissolution and overaging reactions leading to a mechanically unfavorable microstructure. The low Cu/Li alloy 2196 developed a higher process temperatures and exhibited a more evolved precipitation sequence.

© 2019 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

1. Introduction

Al–Cu–Li alloys have been produced for more than 100 years. Alloying with lithium is especially attractive since it increases

* Corresponding author.
E-mail: jannik.entringer@hzg.de (J. Entringer).
https://doi.org/10.1016/j.jmrt.2019.01.014
2238-7856 © 2019 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).
multi-stage heat treatments leading to more balanced properties. The history of Al-Li alloys, as well as the property evolution, was reviewed in detail in [2,3]. Al-Li alloys are of special interest when materials with high specific strength and modulus are required. Consequently, these alloys can be found in many aeronautical and space applications. Prominent representatives are AA 2195, which led to massive weight savings on the external tank of the space shuttle [4], AA 2050, which was used in the modern Airbus fleet [5] and AA 2198, which was used in the manufacturing of the first stage of Falcon 9 rockets [6].

The Cu/Li ratio in Al–Cu–Li alloys is a key parameter with regards to their properties and therefore often focus of research. The development throughout the Al–Cu–Li generations led to a consequent reduction of the Li content, holding it under 2 wt% from most of the 3rd generation alloys. The development of the Cu/Li content is shown in Fig. 1 for a multitude of 3rd generation alloys. Interestingly, the alloy composition development presented in Fig. 1 shows that either high- or low-ratios are preferred. As the precipitation sequence features aspects of both binary Al-Cu and Al-Li systems [7], it seems that alloys are designed to take advantage primarily of one of those systems (which, in the case of low Cu/Li contents Al-Li and high Cu/Li contents Al-Cu). This finding is confirmed by two recent studies investigating the role of Cu/Li in Al-Li [7,8].

Since the role of the different alloying elements was identified as the key to the complex microstructure evolution in Al-Cu-Li alloys, several studies have been conducted focusing on the microstructure evolution [9,10]. The precipitation sequence found for Al-Cu-Li alloys sensitively depends on the Cu/Li ratio, and the main reactions can be summarized as

$$\alpha(\text{SSSS}) \rightarrow \text{GP zones} + \delta' \rightarrow T1 + \delta + \theta' \rightarrow T_B + T_2 + \theta$$  \hspace{1cm} (1)

Jo and Hirano found in 1987 a strong Cu/Li ratio dependence of the precipitation sequence [11] and further proposed a Cu/Li dependent sequence for the Al–Li–Cu system. This shows that the alloys in this study satisfy the following conditions:

For Cu/Li $> 4$ \begin{equation} \alpha(\text{SS}) \rightarrow \text{GP zones} \rightarrow \theta'' \rightarrow \theta' \end{equation} (2)

For Cu/Li $1 - 2.5$ \begin{equation} \alpha(\text{SS}) \rightarrow \text{GP zones} + \delta' \rightarrow \theta' + \delta'' \rightarrow \delta + T1 \rightarrow T1 \end{equation} (3)

The T1 phase consists of Al$_2$CuLi, while $\delta'$ consists of Al$_2$Li, and $\theta'$ of Al$_2$Cu. Although these sequences are often cited, they can only be taken as guidance since several other phases are frequently observed when detailed microstructure analysis is reported, such as T$_2$, T$_B$, S' or $\Omega$ [12,13]. Furthermore, modern third generation Al-Cu-Li alloys are becoming more complex with extending the functional microstructure with elements from Al-Li-Cu, Al-Li-Mg, Al-Li-Zr, Al-Cu-Mg or quaternary Al-Cu-Li-Mg-(Ag) systems. Minor elements can lead to changes in the microstructure [14]. However, the Cu/Li ratio seems to be of most importance and in 2013, Decus et al. [7] investigated the role of Cu/Li ratio in Al-Cu-Li-X alloys by analyzing the microstructure of AA 2196 and AA 2198 in different heat treatments. The findings reveal a different precipitation sequence with more Cu containing precipitates, such as Cu-rich clusters, and $\theta'$ for the high Cu/Li alloy and more Li-rich clusters and $\delta'$ for the low Cu/Li alloy. The T1 phase developed in both alloys but showed later incubation in case of the low Cu/Li ratio alloy.

With the introduction of Friction Stir Welding (FSW) in 1991, a new joining technology was developed, which could deal with hard-to-weld materials. This is the case of aluminum, which usually suffers under the high-heat input induced by conventional fusion welding techniques [15]. FSW follows the principle of intermixing, joining parts in a solid-state condition, whereby the energy for sufficient plastification is generated by the contact between the weld tool and the workpiece, which generates frictional and deformational heat. As the whole working process typically occurs below the melting temperature, significant improvements on the joint properties can be achieved. Comprehensive studies can be found in regularly published reviews on FSW as for example by Threadgill et al. [16]. Bobbin Tool Friction Stir Welding (BT-FSW) is a variant of FSW for structures with limited access, since it forgoes the backing bar via an incorporation of a second shoulder to maintain the required tool pressure. BT-FSW therefore increases the flexibility of the application, although the more complex tooling and process control has hampered industrial applications.

Less than 50 studies on BT-FSW have been published since introduction in 1991. The general increasing publication numbers confirm the success in FSW research, bringing more attention to niche variants, such as BT-FSW or stationary shoulder variants. Most of the investigations about BT-FSW have focused on aluminum alloys, of the type Al-Si-Mg (6xxx) and Al-Cu (2xxx). Interestingly, a significant fraction of the work on the 2xxx series alloys employs Al-Cu-Li alloys confirming the high interest for these types of alloys. One of the first studies on BT-FSW in Al-Li alloys was performed by Skinner in 2003 [17] showing general feasibility on AA 2195 by comparing its mechanical performance to that of
conventional FSW. In 2010, Schneider et al. [18] investigated the correlation of the bulging, the grain-refinement and the strength of AA 2195-T8 welded using BT-FSW. Recently, Wang et al. [19] welded AA 2198 T851 using BT-FSW. The influence of the process parameters on the mechanical properties was investigated showing high mechanical efficiencies up to 80% UTS. Later, Shen et al. [20] revealed the local texture of different weld regions in BT-FSW of the same material. Four subzones were identified, formed vertically in the stirred zone (SZ) holding different textural patterns. In 2015, the concept of stationary shoulder was adapted to BT-FSW [21]. It was applied as semi-stationary shoulder BT-FSW by means of holding the upper shoulder stationary (SSBT-FSW). Employing this concept, such benefits as lower heat input or superior surface finish reported from stationary shoulder FSW were transferred to BT-FSW applications. In a study by Goebel et al. [22] coupons of AA 2198-T851 were produced by SSBT-FSW and compared to the conventional BT-FSW welds in terms of mechanical and thermal analysis. To the authors’ knowledge, no further Al-Cu-Li alloys have been welding using BT-FSW.

Only a small number of studies exist dealing with friction stir welding of AA 2060 alloy [12, 23] or the more established AA 2196 alloy. The literature lacks studies on the weldability and behavior of those alloys when subjected to BT-FSW. Belonging to the same Al-Cu-Li family, AA 2060 and AA 2196 exhibit a significant difference in the Cu/Li ratio (4.67 and 1.45, respectively), which is known to affect the precipitation morphology. Additionally, the influence of the Cu/Li ratio on the precipitation evolution response when exposed to welding thermal cycles has not been studied in detail.

In this study, AA 2060-T8 and AA 2196-T8 have been welded in a butt-joint configuration using identical process parameter and welding conditions. The semi-stationary shoulder bobbin tool friction stir welding (SSBT-FSW) was employed. The influence of the Cu/Li ratio on the materials behavior during welding was analyzed. To this end, thermal cycle analysis and analysis of microstructural features, including the precipitation evolution, was employed to explain the mechanical properties, such as hardness and tensile characteristics, for both alloys. The precipitation evolution in the different welding zones is discussed with respect to the influence of the Cu/Li ratio.

2. Materials and methods

2.1. Materials

The materials used in this study belong to the group of third generation Al-Li alloys. The composition is provided in Table 1 and was determined by X-ray fluorescence spectroscopy plus additional inductively coupled plasma optical emission spectrometry for the light elements Li and Ag. Both materials were received in the T8 temper, i.e., solution heat treated, cold worked and artificially aged. Sheets of AA 2060 T8 were machined to 300 mm × 100 mm × 3 mm, and extrusions of AA 2196 T8 were machined to 200 mm × 100 mm × 3 mm. Prior to welding the joining surfaces have been flat milled and cleaned with ethanol.

2.2. Experimental procedure, testing and analysis

Fig. 2 illustrates the SSBT-FSW process, which was used to join the two workpieces in a butt joint configuration as applied in this study. The upper shoulder is held stationary, while the lower one is connected to the rotating probe. More details on the process and a comparison to conventional BT-FSW are provided in a recent work [22].

All welds were produced on a five-axis kinematic robot system (PKM T805) equipped with a custom designed weld head. The robot has five degrees of freedom provided by three axial and two rotational actuators. Process response data, such as torque, forces and velocities, were recorded with an acquisition rate of 200 Hz.

The tools were dimensioned to a 7 mm diameter, zigzag featured probe and two 15 mm diameter shoulders. The probe was manufactured out of a nickel-cobalt-chromium alloy (MP159™) and the shoulder was made from a molybdenum-vanadium hot work steel (X8CrMoV5-1). The rotating shoulder was symmetrically scrolled (every 60°, half-moon shaped), while the stationary shoulder maintained a flat surface. The process was run in self-reacting mode, meaning a constant force between the tool shoulders was maintained during the process by axial actuators. The welding was performed in butt joint configuration parallel to the longitudinal direction. Welding parameters were set to a rotational speed of 150 RPM, 150 mm/min transverse speed with a constant force of 5.5 kN between the shoulders.

Fig. 2 illustrates the welding process. In a), the rotating tool approaches the workpieces holding a non-rotational upper shoulder. In b), the lower shoulder moves toward the workpiece until in contact with the workpiece (c). After a short dwelling time, the welding tool translates and starts the welding process d).

Prior to any testing, the welds were left at room temperature for two months allowing natural aging processes to settle. No significant strengthening was observed between two and twelve month. For mechanical testing samples were machined by spark erosion to a modified dog-bone-like shape with a parallel gauge length of 50 mm and a width of 12.5 mm, as shown in Fig. 3.

The weld thermal cycle was recorded using K-type thermocouples of 0.5 mm diameter, which were placed at mid-thickness in drilled holes and sealed using a silver conductive paste. The positions were 3.5, 6, 9 and 20 mm away from the weld center toward the advancing side (AS), compare Fig. 3. Additionally, one thermocouple was placed in the

| Table 1 – Base metal: Chemical composition and mechanical properties. |
|--------------------------|----------|----------------|----------------|------------------|-----------------|-----------------|-----------------|-----------------|
| Alloy | Cu/Li ratio | Cu | Li | Zn | Mg | Mn | Ag | Al | UTS |YS | Hardness |
| 2060 | 4.67 | 4.2 | 0.9 | 0.36 | 0.85 | 0.32 | 0.36 | Bal | 516 MPa | 447 MPa | 169 HV |
| 2196 | 1.45 | 2.9 | 2.0 | 0.05 | 0.38 | 0.32 | 0.28 | Bal | 549 MPa | 513 MPa | 169 HV |
center of the weld. This measurement was performed only once, as the thermocouple is destroyed by the rotating probe. Cross-sections were analyzed using a light microscope after process to evaluate the actual thermocouple position with respect to the stirred zone (SZ) border. Tensile tests were carried out on a Zwick & Roell Z100 equipped with a 100 kN load cell. The tests were performed on specimens extracted perpendicular to the rolling direction with a speed of 1 mm/min following ISO 6892-1:2009. The hardness measurements were conducted on a Zwick Vickers hardness tester (LECO, type M-400-H) in accordance with ISO 6507-1:2005 and applying 200 grams for 10 s. Measurements were taken at mid-thickness with a step of 300 μm.

For microstructural analysis, samples were sectioned perpendicular to the welding direction and prepared employing a standard metallographic specimen preparation procedures by means of flat grinding and finish polishing. For microstructural analysis using polarized light microscopy, samples were anodized using a 1 vol% solution of HBF4, known as BARKER solution, at 22 V for 90 s.

Differential scanning calorimetry (DSC) measurements were conducted using a Netzsch DSC 200 F3 Maia®. The base metal and stirred zone samples were prepared as 5 mm diameter disks of approximately 70 mg weight taken from mid-thickness. Due to limitations in the sheet thickness, the heat affected zone (HAZ) samples were adjusted to a mass of 40 mg. HAZ sample thickness was oriented vertically to the sheet surface and taken from the advancing side. The temperature cycle was set to an isothermal step at 25 °C before heating at a rate of 10 K/min from 25 to 590 °C. Samples were corrected via baseline subtraction and normalized to sample mass.

High-energy X-ray diffraction (HEXRD) was performed at the beamline of Deutsches Elektronen- Synchrotron (DESY) in Hamburg, Germany. Samples with a thickness of 3.0 mm were measured in transmission with a beam cross section of 0.175 mm x 0.2 mm. For the HAZ location, the sample was measured at the location of the DSC (HAZ) sample. The specimens were exposed to high-energy X-rays with a photon energy of 100 keV, which correlates to a wavelength of 0.124 Å. Debye-Scherrer diffraction rings were recorded on a 2-dimensional Perkin Elmer XRD 1622 detector with an exposure time of 10 seconds. Subsequent diffractograms were generated by integration and compared to simulated phase diffraction patterns for qualitative phase analysis.
3. Results

3.1. Macrostructural features

Macrographs of cross-sections of both materials are presented in Fig. 4. Complete weld consolidation with no evidence of volumetric defects or other flaws can be observed. The SSnBT-FSW weld shows an asymmetrical hourglass shape weld zone formation widening up toward the rotating, lower shoulder. The weld zones follow the common FSW characteristics (BM: base metal - HAZ: Heat affected zone - TMAZ: Thermo-mechanically affected zone - SZ: stirred zone). The SZ experienced highest shear rates and temperatures leading to fully recrystallized, fine grains. In the BT-FSW, material is sheared and transported by the probe. The opposed, translational directions of workpiece and probe lead to a sharp transition of SZ and TMAZ on the advancing side (AS). Because of material extrusion on the retreating side (RS) around the probe, the SZ is widened toward the RS and exhibits a smooth transition SZ-TMAZ transition.

3.2. Analysis of the mechanical properties

The micro-hardness profiles of the joints produced in both alloys are presented in Fig. 5. Both alloys display a W-shape profile, well-known for high-strength, precipitation-hardened aluminum alloys in peak-aged temper [15]. Highest temperatures are present in the center and lead to severe softening reactions, compare Chapter 3.3. The HAZ undergoes softening until the point of lowest hardness in the TMAZ is reached. After this point, the material becomes harder again until the SZ plateaus in the center of the weld. Highest temperatures are measured in the center of the weld (compare chapter 3.3) leading to severe softening reactions. The plateau is wider than the SZ and reaches into the TMAZ, the transition between the weld zones is not reflected by the hardness values. Low Cu/Li alloy 2196 experiences a higher softening in the location of lowest hardness. Here, the hardness decreases 10% more than in the case of AA 2060. As the weakest location determines the overall performance, this behavior is also seen in the tensile testing, which is presented in Table 2. Low Cu/Li alloy 2196 reached 57% of the base metal yield strength, while high Cu/Li alloy 2060 achieved 68% (70% and 78% of base metal ultimate strength, respectively). All samples fractured in shear mode while necking was observed on both sides of the joints, final failure occurred on the AS in all samples. The final crack follows an approximately 45° path, compensating for the high shearing forces.

3.3. Thermal cycle analysis

The temperature measurements were recorded at mid thickness at various distances from the SZ. Because of the different dimensions of the SZs (compare previous chapter) and to minimize deviations, the actual distance of the thermocouples

| Table 2 – Microhardness and tensile testing results of both materials. |
|-------------------|---------|---------|-------------|---------|---------|--------------|
| Alloy | BM YS/UTS [MPa] | YS efficiency [%] | UTS efficiency [%] | BM hardness [HV] | SZ hardness [HV] | Lowest hardness [HV] |
| 2196 T8 | 518/572 | 56 ± 2 | 67 ± 3 | 170 | 130 | 110 |
| 2060 T8 | 457/512 | 62 ± 1 | 78 ± 2 | 170 | 135 | 128 |
to the SZ border was determined ex situ via metallographic analysis. The SZ area is experiencing highest temperatures during the process, as a result of shear and friction dissipations. From the origin of heat generation close to the rotating tool, heat is dissipated to the surrounding material. Chao et al. [24] investigated the temperature fields around the tool and observed that 95% of frictional heat is transferred into the workpiece. Therefore, the temperatures in the workpiece will represent the heat input of the tool. The temperature evolution of both materials at distances of 0.3 mm (0.4 mm), 2.5 mm, 5.5 mm and 17 mm outside the SZ are shown in Fig. 6 a) and b). Additionally for AA 2060, the temperature was measured in the SZ. Heating rates were calculated between 100 °C and 300 °C for the thermocouples at the position 0.3 mm (0.4 mm). AA 2196 experiences rapid heating with a rate of 37.5 K/s, the temperature reaches a maximum of 465 °C at 0.3 mm outside the SZ and cools down at a rate of 4.3 K/s. For AA 2060, the heating rate is 32.7 K/s, the maximum temperature 450 °C at 0.4 mm from the SZ, and the cooling rate 4.6 K/s. Generally, heating rates increase with vicinity to SZ. In Fig. 7a the maximum temperatures are plotted against distance and show linear behavior up to 5 mm from the SZ. Further distances of temperature measurements do not follow this linear behavior attributing to the changing heat conduction conditions most likely related to the clamping condition.

Fig. 7a compares the maximum temperatures at various positions of both alloys. Additionally, higher heating rates but similar cooling rates are measured for AA 2196, compare Fig. 7b. In the proximity of the SZ, AA 2196 show slightly higher peak temperatures than AA 2060. The SZ temperature measurement in AA 2060 recorded a maximum process temperature of 478 °C. Due to identical welding speed and thermocouple positions, the higher peak temperatures in AA 2196 can only be achieved with higher heating rates. The heating rate is approximately 5 K/s higher than in AA 2060 while the measured cooling rates are similar (4.6 K/s vs. 4.3 K/s). Because the boundary conditions such as welding parameters or clamping situation are hold similar for both materials, these rates will depend on the materials thermal properties such as specific heat capacity and conductivity. As those quantities are not existent for the alloys under investigation, pure metal data for the main alloying elements is taken as indication. The conductivity is lowest for lithium, followed by aluminum and copper (71 < 210 < 385 W/mK) and the specific capacity is the lowest for copper followed by aluminum and lithium (0.39 < 0.90 < 3.31 J/gK) [25]. Following the pure metal properties, alloy AA 2060 will hold high conductivity with a lower capacity while AA 2196 has a low conductivity with a high capacity. A lower conductivity in AA 2196 can attribute to a formation of the heat pool in the SZ while in AA 2060 the heat will be transferred to the outer areas. A more detailed study of the thermal diffusivity of the alloys is needed to understand this critical physical property differences.

3.4. Microstructure evolution

To reveal differences in the precipitation evolution, DSC and HEXRD analysis were conducted in the BM, SZ and HAZ of both materials. The HAZ specimen was taken 6 mm from the center of the weld. BM material was taken in un-welded condition, while the SZ underwent highest temperatures of up
to 478 °C (in the case of AA 2060) and the HAZ microstructure experiences temperatures of approximately 400 °C at 6 mm from the weld center.

Since the Cu/Li ratio is the main difference of the two alloys under investigation, the contribution of other elements as Zn and Mg is not primary considered in this study. Still, Mg contributions are reported to facilitate the T1 nucleation [14,26] and provide strength while Zn is used to offset the loss in corrosion resistance when adding Mg [27].

The dashed lines in Fig. 8a and b represent the DSC signal of the alloys in T8 condition (as received). As T8 temper is at the peak aged stage, the main strengthening precipitate T1 is expected with minor contributions of precipitates formed at low-temperature such as δ' phase and clusters. During heating, the existing phases will dissolve according to their solvus temperature. Both alloys reveal a similar thermogram consisting of three endothermic peaks (A, B, E) indicating dissolution reactions and two exothermic peaks (C, D) indicating phase formation reactions. Peak A (130 °C) is associated with the dissolution of GP zones and fine δ' phases while peak B (210 °C) is correlated with the dissolution the δ' phase [28]. The δ' phase seen at 210 °C is believed to exist during the artificial aging for T8 temper, which is usually done at lower temperatures [26]. While the artificial aging will stabilize the phases found in peak B, fine δ' phases can precipitate during natural aging (peak A). Because of its higher Li content, AA 2196 shows a higher intensity of peak B, a general lower response is measured in AA 2060 at temperatures below 220 °C. At temperatures of approximately 250 °C, the exothermic region C indicates the formation of the main strengthening precipitates such as T1 and δ'-like phases, which form preferably under elevated temperatures [7]. Minor exothermic peaks in region D are assumed to occur due to the formation of equilibrium phases, such as T2 or T5 [28]. The subsequent endothermic dissolution region starting at 270 °C and ending in peak E is correlated with the dissolution of the prior formed T1 precipitates, secondary phases and equilibrium phases. Comparing the both alloys, it can be seen that peak A and B are more pronounced for AA 2196 indicating a higher volume fraction of low temperature precipitates present in the T8 temper. Lastly, the final T1 dissolution is shifted to higher temperature in AA 2196 than in AA 2060, which leads to the assumption that T1 precipitates in AA 2196 develop to a coarser extent, reaching a more stable condition and thus lowering the Gibbs-Thomson effect, which was also seen for higher Cu/Li ratio in Al–Cu–Li alloys [7]. The dissolution temperatures are in agreement with the DSC results found for identical phases summarized by [29].

In Fig. 8a, the precipitate transformation in the SZ is shown. In comparison with the BM, the two alloys behave differently. AA 2196 shows an increase in peaks A and C, while peak B is reduced. AA 2060 shows an increased peak B and C. Peaks in region D disappear for both alloys and peak E develops very similar. The SZ specimen experienced peak temperatures of around 480 °C during the welding process, which lead to severe dissolution reactions of the strengthening phases. Geuser et al. [30] found full dissolution of T1 phases when temperatures exceeded 450 °C in AA 2050. The time-temperature-precipitation diagram for AA 2195 shows full dissolution of all phases above 500 °C [31]. During post-weld, natural aging low-temperature phases have formed, which in return dissolve in the DSC measurement giving rise to peak A and B. Peak A represents the dissolution of GP zones, clusters and fine δ' formed during post-weld natural aging and is pronounced for AA 2196, while peak B indicates the prior presence of δ' phases. The rise of peak A in AA 2196 is striking and represents the higher natural aging response of AA 2196. This was also reported for the similar Cu/Li ratio alloy AA 2199 [28]. Both alloys show a clear raise of peak C compared to the BM. Because of a higher volume fraction of low-temperature dissolution reactions seen in peak A and B (below 210 °C), elements such as Li and Cu are available for the subsequent phase formation reactions observed in peak C. In this instance, phases known to form at elevated temperatures, such as T1 and δ', form. Secondary reactions related to formation of equilibrium phases seen in the BM state (peak D) do not occur. Several reasons can lead to a suppression of T2 and T5 formation seen in the BM. The SZ is in a recrystallized, deformation-free state, which influences the T1 precipitation (during the DSC heat cycle) and the subsequent transformation to the equilibrium phases. Also, equilibrium phases might have formed during the welding process lowering the formation energy during the DSC measurement. Lastly, the higher formation peak C might overlap the much weaker formation peaks of T2 and T5 phases.

Thermograms of the HAZ location are shown in Fig. 8b. The HAZ experienced medium to high temperatures, which are critical with regards to the mechanical properties, see Fig. 5. In detail, the maximum temperature at the HAZ position
was measured as 396 °C and 408 °C for AA 2060 and AA 2196 respectively. The total exposure to temperatures above 250 °C was around 16 s (short intervals below 250 °C were reported to not modify the microstructure significantly [30]). At this temperature, the T1 precipitates are reported to dissolve only partially. Coarsening effects of T1 were observed via small angle X-ray scattering technique only for highly overaged conditions in AA 2196 and AA 2198 [7].

The alloys show an intermediate state between the SZ and the BM state described above. Compared to the BM, peak B is reduced for AA 2196 and slightly increased for AA 2060 indicating an alternating volume fractions of δ′ phase. The decrease of peak C for both alloys is associated with less phase formation reactions owing to the higher volume fraction of over-aged precipitates. This might be related to a coarsening of the T1 precipitate that was also reported in [13,28]. Peaks in region D, which are present in BM but not observed in SZ, are very weak in the HAZ material. This indicates that T1 precipitates partially survive the welding process in this region but at the same time a high volume fraction of equilibrium phases have already been formed during the welding process in the HAZ location (overaging).

As a complementary identification technique of the present phases, the identical locations were subjected to HEXRD analysis. The diffractograms are given in Fig. 9. The major peaks at 2θ positions of 3.04°, 3.51°, 4.96°, 5.82° and 6.08° were indexed as aluminum matrix. No significant shift between the alloys and the welding zones was apparent. Traces of T1 phases are indexed in all welding zones, but certain orientations do diminish from initial BM to welded SZ as seen at 3.8° 2θ angle in both alloys. A reduction of the T1 volume fraction was also observed in the DSC analysis. The highest volume fraction is seen in the BM samples and T1 precipitates detected in SZ and HAZ did survive the welding process or were re-precipitated during the cooling phase after welding. δ′ and δ′ precipitates are generally difficult to detect because of their small mismatch with the Al-matrix [28] and traces of δ′ could only be observed for AA 2196 BM at 1.7°. Equilibrium phases like δ or δ are seen for both alloys in the SZ and HAZ samples while the BM does not match the equilibrium phase pattern. This is also confirmed by the DSC curves of the BM material by the double peak D. Additional equilibrium phases resulting from the T1 phase develop in both alloys during the welding process and are visible in the HAZ and the SZ (5.9°) post welding. Equilibrium phases like T2 and T₈ and remaining T1 phases were detected in the SZ of both alloys indicating partial dissolution and possible reprecipitation reactions.

In the case of defect-free welds, the thermal cycle is the main factor for a reduction in mechanical properties in precipitation-hardened alloys in peak aged temper. Several studies have correlated the precipitation state with the mechanical properties in precipitation hardening aluminum alloys [15].

In the DSC analysis, the low Cu/Li alloy 2196 showed a larger amount of δ′ phases, while AA 2060 exhibits less δ′ reaction and precipitates forming at low temperatures are also related to clustering. As the Mg content is higher in AA 2060 and clustering of Cu–Mg was also found to be likely in Mg containing Al–Cu–Li alloys [7], Al–Cu and Cu–Mg are likely to form. After welding, the low Cu/Li composition (AA 2196) allows more phases to form during natural aging processes, shown by Fig. 8 in peak A. Higher temperatures were measured near the SZ for AA 2196 during the welding process. As result, a higher fraction of T1 precipitates evolve, leading to a larger degradation of mechanical properties. The higher volume fraction of precipitates formed during natural aging for AA 2196 does not contribute significantly to the overall strength as the SZ hardness is similar in both alloys after welding (SZ location). T1 precipitates are the main strength contributors and were estimated to contribute more than 90% to the overall strength in AA 2195 T8 [32] confirming the low secondary phases effect on the hardness observed in the SZ.

It can be seen from Fig. 5a that the low Cu/Li alloy AA 2196 softens considerably more in the location of the lowest hardness than the high Cu/Li alloy AA 2060. In the region of lowest hardness, located 6 mm from the weld center, the peak temperature was approximately 400 °C (396 °C and 408 °C for AA 2060 and AA 2196, respectively) during the welding process, see Fig. 6. The microstructure analysis shows a transient microstructure where partial dissolution and overaging is known to occur. The region of lowest hardness is thus related to beginning dissolution of the main strengthening phases T1 and overaging toward T2 and T₈ phase for both alloys. Low Cu/Li alloy 2196 indicates a higher T1 dissolution in the HAZ area as the local hardness is 10% lower than in high Cu/Li alloy 2060. This might be related to the higher and longer temperature exposure. Extensive dissolution occurred when temperatures rise above 450 °C. This temperature is experienced by the SZ, compare Fig. 6b, for a short time of 2.5 s.
Kinetic reasons hindering full dissolution during welding and reprecipitation after cooling are assumed to occur and result in precipitates identified in post-weld state via HEXRD. This is an interesting finding, as full dissolution of T1 was found by Geuser et al. at holding time of 5 s in AA 2050 above 400 °C [30].

4. Conclusions

The high Cu/Li ratio alloy 2060 T8 and low Cu/Li ratio alloy 2196 T8 were welded using SS–BT-FSW employing identical process parameters. The resulting macrostructure shows a defect-free, asymmetrical hour-glass shape exhibiting the typical welding zones.

Local mechanical properties showed higher softening for low Cu/Li alloy 2196 in the area of lowest hardness. Tensile tests confirmed these results where low Cu/Li alloy 2196 reached 57% of the base metal yield strength, while high Cu/Li alloy 2060 achieved 68% (70% and 78% of base metal ultimate strength, respectively).

The different Cu/Li ratio led to a different precipitation sequence in the base material. A low Cu/Li ratio favors the precipitation of $\beta'$ and retards the nucleation of T1, while a high Cu/Li ratio is strengthened by clustering, as well as early, fine T1 precipitation.

Temperatures of 480 °C in the SZ led to major dissolution of the T1 precipitate and consequent softening of the material. Traces of T1 and several averaged phases such as T2 and $\beta$ have evolved during the welding process and are present in the SZ.

The thermal impact of the welding process led to coarsening and dissolution reactions in the HAZ. The microstructure leading to the highest degradation of mechanical properties microstructure was found at 6 mm from the weld center experiencing temperatures of approximately 400 °C, where partial dissolution and overaging effects are present. AA 2196 softened 9% more, a higher T1 dissolution due to higher process temperatures is assumed. High Cu/Li alloy AA 2060 suggests higher thermal stability and exhibits a more robust microstructure meaning lower loss of functionality of the strengthening precipitate T1 during the welding process.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgments

This work has been conducted within the framework of a technology transfer (Kö BTFSW) project between European Space Agency (ESA) and Helmholtz-Zentrum Geesthacht (HZG) GmbH.

REFERENCES


