The Origin of Coarse Grain Structure in
Friction-Stir Welded AA5083 after Heat Treatment

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Friction stir welding (FSW) takes place in the solid state, thus providing potential advantages of welds of high strength and ductility with fine microstructures. However, post-FSW heat treatment can produce very coarse grains, potentially reducing mechanical properties. AA5083-H18 sheets were friction-stir butt-welded using three sets of welding parameters representing a wide range of heat input. They were then heat treated for 5 minutes at 738 K (465 °C), producing grain sizes exceeding 100 µm near the top weld surfaces, and proceeding toward the bottom surface to various degrees depending on the welding parameters. Electron backscatter diffraction (EBSD), transmission electron microscopy (TEM), scanning electron microscopy (SEM), optical metallography, Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Vickers hardness testing were used to characterize the regions within welds. The mechanism responsible for forming the large grains was identified as abnormal grain growth (AGG). Based on a simple model of grain growth, the heat treatment is found to be near the critical condition for AGG to occur, with the principal determinants being the post-FSW grain size and fraction of low angle boundaries. Other possible factors (second-phase particle volume fractions and sizes, textures, dislocation content, and Mg solute distribution) were not significantly different in AGG and non-AGG regions. Based on these results,
recommendations for changes in alloy, FSW conditions, and post-FSW heat treatment were identified to eliminate AGG.
DEDICATION

This is dedicated to my beloved parents and Chen Xie.
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CHAPTER 1

1. INTRODUCTION

Friction stir welding (FSW) is a relatively new welding technique, invented in 1991.[1] It is a promising solid-state joining process with broad application in joining similar and dissimilar materials which are difficult or impossible to weld through conventional welding techniques. FSW can produce a fine microstructure with fewer gross defects, lower residual stresses, less distortion, better retained mechanical properties, and better dimensional stability. FSW is also a candidate process for joining sheets of dissimilar thickness or composition to create tailor-welded blanks[2-4] that retain the capacity for enhanced ductility and strength via fine grain structure. Recent research focuses on FSW in joining materials such as magnesium alloy and steel.[5-8] Commercialization of FSW, mainly for aluminum alloys, has occurred in the transportation industry, for applications such as automobiles, railway vehicles, ships, and rockets.[9-13]

Solid-phase joining is achieved by FSW through frictional heating and plastic deformation by the rotation of a specially designed non-consumable tool embedded in the workpiece, Fig. 1.[14] A direct consequence of FSW is the severe microstructural change
in the welded region.\textsuperscript{[15-22]} Tool geometry is critical for the material flow, and therefore influences the deformation. Induced by the rotation of the welding tool, the plastic deformation and the friction between the tool shoulder and the workpiece surface provide localized heating,\textsuperscript{[14]} which renders a local temperature for aluminum FSW as high as 0.6$T_m$-0.8$T_m$.\textsuperscript{[23-25]} Peak temperatures have been correlated to combinations of welding parameters $\nu$ (weld feed rates) and $\omega$ (tool rotation speeds) through “heat input parameter” or “heat index”, typically $\omega^2/\nu$\textsuperscript{[23, 26]} or $\omega/\nu$\textsuperscript{[27]}. Three distinct microstructural zones have been identified in the welded region – weld nugget zone, thermo-mechanically affected zone, and heat-affected zone.\textsuperscript{[14]} The weld nugget zone (delineated approximately by the dashed line in Fig. 1c), or “stirred zone”, refers to a bowl-shaped\textsuperscript{[28, 29]} or elliptical\textsuperscript{[30-32]} region that is highly strained and dynamically recrystallized at elevated temperature. The thermo-mechanically affected zone surrounds the weld nugget zone. It experiences more moderate temperatures and strains, and consequently no recrystallization.\textsuperscript{[33-35]} The heat affected zone surrounding the thermo-mechanically affected zone experiences only thermal disturbance during FSW, with possible coarsening of the microstructure.\textsuperscript{[36, 37]} Because of varying FSW temperature and stabilities of second phase particles throughout the weld region, particles may be coarsened,\textsuperscript{[38]} and fully or partially dissolved during FSW.\textsuperscript{[22, 39, 40]} Dynamic recrystallization and texture development occur in the nugget zone,\textsuperscript{[25, 31, 36, 39, 41-43]} while precipitate dissolution and coarsening occur in all three zones.\textsuperscript{[39, 41, 44]} A fine and equiaxed grain structure is obtained after FSW with grain size ranging from 1 $\mu$m to
Ultrafine-grained microstructures with average grain size smaller than 1 μm have also been obtained using special cooling methods.[15, 45]

Post-FSW heat treatment may be carried out for annealing or aging purposes, it may be necessary for further forming (i.e. using FSW tailor-welded blanks for GM’s Quick Plastic Forming process at elevated temperature[46]), or it may be an incidental effect of, for example, the automotive paint-bake hardening cycle. Grain growth to sizes as large as several millimeters can occur during heat treatment in AA7075,[47] AA6061,[28] AA7010,[30] AA7475,[31] and AA2095[29]. Large-grain structures can degrade the mechanical properties of the welds.[48-51]

The formation of these coarse grain structures appears to be discontinuous[30-32, 52, 53] because an obvious initiation step was observed.[32] In such “initiation” step, several much larger grains formed in a surrounding of stable smaller grains. Subsurface region, which directly contacts the welding tool shoulder, is the one mostly reported to initiate coarse grain structure formation.[32, 52, 54] Coarse grains may also form from the bottom of the nugget,[30, 32, 54] and on the edges of nugget to the WN-TMAZ boundaries.[32, 52] And the regions most susceptible to discontinuous grain coarsening are those of smaller grains and higher fraction of low angle boundaries (LAB, misorientation angle < 15°) in the nugget.[30, 52, 55, 56]

Both primary recrystallization and secondary recrystallization are two possible discontinuous processes leading to the coarse grain structure formation. Similar to a
discontinuous phase transformation process, there are also two characteristic steps in primary recrystallization and secondary recrystallization: initiation and growth.\[^{[57-61]}\]

Borrowing the nomenclature from discontinuous phase transformation, the “initiation” is also called “nucleation”. In this paper, “nucleation” and “nuclei” are used in order to be consistent with previous literatures. But it should be noted on that the term “nucleation” as used here involves no nuclei formation through thermal fluctuation or atom by atom construction. The nuclei in primary recrystallization are either recovered cells or subgrains pre-existing in a deformed microstructure;\[^{[60, 61]}\] those in secondary recrystallization are actually grains present already in initial grain structure.\[^{[62, 63]}\]

The driving force of primary recrystallization is the stored strain energy in the form of dislocation content within grains.\[^{[60]}\] It is associated with the formation of new strain-free grains in a deformed matrix, characteristic of the evolution of their boundaries from LAB to high angle boundaries (HAB, misorientation angle > 15°) and the growth of these new set of grains. The formation of coarse grain structure by primary recrystallization is a result of the competing of nucleation rate (\(N\)) and nuclei growth rate (\(G\)). A smaller ratio of \(N/G\) leads to fewer nuclei and therefore a coarser final grain structure after these nuclei grow to impingement.\[^{[59, 64, 65]}\]

The interfacial energy of grain boundaries is the driving force for secondary recrystallization, which takes place after primary recrystallization with normal grain growth inhibited, except for a few favored grains.\[^{[62, 66]}\] In order to distinguish it from
normal grain growth (NGG), it is also commonly called abnormal grain growth (AGG) or discontinuous grain growth.\cite{62, 63, 67} For simplification and clear presentation, the terminology of AGG instead of secondary recrystallization is used here.

AGG can lead to grain size of hundreds of microns or even several millimeters. Very large grains grow more slowly than smaller ones in an ‘ideal grain assembly’ where the energies and mobilities of all boundaries are equal.\cite{62} Only when normal grain growth is inhibited is AGG possible. Therefore, second phase particles, texture, surface effects, and solute segregation which lead to diverse driving forces and mobilities of local boundaries, are important factors leading to AGG.\cite{48-51, 62} Various models of AGG have been proposed, emphasizing the effects of the second phase particles on pinning the grain boundary movement.\cite{48-51, 68}

F. J. Humphreys proposed a model for recrystallization and grain growth in cellular microstructures.\cite{69} It considers a cellular microstructure of a mean grain radius $\bar{R}$, uniform intrinsic grain boundary energy and mobility, and a volume fraction ($f$) of spherical particles of diameter $d$.\cite{69} A particle pinning parameter $\Psi$, is defined as follows:

$$\Psi = 3\bar{R}f / d$$ \hspace{1cm} Eqn. 1

The behaviors of “particular grains” are considered, and they are specified by their size ratios. A size ratio, $X$, is defined as $X = R / \bar{R}$, where $R$ is the radius of a “particular” grain and $\bar{R}$ is the average grain radius.
Microstructural stability is then predicted as shown in Fig. 2. The region bounded by $X_{\text{min}}$ and $X_{\text{max}}$ shows the conditions for the occurrence of AGG. This means that if there is a grain of radius $X^* \bar{R}$ in a matrix of pinning parameter $\Psi$, with $X$ and $\Psi$ plotted within AGG region, this grain will grow abnormally until it reach a size ratio of $X_{\text{max}}$ or impinges on other abnormally growing grains. If $\Psi$ is larger than 0.25 (grey/dashed line in Fig. 2), normal grain growth will be halted, leading to a limiting grain size of $\bar{R}_{\text{lim}} = d / (12 f)$, consistent with other theoretical studies and experimental works.\[^{69-72}\]

However, for particular grains with larger size ratio, additional amount of particles content is needed for inhibiting their growth. Therefore with the increase of pinning parameter $\Psi$, the required size ratio for AGG is also increased, Fig. 2. When $\Psi > 1$ (orange/dashed line in Fig. 2), the particle pinning is so strong that even an infinitely large grain cannot grow any more.

This microstructural stability plot is more easily to be understood if it is divided into two parts: 1) NGG allowed when $\Psi < 0.25$ and 2) NGG inhibited when $\Psi > 0.25$. For NGG allowed part, AGG is also possible when $0.25 > \Psi > 0.1$. If $0.1 > \Psi > 0$, the maximum size ratio $X_{\text{max}}$ is 5, too small to be considered as AGG, and it is actually the broadening of grain distribution. For $0.25 > \Psi > 0$ as a total, $\bar{R}$ can increase as a result of NGG to a level that makes pinning parameter enter into NGG inhibition part. If $\Psi = 0$, there is no particle dispersion, only NGG is operative. For NGG inhibited part, AGG is able to occur when $1 > \Psi > 0.25$ as long as there are grains having size ratio larger than $X_{\text{min}}$. The maximum
sized grain is normally found to be $2.5 \bar{R}$ in a grain structure. More grains can be found with a size smaller than $2.5 \bar{R}$. Corresponding to $X = 2.5$, pinning parameter $\Psi$ equals to 0.6. Therefore AGG is easy in a microstructure with $\Psi$ smaller than 0.6, and get more difficult with the increase of $\Psi$. For $\Psi \geq 1$, no growth will be allowed even for an infinitely large grain.

The determining effect of $\Psi$ can be separated into mean grain size $\bar{R}$ and particle dispersion level $f/d$, as shown in Fig. 3. Lines of various colors in this plot represent different values of pinning condition $\Psi$. For NGG inhibited microstructure with $\Psi > 0.25$, this condition should sit somewhere upper right of the grey line in the plot. As discussed above, nuclei-like grains operate when $0.6 > \Psi > 0.25$, that is the region between grey and blue lines. On the other hand, in a grain structure with $1 > \Psi > 0.6$, AGG nucleation is not likely to occur, but if there is a sufficiently large grain available, this large grain can grow abnormally.

Because of the crucial roles of particles in pinning grain boundary, knowledge of important second-phase particles in FSW AA5083, the material under study, is necessary. In particular, the following particles have been identified in the literature\cite{38, 73-80}:

$(\text{Fe,Mn})\text{Al}_6$, $\text{Al}_2\text{O}_3$, $\text{Mg}_2\text{Si}$, $\text{(Fe,Cr)}\text{Al}_7$, $\text{Mg}_5\text{Al}_8$, $\text{Al}_{12}(\text{Fe,Mn})_3\text{Si}$, and $\text{Al}_{11}\text{Cr}_2$. These particles may be segregated into four categories based on their likely effectiveness as grain boundary pins:

1. Fine and stable particles: dispersoid (Fe,Mn)Al₆ and Al₂O₃. Dispersoid (Fe,Mn)Al₆ particles (< 0.4 µm) are known to be semi-coherent and effective in the pinning of the grain boundary[76, 78, 81] and are susceptible to coarsening above 500 °C.[38, 76] They have a plate-like appearance that has been associated with the development of pancake-shaped grains.[76] Al₂O₃ particles have been identified in FSW weld nugget zones[79, 80], likely arising from the breakup and entrainment of the surface oxide. The melting point of Al₂O₃ is 2327 K (2054 °C),[82] thus these particles are very stable during the post weld heat treatment (465 °C).

2. Coarse, incoherent, constituent particles: Mg₂Si, large (Fe,Mn)Al₆, (Fe,Cr)Al₇. These particles are formed during solidification, with non-uniform distribution.[73] Mg₂Si is thermally stable, with a melting point of 1360 K (1087 °C).[83] The melting temperature of CrAl₇ is 790 °C (1063 K),[84] much higher than the melting temperature of AA5083 (574 °C, or 847 K[85]). Other than (Fe,Mn)Al₆ dispersoids, (Fe,Mn)Al₆ can be particles of several microns.[38] These particles have been reported to be unimportant to grain boundary migration.[74] However, it has also been argued that such particles may be attractive obstacles to the migration of grain boundaries by creating holes in the grain boundaries.[86]

3. Thermally unstable particles: Mg₅Al₈. Mg₅Al₈ is not heat resistant.[83] an aging temperature of 623 K (350 °C) eliminated Mg₅Al₈[87] while it precipitated out during deformation at 608 K (335 °C).[88] The solvus temperature of Mg is 573 K (300 °C). Therefore, it is expected that Mg₅Al₈ is unstable during typical post-
weld heat-treatments at 738 K (465 °C), and thus should have little effect on grain boundary pinning, above approximately 300 °C – 350 °C.

4. Rarely reported particles: Al$_{12}$(Fe,Mn)$_3$Si and Al$_{11}$Cr$_2$. These particles have occasionally but rarely been reported in AA5083, with Al$_{12}$(Fe,Mn)$_3$Si identified as constituents and Al$_{11}$Cr$_2$ identified as dispersoids. Because crystallographic data needed to index these phases through OIM are not available in literature and they cannot be identified unequivocally, they have been ignored in the current EBSD study. However, with their larger Z number than matrix Al, they will be imaged via SEM in backscatter (BSE) mode.

Fig. 4 and Fig. 5* illustrate the role of FSW conditions and heat treatment time on the occurrence of coarse grain structure followed heat treatment. Coarse grain structure is favored by lower FSW temperatures, as predicted by lower heat input or lower heat index, Fig. 4. (As described later, the “heat input” and peak temperatures encountered during FSW increase with higher rotational speed, $\omega$, and lower translational speed, $v$.) For the extreme low-heat-input case shown, i.e. $\omega = 1000$ rpm and $v = 300$ mm/min, coarse grain structure occurs throughout the weld nugget region. For the extreme high-heat case, $\omega = 1500$ rpm and $v = 150$ mm/min, coarse grain structure only takes place on the top surface of the weld nugget. Coarse grain structure is also favored by longer heat treatment times, Fig. 5. It starts from the top region and proceeds

* Fig. 4 and Fig. 5 are taken from reference 89 and used with permission of the Journal of Engineering Materials and Technology - Transactions of the ASME.
downward as the heat-treatment time (at 465 °C) increases. Fig. 4 and Fig. 5 suggest that the top part of the weld nugget has a microstructure which promotes coarse grain structure relative to other areas. No coarse grain structure formation occurs outside of the weld region, even for extended heat treatment times, consistent with the literature for AA7075. This is made apparent by comparing the weld nugget zone in Fig. 1c with the extended heat-treatment regions of coarse grain structure (Fig. 5, Views (7) and (8)).

Dynamic recrystallization dominates the initial fine grain formation during the FSW process in the weld nuggets, producing very fine grains, ranging from 0.1 µm to 0.8 µm. These grains are typically smaller than the original subgrains (about 1 µm), tending to confirm the importance of dynamic recrystallization near the tool pin as established by high energy storage, and large strain and strain gradients, complex strain paths.

Hardness and the yield stress are dependent on the grain size in a polycrystalline metal. An empirical Hall-Petch equation or relationship is found to well relate hardness (H) to grain size (d) in the form

\[ H = H_0 + k_H d^{-1/2} \]  

Eqn. 2

where \( k_H \) and \( H_0 \) are constants, and \( d \) is grain size in the form of average diameter. \( k_H \) is the slope of the plotted straight line of the hardness against the reciprocal of the square root of the grain size \( d \). \( H_0 \) is the hypothetical hardness for grain boundary free metals.

The yield stress (\( \sigma \)) can be related to grain size in a similar form
\[ \sigma = \sigma_0 + k_y d^{-1/2} \]  
Eqn. 3

where \( k_y \) and \( \sigma_0 \) have similar meaning with \( k_H \) and \( H_0 \) but in term of yield stress instead.

Effects of dislocation density and solute content have been found important on the Hall-Petch relationship. \( k_y \) increases with the increase of the dislocation density.\textsuperscript{[38]} In Al-Mg alloy, the increase of Mg content leads to the increase of \( k_y \).\textsuperscript{[92]} The study of Hall-Petch relationship on FSW AA5083 showed that the particle dispersion of the materials renders the hardness mostly dependent on the dislocation pinning by particles and Hall-Petch relationship ineffective.\textsuperscript{[38]}

In order to assess the mechanisms for coarse grain structure formation during heat-treatment of aluminum alloys, aluminum alloy 5083-H18 was friction stir butt-welded using three sets of weld parameters representing a wide range of heat input. Following analysis of the as-welded microstructures, samples were heat treated for 5 minutes at 738 K (465 °C). The material and heat-treat cycle represent those used in elevated temperature forming in the GM Quick Plastic Forming process\textsuperscript{[46]}, a target application. As-welded samples were also heat-treated at other temperatures for various length of time in order to study either the effect of recovery annealing on the coarse grain formation. Aluminum alloy 5083 is also of interest for superplastic forming in transportation industry\textsuperscript{[93-100]}, where rapid grain growth in FSW regions could be detrimental to final service properties.
More background information on the friction stir welding including its concept, heat input, and microstructural establishment, observed annealing behavior of FSW structures, and mechanisms for coarse grain structure are presented in Appendix A.
CHAPTER 2

2. EXPERIMENTAL PROCEDURES

2.1. Materials

Commercial AA5083 is a non-age hardenable aluminum alloy with additions of Mg for solid solution strengthening. Its chemical composition (in weight percent) is 0.40 % max Si, 0.40 % max Fe, 0.10 % max Cu, 0.40 % to 1.0 % Mn, 4.0 % to 4.9 % Mg, 0.05 % to 0.25 % Cr, 0.25 % max Zn, 0.15 % max Ti, and remaining Al.\cite{85} It was obtained in the fully strain-hardened temper (H18), with thickness of from 1.57 mm to 1.63mm. Second phase particles are described in Chapter 1.

2.2. Friction Stir Welding Procedures

AA5083-H18 sheets were welded by Hitachi using a 3D-FSW machine with welding tool made of matrix high speed tool steel.\cite{101} The diameters of the shoulder and the pin are 10 mm and 4 mm respectively. The pin is threaded for increasing the stir effect and
lowering the welding force. The pin length is 0.3 mm less than the sheet thickness and the tool shoulder penetrated approximately 0.2 mm into the workpieces during welding. All the welding directions were aligned with the rolling direction (RD). The as-welded plate with various directions is schematically shown in Fig. 1c.

Table 1. Welding parameters and associated thermal characterization

<table>
<thead>
<tr>
<th>Label</th>
<th>H1</th>
<th>H2</th>
<th>H3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tool rotation speed, ω – rps, rev/s</td>
<td>16.7</td>
<td>16.7</td>
<td>25.0</td>
</tr>
<tr>
<td>Translational rate, v – mm/s</td>
<td>5.0</td>
<td>1.7</td>
<td>2.5</td>
</tr>
<tr>
<td>Heat Index 1, ω²/v² [23, 26] – (rps)²/(mm/s)</td>
<td>56</td>
<td>163</td>
<td>250</td>
</tr>
<tr>
<td>Heat Index 2, ω/v [27] – rps/(mm/s)</td>
<td>3.3</td>
<td>9.8</td>
<td>10.0</td>
</tr>
<tr>
<td>Peak Temp, AS, RS [89] – °C</td>
<td>459, 400</td>
<td>522, 476</td>
<td>526, 500</td>
</tr>
<tr>
<td>Avg. Peak Temp [89] – °C</td>
<td>430</td>
<td>499</td>
<td>513</td>
</tr>
<tr>
<td>Time Above 300 °C (Avg. AS, RS) [102] – s</td>
<td>2.6</td>
<td>10.8</td>
<td>6.5</td>
</tr>
<tr>
<td>Time Above 400 °C (Avg. AS, RS) [102] – s</td>
<td>0.7</td>
<td>5.6</td>
<td>4.1</td>
</tr>
</tbody>
</table>

Key: AS, RS = Advancing side, Retreating side

As shown in Table 1, the three weld conditions are labeled H1, H2, and H3 in order of progressively higher temperatures measured during their welding (and corresponding to the proposed indices). Three combinations of weld translational rates (v) and tool rotation speeds (ω) were utilized to produce welds characterized by a range of “heat inputs” or “heat indexes”. [23, 26, 103] Two such indexes have been proposed, as used in Table 1:

Heat Index 1: [23, 26] \( \frac{\omega^2}{v} \)  
Eqn. 4

Heat Index 2: [27] \( \frac{\omega}{v} \)  
Eqn. 5
These heat indexes have been correlated phenomenologically with observed temperatures and times-at-temperature during FSW. For a given translational speed, a higher tool rotation speed produces more stirring in a spatial region, thus producing higher temperatures. Conversely, for a given rotational speed, a slower travel will produce more heat at a given location.

Table 1 compares measured peak temperatures and the residence times above 300 °C and 400 °C 2 mm from the weld centerline (i.e. at the pin-workpiece interface) and 0.6 mm below the initial top surface of the 1.64 mm sheet. The methodology by which these measurements were made has been described elsewhere. Note that measured times-above-temperature are longer for H2 than for H3 although the peak temperatures are slightly lower. This seems to be reflected better in Heat Index 2, which is nearly equal for the two cases. Simulations predict that the temperatures during FSW near the top surface are higher than in the middle by 7 °C (H1), 19 °C (H2), and 29 °C (H3).

2.3. Post-weld Heat Treatment

Test coupons (10 mm x 25 mm) were sectioned from the welded sheets of H1, H2, and H3 conditions with weld region sitting in the longitudinal center. Coupons were then heat treated for 5 minutes at 738 K (465 °C), and is denoted here as QPF-HT because it represents that used in elevated temperature forming in the GM Quick Plastic Forming
(QPF) process, a target application. Additional test coupons were cut from H2 condition to study alternate heat treatments.

Full annealing of AA5083 (solidus temperature is 847 K, or 574 °C) to achieve the “O” condition is typically done at 618 K (345 °C) for a time the whole sample reaching 345 °C. The coupons of base H18 material were then annealed at 618 K (345 °C) for 1 hr and 380 °C for 1 hr to compare with the QPF-HT.

During heat treatment, an aluminum sheet was preheated in the furnace and coupons were placed on the sheet to promote rapid heat up. The heat treat times were started when the furnace regained the set temperature after placement of the coupons.

2.4. Hardness Testing

As-welded and heat-treated coupons were mounted using Epoxy or conductive Bakelite. Hardness testing was conducted using a Buehler Micromet II digital micro hardness tester along the centerline of the weld from top surface to bottom on the LT-ST plane (see Fig. 1) on the mechanically polished samples, with a load of 50 g force and dwell time of 15 seconds. At least five Vickers microhardness test values were recorded at each distance from the top surface near the centerline and were averaged after excluding the maximum and minimum values in order to minimize the effect of local conditions, such as locating
the indentation on a cluster of particles. Along the thickness, ten positions were measured for Vickers hardness. Therefore, a minimum of 50 hardness measurements were made on each welded sample. For both the as-received and fully annealed base metal, hardness test is similarly done on the sectioned LT-ST plane. All the measurements were then averaged to a single hardness value. More details on the hardness test are in Appendix B.

2.5. Optical Microscopy

Macro- and micro-structural characterization was done on the cross sectioned plane (LT-ST) unless otherwise noted. All samples were successively ground using silicon carbide grinding paper and then polished using diamond compounds (final grit: 1 μm) for optical microscopy (OM). Samples were etched with a solution of 12 mL HCl (conc), 16 mL HNO₃ (conc), 17 mL H₂O, 1 mL HF (48 %), and 4.8 g chromic acid on sample surface for 1 to 4 min.⁷⁰⁵

2.6. Electron Backscatter Diffraction (EBSD)

Samples for electron backscattered diffraction (EBSD) analysis were polished as for optical microscopy and then using 0.05 μm colloidal silica in a vibratory polisher. EBSD
scans were conducted using TSL OIM Data Collection Program on a Philips XL-30 Environmental SEM (ESEM) with field emission gun (FEG), which is equipped with a CCD camera. SEM was operated at 25 kV. Low angle boundary measurement was found to be dependent on the settings of EBSD Collection, especially on step size, the binning of Kikuchi band images, and the minimum cutoff misorientation value. According to the experimental results (see Appendix C), the step size for most of the scans is chosen to be 0.25 µm, the binning for the Kikuchi pattern image to be 4 x 4, and the minimum cutoff value is 3°. Low-angle boundaries (LAB) in this paper are defined as having misorientation angles from 3° to 15°. For most scans the scan area is 50 by 50 µm². Hough peaks of scanned points were collected first. Using the known crystallographic structures of Al, (Fe,Mn)Al₆, Mg₂Si, Mg₅Al₈, and (Fe,Cr)Al₇, and Al₂O₃ phases, the initially collected Hough-peak data were used to generate orientation data for identifying these phases.

The orientation data were analyzed using TSL OIM Analysis Program, with a 5-degree grain tolerance angle, that is, 5 degree is the minimum misorientation for defining the grain boundary. The grain size statistics are generated directly by the TSL OIM Analysis Program. The accuracy and reproducibility of grain sizes were established by performing 3 scans at adjacent regions in specimens of H1-Mid and H3-Mid, and calculating standard deviations of the measured values. More detail in grain size measurement and its scatter determination is in Appendix D.
In grain growth model, mean grain diameter/radius is used. Therefore, grain size in this paper is represented by grain diameter. With the assumption that all grains are spherical, mean 2D grain radiuses, or Equivalent Circle Diameter (ECD) for characterization locations were then calculated from EBSD measured average grain area ($\overline{A}$):[106]

$$ECD = 2 \times (\overline{A} / \pi)^{1/2}$$

Eqn. 6

3D grain diameter ($d$) were converted from ECDs thereafter based on Stereology:[106]

$$d = (2/3)^{-1/2} \times ECD = 1.225ECD$$

Eqn. 7

2.7. Scanning Electron Microscopy (SEM)

Samples for scanning electron microscope (SEM) investigation were prepared as for EBSD. A Philips XL-30 Environmental SEM (with field emission gun) was operated at 15 kV for imaging and an energy dispersive X-ray spectroscopy (EDS) was used for chemical analysis.

A Sirion high-resolution FEG-SEM was operated at 5 kV under backscattered electron (BSE) mode for second phase particle (larger than 0.2 µm) analysis. All photomicrographs were taken under a constant magnification of 1500X, and approximately a total of 3000 particles were analyzed. The resolution of these photomicrographs was such adjusted to make a single pixel size to be 0.016 µm. A low electron voltage of 5 kV was used to reduce the information depth. Compared with the
BSE information depth of about 400 nm at 10 kV, it is only about 120 nm for a 5 kV beam.\cite{107} A large information depth will bias the SEM analysis away from the 2-D condition, resulted in biased results of particle sizes and volume fractions which are calculated based on stereologic theories relating exact 2D and 3D information.\cite{108} More details for SEM setting determination can be found in Appendix C.

2.8. Transmission Electron Microscopy (TEM)

Second phase particles smaller than 0.2 µm were imaged using TEM. Thin-foil disk specimens from various weld locations were mechanically thinned to about 120 µm and then electro-polished using a solution mixture of 70% methanol and 30% nitric acid at about 12 °C. A Philips CM-12 transmission electron microscope (TEM) was used, operating at 120 kV. Convergent Beam Electron Diffraction (CBED) was used for sample thickness estimation. More detail is in Appendix E.

2.9. Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

A Perkin-Elmer Sciex ELAN 6000 Inductively Coupled Plasma Mass Spectrometer (ICP-MS) with laser ablation was used to measure magnesium content. Measurements were made for the H2 and base H18 condition. Samples were cross-sectioned, and measurements were made on ST-LT plane, Fig. 1c. The laser beam was scanned from
top to bottom along the centerline of the H2 weld, and top-to-bottom for the base H18. Materials scanned by laser was ablated, and turned into aerosols. These aerosols were then transported into Argon plasma and ionized under high temperature (about 1000 °C). A mass analyzer was then able to separate ions and quantify the amount of each kind according to their mass-to-charge ratios.
CHAPTER 3

3. EXPERIMENTAL RESULTS

3.1. Grain Growth during Heat Treatment

A typical optical FSW macrostructure, Fig. 1c, shows a basin-shaped dynamically recrystallized weld nugget zone. After heat treatment at 465 °C for 5 min (QPF-HT), etching reveals the extent of coarse grained area, depending on weld parameters, Fig. 6. The lower the heat input is, the more extensive the coarse grained area, spreading from the upper surface. Microstructure analyses focus on the top and middle regions of these three samples to differentiate the conditions favoring coarse grain structure. The centers of all the 50 by 50 µm² scans for top regions are approximately 50 µm below the sample top surface to avoid near-surface effects.

The H2 condition was chosen to further explore the annealing behavior of FSW welds because 1) only part of the weld experienced coarse grain formation during QPF-HT, 2)
this welding condition was determined to be optimal for mechanical properties,[89] and 3) this condition was used to generate Fig. 5[89].

As shown in Fig. 7, coarse grains begin to appear at the top surface at 420 °C for 5-minute annealing. As the annealing temperature increases, the coarse grained area extends downward progressively. For 510 °C and above, the whole weld region becomes coarse grained. A closer examination of the 510 °C, 540 °C, and 560 °C annealing cases shows that the final grain size in the center of the sheet decreases as the annealing temperature increases. Also, elongated coarse grains begin to extend into the base material at these temperatures. This observation differs from an earlier report that coarse grains cannot extend outside of FSW welds into the base metal.[32]

In order to see the grain coarsening behavior and its relationship to FSW process, a plan-view of the H2 weld after QPF-HT is shown in Fig. 8a. A convolution pattern is clearly seen corresponding to the rotation of the welding tool, with the pattern concave in the welding direction. A previous study on the onion ring reported a similar pattern.[35] Particle-rich (dark band) and particle-poor (bright band) regions follow a similar convolution pattern, Fig. 8b. This similarity suggests the importance of particle pinning in the coarse grain formation.
3.2. Hardness Testing

The hardness of H18 is 132 ± 2 Hv. After annealing at 345 °C and 380 °C for 1 hr, the hardness 69 ± 2 Hv and 67 ± 3 Hv respectively. (The error for each Hv measurement is the standard deviation of at least three measurements in closely adjacent regions.) Published hardness of the O condition is approximately 67 Hv.\[38]\]

Plots of hardness values along the weld centerlines of as-welded conditions and after QPF-HT are compared with base H18 and O materials in Fig. 9. There is little variation of hardness laterally (standard deviation of 1 Hv) in the weld zone within the central 4 mm of these welds.\[89]\] For all as-welded conditions, the top region is harder than the middle region by 2-3 Hv. This result contrasts with those for a heat-treatable alloy (AA7050-T7451) where the top region is softer.\[34]\]

After QPF-HT the whole region of FSW-H1 and the upper regions of FSW-H2 and FSW-H3 have coarse grains and become softer by 6-8 Hv while the lower regions of FSW-H2 and FSW-H3 have the same hardness, within 2 Hv. For the H2 case, the hardness readings in the grain coarsening regions were reduced from 75 ± 2 Hv to 69 ± 3 Hv while the non-coarsening regions were 72 ± 1 Hv and 71 ± 1 Hv before and after QPF-HT. QPF-HT has little or no effect on hardness except in the regions where the grain coarsening occurs.
Hv maps for various locations and Hv averages and scatters for each depth for all welding conditions before and after various heat treatments are available in Appendix B.

3.3. Recovery Annealing

A series of two-stage heat treatments was conducted to probe the nature of the driving force for the formation of the coarse grains. Recovery anneals of FSW-H2 material were performed over a range of times and temperatures as follows: 80°C/1hr, 150°C/1hr, 200°C/1hr, 250°C/1hr, 300°C/1hr, 350°C/1hr, 350°C/24hr, 370°C/24hr, and 380°C/24hr. A maximum temperature of 380 °C was used because at this temperature large grains were observed forming at the edge of weld nugget, Fig. 10, as circled in View (3). Micrographs were prepared and hardness values were recorded following Stage 1 annealing and then QPF-HT was performed as Stage 2, with subsequent micrographs prepared. Fig. 10 shows selected micrographs before and after Stage 2 heat treatment. There is no discernible effect of the Stage 1 recovery heat treatment on the formation of coarse grains in the Stage 2 QPF-HT.

After QPF-HT, approximately top 1/3 region in the weld nugget of H2 showed coarse grains but no obvious microstructural change in the remaining part. Therefore, TOP-1/3 is used here to denote the region of graining coarsening; while BOT-2/3 for non-coarsening region. The hardness values within each region are averaged to reveal with
less scatter the difference between these two regions with very different annealing behavior. (The measurements closest to the bottom surface were excluded because they appeared to differ sharply from the rest of the lower region.)

As shown in Fig. 11 and Fig. 12, the hardness of TOP-1/3 drops up to 5 Hv, and getting greater with either a higher temperature for a constant annealing duration or a longer time at a constant temperature. On the other hand, the hardness drop of BOT-2/3 is 1-2 Hv. After 380 °C/24hr annealing, the relatively larger drop of the TOP-1/3 hardness led to a similar hardness on TOP-1/3 and BOT-2/3, 71 ± 1 Hv and 70 ± 1 Hv respectively. 380 °C is already a much higher temperature than the recommended recrystallization temperature (345 °C) for this material.[104] At the higher temperatures and time, full recovery is expected for this alloy.

Consistent with the OM results in Fig. 10, hardness variations of both TOP-1/3 and BOT-2/3 are similar no matter the sample experienced recovery anneal or not, with the TOP-1/3 dropped to about 68 Hv from 75 ± 1 Hv of as welded sample and the BOT-2/3 constant (± 1 Hv) at 71 Hv, Table 2. It is therefore concluded that strain energy stored in the form of dislocations (which are removed by recovery anneals) is not the driving force for the formation of the coarse grains in FSW AA 5083.
Table 2. Hv of FSW-H2 after recovery anneal and QPF-HT according to the recovery anneal conditions

<table>
<thead>
<tr>
<th>No Recovery +QPF-HT</th>
<th>350 °C/1hr +QPF-HT</th>
<th>380 °C/24hr +QPF-HT</th>
</tr>
</thead>
<tbody>
<tr>
<td>TOP-1/3</td>
<td>69 ± 3</td>
<td>67 ± 2</td>
</tr>
<tr>
<td>BOT-2/3</td>
<td>71 ± 2</td>
<td>72 ± 2</td>
</tr>
</tbody>
</table>

3.4. Results from EBSD Analysis

The microstructures of the as-welded specimens are first presented, then those after QPF-HT (465 °C for 5 min), and those after various recovery anneals at the end.

A. As-welded grain structures

An EBSD phase-contrast map, Fig. 13, reveals the grain structure of as-received H18 base material, pancake-shaped of widely varying grain size. Intercepts give $d_1 = 3.0 \, \mu m$ (LT direction) and $d_2 = 1.3 \, \mu m$ (ST direction). Given that grains in rolled sheets are usually larger in the L direction than in LT, the average grains intercept may be approximated as 3 \, \mu m (i.e. average of 3, 1, 5 \, \mu m). Fig. 13 also shows the second-phase particles for which crystallographic structures are known.

Fig. 14 is similar to Fig. 13, but compares weld locations shown in Fig. 6. Recrystallization is made apparent by comparing with Fig. 13. Grains are smaller near
the top of the welds, as compared with the middle. The average grain size, \( d \), determined by the mean equivalent 3D diameter, appear on each figure with uncertainty of \( \pm (0.02-0.04) \times d \), that is, within 4% of the measurement\(^*\). All locations that exhibit coarse grains after QPF-HT (Fig. 14a-d) had as-welded grain sizes less than 3 \( \mu m \) while all those locations that do not show coarse grains (Fig. 14e and f) had as-welded grain sizes greater than 3 \( \mu m \).

Larger grains correspond to higher heat input (i.e. higher peak temperatures or longer times-at-temperature during welding). This can be seen by comparing H1 - H3 (increasing order of heat index) for either upper or middle locations, Fig. 14. The middle regions (lower strains, lower peak temperature) have larger grain sizes than the corresponding upper locations (higher strains, higher peak temperature). This is consistent with the previous observations, i.e. for AA2519\[^{109}\] and AA7010\[^{30}\], where grain sizes decreased from top to bottom of the FSW region. The through-thickness temperature gradients in the current weld are expected much smaller than in those previous cases because the sheet is thinner (1.65 mm here vs. 25.4 mm\[^{109}\] and 6.4 mm\[^{30}\]) and higher rotational speeds were used (1000 and 1500 rpm here vs. 180rpm to 450 rpm\[^{30, 109}\])

This expectation is consistent with FSW thermo-mechanical simulation results for the conditions used here showing less than 30 degrees difference between the top and middle

\(^*\) The uncertainty is determined via three adjacent scans on the H1 middle region and the H3 middle. The standard deviation is calculated for each location. More details on scatter determination are in Appendix D.
regions.\textsuperscript{[102]} Results from these thermo-mechanical simulations appear in the Appendix F. With this relatively uniform temperature profile, the additional deformation near the tool shoulder on the upper region plays an important role in controlling grain size, and therefore grains are smaller there.

The composition of the second phase particles is shown by the color codes on Fig. 13 and Fig. 14. Large particles (in purple) are (Fe,Mn)Al\textsubscript{6}, consistent with reports in the literature.\textsuperscript{[38, 76]} Because sparse, large particles have little affect on AGG,\textsuperscript{[62, 75, 110]} these scan fields were chosen to avoid these large particles, and thus do not show representative numbers of (Fe,Mn)Al\textsubscript{6} particles. Analysis of second phase particles appears in a subsequent section.

B. Grain structures after heat treatment

EBSD characterization was conducted on the top corner region of FSW-H2 to the retreating side after annealing at 380\degree for 24 hr (Fig. 10, View (3)) as mentioned in Section 3.1, where the grain coarsening initiation appears. Those large grains (~ 100 \(\mu m\)) were found to be surrounded by fine grains with a size ratio of about 40. One of those large grains with its surrounding of fine grains is shown in Fig. 15, which is an EBSD grain structure map. Color code indicates the crystal orientation. Therefore an area belonging to a single grain shows a same color.
After QPF-HT, areas of the weld that experienced grain coarsening differ markedly from adjacent areas of non-coarsening, as previously shown in Fig. 4 and Fig. 5. The largest grains after heat treatment at 738 K (465 °C) for 5 min are ~100 µm (Fig. 4a), while grains after elongated heat treatment can be ~500 µm, in Fig. 5, View(8). The border region between coarse grain structure and unchanged fine structure of FSW-H3 after QPF-HT is revealed by EBSD, in Fig. 16 (color code here for the crystal orientation). The boundary between large and small grains is ~ 150 µm below the top surface.

In order to study the change of grain structure for the regions of no coarse grain formation, an EBSD scan on the middle region of the H2 weld after QPF-HT was conducted, shown in Fig. 17 (Fig. 14e is the pre-QPF-HT view). The average grain size is 2.4 µm after heat treatment, compared with 2.3 µm before QPF-HT, the difference lying within experimental scatter.

A region at the top of the weld nugget zone, about 4 to 5 µm thick, has a higher density of second phase particles and irregularly shaped small grains, much smaller than elsewhere (d ≈ 0.3 µm), Fig. 18. The contrast with the adjacent region with coarse grains (d ~100 µm), is marked: a difference of 2.5 orders of magnitude of grain size. Although the peak temperature is likely highest in this surface nearest the tool shoulder, it would also be expected to cool the most rapidly after the passing of the tool and have experienced highest amount of strain. Most importantly, the high area fraction of second phase particles presumably inhibits grain growth. The concentration of second phase particles is likely related to segregation of Mg and Si by the severe deformation, as well as
entrained weld debris such as surface oxides. Additional studies on this region using SEM will be shown later.

The entire weld region of FSW-H1 forms coarse grains during QPF-HT, with or without a recovery anneal before QPF-HT (Fig. 6a). EBSD study on these weld nuggets after QPF-HT showed three regions of very different grain sizes in the through-thickness direction, as shown in Fig. 19 (color code for the crystal orientation). The grain structure in Fig. 19 was formed by recovery anneal at 370 °C for 7 days as the Stage 1 and followed by QPF-HT as the Stage 2. The three regions can be distinguished as: 1) the small grains at the top ($d \sim 20 \mu m$, up to a distance of 30 µm from the top surface), 2) medium grains ($d \sim 60 \mu m$), 30 µm - 100 µm below the surface, and 3) large grains ($d \sim 500 \mu m$) throughout the rest of sheet thickness. Two border lines dividing these three regions are clearly observable. If the ultra-fine grain structure directly beneath the top surface is included, four regions of corresponding to grain size can be discerned.

C. Second phase particles characterized using EBSD

The effects of particle shape and distribution are important factors in recrystallization and grain growth via changing the boundary mobility\cite{110}, with the largest influence from high densities of particles of size smaller than 1.0 µm.\cite{62, 75, 87} Second phases can be resolved by EBSD phase imaging, e.g. Fig. 13, Fig. 14, Fig. 17, and Fig. 18. (Note that the analyzed areas were chosen to avoid the large, sparse constituent particles.) Shown by
these figures, the advantage of this technique is its ability to reveal the phase of each particle, and specify the location and estimate the amount of each second phase. The quantitative analysis was done with the data achieved from EBSD and results are collected in Appendix G. However, the resolution of EBSD for aluminum sample using FEG-SEM is about 20 nm along the sample tilt axis, and with a 70° sample tilt needed for EBSD scan, the resolution perpendicular to the tilt axis is about 3 times larger.\[112\] Compared with the size of smallest particles in AA5083 (~ 30 nm), the resolution of EBSD is not enough for particle content quantification. Quantification of second phase particles was conducted using TEM and SEM instead, and shown later.

E. Misorientation and distribution and low angle boundary (LAB) analysis

The kinetics of recrystallization and grain growth has been associated with the character of the grain boundaries involved.\[61,113\] Grain-to-adjacent-grain misorientation angle (θ) distributions over a range of 5° to 65° are shown in Fig. 20 as compared with a random texture\[114\]. Fig. 20a illustrates the differences among the FSW locations, with top locations shown in red/solid lines and middle locations shown in blue/dashed lines. The top region is characterized by more low angle boundaries (LAB) (5° to 15°) than the middle region. Both regions have more LAB than a random distribution,\[114\] as shown. Fig. 20b shows that 5083-O annealed material has misorientations corresponding to a random texture, while 5083-H18 has predominantly low-angle boundaries. Comparison
of Fig. 20a and Fig. 20b suggests that deforming an annealed specimen introduces an
excess of LAB as compared with random, whether the deformation is by rolling or FSW.

Fig. 20c summarizes selected information in Fig. 20a and b. The number fraction of
boundaries for each misorientation is adjusted by subtracting the fraction expected for a
random distribution, thus representing the deviation from random. The annealed 5083-O
is nearly random while the middle FSW regions, top FSW regions, and heavily rolled
5083-H18 progressively exhibit higher proportions of lower-angle boundaries (5° - 20°)
and lower proportions of higher-angle boundaries (30° - 50°). The order is consistent
with the hardness distribution. As shown in Fig. 9, the upper FSW regions are harder
than the middle regions, with both being modestly harder than 5083-O and much softer
than 5083-H18.)

The as-welded grain sizes and fractions of LAB for all six characterization locations are
plotted in Fig. 21. LAB here is defined to be boundaries with misorientation from 3° to
15°. This plot shows a location of smaller as-welded grain size generally has a higher
fraction of LAB. Regions (in red) of smaller grain size and higher LAB% are more
susceptible to coarse grain structure formation.
3.5. Characterization Results from SEM and TEM

EBSD can identify the phases of particles but not accurate for quantifying sizes and volume fractions of particles of roughly the step size used. SEM and TEM were used to quantify these aspects. The ultra-fine grain structure on the subsurface region was further studied by SEM.

A. SEM results

Particles larger than 0.2 μm were measured using SEM on all six as-weld locations plus H2-Top after QPF-HT (465 °C for 5 min). Table 3 summarizes the volume fractions and average spherical equivalent diameters for each location, including the estimated uncertainties. The volume fraction of second phase particles is equal to the area fraction of particles imaged in 2D via SEM. The average spherical diameter was obtained from the average equivalent circle size of all particles by multiplying 0.816, the factor determined stereologically relating 2D circle diameter to 3D spherical diameter. Five adjacent fields were taken for each location. Particle size and volume fraction values for all fields of a location were averaged to obtain the one collected in the table, with standard deviations correspondingly determined.
The total volume fractions of all particles in various locations are all about 1.8%, relatively the same location by location. A difference in particle average size between the top and middle regions exists, even though it is small. Particles in the top regions are 0.04-0.05 µm larger than those in the middle. This can be caused by a higher temperature on the top region during FSW leading to particle coarsening on this region. Larger particle sizes are also found in higher heat-input welds (H2 and H3) than H1, further supporting the effect of welding temperature on the as-weld particle size.

Table 3. Volume fractions and mean spherical diameters for various locations from SEM

<table>
<thead>
<tr>
<th></th>
<th>H1_Top</th>
<th>H1_Mid</th>
<th>H2_Top</th>
<th>H2_Mid</th>
<th>H3_Top</th>
<th>H3_Mid</th>
<th>H2_Top (AH)</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Vol. Fraction</strong></td>
<td>0.015</td>
<td>0.017</td>
<td>0.017</td>
<td>0.020</td>
<td>0.020</td>
<td>0.019</td>
<td>0.021</td>
<td>0.018</td>
</tr>
<tr>
<td>(f)</td>
<td>±0.004</td>
<td>±0.006</td>
<td>±0.002</td>
<td>±0.008</td>
<td>±0.006</td>
<td>±0.004</td>
<td>±0.009</td>
<td>±0.003</td>
</tr>
<tr>
<td><strong>Mean Spherical</strong></td>
<td>0.42</td>
<td>0.38</td>
<td>0.45</td>
<td>0.40</td>
<td>0.43</td>
<td>0.39</td>
<td>0.43</td>
<td>0.41</td>
</tr>
<tr>
<td><strong>Equiv. Diam.</strong></td>
<td>±0.03</td>
<td>±0.03</td>
<td>±0.04</td>
<td>±0.02</td>
<td>±0.05</td>
<td>±0.03</td>
<td>±0.03</td>
<td>±0.03</td>
</tr>
<tr>
<td>(d, µm)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The ultra-fine grain structure region right below the top surface revealed by EBSD observation (Fig. 17b) was further studied by SEM characterization. Crystal orientation contrast under BSE mode is able to reveal grain structure. Fig. 22a was taken from such a region of H3 after QPF-HT. The thickness of this ultra-fine grain structure is about 20 µm. Below this region, it is a part of coarse grain structure (about 150 µm thick for H3 after QPF-HT).
A closer examination revealed that a band of high-density particles showing black (indicating a lower average atomic number than matrix aluminum) sitting between the ultra-fine grain region and coarse grain region. EDS analysis was conducted on these black particles as labeled in Fig. 22a. The largest particle, particle 1, mainly contains Mg and Si, Fig. 22b. Similar to this, black large particles (larger than 2 µm) analyzed on other regions measured to be high Mg- and Si-containing particles as well. So they probably are MgSi$_2$ particles. This result is consistent with the literature that MgSi$_2$ are large constituent particles.\textsuperscript{[73, 74]} Smaller black particles, particle 2 and 3, contain more oxygen and Mg than matrix, Fig. 22c. They are then probably Al$_2$O$_3$ and Mg$_5$Al$_8$.

Additionally, by visual comparison on micrographs in Fig. 23, taken from as-weld H3 under exactly same SEM setting, higher amount of particles can be found within this ultra-fine grain region than other regions. Microstructure from ultra-fine grain region is shown in Fig. 23a. Fig. 23b shows the microstructure of a region 30 µm below (outside of the ultra-fine grain region but in the grain coarsening region). Simple visual examination can tell the grain size is smaller in Fig. 23a than Fig. 23b. Bright particles (higher average Z value than Al) are clearly shown in both micrographs. The size of these particles in Fig. 23a is smaller than Fig. 23b and the amount is larger, consistent with previous EBSD characterization, Fig. 17b. This suggests a stronger particle pinning on the ultra-fine grain region.
B. TEM results

FSW-H2 and FSW-H3 were selected for further characterization using TEM because of the apparently different annealing phenomena during QPF-HT between the top and middle regions. (FSW-H1 showed similar grain sizes after QPF-HT at top and middle sections) TEM micrographs are shown in Fig. 24. Because of the projection nature of TEM imaging and following the general assumption that all particles are spherical, the measured sizes can be taken as 3D sizes. All quantification done with TEM data excludes particles with diameter larger than 200 nm. Particle size and volume fraction values for all fields of a location were averaged to obtain the one collected in Table 4, with standard deviations correspondingly determined.

Within the scatter, the volume fractions are the same between the locations in a single weld and between different welds. The mean particle sizes show no change within scatter as well.

<table>
<thead>
<tr>
<th></th>
<th>H2_Top</th>
<th>H2_Mid</th>
<th>H3_Top</th>
<th>H3_Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vol. Fraction (f)</td>
<td>0.012±0.004</td>
<td>0.012±0.007</td>
<td>0.010±0.007</td>
<td>0.011±0.003</td>
</tr>
<tr>
<td>Mean Spherical</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Equiv. Diam. (d, µm)</td>
<td>0.10±0.01</td>
<td>0.09±0.02</td>
<td>0.10±0.01</td>
<td>0.09±0.03</td>
</tr>
</tbody>
</table>
The low strain energy nature of as-welded regions, determined previously through the hardness testing and recovery annealing, is further supported by TEM characterization on dislocation contents, Fig. 25. Dislocations were found in both top and middle regions of H2 and H3 (Fig. 25a, b, c, and d). However, if their dislocation structures are compared with Base H18’s (Fig. 25a, b, c, and d vs. Fig. 25e), the dislocation densities in FSW regions are much lower. The dislocation density of FSW regions can be estimated to be about $10^{12}$ m$^{-2}$, compared with the documented values of $10^{10}$ to $10^{12}$ m$^{-2}$ for an annealed metal and ~ $10^{16}$ m$^{-2}$ for heavily cold-worked metals.\textsuperscript{[64]}

C. Particle dispersion levels ($f/d$)

Data obtained from TEM and SEM was analyzed based on the theoretical treatment on particle dispersion level shown in the Appendix H.

(1) \textbf{Particles > 200 nm via SEM.} Particle sizes obtained via SEM are 2D ones. The 3D size for each particle is not possible to obtain directly from SEM. A classic way to treat such data is Size Distributions Unfolding.\textsuperscript{[108]} The measured data is presented in histogram form with the bin width $\delta$. In most cases 10 or 15 bins are used and spaced in size up to the largest one. For larger-sized particles, they have probability to be cut to show smaller circles with the probability corresponding to the circle size. Therefore the particle numbers measured for the bins of smaller sizes in SEM particle size distribution have contributions from larger particles. A coefficient matrix should be generated to
convert SEM measured 2-D size distribution to 3-D distribution. However, it can be shown that for the purpose here to obtain $f/d$ this conversion is unnecessary.

Data for H2-M is used for illustration. With the sizes smaller than 200 nm cut off, measured 2D sizes were grouped into 15 bins. The distribution is plotted in Fig. 26. As shown from the plot, most of the particles are in the bin of the smallest size. Similar distributions were also found in other locations. The relatively low frequencies for the larger particles in the size distribution measured by SEM lead to the small contribution to the number in the bins of 250 nm and 750 nm from these larger particles (less than ~ 5%). Effect of such a small change is limited. Therefore, SEM measured particles can be grouped into 15 bins. Assuming that all particles within a single bin have a same particle size, the 3D size of each group was calculated to be $\frac{d}{0.816}$ ($\bar{d}$ is the average 2D equivalent diameter and the factor 0.816 is the ratio of the average diameter from the intersection circles to the actual spherical diameter\textsuperscript{[108]}). Subsequent calculations of $f/d$ can be separated into two methods based on two different equations derived in the Appendix H.

\textit{Method 1: Based on the f/d of each bin}

The total $f/d$, $\frac{f^{Tot}}{d^{Eff}_Z}$, can be obtained from:
\[
\frac{f_{\text{Tot}}}{d_{Z}^{\text{Eff}}} = \frac{f_1}{d_1} + \frac{f_2}{d_2} + \ldots + \frac{f_{15}}{d_{15}}
\]  
Eqn. 8

with adding the \( f/d \) of 15 sets (or bins) of particles. The volume fraction of particles belonging to each bin is obtained from SEM measurement directly, that is, the area fraction of the particles having sizes within the size range of each bin. The \( \frac{f_{\text{Tot}}}{d_{Z}^{\text{Eff}}} \) values of all locations calculated via this method are shown in Table 5.

Table 5. Zener particle size measured by SEM via Method 2 and calculated dispersion level

<table>
<thead>
<tr>
<th></th>
<th>H2-Top</th>
<th>H2-Mid</th>
<th>H3-Top</th>
<th>H3-Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{f_{\text{Tot}}}{d_{Z}^{\text{Tot}}} ) (( \mu \text{m}^{-1} ))</td>
<td>0.025</td>
<td>0.024</td>
<td>0.027</td>
<td>0.028</td>
</tr>
</tbody>
</table>

Method 2: Based on the calculation of effective Zener \( d \)

As shown in the Appendix H, the effective Zener diameter, \( d_{Z}^{\text{Eff}} \), for all particles can be obtained by

\[
d_{Z}^{\text{Eff}} = \sum_{i=1}^{n} f_i / \sum_{i=1}^{n} \frac{f_i}{d_i} = \sum_{i=1}^{n} n_i \times d_i^3 / \sum_{i=1}^{n} n_i \times d_i^2
\]  
Eqn. 9
\( f^{\text{Tot}} \) is the area fraction of all particles with 2D diameter larger than 200 nm. Therefore, \( \frac{f^{\text{Tot}}}{d^{\text{Tot}}} \) is ready to be calculated. The values of all locations are shown in Table 6.

Table 6. Zener particle size measured by SEM via Method 1 and calculated dispersion level

<table>
<thead>
<tr>
<th></th>
<th>H2-Top</th>
<th>H2-Mid</th>
<th>H3-Top</th>
<th>H3-Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>( f^{\text{Tot}} )</td>
<td>0.017±0.002</td>
<td>0.020±0.008</td>
<td>0.020±0.006</td>
<td>0.019±0.004</td>
</tr>
<tr>
<td>( d^{\text{Tot}} ) (µm)</td>
<td>1.57</td>
<td>3.59</td>
<td>2.53</td>
<td>2.30</td>
</tr>
<tr>
<td>( \frac{f^{\text{Tot}}}{d^{\text{Tot}}} ) (µm(^{-1}))</td>
<td>0.011±0.001</td>
<td>0.006±0.002</td>
<td>0.008±0.002</td>
<td>0.009±0.002</td>
</tr>
</tbody>
</table>

(2) **Particles < 200 nm via TEM.** Particles smaller than 200 nm for \( f/d \) quantification were measured by TEM. Two assumptions are made when processing TEM data: 1) all particles are spherical; 2) measured sizes are the real 3D particle sizes. \( V^{\text{Tot}}_{\text{ppt}} \) was obtained by adding up the volume of each particle calculated from its diameter. The volume fraction can be calculated by:

\[
f^{\text{Tot}} = \frac{V^{\text{Tot}}_{\text{ppt}}}{At}
\]

Eqn. 10

where, \( A \) is the area of material measured and \( t \) is the TEM foil thickness.
Following the Method 1 of SEM particle treatment, these particles were grouped into a bin of size range of 0 - 200 nm. Average particle size is used here for $f/d$ calculation for these particles. The $\frac{f_{\text{Tot}}}{d_{\text{eff}}}$ values for all locations are shown in Table 7.

Table 7. Zener size measured by TEM and the calculated dispersion levels

<table>
<thead>
<tr>
<th></th>
<th>H2_Top</th>
<th>H2_Mid</th>
<th>H3_Top</th>
<th>H3_Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vol. Fraction ($f$)</td>
<td>0.012±0.004</td>
<td>0.012±0.007</td>
<td>0.010±0.007</td>
<td>0.011±0.003</td>
</tr>
<tr>
<td>$d$ (µm)</td>
<td>0.10±0.01</td>
<td>0.09±0.02</td>
<td>0.10±0.01</td>
<td>0.09±0.03</td>
</tr>
<tr>
<td>$\frac{f_{\text{Tot}}}{d}$ (µm$^{-1}$)</td>
<td>0.124</td>
<td>0.140</td>
<td>0.105</td>
<td>0.121</td>
</tr>
</tbody>
</table>

(3) $f/d$ Combination from SEM and TEM. According to the Appendix H, the above results can be combined through adding up the $f/d$ values from SEM and TEM. The total dispersion levels of all locations are shown in the Table 8.

Table 8. Total dispersion levels for various characterization locations

<table>
<thead>
<tr>
<th></th>
<th>H2-Top</th>
<th>H2-Mid</th>
<th>H3-Top</th>
<th>H3-Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>TEM</td>
<td>0.124</td>
<td>0.140</td>
<td>0.105</td>
<td>0.121</td>
</tr>
<tr>
<td>$f_{\text{Tot}} / d$ (µm$^{-1}$)</td>
<td>0.025</td>
<td>0.024</td>
<td>0.027</td>
<td>0.028</td>
</tr>
<tr>
<td>SEM Method 1</td>
<td>0.011</td>
<td>0.006</td>
<td>0.008</td>
<td>0.009</td>
</tr>
<tr>
<td>$f_{\text{Tot}} / d_{\text{Tot}}$ (µm$^{-1}$)</td>
<td>0.149</td>
<td>0.164</td>
<td>0.132</td>
<td>0.149</td>
</tr>
<tr>
<td>SEM Method 2</td>
<td>0.135</td>
<td>0.146</td>
<td>0.113</td>
<td>0.130</td>
</tr>
<tr>
<td>Total $f/d$ (µm$^{-1}$) – Method 1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total $f/d$ (µm$^{-1}$) – Method 2</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
In Humphreys’ Model, mean grain radius instead of grain diameter is used. 3D grain radiiuses and pinning parameters of the top and middle regions of H1 and H2 are summarized in Table 9.

Maximum size ratios in Table 9 of these locations were obtained via EBSD through dividing the largest grain radius (2D) by the mean grain radius (3D radius converted via Equation 7), assuming the largest grain radius is the largest grain cut by the sectioning plane forming the largest sectioning area. The size ratio is consistent with the theoretical value of 2.5 for an ideal microstructure. The smaller values of middle regions can be caused by less grains measured (~200 in the middle vs. ~900 in the top). This result is different from others in literature which reported that larger size ratio in FSW reaching 4.5. This is probably a more thorough dynamic recrystallization occurred during FSW here with a higher tool rotation rate (1000-1500rpm here vs. 180-450rpm). Therefore, it is reasonable to follow the assumption in the grain stability model that the largest grain in the initial microstructure is 2.5 times larger than the mean grain size.
Table 9. Calculated pinning parameters of each second phase particles and the total parameters for top (denoted as -Top) and middle (denoted as -Mid) of all welding cases

<table>
<thead>
<tr>
<th></th>
<th>H2_Top</th>
<th>H2_Mid</th>
<th>H3_Top</th>
<th>H3_Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean Grain Radius (µm)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>1.8</td>
<td>1.2</td>
<td>2.0</td>
</tr>
<tr>
<td>Maximum Size Ratio</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.4</td>
<td>2.1</td>
<td>2.4</td>
<td>2.2</td>
</tr>
<tr>
<td>f/d (µm⁻¹)</td>
<td>TEM</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.124</td>
<td>0.140</td>
<td>0.105</td>
<td>0.121</td>
</tr>
<tr>
<td></td>
<td>SEM Method 1</td>
<td>0.025</td>
<td>0.024</td>
<td>0.027</td>
</tr>
<tr>
<td></td>
<td>SEM Method 2</td>
<td>0.011</td>
<td>0.006</td>
<td>0.008</td>
</tr>
<tr>
<td></td>
<td>Total Method 1</td>
<td>0.149</td>
<td>0.164</td>
<td>0.132</td>
</tr>
<tr>
<td></td>
<td>Total Method 2</td>
<td>0.135</td>
<td>0.146</td>
<td>0.113</td>
</tr>
<tr>
<td>Ψ – Method 1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.447</td>
<td>0.886</td>
<td>0.475</td>
<td>0.894</td>
</tr>
<tr>
<td>Ψ – Method 2</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.405</td>
<td>0.788</td>
<td>0.407</td>
<td>0.780</td>
</tr>
</tbody>
</table>

3.6. ICP-MS Results

As shown in Fig. 27 by ICP-MS, the magnesium content in the weld is the same with that in the original base material and there is also no change in Mg content along the thickness.
CHAPTER 4

4. DISCUSSION

4.1. Establishment of FSW Microstructures

The microstructures in the FSW weld zones can be interpreted in terms of dynamic recrystallization and recovery, with possibly some grain growth, as influenced by the severe deformation and high temperatures generated during the FSW process. The most severe deformation and highest temperatures occur near the top surface while stirring. (The larger strain experienced at the top is consistent with the grain boundary misorientation distribution, Fig. 20.)

Aluminum alloys mainly dynamically recover during hot working at a temperature below 450 °C\cite{115,116} at strains up to 5\cite{115,116}. In the presence of large second-phase particles, dynamic recrystallization by nucleation and grain growth may also occur at these temperatures.\cite{116,117} Dynamic recrystallization starts with the nucleation of nano-scale grains in regions of high strain and stress in the weld zone.\cite{25,45,90,91} These nuclei can
grow and coarsen to a size of several microns after heating 1-4 min at 350-450 °C,[91]
matching the observed grain sizes in the current study. Grain growth relies on grain
boundary mobility, which is impeded by the presence of closely-spaced second-phase
particles.[118]

The relative grain sizes within the weld zone can be understood in terms of temperatures
and deformation. Relative to the middle regions, the top regions experience higher
strains (as shown by simulation[102] and by %LAB, Fig. 20) and modestly higher
temperatures simultaneously, thus promoting rapid nucleation of fine grains.
(Simulations show that the upper surface experience peak temperatures 7-30 degrees C
higher than the middle plane under H1, H2, and H3 welding conditions, Appendix F.[102])
With the relatively small temperature difference to the welding temperature (~500 °C),
the larger deformation on the top leads to finer grains there, consistent with other
reports.[90] Thus similar grain sizes (Fig. 14a, b, and c) are found in the top regions for
three cases after dynamic recrystallization.

At a single weld location, the effect of heat input may be predicted. Comparing H1 and
H2, for example, only the tool traverse rate is changed. For H2, there will be more
stirring (straining) of each element in the weld zone, with concurrently higher peak
temperatures. The time above a certain temperature (300 °C – 400 °C) is 4-6 times
longer for H2 (Table 1), which allows more time for grain coarsening relative to H1. As
expected, there is little difference in grain size between H2 and H3, which have similar
heat inputs.
4.2. Effect of Grain Size on Hardness

The hardness drop of the H2 top region after QPF-HT (Fig. 9b) indicates the effect of grain size on hardness on the other hand. A previous study shown the Hall-Petch slope $k_H$ of AA 5083 is $14 \text{ Hv } \mu\text{m}^{-1/2}$.[119] In the studied FSW locations, hardness of H2-Mid was shown to be constant ($\pm 2 \text{ Hv}$) against heat treatment (without coarse grain formation). The $H_0$ is then determined from H2-Mid ($d = 3.5 \mu\text{m}; Hv = 72 \text{ Hv}$) to be $65 \text{ Hv}$.

<table>
<thead>
<tr>
<th></th>
<th>Grain Size ($\mu\text{m}$)</th>
<th>Hardness (Hv) Predicted</th>
<th>Expl. Hardness Avg. (Hv)</th>
<th>Scatter</th>
</tr>
</thead>
<tbody>
<tr>
<td>H2 Top-1/3</td>
<td>2</td>
<td>75</td>
<td>75</td>
<td>2</td>
</tr>
<tr>
<td>H2 Mid-2/3</td>
<td>3.5</td>
<td>72</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>H2 Mid+QPF</td>
<td>3.7</td>
<td>72</td>
<td>71</td>
<td>1</td>
</tr>
<tr>
<td>H2 Top+380°C/24hr</td>
<td>4</td>
<td>72</td>
<td>71</td>
<td>1</td>
</tr>
<tr>
<td>H2 Mid+380°C/24hr</td>
<td>6.1</td>
<td>71</td>
<td>70</td>
<td>1</td>
</tr>
<tr>
<td>FSW+QPF</td>
<td>80</td>
<td>67</td>
<td>69</td>
<td>2</td>
</tr>
<tr>
<td>Base H18+380°C/24hr</td>
<td>6.6</td>
<td>70</td>
<td>67</td>
<td>3</td>
</tr>
</tbody>
</table>

As shown in Table 10, within the scatter, the predicted hardness of H2 FSW regions, both before and after heat treatment, are consistent with the measured values. It indicates the H2-Top hardness difference between $380^\circ\text{C} / 24 \text{ hr}$ recovery anneal could be caused by the increase of grain size as well, in addition to the decrease of the stored strain energy.
After recovery anneal, the grain size increase from 4 to ~80 µm on the H2 top region after QPF can be fully accounted for the hardness decrease. Within the scatter, the hardness of annealed base metal (after 380°C / 24 hr) is also correctly predicted.

4.3. Origin for Coarse Grain Structure

Coarse grain structure formation during post-weld annealing has been found in various friction-stir welded alloys. Even though in some literature, AGG has been used extensively to describe this phenomenon, no clear evidence has been given to prove it.\textsuperscript{[29, 30, 56]} Recrystallization has also been argued as the origin of the coarse grain structure in FSW.\textsuperscript{[30, 52]} The mechanism for the coarse grain structure formation is discussed here.

A. Mechanism for coarse grain structure formation

As suggested by the previous study, the coarse grain structure originates from a region typical of a deformed microstructure,\textsuperscript{[30]} which is also similarly observed in our investigation. All the top regions of three cases have a large portion of low angle grain boundaries, in comparison to the middle regions (Fig. 20). Therefore, it was previously proposed that the coarse grain structure is initiated by recrystallization in this deformed region leading to several larger grains than surrounding grain structure.\textsuperscript{[30]} However,
these deformed structures are more likely providing additional driving force for the coarse grain formation.

As previously proved that the hardness difference between the FSW and fully annealed base O material ($\Delta \sim 7 \text{ Hv}$) is only a small fraction of the difference between the fully hardened base H18 and the O materials ($\Delta \sim 65 \text{ Hv}$). The difference between the top and middle regions is even smaller ($\Delta \sim 3 \text{ Hv}$). Additionally, the recovery anneals removing the strains showed no effect on the coarse grain formation process and amount, Fig. 10 and Table 2. Therefore, stored strain energy is unlikely the driving force for the coarse grain formation and recrystallization can be ruled out.

This leaves normal grain growth (NGG) and abnormal grain growth (AGG) as possible mechanism. The most distinguishable difference is that in NGG grain structure coarsens uniformly and similar behavior can be found location by location; while in AGG, a discontinuous process, several grains grow excessively within a finer-grain matrix which varies little during the process. Therefore an easy way to determine the mechanism is NGG or AGG is whether there are large grains grow preferentially. An un-uniform grain structural evolution was found in the retreating side of FSW-H2 weld experienced annealing at 380 °C for 24 hr, Fig. 15. Therefore, the mechanism for the coarse grain structure formation should be boundary energy-driven AGG.
B. Factors leading to the occurrence of AGG

Further study was conducted to seek for the microstructural factors leading to the easy occurrence of AGG in FSW rather than in base metals. Among all possible factors introduced in Chapter 1, particles are effective in pinning boundaries, and many AGG cases in literature are related with particle pinning effect, even when there are other factors existing.\(^{[62, 63]}\) Quantification of particle contents in various FSW locations has been performed. As shown previously in Table 8, the particle dispersion levels are similar (± 0.01 \(\mu\)m\(^{-1}\)) location to location and also between different welds.

However, the small (Fe,Mn)Al\(_6\) particles, with the size of smaller than 0.2 \(\mu\)m, are unstable under high temperature (> ~ 500 °C), susceptible to coarsening.\(^{[76]}\) Sato et al. referred to the solvus temperature of (Fe,Mn)Al\(_6\) of 908 K (635 °C),\(^{[38, 120]}\) which is higher than the solidus temperature of AA5083, a conclusion was then suggested that (Fe,Mn)Al\(_6\) would not dissolve during FSW.\(^{[38]}\) However, this solvus temperature value is of bulk alloy, when the size reaches as small as 30 nm, this temperature is calculated to be about 80 K lower because of the size-dependent thermal properties of small particles.\(^{[121]}\) Therefore it is still possible the nano-sized (Fe,Mn)Al\(_6\) will dissolve during FSW.

Solute segregation on the grain boundary is also a way to pin the grain boundary, and it is therefore suggested that Mg content change and its redistribution by FSW may lead to
AGG.\textsuperscript{[63]} As shown by ICP-MS results, Mg amount (represented by the signal ratio of Mg to Al) is almost constant along the weld centerline from top to bottom and also reveal no difference to the Mg distribution on the base metal, Fig. 27. The observed AGG is unlikely induced by the depletion and redistribution of Mg content during FSW.

Texture is another important factor for AGG, observed in many materials including aluminum. Texture induced AGG requires that there is a strong and single orientation texture component in the initial microstructure.\textsuperscript{[62]} A microstructure of more than two components is unlikely to show AGG induced solely by texture. After AGG, large grains orient similarly and a secondary texture is normally formed.\textsuperscript{[63]} However, all the as-welded locations show more than one texture components, Fig. 28. Compared with the base O (Fig. 28g), texture in FSW is not strong as well. Therefore, it is unlikely that the AGG observed in FSW here is solely induced by texture.

Microstructural factors with strong correlation with AGG here in FSW were found to be grain size and fraction of LAB. As shown in Fig. 6 and Fig. 14, AGG locations are those have as-welded grain size less than 3 \(\mu\text{m}\). H2-Mid and H3-Mid region have as-welded grain size larger than 3 \(\mu\text{m}\) showing no AGG during QPF. A higher fraction of LAB may also more likely lead to a discontinuous grain growth process as predicted by a previous theoretical treatment.\textsuperscript{[57]} The AGG regions have %LAB larger than 18%; those non-AGG regions smaller than 18%.
C. Application of grain growth model

With the pinning parameters and grain sizes in Table 9, predicted stabilities at all locations are shown in Fig. 29 (Fig. 29 is an enlarged region of Fig. 3). Each location is represented by a bar, with the ends of the bar being the two $f/d$ values from Method 1 and Method 2. Red bars are locations that exhibited AGG after QPF-HT, blue ones represent locations that do not show AGG during QPF. As shown, two top regions well entered into the AGG nucleation domain in this grain growth plot, expected to show AGG readily. For middle regions, they sit in the AGG difficult region. This model predicts these middle regions are not able to nucleate readily. This is also consistent with experimental observation. Similarly, the conditions of all locations plotted into Fig. 30. The top regions are in the AGG domain, and AGG is ready to occur. While the middle regions are in No Growth domain.

Also predicted by the model from Fig. 29, AGG in middle region will occur if large enough grain existing. In fact it shows AGG or non-AGG of H2_M depending on time (Fig. 5). After 1-hour anneal, those coarse grains nucleated from the top region has propagated down to the middle regions. The size ratio reaches a value of about 50. As shown by Fig. 30, these coarse grains on the middle region satisfy AGG condition (the bar labeled as H2-M+465°C/24hr). They are expected to be able to keep on growing abnormally, seen experimentally as well. Therefore, grains in the middle regions formed after elongated anneal at 465 °C are about 5 times larger than those in the top region.
because easy nucleation in top regions generates more secondary grains growing to impingement.

Middle regions are also expected ready to enter into nucleation domain and to nucleate AGG with the decrease of particle content, along the x-axis in both Fig. 29 and Fig. 30, that is, \( f/d \) and \( \Psi = 3\sqrt[d]{f/d} \) respectively. Particle coarsening can also occur during high temperature annealing. The dissolving of \( \text{Mg}_5\text{Al}_8 \) occurs at about 350°C and coarsening of \( \text{MnAl}_6 \) at temperatures higher than 500 °C. The reduction of \( f/d \) and \( \Psi \) by the increase of \( d \) can result in the fully entering of \( H2_M \) into AGG domain in microstructure stability plot. This is consistent with the observation of annealing experiments that increase the annealing temperature to 510 °C (a 40-degree increase) leads to AGG nucleation in middle region of \( H2 \), Fig. 7 View(3).

It was reported that coarse grain structure is limited in weld region.\(^{[32]}\) In our experiment on \( H2 \) sample, it is clearly shown some large grains can extend out to base metal, or more accurately to the TMAZ region, at samples annealed at temperature higher than 510 °C in Fig. 7. This phenomenon is not found at 465 °C even after elongated annealing time, Fig. 5. If the particle dispersion level in the base metal is assumed to be the average of the measured values for FSW locations, that it, 0.093 µm\(^{-1}\), \( \Psi \) is then equal to 0.92 (3D grain radius of base metal after recrystallization is 3.3 µm). From Fig. 2, for such a pinning parameter, a very large size ratio (~ 100) is needed for AGG occurrence, which is not readily available during QPF-HT. But when \( f/d \) is changed by higher temperature annealing, \( \Psi \) can be reduced to a value much smaller than 1.00. According to the model
(Fig. 2), required size ratio for AGG drops fast with the decrease of $\Psi$ and for example the required ratios are 10 and 6.7 respectively for $\Psi$ equal to 0.9, and 0.85. Therefore it is seen large grains grew into base metal in Fig. 7 View (1), (2), and (3).

The fact that the observed subsurface ultra-fine grain structure remains almost the same after QPF-HT can actually be related to the increased amount of boundary-pinning particles. The size ratio on the border between fine grain structure ($\sim$ 0.3 $\mu$m) and coarse grain structure ($\sim$ 100 $\mu$m) is about 300 (Fig. 17). A higher density of low-Z particles (probably Al$_2$O$_3$ and MgSi$_2$ as shown in Fig. 22) on a narrow band can be attributed to the enhanced inhibition of the AGG expanding to subsurface region. Additionally, within this subsurface region, the particle content is significantly larger than the rest of weld region from both SEM (Fig. 23) and EBSD (Fig. 17) characterizations, especially apparent in SEM micrographs of Fig. 23 under a high magnification of subsurface region of ultra-fine structure (within 20 $\mu$m to the top surface) and normal top region (about 50 $\mu$m away from the top surface), even though a quantitative comparison is difficult from these figures because of the SEM resolution.

D. The rapid and critical natures of AGG in FSW

For many previously observed AGG cases, there is always a significantly long period of incubation before AGG. These observations lead to a characteristic feature of AGG concluded in a review that AGG initiation is sluggish, on the order of hours.
However, the experiments on coarse grain morphology vs. annealing time on the FSW specimens showed that AGG forms within a minute and propagates to cover the top 1/3 region within 5 min during QPF. This may be caused by following reasons:

1. The microstructural conditions of top regions well entered into AGG nucleation domain (Fig. 29), in comparison with other cases that: 1) dissolution of particle needed to make condition enter into AGG nucleation region,\textsuperscript{[71]} 2) NGG needed for microstructure reaching the limiting grain size and then inhibition of NGG occurs, leading to the entering of the microstructural conditions into AGG nucleation region.

2. Because of the small grain sizes (~ 3 µm), the FSW regions are energetic. The driving force for the grain growth is therefore can be high.

It was also found that only annealing below 370 °C did not have any AGG occur. At 380 °C annealing, several large grain formed. A higher temperature at 510 °C for 5 min, a thin layer of coarse grains formed. Such a critical temperature for AGG is similar the one for the dissolving of Mg₅Al₈ (at about 350 °C). It is possible that the dissolving of this phase results in the microstructural conditions moving left in Fig. 29, and entering into nucleation domain. As for the middle region of H2, which does not nucleate at 465 °C, it did show nucleation during higher temperature annealing (510 °C, 540 °C, and 560 °C), Fig. 7. It can be attributed to the MnAl₆ coarsening, which occurs under these temperatures according to literature\textsuperscript{[76]}. 
4.4. Possible Methods to Avoid AGG

From the above discussion, the coarse grain structure formation is an AGG process and originated by microstructural conditions, most importantly second phase particle pinning, grain sizes, and %LAB (having a consistent trend with grain size, Fig. 21). Two of them are controllable: the average grain size (%LAB is associated with grain size in FSW, Fig. 21) and the pinning parameter \( \frac{f}{d} \). It is corresponding to moving the microstructural condition to up/right in Fig. 3, or making \( \Psi > \) critical \( \Psi (= 0.6) \). Possible methods for avoiding AGG are listed as follows:

1) Increase heat input during FSW

As shown in Table 9 and Fig. 29, the welding condition does not change particle pinning parameters greatly. But the grain size increases (%LAB decreases) with the increase of heat input as shown previously. Within this small range, AGG phenomenon is very dependent on average grain size. An increase of grain radius from 1 \( \mu m \) to 2 \( \mu m \) leads to this microstructure stable against the AGG nucleation, and a further increase to 3 \( \mu m \) will make all grains unable to growth, that is, no growth can occur even for a pre-existing infinitely large secondary grain.

It was shown by previous study, grain size does not always increase with the increase of tool rotation speed (heat input increase).\cite{122} This is probably because of the final grain size in the microstructure determined by combined effect of dynamic deformation,
recrystallization, recovery, and grain growth. Increased tool rotation speed increases the metal temperature favoring grain growth to larger grains and also induces deformation promoting dynamic recrystallization to reduce grain size at the same time. This contradictory effect of tool rotation may lead to the observed initial increase of grain size and a later decrease with the increase of the tool rotation speed. Therefore, the welding conditions for avoiding the AGG in the whole weld can be found experimentally and indicated by the as-welded grain structure. From the experiments conducted for AA5083 welds, the grain size increase with the increase of welding heat input. For this material, the as-welded microstructure of 3D grain radius larger than 2 µm (diameter > 4 µm) is expected to be stable against AGG during QPF-HT.

In addition to the heat directly from the welding process, more heat can be added through, e.g. resistance heating or radiation. Grains can grow to larger size at such higher temperature or longer times-at-temperatures provided by the additional heat sources.

2) Modification of alloy content

It is also possible to use a larger f/d of thermal stable particles to prevent the AGG. For H2_T and H3_T of grain radius equal to about 1 µm, in which AGG occurs during QPF-HT, more small pinning particles should be induced in to obtain an f/d of about 0.22 µm⁻¹ for avoiding AGG (Fig. 29). As reported in literature an addition of Mn, the major dispersoid-forming element in AA5083, can be a desirable method, even though this may not be as effective as the addition of Zr because the addition of Mn contributes most to
the formation of MnAl$_6$ particle larger than 2 µm instead of submicron dispersoids.\textsuperscript{[87]} Zr was on the other hand found to form finer ZrAl$_3$.\textsuperscript{[87]} In addition, coherent ZrAl$_3$ is very effective in pinning boundaries showing drag force twice that of incoherent particles.\textsuperscript{[87, 110]}

3) Reduction of QPF temperature

The critical nature that coarse grains form only at temperature higher than 380 °C for long duration (24 hr) and only at temperature higher than 420 °C for short duration (5 min) may suggest another method: lower the QPF temperature to less than 420 °C for its 5 min anneal. During this heat treatment condition, no coarse grains will form.
The following conclusions apply to friction-stir welded AA5083 (originally H18), post-friction stir welding (FSW) heat-treated 465 °C for 5 minutes:

1. Grains of sizes exceeding 100 μm are formed, starting near the top sheet surface.

2. The mechanism for forming the coarse grains is abnormal grain growth (AGG), driven by the reduction of grain boundary area. This conclusion differs from some reports in the literature concluding that recrystallization is responsible for such large grains.\[30, 52]\]

3. AGG is favored by higher strains and lower temperatures (i.e. lower heat input indices). Weld conditions with high heat input show coarse grains in only the upper region of the weld zone; low heat input welds show large grains throughout the weld zone.
4. The presence and critical nature of AGG for the post-FSW microstructures is consistent with a simplified theory of grain growth appearing in the literature.\textsuperscript{[69]}

5. The heat treatment is near the critical condition for AGG to occur. Small changes in microstructure produce regions of AGG adjacent to regions of no grain changes. The critical microstructural parameters for AGG have been established as follows:

   a) As-welded grain radius < 1.5 µm
   b) %LAB > 18 %
   c) Ψ < 0.6

Within measurement accuracy, there are no differences in second-phase particle sizes, volume fractions, or solute content (Mg) in AGG vs. non-AGG regions. Textures are different, but do not clearly favor or inhibit AGG.

6. AGG cannot be avoided by a prior recovery anneal.

7. The critical temperature for “rapid” (within a few minutes) AGG is likely related to the dissolution or coarsening temperatures for second phase particle – Mg\textsubscript{5}Al\textsubscript{8} (dissolving at about 350 °C). The transition of non-AGG middle region to AGG at higher temperature (510 °C - 560°C vs. 465 °C) is attributed to the coarsening of dispersoid – MnAl\textsubscript{6} (coarsening at > 500 °C).
8. AGG is not observed for the H18 base metal under any annealing condition, except as growth from an FSW-affected region.

9. The distribution of grain size through the thickness of a FSW weld zone, among other microstructural characteristics, is determined by a complex combination of strain and time-at-temperature, both of which are larger near the top surface. These conditions produce higher hardness (typically 75 vs. 70 Hv), smaller grains (typically, 2.0 µm vs. 3.5 µm), and higher fraction of LAB (25% vs. 16%), as compared with middle sections of the same welds.

10. Based on the above conclusions, the following strategies may prove successful in avoiding AGG:

   a) $\Psi > \Psi^* (= 0.6)$
      
      – Increase welding heat input to obtain large as-welded grain size and lower %LAB.
      
      – Add additional heating sources to increase welding temperature for obtaining larger as-welded grain size.
      
      – Change the alloy composition to achieve more stable pins at post-weld heat treatment temperature.

   b) Lower the QPF-HT temperature to less than 380 °C.
APPENDIX

APPENDIX A: ADDITIONAL BACKGROUND REVIEW

A.1. Introduction to Friction Stir Welding (FSW)

Conventional fusion welding of high-strength aerospace aluminum alloys is difficult to process, such as the 2XXX and 7XXX series, and most of 7XXX are actually non-weldable.\[32, 35, 123\] Because of the poor solidification microstructures and defects produced in the welded region and the subsequent significant decrease in mechanical properties, the welding process has been used limitedly in joining the structures of these alloys.\[36\] In 1991, as an innovative solid-state joining technique, friction stir welding (FSW) was invented at The Welding Institute (TWI) of the United Kingdom with aluminum alloys used as the initial application target.\[14, 124\] There is no melting and recasting in this solid-state welding process, therefore the problems associated with traditional fusion welding do not exist in friction-stir welds, and those traditionally non-weldable aluminum alloys can be successfully joined by FSW.\[34, 39, 125\]
As a promising solid-state joining technique, properly controlled FSW can produce a fine and equiaxed recrystallized grain structure (grain size $d = 0.5–10 \, \mu m$)\cite{18, 22, 25, 31, 36} with fewer gross defects\cite{126}, better dimensional stability,\cite{127} less distortion,\cite{127} lower residual stress,\cite{14} and better retained mechanical properties\cite{14, 31}. FSW is also a candidate process for joining sheets of dissimilar thickness or composition to create tailor-welded blanks\cite{2-4} that retain the capacity for enhanced ductility and strength via fine grain structure. Recent research focuses on FSW in joining materials such as magnesium alloy and steel.\cite{5-8} Commercialization of FSW, mainly for aluminum alloys, has occurred in the transportation industry, for applications such as automobiles, railway vehicles, ships, and rockets.\cite{9-13}

The superior advantage of FSW in producing fine grain structure led to the development of friction stir processing (FSP), holding the same basic concept to that of FSW but working on a monolithic workpiece with the purpose of modifying the microstructure to enhance the superplastic properties.\cite{128} AA7075\cite{128} and AA2024\cite{122} have been friction-stir processed to successfully obtain high-strain rate superplasticity.

A. Basic Concept of FSW and the Macrostructure Established by FSW

The basic concept of FSW is illustrated in Fig. 1.\cite{14} A specially designed non-consumable welding tool composed of pin and shoulder has its pin embedded into the workpiece with its shoulder contacting the surface of the workpieces to be joined. The fast rotation of the welding tool produces the plastic deformation and frictional heating.
The heating softens the materials to be welded. The intense plastic deformation by a combined effect of rotation and forward translation of the welding tool results in a solid-state joining at an elevated temperature at $0.6T_m - 0.8T_m$. In such a coupled thermal and mechanical working process, the welding tool serves to generate heat, move and mix material to establish a joint, and constrain softened material under the shoulder. The hot, plasticized material is then transported around the weld by the combined effect of the rotation and translation of the welding tool and is deposited on the back side of the tool. A direct consequence of FSW is the severe microstructural change in the welded region, established by such a material flow and the temperature history. However, the current understanding of the material flow during FSW is limited because of its complexity. Tool geometry is critical for the material flow, and therefore influences the deformation. Two very important welding parameters are tool rotation speed ($\omega$, in rpm) and traverse speed ($v$, in mm/min) in the welding direction along the joint line, Fig. 1.

As shown in Fig. 1c, the post-weld macrostructure is commonly divided into three different zones based on their distinct microstructural evolution during FSW: weld nugget (WN), thermo-mechanically affected zone (TMAZ), and heat-affected zone (HAZ). The weld nugget, or “stirred zone” (delineated approximately by the dashed line in Fig. 1.1b), which can be bowl-shaped or elliptical, is highly deformed and dynamically recrystallized at high temperature during FSW. This zone widens near the top surface because of the significant frictional and deformational effect from the tool shoulder; therefore it is sometimes separated into two zones, with the subsurface
region being called the shoulder contact zone\textsuperscript{[56]}. The thermo-mechanically affected zone surrounds the weld nugget. It experiences lower temperatures and less deformation, and consequently no recrystallization.\textsuperscript{[33-35]} The heat affected zone surrounding the TMAZ only experiences thermal disturbance during FSW, and possible modification of the microstructure.\textsuperscript{[36, 37]} The advancing side (AS) refers to the side where (right-hand side in Fig. 1a and left-hand side in Fig. 1b and Fig. 1c) the tool traverse direction and the tangential direction of the rotating tool’s edge are the same; the other side with these two directions opposite is retreating side (RS).

B. Heat Input and Temperature Field of FSW

Induced by the rotation and translation of the welding tool, the plastic deformation and the friction between the tool shoulder and the workpiece surface provide localized heating,\textsuperscript{[14]} resulting in a local temperature increase for aluminum to as high as $0.6T_m-0.8T_m$.\textsuperscript{[23-25]} Peak temperatures have been correlated to combinations of welding parameters $\nu$ (weld feed rates) and $\omega$ (tool rotation speeds) through a “heat input parameter” or “heat index”, typically $\omega^2/\nu$\textsuperscript{[23, 26]} or $\omega/\nu$\textsuperscript{[27]}.

The plastic deformation and the frictional heating are so inhomogeneous that the peak temperatures also vary locally.\textsuperscript{[39, 89, 127]} Efforts have been made to understand the temperature history and field, either through direct measurement by embedded thermocouples\textsuperscript{[24, 127]} or through correlating the as-weld microstructures to the known aging curves\textsuperscript{[20, 125]}. The temperature field was measured to be asymmetric in the weld
nugget, with the peak temperature slightly higher on the advancing side than the retreating side\textsuperscript{[89, 127]} and the top region hotter than the middle and bottom regions\textsuperscript{[39]} It is also reported that the temperature field is nearly isothermal within the weld nugget.\textsuperscript{[24]}

A.2. Microstructure in FSW

The combined effect of severe plastic deformation and elevated temperature during friction-stir welding also leads to remarkable microstructural evolution. Dynamic recrystallization,\textsuperscript{[22, 30, 36, 52, 125]} breaking-up of second phase particles in the nugget,\textsuperscript{[38]} and microtexture development\textsuperscript{[53, 56, 130, 131]} occur in the nugget zone,\textsuperscript{[25, 31, 36, 39, 41-43]} while precipitate dissolution and coarsening occur in all three zones.\textsuperscript{[20, 36, 37, 39, 41, 44]} A fine and equiaxed grain structure is obtained after FSW with grain size ranging from 1 µm to 10 µm.\textsuperscript{[15, 36, 37, 39, 41, 44]} Ultrafine-grained microstructures with average grain size smaller than 1 µm have also been obtained using special cooling methods.\textsuperscript{[15, 45]}

Observations of dislocation density in weld nugget zone vary. A high dislocation density was reported in AA7050,\textsuperscript{[34, 36]} a low dislocation density was found AA5083 welds by comparison;\textsuperscript{[119]} and the dislocation density in the AA6063 FSW weld nugget zone was reported to be relatively low while a few of the grains contained a high density of dislocations.\textsuperscript{[20]}
The established microstructure is different, not only between various macrostructural zones but also locally in the weld nugget. These microstructural factors are important for understanding post-weld annealing behavior.

A. Grain structure

It has been generally accepted that fine and equiaxed grains are obtained in the weld nugget through dynamically recrystallization during FSW/P. The grain aspect ratios are found to be 1.01 in the nugget center and 1.26 near the top surface, indicating the effect of additional deformation from the tool shoulder.

Welding temperature, crucial for as-weld grain size, is a function of the ambient temperature and the welding parameters. Submicron grain structure has been achieved through liquid nitrogen cooling during FSW. Either the increase in the tool rotation rate at a constant traverse speed or the decrease in the traverse speed at a constant tool rotation rate produced larger average grain sizes, consistent with the effect of these welding parameters on the welding temperature. However, it was also reported that with a constant tool traverse rate, grain size in FSP AA2024 increased with the increase of the rotation rate from 200 rpm ($d \sim 2 \mu m$) and peaked at 600 rpm ($d \sim 6 \mu m$), and then decreased to $\sim 3 \mu m$ at 800 and 1000 rpm. This suggests that the temperature is not the only factor determining the post-weld grain size; the effect of strain and strain rate during FSW/P should be also considered. More quantitative study is needed on the dependence of grain size on the welding temperature, strain and strain rate.
Average grain sizes within the weld nugget differ horizontally, with grain size on the retreating side smaller than in the nugget center and on the advancing side. Either a coarser or finer grain structure than in the nugget center can be found on the advancing side.\cite{52,136} On the other hand, the results of vertical grain size variation are contradictory in the literature. A decreasing tendency from top to bottom has been shown in 6.4-mm thick AA7010\cite{30}, 7-mm thick AA2024-T3,\cite{35} and 25.4-mm thick AA2519\cite{109}. Conversely, an average grain size increase from the top to bottom in 4-mm thick AA1100 has been reported.\cite{52} This difference can be associated with the temperature profile and the severity degree of plastic deformation. Fast heat dissipation to the backing anvil beneath the workpieces leads to a lower temperature and a shorter exposure time. The grain growth at the bottom region is then retarded. But for a thinner workpiece, temperature profile is more uniform vertically. The effect of additional deformation from the tool shoulder is therefore more significant, leading to finer grain structures at the top region and the subsurface region right below the shoulder.

Additionally, a broader grain size distribution was found in a friction-stir welded microstructure of $R_{\text{max}} \approx 4.5 \overline{R}$\cite{30} than $R_{\text{max}} \approx 2.5 \overline{R}$ (\overline{R} is the average grain size) for an ideal grain distribution\cite{69}. A wider grain size distribution is favored for abnormal grain growth.\cite{69}
B. Deformed state of FSW microstructure

Different degrees of recovery in the grains close to each other in the nugget have been found by transmission electron microscope (TEM) studies in various aluminum alloys, such as AA2195,\textsuperscript{[137]} AA6063,\textsuperscript{[126]} and AA7075\textsuperscript{[36]}. Grains with high density of dislocations can be found adjacent to regions in which there are recovered structures with subgrain boundaries and a relatively low density of dislocations.

Another indicator of deformation severity is the fraction of low angle boundaries (LAB), which are considered to be of arrays of dislocations and whose energy and mobility are functions of their misorientation angles.\textsuperscript{[57, 118]} A misorientation angle of 15° is typically considered the transition from LAB to high angle boundary (HAB),\textsuperscript{[52, 57, 138]} whose energy and mobility on the other hand are generally constant, except for coincidence site lattice (CSL) boundaries\textsuperscript{[118]}. The emergence of high resolution electron backscatter diffraction (EBSD), in conjunction with a field emission gun scanning electron microscope (FEGSEM), provides a powerful method for characterizing grain/subgrain boundaries and quantitatively measuring boundary misorientation distribution.\textsuperscript{[112, 139, 140]}

EBSD maps showed a higher fraction of LAB (%LAB) in FSW/P nuggets, varying from 10% to 40% in previous reports,\textsuperscript{[30, 52, 55, 56]} than that in a microstructure with random grain assembly (\~5\%\textsuperscript{[106]}). The subsurface region, which directly contacts the weld shoulder during FSW/P, is in a more deformed state with higher %LAB\textsuperscript{[52, 55]}, which can be attributed to additional deformation from the tool shoulder and faster quenching. The
bottom region at the root of the welding tool pin sometimes has a higher %LAB,[30, 55] probably produced by insufficient deformation[30] or by lowered temperature caused by the cold backing anvil acting as a heat sink. Additionally, both edges of the nugget to the WN-TMAZ boundaries are also of higher %LAB,[52, 55, 56] indicating these regions are more deformed than the nugget center.

C. Microtexture

A vertical gradient of microtexture along the weld centerline has been reported in the weld nugget.[53, 130] After FSW on AA6063 base metal with the Goss orientation as dominant texture component, a single {110}<001> component was found on the top region within a depth of 0.5mm. Two components, {114}<221> and {110}<001>, co-exist on the middle and bottom regions. ([hkl] designates the crystallographic plane perpendicular to the welding direction; <hkl> designates the crystallographic direction parallel to the normal direction of the metal surface).[53] Two other studies on AA2024[56] and AA7075[141] showed sharp textures with two or more components, leading to a high fraction of HAB (85-90%).

D. Second phase particles

The precipitation behavior of heat treatable aluminum alloys during FSW is dependent on the specific alloys to be joined.[20, 22] Dissolution and re-precipitation has been reported in AA7075 during FSW, and the cooling rate is so low that large precipitates formed.[22]
However, because of the low quench sensitivity of AA6063, only precipitate dissolution occurred in the weld nugget of this material.\textsuperscript{[20]}

As for different macroscopic zones, previous studies consistently showed different precipitation behaviors because of different thermal cycles experienced in these zones during FSW.\textsuperscript{[20, 36, 37]} Peak temperature in HAZ is lower than the solution temperature, therefore severe particle coarsening occurs in this zone. In TMAZ, of a higher peak temperature, both precipitate coarsening and dissolution occur, and re-precipitation takes place heterogeneously on the dislocations and subgrain/grain boundaries during cooling. In the weld nugget, the peak temperature is so high that precipitates can be fully dissolved into solid solution during FSW/P and re-precipitated out during subsequent cooling.

In non-heat treatable aluminum alloy AA5083, the peak welding temperature is not high enough to dissolve small particles, mainly Al\textsubscript{6}(Mn,Fe).\textsuperscript{[38]} The coarsening of these particles and the break-up of large constituent particles to small ones resulted in a higher volume fraction and larger average particle diameter of small pinning particles in the weld nugget than in the parent metal.
A.3. Observed Annealing Behavior of FSW Structures

Post-FSW heat treatment may be carried out for several reasons. It may be conducted for further forming (i.e. using FSW tailor-welded blanks for GM’s Quick Plastic Forming process at elevated temperature\cite{46}); or it may be an incidental effect of, for example, the automotive paint-bake hardening cycle. Grain growth to sizes as large as several millimeters can occur during heat treatment in both heat-treatable alloys, such as AA7050\cite{32}, AA6082\cite{142} and AA2024\cite{56} non-heat treatable aluminum alloy, such as AA5083\cite{89} and also commercial pure AA1100\cite{52} of low particle density.

The formation of these coarse grain structures was found to be discontinuous\cite{30-32, 52, 53} because an obvious initiation step was observed\cite{32}. In this “initiation” step, several much larger grains formed in an area surround by stable smaller grains. This “initiation” step is referred to as “nucleation” in many studies, borrowing the nomenclature from the phase transformation process\cite{59-61}. The subsurface region, which directly contacts the welding tool shoulder, is the one most often reported to initiate coarse grain growth\cite{32, 52, 54}. Coarse grains may also form from the bottom of the nugget\cite{30, 32, 54} and on the edges of the nugget to the WN-TMAZ boundaries\cite{32, 52}. The regions most susceptible to discontinuous grain coarsening are those consisting of smaller grains and higher %LAB in the nugget\cite{30, 52, 55, 56}.

Previous study on AA5083, shown in Fig. 2 and Fig. 3\cite{89} illustrated the role of FSW conditions and heat-treatment time on the occurrence of coarse grain structure. Coarse
A two-stage discontinuous process of grain structure evolution can be primary recrystallization, secondary recrystallization, or discontinuous subgrain growth. Discontinuous subgrain growth, only associated with the low angle grain boundary
migration only,\textsuperscript{57, 143} is not normally observable. It would be easily overwhelmed by \textit{primary recrystallization}, considering the latter is about 100 times faster, or it would behave as the nucleation stage for \textit{primary recrystallization} in a large orientation-gradient microstructure.\textsuperscript{57, 144} Further discussion on discontinuous subgrain growth will be avoided in this paper.

Primary recrystallization and secondary recrystallization can be distinguished by their driving force.\textsuperscript{62, 66} The driving force of primary recrystallization is the stored strain energy in the form of dislocation content within grains.\textsuperscript{60} It is associated with the formation of new strain-free grains in a deformed matrix, characteristic of the evolution of their boundaries from LAB to HAB, and the growth of this new set of grains. By contrast, the interfacial energy of grain boundaries is the driving force for secondary recrystallization, which takes place after primary recrystallization with normal grain growth inhibited, except in several selected grains.\textsuperscript{62, 66} Those grains grow excessively to diameters significantly larger than the surrounding grain size, being able to reach the order of several millimeters. Because this process involves no new grain formation (bounded by HAB), and in order to distinguish it from normal grain growth (NGG), it is sometimes called “abnormal grain growth” (AGG) or “discontinuous grain growth”.\textsuperscript{62, 63, 67} For simplification and clear presentation, the terminology of AGG instead of secondary recrystallization is used here.

Nucleation and growth are two characteristic steps in a discontinuous-phase transformation process.\textsuperscript{57, 58} Similar steps also exist in primary recrystallization and
AGG.\textsuperscript{[59-61]} However, unlike in phase transformation, there is no new phase (different from the matrix in structure or composition) formed during either primary recrystallization or AGG.\textsuperscript{[60, 61]} Borrowing the nomenclature from discontinuous phase transformation, the “initiation” step of primary recrystallization and AGG is also referred to as “nucleation”. In this paper, “nucleation” and “nuclei” are used in order to be consistent with the previous literature. But it should be noted that nucleation here involves no nuclei formation through thermal fluctuation and atom-by-atom construction. The nuclei in primary recrystallization are either recovered cells or subgrains pre-existing in a deformed microstructure;\textsuperscript{[60, 61]} those in AGG are actually grains already present in the initial grain structure.\textsuperscript{[62, 63]} Both primary recrystallization and AGG can produce a coarse grain structure. Their mechanisms on this are introduced in the following sections.

A. Primary recrystallization

The formation of coarse grain structure by primary recrystallization is actually a result of the competing of nucleation rate (\(N\)) and nuclei growth rate (\(G\)). A smaller ratio of \(N/G\) leads to fewer nuclei and therefore a coarser final grain structure after these nuclei grow to impingement.\textsuperscript{[59, 64, 65]}

Critical strain effect in the primary recrystallization is one of the methods to produce coarse grain structure.\textsuperscript{[59, 64, 65]} Critical strain is defined to be the minimum amount of strain needed to have primary recrystallization occur within reasonable length of
annealing time.\cite{59,64} Within such length of time, primary recrystallization can not occur if the experienced deformation is below the critical strain. The formation of the coarse grain structure is because the decrease of nucleation rate ($\dot{N}$) is more rapid than the decrease of the nuclei growth rate ($\dot{G}$) with the decrease of the induced amount of strain.\cite{59,64} At the critical strain, the ratio ($\dot{N}/\dot{G}$) of the nucleation rate to the nuclei growth rate is the smallest, resulted in minimum amount of nuclei grow to impingement. Therefore, coarse grain structure can be formed.

B. Abnormal grain growth

AGG is a grain structure coarsening process involving the growth of several grains with the rest of the grains relatively unchanged. Two necessary conditions for AGG are: 1) the inhibition of normal grain growth and 2) growth of several grains is allowed by the microstructural conditions.\cite{63} The character of heterogeneity associated with AGG is actually the result of the un-uniformity of boundary migration. Only limited number of boundaries (belonging to nuclei) can move with most of others relatively stable. AGG is sometimes called “exaggerated grain growth” because it is common to see grains as large as millimeters or even centimeters produced by AGG.\cite{63} According to various NGG inhibition forms, AGG can be classified as follows:

1) Particle-induced inhibition
As shown by Zener about half a century ago a force can be exerted by particles on boundaries, this force is balanced with the driving force for the grain growth, and with enough amount of particle pinning force the normal grain growth is then able to be inhibited. Particles as the source for the grain growth inhibition is frequently observed and the theoretical studies were also abundantly carried out.\textsuperscript{[69-72, 145, 146]} Humphreys’ grain growth model as a generally successful model will be introduced in next section.

2) Texture-induced inhibition

In a grain structure with a pronounced texture, there is a high probability that the neighboring grains have similar orientation, leading to a high fraction of low angle boundary (LAB, $\theta_{\text{mis}} < 15^\circ$). The low-energy nature of LAB leads to low stored energy for providing NGG driving force. In addition, the mobility of LAB is low. Therefore, a highly textured grain structure tends to have a sluggish normal grain growth rate. Once there is a grain with normal high angle boundary (HAB, $\theta_{\text{mis}} > 15^\circ$, high mobility and high energy) or special boundary (high mobility and low energy) and carrying a size advantage, its growth can be much faster than the rest of grains. This grain is then appeared to be one of the selective grains to grow preferentially, and abnormal grain growth occurs.

3) Solute-induced inhibition

Solute segregation normally occurs in solid solution alloys. Solute atoms either tend to move toward or away from the grain boundaries according to their
interaction energy with the matrix atoms. The migration of grain boundary breaks the symmetry of the solutes distribution on the two sides of grain boundary. This asymmetry will in turn exert a force on the boundaries, leading to the pinning of boundary movement. Even though solute-induced inhibition is seldom reported to be the cause for AGG, above mentioned mechanism is still operative and should be considered.\textsuperscript{[63]}

4) Surface effect in thin samples

When the thickness of the sample is close to the dimension of the inside grains, the pinning force from boundary grooves on the sample surface turns to be important.\textsuperscript{[62, 63]} Boundary migration will be inhibited because it will increase the total boundary area if it moves away from the grooves. Additionally, for thin samples under specific atmosphere, orientation-dependent surface energy stands out to be an important driving force for grain growth.\textsuperscript{[62, 63, 67]} Grains with low surface energy grow as AGG nuclei when most other grains stay relatively unchanged.
APPENDIX B: HARDNESS MEASUREMENT, HV MAP, AND SCATTER DETERMINATION

Hardness testing was conducted using a Buehler Micromet II digital micro hardness tester along the centerline of the weld from top surface to bottom on the LT-ST plane (see Fig. 1b) on the mechanically polished samples, with a load of 50 g force and dwell time of 15 seconds. At least five Vickers microhardness test values were recorded at each distance from the top surface near the centerline and were averaged after excluding the maximum and minimum values in order to minimize the effect of local conditions, such as locating the indentation on a cluster of particles. The indentation size is about 40x40 µm². Limited by the indentation size and the distance required to be displaced for the next hardness test (at least 2.5 times of the diagonal), hardness of ten through-thickness depth positions were measured for Vickers hardness. Therefore, 5 x 10 hardness measurements were made on each welded sample. Fig. 31 schematically shows the small center region for the Hv measurement. Hardness maps, calculated averages, and scatters of various FSW locations are shown from Table 11 to Table 21. For both the as-received and fully annealed base metal, hardness test is similarly done on the sectioned LT-ST plane. All the measurements were then averaged to a single hardness value.
Table 11. As-weld H1 Hv Map (Average $\sigma = 1.1$)

<table>
<thead>
<tr>
<th>Depth (mm)</th>
<th>Mesure 1</th>
<th>Mesure 2</th>
<th>Mesure 3</th>
<th>Mesure 4</th>
<th>Mesure 5</th>
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Table 12. As-weld H2 Hv Map (Average $\sigma = 0.6$)

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Table 13. As-weld H3 Hv Map (Average $\sigma = 1.2$)

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Table 14. H1 after APF-HT Hv Map (Average $\sigma = 1.2$)

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Table 15. H2 after APF-HT Hv Map (Average $\sigma = 0.9$)

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Table 17. Hardness and Scatter of TOP-1/3 and MID-2/3 for As-weld H2

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Table 18. Hardness and Scatter of TOP-1/3 and MID-2/3 for H2 after QPF-HT

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Table 19. Hardness and Scatter of TOP-1/3 and MID-2/3 for H2 after Annealing at 350°C for 1hr

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Table 20. Hardness and Scatter of TOP-1/3 and MID-2/3 for H2 after Annealing at 350°C for 24hr

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Table 21. Hardness and Scatter of TOP-1/3 and MID-2/3 for H2 after Annealing at 380°C for 24hr

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APPENDIX C: EBSD SETTING DETERMINATION FOR LAB MEASUREMENT

This will explain the choosing of binning setting, step size, and cutoff value for LAB. Images of EBSD Kikuchi patterns were usually binned before doing Hough Transform. Binning setting is different for the cameras used to collect pattern. There are four settings I can choose from, i.e. 1x1, 2x2, 4x4, and 8x8. The term binning refers to the averaging of pattern image pixels, and effect of it is reducing the image resolution without changing its size. For example, under a 4x4 binning, a total of 16 nearby 4 by 4 array of pixels will be averaged together into a single pixel. Therefore both resolution and the image size are reduced. The purpose of this performance is: 1) to speed up the processing by decreasing the amount of data to be processed by computer, and 2) to increase the intensity of the pattern image.

Because of the first effect of binning, a large binning requires less dwell time at a single scanning point, which will also reduce the total time to run EBSD scan. However, lower resolution of pattern image leads to a less accurate Hough peak position determination. This will in turn affect the accuracy of orientation determination. A better image
resolution (or smaller binning) would be a better choice for LAB measurement. Unfortunately, the binning setting is not provided in many previous studies.

EBSD scans were conducted on a fully annealed base metal (annealed at 380 °C for 24 hr). At a constant step size of 0.25μm, scans were run at three different binning setting: 2 x 2, 4 x 4, and 8 x 8. Pattern images after binning and total scanning times are shown in Table 23. 4.2hr, 2.3 hr, and 1.7 hr are the scanning time for binning 2 x 2, 4 x 4, and 8 x 8 respectively.

Results of LAB analysis was found to be very dependent on binning setting and the definition of LAB, Table 22. Because a higher resolution pattern image can give a more accurate %LAB measurement, it can be assumed that a smaller %LAB value is measured with a smaller binning is correct. However, a big fluctuation on the %LAB value with the change of binning if the LAB is defined to be 2°-15°, leads to this definition not trustable.

Therefore during processing EBSD data, 3°-15° is defined to be LAB. 4x4 binning is used for most cases. This is performed mostly from the time-efficiency point of view. For such a well annealed sample, 2.3 hr is enough. A longer scanning time will be needed for FSW regions with a more deformed state and a finer grain structure. For a scan on FSW, a typical time is 6hr under 4x4 setting. It will be too long if 2x2 binning is used. Additionally, absolute %LAB value measurement is not the thing we attempted to do. Firstly, limited by the technique itself, no matter LAB defined to be 3°-15° or 3°-15°, there are always boundaries missing in the measurement. Secondly, for the purpose of
this paper, as long as the setting for all the measurement is same, a reasonable comparison can be mad among various locations.

Table 22. Grain size and %LAB(5°-15°, 3°-15°, and 2°-15°) according to binning setting

<table>
<thead>
<tr>
<th>Binning</th>
<th>%LAB (5°-15°)</th>
<th>%LAB (3°-15°)</th>
<th>%LAB (2°-15°)</th>
<th>Grain Size (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2x2</td>
<td>3.4%</td>
<td>5.3%</td>
<td>8.6%</td>
<td>4.41</td>
</tr>
<tr>
<td>4x4</td>
<td>3.6%</td>
<td>6.5%</td>
<td>13.2%</td>
<td>4.32</td>
</tr>
<tr>
<td>8x8</td>
<td>3.9%</td>
<td>7.1%</td>
<td>11.9%</td>
<td>4.46</td>
</tr>
</tbody>
</table>
Table 23. EBSD pattern images and total scanning time for a 50x50 μm² scan according to binning setting

<table>
<thead>
<tr>
<th>Binning</th>
<th>Pattern Image</th>
<th>Total time for a 50x50 μm² scan (hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 x 2</td>
<td><img src="image" alt="Pattern Image" /></td>
<td>4.2</td>
</tr>
<tr>
<td>4 x 4</td>
<td><img src="image" alt="Pattern Image" /></td>
<td>2.3</td>
</tr>
<tr>
<td>8 x 8</td>
<td><img src="image" alt="Pattern Image" /></td>
<td>1.7</td>
</tr>
</tbody>
</table>
APPENDIX D: EBSD GRAIN SIZE CHARACTERIZATION AND SCATTER DETERMINATION

EBSD scans were conducted using TSL OIM Data Collection Program on a Philips XL-30 Environmental SEM (ESEM) with field emission gun (FEG), which is equipped with a CCD camera. SEM was operated at 25 kV and with a beam spot size of 5. The step size for the scans is set to be 0.25 µm and a scan area of 50 by 50 µm² for grain structure determination, the binning for the Kikuchi pattern image to be 4 x 4. Hough peaks of scanned points were collected first. Using known the structures of Al, (Fe,Mn)Al₆, Mg₂Si, Mg₅Al₈, and (Fe,Cr)Al₇, and Al₂O₃ phases, the initially collected Hough-peak data were used to generate orientation data for identifying these phases.

The orientation data were analyzed using TSL OIM Analysis Program, with a 5-degree grain tolerance angle. The grain size statistics are generated directly by the TSL OIM Analysis Program. The area of each grain (Aᵢ) is collected, and then an average area of all grains (a total of N grains) was calculated from all grains ($\bar{A} = \frac{1}{N} \left( \sum_{i=1}^{N} A_i \right)$). The mean equivalent 3D grain diameters were then calculated as shown in Chapter 2.
The accuracy and reproducibility of grain sizes were established by performing 3 scans at adjacent regions in specimens of H1-Mid and H3-Mid. Scans from H1-Mid are shown in Fig. 32 with grain sizes shown on the upper right of each map. And scans from H3-Mid are in Fig. 33. Grain sizes of the adjacent locations are collected into Table 24. The average sizes and the standard deviations were then calculated and are listed to the right of the table.

Table 24. Grain sizes from EBSD scans on adjacent area of H1 and H3 middle regions for grain size scatter determination (µm)

<table>
<thead>
<tr>
<th></th>
<th>Measure 1</th>
<th>Measure 2</th>
<th>Measure 3</th>
<th>Average</th>
<th>Stddev</th>
</tr>
</thead>
<tbody>
<tr>
<td>H1-Mid</td>
<td>1.62</td>
<td>1.70</td>
<td>1.63</td>
<td>1.65</td>
<td>0.04</td>
</tr>
<tr>
<td>H3-Mid</td>
<td>2.59</td>
<td>2.45</td>
<td>2.65</td>
<td>2.56</td>
<td>0.10</td>
</tr>
</tbody>
</table>
APPENDIX E: TEM AND SEM PARTICLE CHARACTERIZATION

In order to apply Humphreys’ Grain Growth model, TEM and SEM is used here for the quantification of size and volume fraction of second phase particles. Because of the limitation of the SEM resolution, particles with circle diameter larger than 0.2 µm were measured by SEM. On the other hand, the measurement volume of TEM is very small only about 10 µm\(^3\). Therefore the statistics for larger particles (\(> ~ 0.5 \text{ µm}\)) is not good enough for quantification. A combination of these two techniques is used here. The justification of the combination, the using of the cutoff value of 0.2 µm, and the quantification of the particle dispersion level (\(f/d\)) will be shown in the following Appendix.

E.1. TEM Characterization

Thin-foil disk transmission electron microscopy (TEM) specimens cut from various weld locations of the welds were first mechanically thinned to about 120 µm and then electro-polished using a solution mixture of 70% methanol and 30% nitric acid at about 12 °C. A Philips CM-12 transmission electron microscope (TEM) was used, operating at 120 kV.
Convergent Beam Electron Diffraction (CBED) was used for sample thickness estimation. Magnification is 28000X for each photomicrograph. These micrographs were then scanned into digitized pictures with an image resolution of 400 dpi. A typical micrograph is shown in Fig. 34a, which is from H2-Mid. Two more micrographs showing the H2-Top and H2-Mid regions are shown in Fig. 24. These digitized micrographs were then processed and bi-level thresholded to show second phase particles using FoveaPro imbedded in Photoshop. Those touching particles were separated by hand. The micrograph after image processing is shown in Fig. 34b. Assuming all particles are spherical, size and the volume fraction of each particle were collected for further analysis. About 80 particles were typically recorded for each location.

E.2. SEM Characterization

A Sirion high-resolution FEG-SEM was operated at 5 kV under backscattered electron (BSE) mode for second phase particle analysis. According to literature,[38, 73-80] (Fe,Mn)Al<sub>6</sub>, Al<sub>2</sub>O<sub>3</sub>, Mg<sub>2</sub>Si, (Fe,Cr)Al<sub>7</sub>, Mg<sub>5</sub>Al<sub>8</sub>, Al<sub>12</sub>(Fe,Mn)Si<sub>3</sub>, and Al<sub>11</sub>Cr<sub>2</sub>, can be existing in AA5083. Z-contrast is the most important image contrast in BSE mode. Therefore Fe, Cr, and Mn-containing particles (larger average Z, showing bright) can be revealed and characterized using BSE detector.

Particles of smaller average Z: Mg<sub>5</sub>Al<sub>8</sub>, Mg<sub>2</sub>Si, and Al<sub>2</sub>O<sub>3</sub>, are not able to be quantified readily. However, because Mg<sub>5</sub>Al<sub>8</sub> is thermally unstable,[83] Mg<sub>5</sub>Al<sub>8</sub> may dissolved already at QPF-HT temperature as described in the Introduction Chapter. Incoherent
constituent Mg$_2$Si particles were coarse and non-uniform distributed\textsuperscript{[73]}, and have been reported to be unimportant in pinning grain boundary.\textsuperscript{[74]} Even through Al$_2$O$_3$ particles identified by EBSD are about 0.4 µm (see Table 26), previous study showed most of Al$_2$O$_3$ particles are less than 0.1 µm. Therefore, the missing of Mg$_5$Al$_8$, Mg$_2$Si, and Al$_2$O$_3$ in SEM characterization under BSE mode can be insignificant.

It was noticed that cutting off particles with 2-D equivalent diameter less than 0.2 µm is not equivalent to the initial purpose of cutting off particles less than 0.2 µm in 3-D, because any particle with size larger than 0.2 µm can have a 2-D intersection size less than 0.2 µm. The result of this is a slight overestimate on the average size. The amount of the overestimation depends on many factors, especially particle size distribution and particle shape. Therefore it is difficult to know the exact amount.

Additionally, the 3-D and 2-D conversion is based on exact Stereology. However, the SEM measurement is not an exact 2-D measurement. Firstly, BSE signal comes from a certain depth depending on the sample composition and energy of electron beam. The BSE information depth can be estimated from the Kanaya-Okayama (KO) Range, about 30% of KO Range. Therefore for an aluminum sample, the BSE information depths for 10 kV and 5kV beam are about 400 nm and 120 nm respectively.\textsuperscript{[107]} A particle of size smaller than the dimension of information depth sitting within the information depth is actually imaged by projection like under TEM. Secondly, the final polish step using
colloidal silica solution which is base in vibratory polisher may remove matrix aluminum first leaving some particle protruding out. This is confirmed by microphotographs under secondary electron (SE) mode. The edge of some particles is brighter than the center region. This is believed to be the effect of diffusion contrast on the edges, one of the types of topographic contrast. These two effects will enlarge the particle size and volume fraction as well, but again difficult to estimate the amount.

On the other hand, some black regions can be seen under SE mode with shape similar to particles. They are normally large ones (1-10 µm), Fig. 22. These are believed to be caused by the removing of particles during polishing. This effect can result in smaller measured sizes and smaller volume fractions. All these above mentioned issues are not easy to avoid, but it is assumed here that the effects of them neutralized themselves that the method used here is still a good estimate for the diameter and volume fraction of particle larger than 0.2 µm.

As mentioned above, for an aluminum sample the BSE information depths for 10 kV and 5 kV beam can be estimated to be about 400 nm and 120 nm respectively. The larger information depth, the more the measurement deviates from the exact 2D condition. Therefore a small voltage of 5kV is used here.

For each location, at least 5 micrographs were taken with a magnification of 1500X, i.e. about 3000 particles measured at each location. The size of each pixel is 0.016 µm.
Limited by the resolution of SEM, only particles larger than 0.2 µm were characterized by SEM. Particles smaller than 0.2 µm, TEM is used for the characterization.

A general procedure is as follows. 1) Adjust the microscope to optimum condition at 10 kV under BSE mode and set the magnification to 1500X. 2) At each location, take 5 digitized micrographs on regions nearby (but no overlapping). 3) Using FoveaPro imbedded in Photoshop, digitized micrographs were then processed and bi-level thresholded to show second phase particles. 4) 2-D equivalent circle diameter and the area of each particle was recorded for further analysis. A typical micrograph taken from H2-Top is shown in Fig. 35a. Its corresponding micrograph after the image processing is shown in Fig. 35b, from which the size of each particle and the total volume fraction are readily achievable.
APPENDIX F: THERMO-MECHANICAL SIMULATIONS OF FSW FOR AA5083-H18

(* Written by Dongun Kim from SNU. This is included here for the completeness of the discussion)

The commercial computational fluid dynamics (CFD) code, FLUENT 6.2, is being used to solve the continuity, momentum and energy equations, which are expressed in terms of non-linear partial differential equations based on the finite volume method (in the Eulerian formulation). The flow behavior of the material is assumed to be of temperature-dependent rigid-perfect-plasticity based on the Mises yield function. The temperature-dependent yield stress for AA5083-H18 used in this work is shown in Table 25 (experimental data provided by OSU). The measured properties of AA5052-H32\cite{148} were proportionally modified for use for AA5083-H18, considering the ratio between the room temperature data.\cite{148} In order to improve the result compared with the experimental one, the modified thermal conductivity and heat capacity were used.

The conduction heat transfer coefficient of 30 W/m$^2$·°C is used for the top and sides of the workpiece which is typical for natural conduction between aluminum and air.\cite{149} As for the bottom of the work-piece, the conduction coefficient is assumed to be 2000
96 W/m²·°C just below the tool assuming that heat transfer increases in that region because of the mechanical pressure applied to the pin.\(^{[150]}\) For the remaining bottom surface, 200 W/m²·°C is assumed.

In the Eulerian approach, the rotating tool is fixed in space while the material flows along the weld direction. Three welding conditions were simulated: 1000 rpm by 1.7 mm/s, 1000 rpm by 5.0 mm/s, and 1500 rpm by 2.5 mm/s. Laminar and incompressible flow are assumed. At the tool surface, the material is assumed to rotate at the tool rotating speed (therefore, heat generation by the interfacial friction is zero under this assumption), without other realistic boundary conditions to prescribe the tool rotation condition, which renders some of simulation results not so credible.

Table 25. Temperature dependent yield stress (OSU data) AA5083-H18 (\( \dot{\varepsilon} = 10^{-2.5} \))

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Yield Stress (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>440</td>
</tr>
<tr>
<td>100</td>
<td>437</td>
</tr>
<tr>
<td>200</td>
<td>364</td>
</tr>
<tr>
<td>300</td>
<td>181</td>
</tr>
<tr>
<td>400</td>
<td>61</td>
</tr>
<tr>
<td>410</td>
<td>20 (assumed)</td>
</tr>
<tr>
<td>540</td>
<td>20 (assumed)</td>
</tr>
<tr>
<td>590</td>
<td>0 (assumed)</td>
</tr>
</tbody>
</table>
APPENDIX G: SECOND PHASE PARTICLE CHARACTERIZATION USING EBSD

The effects of particle shape and distribution are important factors in recrystallization and grain growth\textsuperscript{110}, with the largest influence from high densities of particles of size smaller than 1.0 µm.\textsuperscript{62, 75, 87} Second phases can be resolved by EBSD phase imaging, Fig. 14. (Note that the analyzed areas were chosen to avoid very large, very sparse constituent particles.) It is apparent that the number and area fraction of particles varies inversely with grain size. Thus, the upper regions have more particles than the middle regions and for either region the lower heat-input cases have more particles (with the H2-Mid specimen showing exceptionally low second-phase content).

The visual impression from Fig. 14 are quantified and confirmed by Table 26 and Table 27. Table 26 shows that the particle sizes are approximately the same for all cases, while Table 27 shows that the area fractions of particles are higher in the top regions and low heat-input cases, with again H2-M showing exceptionally low second-phase content. The scatter for measured particle sizes (standard deviation) for the particle sizes is 0.01,
excluding (Fe,Mn)Al$_6$. The larger scatter (= 0.05) for (Fe,Mn)Al$_6$ particles occurs because of a bimodal distribution of the particle sizes.

Table 26. Average sizes of second-phases particles at various locations (In µm; standard deviation: 0.01 µm)

<table>
<thead>
<tr>
<th>Group #</th>
<th>Phase</th>
<th>H1-T (AGG)</th>
<th>H1-M (AGG)</th>
<th>H2-T (AGG)</th>
<th>H2-M</th>
<th>H3-T (AGG)</th>
<th>H3-M</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(Fe,Mn)Al$_6$</td>
<td>0.68</td>
<td>0.45</td>
<td>0.52</td>
<td>0.66</td>
<td>0.50</td>
<td>0.53</td>
</tr>
<tr>
<td></td>
<td>Al$_2$O$_3$</td>
<td>0.49</td>
<td>0.39</td>
<td>0.40</td>
<td>0.35</td>
<td>0.38</td>
<td>0.38</td>
</tr>
<tr>
<td>2</td>
<td>Mg$_2$Si</td>
<td>0.50</td>
<td>0.43</td>
<td>0.40</td>
<td>0.37</td>
<td>0.39</td>
<td>0.42</td>
</tr>
<tr>
<td></td>
<td>(Fe,Cr)Al$_7$</td>
<td>0.52</td>
<td>0.43</td>
<td>0.40</td>
<td>0.38</td>
<td>0.39</td>
<td>0.39</td>
</tr>
<tr>
<td>3</td>
<td>Mg$_5$Al$_8$</td>
<td>0.52</td>
<td>0.44</td>
<td>0.43</td>
<td>0.39</td>
<td>0.40</td>
<td>0.40</td>
</tr>
<tr>
<td>Avg.</td>
<td></td>
<td>0.54</td>
<td>0.43</td>
<td>0.43</td>
<td>0.41</td>
<td>0.43</td>
<td>0.42</td>
</tr>
</tbody>
</table>

Average Top: 0.46  Average Mid: 0.43  Average AGG: 0.45  Average No-AGG: 0.43

Table 27. Area fractions of second-phases particles at various locations (Standard deviation: 0.002)

<table>
<thead>
<tr>
<th>Group #</th>
<th>Phase</th>
<th>H1-Top (AGG)</th>
<th>H1-Mid (AGG)</th>
<th>H2-Top (AGG)</th>
<th>H2-Mid</th>
<th>H3-Top (AGG)</th>
<th>H3-Mid</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(Fe,Mn)Al$_6$</td>
<td>0.010</td>
<td>0.014</td>
<td>0.016</td>
<td>0.002</td>
<td>0.013</td>
<td>0.014</td>
</tr>
<tr>
<td></td>
<td>Al$_2$O$_3$</td>
<td>0.024</td>
<td>0.017</td>
<td>0.023</td>
<td>0.004</td>
<td>0.016</td>
<td>0.013</td>
</tr>
<tr>
<td>2</td>
<td>Mg$_2$Si</td>
<td>0.030</td>
<td>0.018</td>
<td>0.024</td>
<td>0.008</td>
<td>0.015</td>
<td>0.010</td>
</tr>
<tr>
<td></td>
<td>(Fe,Cr)Al$_7$</td>
<td>0.016</td>
<td>0.022</td>
<td>0.015</td>
<td>0.006</td>
<td>0.012</td>
<td>0.014</td>
</tr>
<tr>
<td>3</td>
<td>Mg$_5$Al$_8$</td>
<td>0.045</td>
<td>0.024</td>
<td>0.031</td>
<td>0.011</td>
<td>0.022</td>
<td>0.009</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>0.125</td>
<td>0.095</td>
<td>0.109</td>
<td>0.031</td>
<td>0.078</td>
<td>0.060</td>
</tr>
</tbody>
</table>

Average Top: 0.021  Average Mid: 0.012  Average AGG: 0.019  Average No-AGG: 0.009

Energy dispersive X-ray spectroscopy (EDS) analysis of particles larger than 3 µm in diameter was carried out. It reveals predominantly Al, with significant Mn and Fe, and
very little Mg in these particles, consistent with the literature, and with EBSD phase contrast maps, Fig. 14. Similar analysis of the matrix shows predominantly Al with Mg content higher than in the particles.
APPENDIX H: THEORETICAL TREATMENT ON

PARTICLE DISPERSION LEVEL (F/D) ON A PARTICLE

DISPERSION WITH VARIOUS SIZES

In literature for grain growth and recrystallization models, a single set of mono-sized particles are normally assumed for simplification. However, this is impossible for a real material, in which particles within a large range of sizes can be found. In the materials under study here, AA5083, particle size is from 30 nm up to 10 μm. How to treat f/d is therefore raised as an important issue when applying grain growth and recrystallization models. However, in many literatures, an average d is used for calculating dispersion lever. It can be shown as follows that this is not accurate.

In a hypothetical material of volume $V_{Tot}$, there are $n$ sets of particles. For $i^{th}$ set of particles $(i = 1, 2 \ldots n)$, diameter is $d_i$, and total particle number is $n_i$.

(1) For a random distribution of mono-sized particles of diameter, $d$, and volume fraction, $f$, the Zener pinning pressure is in the form of $P_z = \frac{\alpha f \gamma}{d}$, where $\alpha$ is constant and $\gamma$ is the grain boundary energy. If there are $n$ sets of particles, $d_i$
with a volume fraction of \( f_1 \), and \( d_2 \) with a volume fraction of \( f_2 \), and \( d_n \) with a volume fraction of \( f_n \), the total Zener pressure \( P_{Z_{tot}} \):

\[
P_{Z_{tot}} = \alpha \gamma \frac{f_{Tot}}{d_{Eff}} = \alpha \gamma \left( \frac{f_1}{d_1} + \frac{f_2}{d_2} + \cdots + \frac{f_n}{d_n} \right)
\]

Eqn. 11

Therefore:

\[
\frac{f_{Tot}}{d_{Eff}} = \frac{f_1}{d_1} + \frac{f_2}{d_2} + \cdots + \frac{f_n}{d_n} = \sum_{i=1}^{n} \frac{f_i}{d_i}
\]

Eqn. 12

And the effective Zener particle size for a set of particles with various sizes can be shown as:

\[
d_{Eff} = \sum_{i=1}^{n} f_i / \sum_{i=1}^{n} \frac{f_i}{d_i}
\]

Eqn. 13

It is not a simple average size of all particles. The effective Zener size is related to the volume fraction of each set of particles.

(2) Following the general assumption that all particles are spherical, for the \( i^{th} \) set of particles \((i = 1, 2 \cdots n)\) with a diameter of \( d_i \), its volume fraction, \( f_i \), can be obtained through:

\[
f_i = \frac{n_i \times \frac{4}{3} \pi \left( \frac{d_i}{2} \right)^3}{V_{Tot}}
\]

Eqn. 14

Therefore,
Equation 15

\[ f_{\text{Tot}} = \sum_{i=1}^{n} f_i \frac{n_i \times 4 \pi \left( \frac{d_i}{2} \right)^3}{V_{\text{Tot}}} \]

\[ \sum_{i=1}^{n} f_i d_i = \sum_{i=1}^{n} \frac{n_i \times 4 \pi d_i^2}{V_{\text{Tot}}} \]

\[ d_{\text{Eff}}^Z = \frac{\sum_{i=1}^{n} f_i}{\sum_{i=1}^{n} \frac{f_i}{d_i}} = \frac{\sum_{i=1}^{n} n_i \times d_i^3}{\sum_{i=1}^{n} n_i \times d_i^2} \]

Equation 20 shows that if all particles are spherical with a size range, the effective particle size is determined by both the sizes and the number of particles of each size. And apparently, this effective size is not the average size especially when the size range is large. \( d_{\text{Eff}}^Z \) is getting closer to the average \( d \) when the size range is getting smaller.
APPENDIX I: THE OVERLAPPING OF PARTICLE SIZE DISTRIBUTIONS BY SEM AND TEM

In order to justify the particle size cutoff value of 200 nm and test the consistency of TEM and SEM particle size characterization, following analysis on the particle size distributions measured by SEM and TEM were performed. In order to simplify the analysis and discussion, all particles were assumed to be spherical.

Particles imaged by TEM are through projection type. Therefore, TEM shows the real sizes of the particles. However, SEM micrographs were taken on a 2D-section of the sample. 2D equivalent circle size can be obtained from each particle shown on the polished plane. In order to compare the distributions measured by two different techniques, a conversion of particle size either from 2D to 3D or 3D to 2D is needed.

TEM measured 3D particle diameters distributions were chosen to be converted to 2D diameters distribution here for reasons as follows. (1) SEM measured particles have a large size range of 0.02 µm up to about 7 µm (even though credibility of the sizes of small particles is low). On the other hand, the measured size range for TEM is about
from 20 nm to 400 nm. For a reasonable distribution comparison, the bin size should be at least 50 nm, with which there will be 8 bins for a TEM measured distribution. But for such a bin size, there will be 140 bins for a SEM distribution. Because information of 30 particles is needed for each bin, about 40000 particles should be included for a distribution. (2) The classical particle size distribution unfolding method will have error built up to the bins of the smallest particle sizes, where are the ones we will use for the comparison with the TEM measurement. (3) The conversion of 3D to 2D distribution is, on the other hand, much easier and more credible.

The data obtained from the middle region of H2 sample is used here for illustrating the process. For other locations, the results will be shown at the end of this appendix. There are a total of 92 particles measured by TEM on this region. Steps are listed as follows.

1. A particle size distribution is firstly drawn, in Fig. 36, in the form of particle numbers as a function of particle size. Particles were binned into 10 classes with the bin size is 50 nm, and size shown in figure is the medium of the bin range. That is, particles with a size of 0 nm to 50 nm fall into the first bin labeled as \( d = 25 \) nm; particles of 50 nm to 100 nm into the second bin labeled as \( d = 75 \) nm, and so on. There are a total of 10 bins.

2. For the distribution above, it can be simplified to assume there are ten sets of particles with size from 25 nm to 475 nm. For a single set of particles, if treated
as mono-sized spheres, the probability of cutting a sphere of diameter of D within a given interval can be shown as follows.

For a distribution of k bins, the diameter interval is \( \Delta d = D/k \); and:

\[
d_i = \Delta d
\]

\[
d_i = i \cdot \Delta d \quad (i = 1, 2, \ldots, k)
\]

\[
d_k = k \cdot \Delta d = D
\]

The probability of cutting circle within the diameter interval of \( d_{i-1} \) to \( d_i \) is then:

\[
p_i = \left( \sqrt{D^2 - d_{i-1}^2} - \sqrt{D^2 - d_i^2} \right)/D
\]

Eqn. 18

Therefore,

\[
p_i = \sqrt{1 - (i-1)^2/k^2} - \sqrt{1 - i^2/k^2}
\]

Eqn. 19

For various number of bins (or k values), there is a different set of \( p_i \) values. For the purpose here, \( p_i \) values for \( k = 1, 2, 3 \ldots 10 \) were tabulated here in Table 28. The sum of each column is 1.

For each set of mono-sized particles, there will be k bins of sectioned 2D circle sizes. For the bin of largest size, the probability is \( p_k \). For example, if we divide sectioned 2D circle sizes into 10 bins, the probability found for the bin of the largest size is \( p_{10} \), equal to 0.4359 shown in Table 28. Because the number of each set of mono-sized particles is known (shown in distribution of 3D sizes) and
assuming they will all show in a 2D section, the probability for the bin of largest size, $p_k$ in each column of $k$, should be 1. And the rest of the probability is then turned into $p_{k-1}/p_k$. A new matrix, denoted as $M$, is obtained and shown in Table 29.

Table 28. Tabulated probability values for various bin number $k$

<table>
<thead>
<tr>
<th></th>
<th>$k = 1$</th>
<th>$k = 2$</th>
<th>$k = 3$</th>
<th>$k = 4$</th>
<th>$k = 5$</th>
<th>$k = 6$</th>
<th>$k = 7$</th>
<th>$k = 8$</th>
<th>$k = 9$</th>
<th>$k = 10$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_1$</td>
<td>1.0000</td>
<td>0.1340</td>
<td>0.0572</td>
<td>0.0318</td>
<td>0.0202</td>
<td>0.0140</td>
<td>0.0103</td>
<td>0.0078</td>
<td>0.0062</td>
<td>0.0050</td>
</tr>
<tr>
<td>$p_2$</td>
<td>0.8660</td>
<td>0.1975</td>
<td>0.1022</td>
<td>0.0633</td>
<td>0.0432</td>
<td>0.0314</td>
<td>0.0239</td>
<td>0.0188</td>
<td>0.0152</td>
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<tr>
<td>$p_3$</td>
<td>0.7454</td>
<td>0.2046</td>
<td>0.1165</td>
<td>0.0768</td>
<td>0.0548</td>
<td>0.0412</td>
<td>0.0322</td>
<td>0.0259</td>
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</tr>
<tr>
<td>$p_4$</td>
<td>0.6614</td>
<td>0.2000</td>
<td>0.1207</td>
<td>0.0829</td>
<td>0.0610</td>
<td>0.0470</td>
<td>0.0374</td>
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<tr>
<td>$p_5$</td>
<td>0.6000</td>
<td>0.1926</td>
<td>0.1208</td>
<td>0.0854</td>
<td>0.0643</td>
<td>0.0505</td>
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<tr>
<td>$p_6$</td>
<td>0.5528</td>
<td>0.1848</td>
<td>0.1192</td>
<td>0.0861</td>
<td>0.0660</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>$p_7$</td>
<td></td>
<td>0.5151</td>
<td>0.1773</td>
<td>0.1168</td>
<td>0.0859</td>
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<tr>
<td>$p_8$</td>
<td></td>
<td>0.4841</td>
<td>0.1704</td>
<td>0.1141</td>
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<td>$p_9$</td>
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<td>0.4581</td>
<td>0.1641</td>
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<tr>
<td>$p_{10}$</td>
<td></td>
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<td>0.4359</td>
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</table>

Table 29. Coefficient matrix $M$

<table>
<thead>
<tr>
<th></th>
<th>$k = 1$</th>
<th>$k = 2$</th>
<th>$k = 3$</th>
<th>$k = 4$</th>
<th>$k = 5$</th>
<th>$k = 6$</th>
<th>$k = 7$</th>
<th>$k = 8$</th>
<th>$k = 9$</th>
<th>$k = 10$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bin 1</td>
<td>1.0000</td>
<td>0.1547</td>
<td>0.0767</td>
<td>0.0480</td>
<td>0.0337</td>
<td>0.0253</td>
<td>0.0199</td>
<td>0.0162</td>
<td>0.0135</td>
<td>0.0115</td>
</tr>
<tr>
<td>Bin 2</td>
<td>1.0000</td>
<td>0.2649</td>
<td>0.1545</td>
<td>0.1055</td>
<td>0.0782</td>
<td>0.0610</td>
<td>0.0494</td>
<td>0.0411</td>
<td>0.0349</td>
<td></td>
</tr>
<tr>
<td>Bin 3</td>
<td>1.0000</td>
<td>0.3093</td>
<td>0.1942</td>
<td>0.1389</td>
<td>0.1064</td>
<td>0.0851</td>
<td>0.0703</td>
<td>0.0593</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bin 4</td>
<td>1.0000</td>
<td>0.3333</td>
<td>0.2183</td>
<td>0.1609</td>
<td>0.1260</td>
<td>0.1026</td>
<td>0.0859</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bin 5</td>
<td>1.0000</td>
<td>0.3484</td>
<td>0.2345</td>
<td>0.1764</td>
<td>0.1404</td>
<td>0.1158</td>
<td></td>
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<td></td>
<td></td>
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<tr>
<td>Bin 6</td>
<td>1.0000</td>
<td>0.3587</td>
<td>0.2462</td>
<td>0.1880</td>
<td>0.1515</td>
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<tr>
<td>Bin 7</td>
<td>1.0000</td>
<td>0.3663</td>
<td>0.2550</td>
<td>0.1970</td>
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<td></td>
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<tr>
<td>Bin 8</td>
<td>1.0000</td>
<td>0.3720</td>
<td>0.2619</td>
<td></td>
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<td>Bin 9</td>
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<tr>
<td>Bin 10</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.0000</td>
</tr>
</tbody>
</table>
Therefore the particle numbers of each bin for a calculated 2D distribution is then obtained with above matrix multiplied by a vector of particle number \( \{N_i\} \) (\( i = 1, 2...10 \)):

\[
\{D_{2D}\} = M \ast \{N_i\}
\]

\( D_{2D} \) is then plotted into Fig. 37. Compared with original one (blue), the number is increased in the bins of smaller sizes.

Such size distribution showing particle numbers as a function of particle size can be converted to area fraction as function of particle size distribution considering the volume fraction of the particles should be the same for both 2D area fraction and 3D volume fraction. And \( f \) as a function of \( d \) is shown in Fig. 38.

3. SEM measured 2D particle equivalent diameters were binned in a similar condition with 50 nm as the bin size and \( f \) of all the particles in each bin added up as the total \( f \) for that specific bin. And then TEM and SEM distributions were plotted together into Fig. 39 for comparison. Particles larger than 450 nm, TEM have not measured any particle. It is because the very limited volume of material has been measured in TEM, particles with larger size can not been measured with statistic correctness. This may be also the reason for the sudden jump of the TEM fv for the bin of 425 nm in Fig. 39. For the end of small particles, the amount measured by TEM is more than SEM. This can be the result of the limited resolution of SEM. Some particles are missing in SEM characterization.
However, the middle section from 150 nm to 400 nm shows a similar distribution with \( f_v \) relatively coincident. The cut off value used in this paper is 200 nm, and within the range of coincidence. Therefore, it is a proper cutoff value.
APPENDIX J: FIGURES

Fig. 1. (a) Schematic diagram showing FSW and its basic process definitions,[14] (b) schematic diagram for as-FSW plate shows various directions: long transverse direction (LT), short transverse direction (ST), and rolling direction (RD); (c) optical micrograph for etched ST-LT plane shows weld nugget zone (WNZ), and the positions of the top and middle sites for microstructural analysis.

(continued)
(Fig. 1: continued)
Fig. 2. Grain structure stability plot showing predicted regions corresponding to grain growth mechanism. $\Psi$ and $X$ are defined by Eqs. 1 and 2, Humphreys’ Theory. [69]
Fig. 3. Grain structure stability plot separating mean grain radius and $f/d$, and showing predicted regions corresponding to grain growth mechanism.\textsuperscript{[69]}
Fig. 4. Grain morphology after heat treatment (465 °C, 5 minutes) for various welding conditions. [89]
Fig. 5. Grain morphology evolution according to heat treatment time duration (at 465 °C).\textsuperscript{[89]} Weld conditions: 1000 rpm, 1.7 mm/s.\textsuperscript{[89]} Scale marker added.
<table>
<thead>
<tr>
<th>Sample</th>
<th>Macrostructure after heat treatment at 738 K (465 °C) for 5 min</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>H1</strong></td>
<td>Low heat input (1000 rpm, 5.0 mm/s)</td>
</tr>
<tr>
<td></td>
<td><img src="image1.png" alt="Image" /></td>
</tr>
<tr>
<td><strong>H2</strong></td>
<td>Medium heat input (1000 rpm, 1.7 mm/s)</td>
</tr>
<tr>
<td></td>
<td><img src="image2.png" alt="Image" /></td>
</tr>
<tr>
<td><strong>H3</strong></td>
<td>High heat input (1500 rpm, 2.5 mm/s)</td>
</tr>
<tr>
<td></td>
<td><img src="image3.png" alt="Image" /></td>
</tr>
</tbody>
</table>

Fig. 6. Optical micrographs of FSW cross-sections after heat treatment of 738 K (465 °C) for 5 min: (a) H1 weld condition, (b) H2 weld condition, (c) H3 weld condition. Etching was done as described in the text.
Fig. 7. Grain morphology evolution according to heat treatment temperature (for a constant duration of 5 min). Weld conditions: 1000 rpm, 1.7 mm/s.
Fig. 8. Plan view of microstructures: (a) optical micrograph of etched H2 after coarse grain formation during QPF-HT. (Weld conditions: 1000 rpm, 1.7 mm/s); (b) OM image of etched FSW AA2024 showing distribution of particles. (White bands are of particle-poor regions and black ones of particle-rich regions.)\textsuperscript{[35]}
Fig. 9. Hardness plot with y-axis partly chopped, comparing the hardness of (a) H1, (b) H2, and (c) H3 welds along the centerline both before and after QPF-HT to those of the base H18, O annealed (at 380 °C for 1 hr).
(Fig. 9: continued)

(b)
(Fig. 9: continued)

![Graph showing Hv values versus distance from top surface for different samples.]

- **Base O**
- **FSW-H3+QPF-HT**
- **FSW-H3**
- **Base H18**

Distance From Top Surface vs. Hv
Fig. 10. Grain structures after 2-stage heat treatments: (1) coarse grain structure formed on the top region after QPF-HT only with no pre-recovery anneal performed at stage 1; (2) similar coarse grain structure formed on the top region after 24 hr at 370 °C as stage-1 recovery anneal and QPF-HT as stage 2; (3) several large grains formed on the top-left region of the weld to the retreating side (marked by red dashed circle); and (4) similar coarse grain structure formed on the top region after 24 hr at 380 °C as stage-1 recovery anneal and QPF-HT as stage 2.
Fig. 11. Hardness change of H2 TOP-1/3 (red) and BOT-2/3 (blue) according to the recovery anneal duration at 350 °C.
Fig. 12. Hardness change of H2 TOP-1/3 (red) and BOT-2/3 (blue) according to the temperature of recovery anneal for a constant duration of 24 hr.
Fig. 13. Pancake-shaped 5083-H18 base material EBSD phase-contrast map. ($d = \text{average grain size}$).
Fig. 14. Phase-contrast EBSD maps showing before heat-treatment grain structures in the weld nugget zone for three sets of weld conditions H1, H2, H3. Figures (a) - (c) are for locations near the top surface of the sheet, Figure (d) - (f) are corresponding ones in the middle of the sheet. $d$ is the mean grain size from average linear intercept.

(continued)
(Fig. 14: continued)

(c) H3 Top (AGG) \( d = 2.3 \mu m \)

(f) H3 Middle \( d = 4 \mu m \)
Fig. 15. EBSD grain structure map showing one of the large grains formed after annealing at 380 °C for 24 hr, emerging on the upper-left region of the weld nugget (refer to Fig. 8, View (3))
Fig. 16. EBSD grain structure map taken on the border between the coarse and the fine grain structure after QPF-HT, showing coarse grains (~ 100 µm) on the upper half of the figure and fine grains (~ 2 µm) on the lower half.
Fig. 17. EBSD phase-contrast maps of H2 middle region after QPF-HT
Fig. 18. EBSD phase-contrast maps of H2 after QPF-HT near top surface, showing very fine grains and coarse grains. Top surface layer with ultra-fine grains is about 5 µm thick.
Fig. 19. Coarse grain structure on the whole welded region after 7 days at 370 °C as stage-1 recovery anneal and QPF-HT as stage 2; (a) OM micrograph showing the coarse grain structure formed on the whole weld nugget; (b) EBSD grain structure map showing three different regions across the thickness with different grain sizes.
Fig. 20. Comparison of grain misorientation angles: (a) H1-H3, top and middle vs. random, (b) base materials vs. random, (c) deviation from random: average top (H1-H3), average middle (H1-H3) vs. base materials.
Fig. 21. The grain size and LAB% of various characterization locations. (Locations having coarse grains formation during QPF-HT are in red; locations without coarse grain formation in blue)
Fig. 22. (a) SEM micrograph taken under BSE mode showing grain structure and particle content on the H2 top surface after QPF-HT, showing the ultra-fine grain structure region, the coarse grains (~100 µm) below them, and the particle-rich band between them in the middle. Several particles are marked for further EDS analysis. (b) The EDS result for particle 1 showing increased amount of Si; (c) the EDS results for particle 2 and 3, in comparison with the matrix.
(Fig. 22: continued)
Fig. 23. SEM micrograph showing as-weld H3: (a) top region of ultra-fine structure (b) top region about 30 µm below the ultra-fine structure.
Fig. 24. TEM micrographs showing the particle contents in: (a) the top and (b) the middle region of H2; (c) the top and (d) the middle region of H3.
Fig. 25. TEM micrographs showing the dislocation contents: (a) the top region of H2 sample; (b) the middle region of H2 sample; (c) the top region of H3 sample; (d) the middle region of H3 sample; and (e) the H18 base metal.
(Fig. 25: continued)

Base H18
Fig. 26. 2D particle size distribution of H2 middle region from SEM
Fig. 27. ICP-MS result showing the Mg/Al signal ratio as a function of through-thickness depth. (blue: Base H18; red: H2 weld)
Fig. 28. Pole figures of all six characterization locations: (a) H1-Top, (b) H1-Mid, (c) H2-Top, (d) H2-Mid, (e) H3-Top, (f) H3-Mid, (g) base O.

(continued)
(Fig. 28: continued)
Fig. 29. The stability of each characterized locations predicted by the Humphreys’ Model with the effects of mean grain radius and f/d separated.
Fig. 30. The stability of each characterized locations predicted by the Humphreys’ Model.
Fig. 31. Schematic showing the location in weld nugget for hardness testing
Fig. 32. EBSD scan of adjacent area on the middle region of H1 for grain size scatter determination
Fig. 33. EBSD scan of adjacent area on the middle region of H1 for grain size scatter determination.
Fig. 34. TEM micrograph showing particles in H2-Mid: a) before image process, b) after image process using FoveaPro
Fig. 35. (a) A typical SEM image taken for characterizing second phase particles. (b) the same image processed using FoveaPro
Fig. 36. Particle size distribution from TEM measurement on H2 middle region
Fig. 37. Comparison between as-measured 3D size distribution and converted 2D distribution
Fig. 38. Converted 2D size distribution of $f$ vs. $d$
Fig. 39. Comparison of size distributions from TEM (red) and SEM (blue)
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