Friction Stir Processing of Nickel-base Alloys

DISSERTATION

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ABSTRACT

Friction stir processing (FSP), a derivative of the recently developed friction stir welding process, is a localized solid-state thermomechanical process used for microstructural modification. Location-specific microstructural enhancement resulting from FSP can be used to improve the properties and performance of a component only where necessary thus reducing cost associated with producing a component entirely from a high-performance material. FSP has been limited to relatively low temperature alloys systems, e.g., aluminum and magnesium-base alloys. Recent tool material developments have enabled FSP to be applied to more refractory materials, such as Ni-base alloys. This study focuses on the unique microstructures in Ni-alloys including Alloy 201 (Ni-201), Inconel® Alloy 600 (IN600), Haynes Alloy 282, Haynes Alloy 214, Mar-M247, and René 41 that result from FSP and additive friction stir processing (AFSP), an adaptation of FSP in which material additions are incorporated to further enhance site-specific properties.

Comprehensive autogenous FSP process parameter development was performed for Ni-201 and IN600 to determine process windows. Analysis of process force output revealed defect-free runs were characterized by pseudo-steady state process forces. For both materials, overall measured process forces decreased as the FSP heat input was increased. Microstructural examination of both alloys after FSP reveals considerable microstructural refinement in the stir zone (SZ). Equiaxed, recrystallized grains, with
average grain sizes ranging from 8 to 27 µm were observed. Grain refinement along with the extent of recovery and recrystallization varied based on the applied process parameters.

Additive friction stir processing was used to locally create γ’-strengthened surface layers on non-age hardenable substrates. Superalloys with varied γ’-former contents were deposited on IN600 substrates using fusion-based deposition techniques and subsequently friction stir processed. Alloy 282 and Alloy 214 surface layers on IN600 substrates formed fine distributions of secondary γ’ following an abbreviated post-processing heat treatment. Microhardness measurements and metallographic analyses indicate homogenization and enhanced heat treatment response results from AFSP, especially for AFSP Haynes 282/IN600. Electron backscatter diffraction misorientation and transmission electron microscopy analyses suggest that the degree of stored energy (i.e., dislocation density) for SZ material is inversely proportional to AFSP heat input.

Additive friction stir processing of high-γ’ fraction laser deposited superalloy layers formed fine dispersions of secondary γ’ in the as-processed condition. Consolidation of deposition discontinuities and considerable grain refinement was observed within the additive material-containing SZ. Some near-surface regions in AFSP Mar-M247/IN600 exhibited nano-crystalline grains (d_{avg} = 340 nm) with microhardness values exceeding 750 HV. A model for the supersolvus microstructural development of high-γ’ fraction superalloys is presented.

Friction stir processing was also applied as a buttering technique to modify the base metal microstructure of IN600 prior to fusion welding. Grain refinement from FSP
increased the extent of epitaxial nucleation and led to grain refinement of the weld metal (WM). Grain size within the pre-treated heat affected zone (HAZ) was also reduced by a factor of three near the fusion boundary relative to unprocessed base metal. Simultaneous WM and HAZ grain refinement is unique to FSP and is not otherwise achievable using conventional refinement techniques.
To my parents and friends for their unending support.
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<tbody>
<tr>
<td>AFSP</td>
<td>additive friction stir processing</td>
</tr>
<tr>
<td>AS</td>
<td>advancing side</td>
</tr>
<tr>
<td>BM</td>
<td>base material</td>
</tr>
<tr>
<td>BSE</td>
<td>backscatter electron</td>
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<tr>
<td>CCT</td>
<td>continuous cooling transformation</td>
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<tr>
<td>CDRX</td>
<td>continuous dynamic recrystallization</td>
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<tr>
<td>DDRX</td>
<td>discontinuous dynamic recrystallization</td>
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<tr>
<td>DMD</td>
<td>direct metal deposition</td>
</tr>
<tr>
<td>DRX</td>
<td>dynamic recrystallization</td>
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<tr>
<td>EBSD</td>
<td>electron backscatter diffraction</td>
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<tr>
<td>EDS</td>
<td>energy dispersive spectroscopy</td>
</tr>
<tr>
<td>FCC</td>
<td>face-centered cubic</td>
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<tr>
<td>FSP</td>
<td>friction stir processing</td>
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<tr>
<td>FSW</td>
<td>frictions stir welding</td>
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<tr>
<td>FWHM</td>
<td>full width at half max</td>
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<tr>
<td>GDRX</td>
<td>geometric dynamic recrystallization</td>
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<tr>
<td>GROD</td>
<td>grain reference orientation deviation</td>
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<tr>
<td>GTAW</td>
<td>gas tungsten arc welding</td>
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<tr>
<td>HAADF</td>
<td>high-angle annular dark field</td>
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<tr>
<td>HAZ</td>
<td>heat affected zone</td>
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<tr>
<td>HCP</td>
<td>hexagonal close-packed</td>
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<tr>
<td>HI</td>
<td>heat input</td>
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<tr>
<td>HRSEM</td>
<td>high-resolution scanning electron microscopy</td>
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<td>HV</td>
<td>Vickers hardness</td>
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<tr>
<th>Abbreviation</th>
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<tr>
<td>IN</td>
<td>Inconel® alloy</td>
</tr>
<tr>
<td>IPM</td>
<td>inches per minute</td>
</tr>
<tr>
<td>KAM</td>
<td>kernel average misorientation</td>
</tr>
<tr>
<td>LENS</td>
<td>laser engineered net-near shaping</td>
</tr>
<tr>
<td>LOM</td>
<td>light optical microscopy</td>
</tr>
<tr>
<td>MMC</td>
<td>metal matrix composite</td>
</tr>
<tr>
<td>ODS</td>
<td>oxide dispersion strengthened</td>
</tr>
<tr>
<td>PCBN</td>
<td>polycrystalline cubic boron nitride</td>
</tr>
<tr>
<td>PSB</td>
<td>persistent slip bands</td>
</tr>
<tr>
<td>PWHT</td>
<td>post-weld heat treatment</td>
</tr>
<tr>
<td>RS</td>
<td>retreating side</td>
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<tr>
<td>SAC</td>
<td>strain age cracking</td>
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<tr>
<td>SAED</td>
<td>selected area electron diffraction</td>
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<tr>
<td>SE</td>
<td>secondary electron</td>
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<tr>
<td>SEM</td>
<td>scanning electron microscopy</td>
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<tr>
<td>SFE</td>
<td>stacking fault energy</td>
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<tr>
<td>STEM</td>
<td>scanning transmission electron microscopy</td>
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<tr>
<td>SZ</td>
<td>stir zone</td>
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<tr>
<td>TEM</td>
<td>transmission electron microscopy</td>
</tr>
<tr>
<td>T&lt;sub&gt;m&lt;/sub&gt;</td>
<td>melting temperature</td>
</tr>
<tr>
<td>TMAZ</td>
<td>thermomechanically affected zone</td>
</tr>
<tr>
<td>TTT</td>
<td>time-temperature-transformation</td>
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<td>WM</td>
<td>weld metal</td>
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CHAPTER 1: INTRODUCTION

The physical properties of a material determine the fitness for a specific purpose or application. In demanding applications, costly high-performance materials are often implemented to satisfy the design requirements. However, the properties of such high-performance materials may only be required in certain locations within a component. The development of site-specific material modification techniques allow for local enhancement of material properties and performance. The implementation of location specific properties is a promising prospect to reduce cost while simultaneously improving component performance. As an example, consider a solid solution strengthened Ni-alloy component used at high temperature. Regions of the component subject to high local stresses are susceptible to decreased creep life. Site-specific material modification techniques can be used to locally modify microstructure and constituent phases such that a superalloy is created locally and therefore improving robustness at elevated temperature. As a result, economic benefit is gained by locally modifying to create location specific properties only where necessary rather than producing the entire component from a costlier material.

Friction stir processing (FSP) is a solid-state local microstructural modification technique used to create location-specific properties. As a result of recent process developments to increase temperature capabilities of FSP, the application to high T_m materials including nickel-base alloys has become of great interest. This broader application of FSP beyond low T_m materials (e.g., aluminum and magnesium alloys) to high T_m materials holds many potential benefits. Potential benefits are especially apparent when considering the use of high T_m materials (which includes steels, Ni-base alloys, and Ti-alloys to a lesser degree) is extremely widespread and represents more than 80% of the welded materials [1]. Location-specific property enhancement using FSP is
further enhanced by the addition and in situ reaction of additional materials during processing. However, friction stir processing, by nature, is an autogenous process that does not typically utilize the addition of filler material. Additive techniques to alter the chemistry and constituent phases using FSP do not exist for Ni-alloys and require exploration and development.

Other additive material methods exist to create location specific properties; however, the resulting additive region microstructures often are vastly different than what can be obtained (or is desireable) via FSP. The solid state nature of the FSP process results in a wrought, recrystallized microstructure compared to solidification microstructures associated with fusion based additive techniques such as laser engineering near net shaping (LENS), thermal spray, etc. Furthermore, even some solid state additive process such as cold spray, selective laser sintering, and laminated object manufacture cannot produce fully-consolidated, wrought microstructures that are characteristic of FSP.

Friction stir processing of Ni-alloys also has other benefits beyond additive applications. With respect to microstructure and final properties, FSW is an advantageous solid state joining technique, especially for light metals. However, it is not always feasible to apply FSW to all joining applications. Many aspects of the FSW process lead to implementation difficulties including large capital costs for machinery and tools, fixturing concerns, and somewhat limited joint design. This is especially true for high-$T_m$ materials exhibiting high elevated-temperature flow strength. Consequently, many structures still require final joining to be performed using fusion welding processes rather than FSW. As an alternative to FSW, FSP serves as a high value-added process if the workpiece microstructure can be locally modified only where necessary prior to fusion welding such that the final weld properties are improved and/or weldability problems are prevented. Such application of FSP to Ni-alloys to locally enhance weldability is analogous to ‘buttering’ techniques commonly used in fusion welding. However, the potential benefits of FSP as a ‘pre-treatment’ to base material prior to fusion welding are relatively unknown. Fundamental aspects pertaining to FSP pre-
treatment including microstructural evolution, stability, and process parameter relationships, require understanding before application. This is especially true with regard to high $T_m$ materials.

While site-specific material modification resulting in location-specific properties has numerous potential benefits for Ni-base alloys, there exists a large knowledge gap. This document represents the initial steps in understanding the microstructural evolution, material flow, kinetic enhancement, process parameter relationships, and potential property enhancement of FSP and AFSP of Ni-base alloys. Microstructural characterization of several FSP/AFSP Ni-alloys and alloy combinations of varied compositional complexity was performed using several techniques. While FSP regions are comprised of several microstructural domains, this study predominantly focuses on stir zone microstructural evolution for six different Ni-base alloys at varied process parameters. The results of detailed microstructural analyses combined with physical simulation serve as a foundation for subsequent studies and applications.
CHAPTER 2: BACKGROUND

2.1 Friction Stir Processing

Within a relatively short span of time since its inception in 1991 [2] friction stir welding (FSW) and its more recent derivative, friction stir processing (FSP), have been the focus of considerable research effort [3-5]. Friction stir welding is considered one of the most significant joining developments within past decades due to its energy efficiency, environmental friendliness, resulting weld properties [6]—all attributed to the solid state nature of the process. Friction stir welding has been applied successfully in a wide array of engineering applications including automotive [7, 8], energy [9], railway [10], aerospace [11], and military [12]. Friction stir welding and FSP are both solid state techniques that involve a non-consumable rotating tool plunged into the workpiece. The role of the tool is to create frictional heat by contact with the workpiece. Frictional heat softens the workpiece material, and the traversing rotating tool moves material along the joint under an applied load, thus bonding two faying surfaces. Finally, the tool is retracted leaving a solid-state bond [8]. Figure 2.1 shows a schematic of the FSW process.
Severe plastic deformation of the workpiece beneath the tool during FSW combined with high homologous temperatures near the solidus temperature leads to considerable microstructural changes within the periphery of the tool. Notable changes include recrystallization, homogenization, and second phase comminution (i.e. particle breakup). Mechanistically, FSP is identical to FSW; however, the former is not used for joining. Moreover, the depth of FSP-affected material can be varied from very-near surface to through-plate-thickness simply by varying the tool pin length and geometry.

Friction stir processing takes full advantage of the efficiency, versatility, and location-specific microstructural modification of FSW without creating a weld. The first results of FSP were published by Mishra et al. [13] nearly ten years after FSW was first patented. Since this work, FSP has been used primarily on Al-alloys to enhance superplastic formability, improve fatigue life, homogenize and refine castings, improve formability, heal cracks, harden surfaces, and fabricate surface composites [6].
2.2 **Characteristics of Friction Stir**

2.2.1 **Characteristic Friction Stir Regions**

Friction stir processing/welding leads to the development of microstructural domains with nomenclature unique to the process. Figure 2.2 shows a cross section of through-thickness FSP AA6061. Far away from the FSP region is an unaffected portion of material not affected by thermal cycle or deformation that is referred to as the base material (BM). Closer towards the center region is the heat affected zone (HAZ). This mechanically undeformed region of material is affected only by the FSP/W thermal cycle. Microstructural changes as a result of the thermal excursion can include grain coarsening, recovery of cold work, and precipitate dissolution/coarsening depending on HAZ location, i.e., local thermal cycle. Often the HAZ is subdivided into several sub-regions based on microstructure.

![Figure 2.2. Friction stir processing run on AA6061 demonstrating typical regions within processed zone. Sample etched using Keller’s reagent.](image)

The center of the FSP/W region is known as the stir zone (SZ). In this zone, material undergoes severe plastic deformation at elevated temperature such that recrystallization occurs. High homologous temperatures are experienced within the SZ
during the FSP/W process. Temperatures range from $0.6T_m$ [6] to $0.99T_m$ [14]. As a result of large strains at high temperature, grains within the SZ are often characteristically highly refined compared to the base material due to dynamic recrystallization. The region of material between the SZ and the HAZ is the thermomechanically affected zone (TMAZ). This zone is affected by the FSP/W thermal cycle as well as deformation; however, temperature and strain within this region is not sufficient to promote the extent of recrystallization occurring in the SZ. The size of the TMAZ often depends on FSP/W parameters and the elevated temperature flow stress characteristics of the workpiece [15]. Materials with highly temperature dependent flow stress characteristics may exhibit significantly smaller TMAZ regions compared to the AA6061 sample shown in Figure 2.2.

Proper orientation of the FSP/W region requires understanding of the advancing and retreating convention of the FSP/W region. The advancing side (AS) of the FSP/W region denotes the side where the tool rotation direction is the same as the travel direction. The retreating side (RS) is characterized by the opposite relationship. Advancing side and RS are marked accordingly in Figure 2.2 for a clockwise rotating tool. Strain and temperature distribution within the SZ can vary greatly between the AS and RS [16].

2.2.2 Material Flow

Compared to bulk hot working and severe plastic deformation processes such as hot rolling, equal channel angular pressing (ECAP), accumulated roll bonding, etc, the nature of material flow in friction stir is rather complex. The complex nature of material flow causes plasticized swept by the tool to deposit material within band-like structures behind the moving tool. This creates a unique surface appearance to FSP processed regions that slightly resembles machining marks remaining after processes such as milling. Figure 2.3 shows the typical surface appearance of the top surface of a friction stir processed region. This region is identical in appearance to a friction stir weld.
Bands visible on the surface of a FSP region are affected by FSP process parameters, namely the tool traverse velocity and the rotation rate of the tool. The spacing of the bands is approximately equal to the tool traverse velocity divided by the tool rotation speed (Figure 2.4).

The complex material flow occurring during FSP is asymmetric about the centerline of the process region [16]. This asymmetric flow, which is a function of
workpiece flow behavior, processing parameters, and tool geometry, affects the microstructural development and distribution of microstructural modification.

The nature of material flow during FSP/W is chaotic therefore modeling such behavior is not straightforward. Most knowledge of material flow during FSP/W has been gathered from examination of dissimilar embedded tracer materials [16, 17]. Such tracer materials are translated within the complex flow field and positions are noted upon examination post-FSP/W. Although not entirely definitive, some empirically-based qualitative models such as the Nunes kinematic model [18] were constructed to better explain the material flow within the SZ and the TMAZ. This model proposes two operative flow currents: ‘straight through’ flow and ‘maelstrom’ flow. The straight through material flow occurs on the retreating side of the FSP/W tool and results in metal picked up by the front of the tool which is then deposited on the trailing edge of the tool. Figure 2.5A shows a schematic of straight-through flow proposed in the Nunes model. Straight through flow as shown in Figure 2.5A represents plastically deformed material away from the pin/shoulder that does not undergo rotational translation. Depending on the proximity to the rotating tool, plastically deformed material will undergo rotational translation about the rotating tool as depicted in Figure 2.5B.
The ‘maelstrom’ current, proposed as the second operative mechanism in the Nunes kinematic model, refers to convective-like material flow that occurs predominantly tool advancing side (AS) (Figure 2.5C). Some material residing on the tool AS experiences rotational flow about the rotating tool and is trapped by the radial influx of material. This radial influx of material is a direct result of the vortex flow created by the threaded pin tool used in the Nunes study. The material from the top of the plate on the AS is driven downward towards the bottom of the plate by maelstrom flow. This downward material flow described in the work by Nunes is a result of the handedness of the threads used for the tool pin feature. A pin with the thread pitch reversed would result in the reversal of material flow, i.e., material flow from bottom to the top of the plate. Material flow from the bottom to top of the plate has a tendency to form wormhole defects [19]
The complex interaction and combination of the two or more material flow currents has been used to describe the formation and texture of ‘onion-ring’ SZ features (Figure 2.6). While these onion ring features are generally considered to not affect the mechanical properties of the resulting weld, definitive explanations as to the origin still do not exist [20].

![Figure 2.6. Onion ring features in A.) FSW AA2195-T81 [18], B.) FSW 6061[20]](image)

2.3 Stir Zone Microstructural Evolution

Deformation of material beneath the friction stir tool leads dynamic recrystallization, i.e., nucleation of new grains occurs during the deformation rather than post-deformation as with static recrystallization [21]. Several dynamic recrystallization mechanisms have been proposed; however the vast majority of friction stir-related studies concerning recrystallization focus primarily on Al-alloys. During continuous dynamic recrystallization (CDRX), the formation of high angle grain boundaries results from the constant accumulation of dislocations arising from strain during FSP/W. The progressive accumulation of strain drives rearrangement of dislocations into subgrains. Such rearrangement increases the misorientation angle of subgrain boundaries. The exact mechanism by which subgrain rotation occurs is not well understood, however it is
observed most often in systems that have inhibited dislocation motion by mechanisms such as solute drag or lack of slip systems (e.g., Mg-alloys) [21]. The subgrain rotation process continues with continued deformation, and leads to the development of high angle misorientation between subgrains and eventually leads to new grains. During rotation, very little boundary migration occurs therefore recrystallized grains formed via CDRX are on the order of subgrain size [6]. Figure 2.7 schematically demonstrates the formation of and rotation of subgrains formed along preexisting high angle grain boundaries characteristic of the CDRX process.

![Figure 2.7. Schematic of CDRX: dislocation accumulation near a grain boundary causing gradual subgrain rotation and the eventual formation of new grains with high angle misorientations (adapted from Humphries and Hatherly [21])](image)

Dynamic recrystallization may also occur by a different mechanism, known as discontinuous dynamic recrystallization (DDRX), with characteristics of ‘classical’
recrystallization with defined nucleation and growth stages. By this mechanism, applied strain introduces dislocations to the system just as in CDRX; however, a nucleation step exists in which new grains nucleate at the prior high angle grain boundaries. As continued deformation occurs, the dislocation density increases in the new grains and reduces the driving force for further growth [21]. As a result, regions with low dislocation density will grow at the expense of regions with high dislocation density. Eventually, a uniform, equiaxed grain structure with a limited grain size (known as the saturation value [22]) that varies on strain rate and temperature. Figure 2.8 illustratively demonstrates the evolution of recrystallized microstructure during DDRX.

Figure 2.8. Schematic representation of DDRX: nucleation of new grains along the larger initial grain boundaries. Continual formation of grains forms a necklace structure along the original grain boundaries. Eventually the material becomes fully recrystallized. Grain growth of recrystallized grains is shown as the last step in the schematic (adapted from Humphries and Hatherly [21])
Other dynamic recrystallization mechanisms such as geometric dynamic recrystallization (GDRX) occur when material is deformed in such a way that tortuous high angle grain boundaries impinge on each other during deformation. Points of contact between high angle grain boundaries will annihilate and in effect ‘pinch’ into new grains. Like other dynamic recrystallization mechanisms, GDRX will also lead to an equiaxed microstructure [23]. This mechanism is depicted schematically in Figure 2.9.

![Diagram of GDRX](image)

Figure 2.9. Schematic representation of GDRX. A.) Strain is initially applied. B.) At higher strain values, high angle grain boundaries impinge which leads to the formation of equiaxed grains at temperatures sufficient for boundary mobility.

Naturally it is difficult to observe the different recrystallization mechanisms directly, especially during FSP/W. Characterization of grain boundary character using electron backscatter diffraction (EBSD) and/or transmission electron microscopy (TEM) has been used to propose operative recrystallization mechanisms during FSP/W. Jata and Semiatin [24] suggest continuous dynamic recrystallization (CDRX) as the operative mechanism in FSW of a Al-Li-Cu alloy. The authors propose the low-angle misorientation subgrains in the base material are replaced by high-angle misorientation
grains by a continuous increase in misorientation during deformation. The authors state dislocation glide is responsible for the gradual rotation of subgrains. Subsequent studies, however, have noted that the recrystallized grain size in the SZ is actually smaller than the subgrains found in the base material [25]. Therefore it is unlikely that the fine equiaxed grains in the SZ are the direct result of rotation of base material subgrains. From characterization of microstructural characteristics of FSW AA7050, Su et al [25] proposes a mechanism similar to Jata and Semiatin [24] while considering subgrain evolution during dynamic recovery. The evolution of the stir zone microstructure can be broken down into 4 major phases [25]:

1.) Dislocation Introduction: Deformation at elevated temperature during FSP/W imparts a large amount dislocations. The elevated temperature leads to coarsening.

2.) Dynamic Recovery: Dislocations organize into a network of very small subgrains. During this stage, the subgrain boundaries represent low-angle boundaries.

3.) Continuous Dynamic Recrystallization: Continued thermomechanical input during the FSP/W process imparts additional dislocations into the material due to strain incompatibility between neighboring subgrains. Meanwhile, the geometrically necessary dislocations are continually absorbed into the subgrain boundaries. This continuous introduction and absorption of dislocations during deformation coarsens the subgrains and increases the misorientation between adjacent subgrains.

4.) Post-Dynamic Recrystallization and Partial Recovery: After DRX, additional deformation during FSP/W introduces additional dislocations in the new, recrystallized grains. Some dislocations are eliminated during the remainder of the FSP/W thermal cycle.
Recrystallization studies focusing on FSP/W material (predominantly Al-alloys) in general rule out DDRX based on post-FSP/W microstructural evaluation. However, some studies have characterized FSP/W microstructures ‘frozen’ in the non-equilibrium, deformed state by use of rapid quenching behind the tool or ‘plunge and extract’ methods [6]. In these studies, equiaxed nanoscale grains (25-100 nm) with high angle misorientation were observed. It has been proposed that DDRX is the operative mechanism for FSP/W conditions leading to nano-scale microstructures.

2.3.1 Influence of Stacking Fault Energy on Recrystallization Mechanism

The dissociation of a perfect dislocation in any FCC metal creates two dislocation partials which repel one another. Dislocation partials separate due to a repelling force that is proportional to the shear modulus (G) of the material. The dissociation of dislocation partials creates stacking fault (a two-dimensional local error in the stacking sequence of a crystal) between the partials. The planar stacking fault, which separates the partials, is a region with locally higher energy relative to the surrounding lattice with a perfect stacking sequence. The energy associated with maintaining separation of the partials via the stacking by some distance (d) is an energy per unit area known as the stacking fault energy (SFE, \( \gamma \)). Ukei et al [26] relate the equilibrium separation distance (d) to SFE (\( \gamma \)) by the following relationship:

\[
d = \frac{Ga^2}{24\pi\gamma}
\]  

(1)

where ‘a’ is the lattice parameter and ‘G’ is the shear modulus. The stacking fault energies of various metals and alloys are shown in Table 2.1. Some variation in the values for pure metals arises from the assumptions made for the elastic constants used to
calculate SFE from measurements taken from TEM analyses. In general, materials with high values of SFE will have corresponding small separation distances associated with dislocation partials. As a result climb and cross slip occurs more easily compared to materials with low SFE. Low stacking fault energy materials have smaller energy penalties associated with the dissociation of partials widely spaced apart. The very large partial separation distance hinders the cross slip and climb and makes processes such as recovery more difficult [21].
Table 2.1. Compiled stacking fault energy values for various materials and alloys

<table>
<thead>
<tr>
<th>Material</th>
<th>Stacking Fault Energy (mJ/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dieter [27]</td>
<td></td>
</tr>
<tr>
<td>Brass</td>
<td>&lt;10</td>
</tr>
<tr>
<td>303 Stainless Steel</td>
<td>8</td>
</tr>
<tr>
<td>304 Stainless Steel</td>
<td>20</td>
</tr>
<tr>
<td>310 Stainless Steel</td>
<td>45</td>
</tr>
<tr>
<td>Silver</td>
<td>~25</td>
</tr>
<tr>
<td>Gold</td>
<td>~50</td>
</tr>
<tr>
<td>Copper</td>
<td>~80</td>
</tr>
<tr>
<td>Nickel</td>
<td>~150</td>
</tr>
<tr>
<td>Aluminum</td>
<td>~200</td>
</tr>
<tr>
<td>Benyoucef et al. [28]</td>
<td></td>
</tr>
<tr>
<td>Nickel</td>
<td>140</td>
</tr>
<tr>
<td>Ni-26Cr (wt%)</td>
<td>76</td>
</tr>
<tr>
<td>Ni-26Cr-10Co (wt%)</td>
<td>28</td>
</tr>
<tr>
<td>Pettinari et al. [29]</td>
<td></td>
</tr>
<tr>
<td>MC2 Superalloy</td>
<td>21-28</td>
</tr>
<tr>
<td>Ni-26Cr-2Mo-2W-4Re</td>
<td>32 ± 3</td>
</tr>
<tr>
<td>Ni-26-2Mo-2W-4Ru</td>
<td>31 ± 3</td>
</tr>
</tbody>
</table>

Materials with high SFE (such as pure aluminum) recover easily during deformation due to the ease of dislocation climb and cross slip. As a result of the ease of recovery, it is difficult to develop dislocation densities sufficiently high for ‘conventional’ dynamic recrystallization via nucleation and growth of new grains along preexisting grains, i.e., DDRX. Instead, dynamic recrystallization for low SFE materials
occurs by CDRX, i.e., progressive lattice rotations in which there is no clearly defined nucleation step [22]. Conversely, low SFE materials (such as pure Cu) are able to achieve high dislocation densities during deformation due to the slow recovery rate. This causes dynamic recrystallization to occur in a discontinuous manner (hence DDRX) in which there are clear nucleation and growth stages [22].

The presence of solute atoms in an alloy also affects the operative dynamic recrystallization mechanism directly. In general, solute content lowers the SFE for an alloy [26] (see Table 2.1). As a result, increasing solute content will promote dynamic recrystallization by reducing the ease of recovery. Mechanisms such as DDRX will become increasingly more favorable as the alloying content increases. Alloying content also has the effect of retarding boundary mobility [21]. Reduced boundary mobility has the effect of retarding the onset of dynamic recrystallization to higher applied strains or higher temperatures. However, the solute atom itself is not expected to affect the operative dynamic recrystallization mechanism directly [26].

No studies on recrystallization mechanisms during FSP/W of Ni-base alloys exist. However, it is expected that some mechanisms occurring during dynamic recrystallization of Al-alloys during FSP/W may not operate similarly during FSP/W of Ni-base alloy based solely on the expected difference in SFE for the different alloy systems. In a study by Song et al. [30] it is suggested that a possible DDRX mechanism occurring in FSP of Ni-base alloys may result in a microstructure with finer grains compared to a FSP/W material with high stacking fault energy. However, no in-depth discussion or microstructural evidence was provided by the authors as to the operative dynamic recrystallization mechanism.

Very recent work by Mironov et al. [31] explored the recrystallization mechanism of FSP superaustenitic stainless steel, which has a low stacking fault energy. Depending on the location within the FSP process region, observations performed using EBSD suggest competition between both discontinuous and continuous dynamic recrystallization mechanisms. For some regions of the stir zone, low angle grain boundary fraction appears high with low angle grain boundaries arranged in a periodic
array forming a substructure which is characteristic of continuous dynamic recrystallization. However within other regions of the SZ, predominantly near the extremities of the SZ, small equiaxed grains decorate elongated grains thus forming a necklace-type structure. The small equiaxed recrystallized grains do not contain any low angle boundaries. Evidence of necklace structures within these regions suggests that DDRX is the operative mechanism. Because different mechanisms appear to be operative, it can be suggested that the exact recrystallization mechanism depends not only on stacking fault energy but the local temperature and strain condition which is characteristically heterogeneous in its distribution for FSP.

2.4 Kinetic Enhancement Via Friction Stir Processing

The kinetics of many mobility-limited processes is enhanced during FSP due the elevated temperature nature of the process. Additionally, a constant input of defects (in the form of dislocations) into the structure during severe plastic deformation can accelerate the kinetics for many processes, especially if dense dislocation populations are not completely eliminated by recovery and recrystallization process.

Several studies have explored possible practical benefits of enhanced formation kinetics during FSP. Hsu and coworkers. [32] took advantage of the in situ reaction between Al and Cu during FSP to create an Al-Al$_2$Cu nanodispersed composite from a sintered powder billet. Hsu et al., attribute the increased reaction rate to diffusional enhancement provided during FSP, however, they fail to elaborate.

For some systems, enhanced diffusion during FSP has been attributed to the formation of small amounts of liquid present in a process that is generally regarded as solid state. In the fabrication of an Al$_3$Ti nanoscale composite starting with Ti and Al powder, the exothermic reaction of the starting powders provided sufficient additional heat during FSP sufficient to induce localized melting. Very rapid reaction between Al and Ti powder particles during FSP was attributed to the stirring action, which removed the Al$_3$Ti reaction product from the interface thus promoting frequent direct contact
between Al and Ti. Formation of this nanocomposite material occurred significantly more rapidly compared to conventional fabrication methods such as sintering.

In similar study by Chuang et al [33], a composite MgAlZn intermetallic alloy was fabricated by FSP starting with an elemental powder compact. Similar rapid reaction was attributed to the formation of small amounts of liquid during stirring. Evidence of liquid formation was observed in the microstructure by cooling the workpiece behind the moving FSP tool with liquid N₂.

Little is known about the enhancement of second phase formation kinetics during FSP, especially for high-Tₘ systems. Although not necessarily beneficial, Park et al., discovered interesting results regarding the formation kinetics of sigma phase (σ) in FSW 304 stainless steel [34]. The formation of σ phase typically causes undesirable degradation of mechanical properties in stainless steels [35]. Sigma phase formation is typically a very sluggish process and typically requires long aging times on the order of hours [35]. Figure 2.10 shows threshold times for σ formation as a function of chromium content. In the Park study [34], σ phase was observed on the AS of as-FSW 304 stainless steel. The observation made by Park is quite surprising because of the very short FSW thermal cycle duration (approximately 30 seconds) within the σ-susceptible temperature range. The presence of ferrite, which enhances σ formation, combined with large shear strains experienced during stirring was proposed by Park et al. as the mechanism controlling rapid σ formation. However, elaboration of the mechanism was not presented. Although not explicitly discussed in the study, the morphological distribution of σ appears to further strengthen the latter portion of the Park et al. theory regarding enhanced kinetics due to large shear deformations. Lamellar structures of σ phase were formed only within certain SZ regions. These lamellar regions are similar in appearance to onion ring features typically observed in Al and Mg alloys [3, 20]. This distribution of σ appears to correspond to the gradients in strain and/or strain rate that also lead to onion ring formation. During the relatively short FSW/P cycle, σ appears to form only in the SZ onion ring lamellae which apparently received a significant level of mechanical work.
From FSW experiments conducted in AA6061 and AA7075, Krishnan et al. [20] postulated that the formation of lamellar onion ring bands in the SZ are related to cyclic heating followed by extrusion around the pin around the RS. It can be assumed that the thermomechanical history resulting from a discontinuous cyclical deformation creates a locally heterogeneous distribution of strain that ultimately affects recrystallization. Such heterogeneous distributions of strain presumably will lead to regions with locally varied dislocation density. For the formation of secondary phases in which nucleation is rate-limited by diffusion, the nucleation kinetics are enhanced within regions with high stored energy due to the enhancement in diffusivity resulting from dislocations. Stored energy from cold work has been shown to increase the kinetics of formation for $\gamma'$ in Ni-base superalloys [36, 37] but not $\sigma$ phase [38]. The nucleation of sigma phase, however, has been shown to be rate-limited by large-scale atomic movements rather than diffusivity (of Cr) [38]. While FSP/W has the potential to increase dislocation density via plastic deformation, it is possible that the kinetic enhancement of $\sigma$ phase formation is due to large-scale atomic movements associated with moving recrystallization fronts during dynamic recrystallization that occurs in FSW 304 stainless steel.
2.4.1 Defect Structure and Distribution in Friction Stir Processed Materials

Fundamental issues related to the evolution of friction stir microstructure including recrystallization, defect structure, and precipitation phenomena were investigated in great detail by Su et al. [25]. In the Su study, TEM was used to observe grain, dislocation distribution, and precipitate morphologies in FSP AA7050. TEM analyses of the SZ region suggested varied dislocation density. Intragranular dislocation density appeared to vary on the local extent of recovery. New recrystallized grains had very low dislocation densities, as expected. Regions of grains containing high dislocation densities were attributed to a combination of plastic deformation occurring post-
recrystallization and incomplete recovery/recrystallization during the cooling portion of the thermal cycle when tool has passed.

While containing an overall lower precipitate density compared to the base material, precipitates within the SZ of AA7050 (predominately $\eta$ with some $\eta'$) appeared to depend strongly on local dislocation structure. Stir zone regions with high dislocation density contained a uniform dispersion of 60-100 nm precipitates whereas lower dislocation density regions had significantly fewer precipitates. The authors suggest nucleation of precipitates occurs heterogeneously on dislocation pile ups or subgrain boundaries rather than homogeneously as in the base material. Moreover, the precipitate growth (predominantly the equilibrium phase, $\eta$) on dislocations appears to be more extensive than in the BM.

The discussion of dislocation density based on TEM analyses by Su et al. [25] was entirely qualitative. A later study by Woo and coworkers. [39] aimed to quantify dislocation densities in various regions of FSP AA6061 using X-ray peak profile analysis. The technique relies on peak broadening of X-ray diffraction peaks by the presence of subgrain boundaries and dislocations in a material. The average dislocation density within the SZ was lower than any other region measured. The SZ dislocation density was found to be $1.7 \times 10^{14}$ m$^{-2}$ compared to $4.5 \times 10^{14}$ m$^{-2}$ of the BM. The authors attribute the lower SZ dislocation density to recrystallization events during stir processing. The continuous introduction of dislocations into the material due to thermomechanical deformation during FSP/W is likely balanced the elimination of dislocations during subgrain growth and eventual increase in the subgrain misorientation [21]. This increasing misorientation between subgrains leads to the formation of new recrystallized grains. This continuous dynamic recrystallization process, as described in great detail by Su et al. [25], reduces SZ material dislocation density as measured by Woo et al. [39]. Results in the Woo study were further validated by Steuwer et al. [40] with small-angle X-ray scattering using a synchrotron source. In that study, diffraction peak broadening, as quantified by measuring the peak full width at half max (FWHM), had lowest measured values within central regions of the stir zone for FSP AA7010. The
lack of strain broadening associated with the narrow measured diffraction peaks (i.e., low FWHM values) are indicative of low dislocation density, characteristic of completely recrystallized materials. Full width at half max values gradually increased in locations from the center SZ out towards the TMAZ where dislocation density is assumed to be high due to incomplete recovery/recrystallization processes.

One can conclude that dislocation distribution after FSP is rather heterogeneous with the overall dislocation density lower on average compared to the BM (hardenable Al-alloy in artificially aged condition). For materials other than Al-alloys, limited FSP/W studies exist that examine microstructural development with emphasis on dislocation development and annihilation. Materials such as Ni-base alloys differ from Al-alloys with respect to the relative ease of dislocation cross slip and climb [41], as indicated by lower SFE. Further study is required for low SFE materials. Fully understanding precipitate formation enhancement during additive FSP of Ni-base alloys requires a better understanding of the interplay between thermal cycle, and deformation behavior, and recovery/recrystallization processes.

2.5 Nickel-base Alloys

Ni-alloys represent a diverse group of important engineering alloys. Ni-base alloys are often selected on the basis of superior corrosion resistance, thermal stability, and high temperature mechanical properties. Ni-base alloy compositions are divided into four categories which include commercially pure, solid-solution strengthened, precipitation strengthened, and specialty alloys. Figure 2.10 graphically demonstrates the classification scheme of Ni-base alloys.
Commercially pure Ni-alloys contain >99wt% Ni. Notable alloys in this category include Alloy 200 and 201 which are used primarily for corrosion resistance in high pH environments [43]. Commercially pure Ni-base alloys have characteristically low strength, hardness, and are highly formable.

Solid-solution strengthened alloys contain appreciable quantities of substitutional alloying elements, such as Cr, Mo, Fe, Cu, W, etc., which significantly affects alloy properties such as strength, hardness, ductility, electrical, coefficient of thermal expansion, shape memory effects, and magnetic properties beyond those of commercially pure alloys. Inconel® Alloy 600 is an example of a structural Ni-Cr-Fe solid-solution strengthened alloy used extensively in applications requiring heat and oxidation resistance such as chemical processing, heat treating equipment, aerospace, and power generation. Maximum ultimate tensile and yield strengths of solid-solution alloys approaches 120 ksi (830 MPa) and 50-70 ksi (345-480 MPa), respectively [44].
Ni-base superalloys represent a category of precipitation-strengthened alloys with very high strength (UTS>200 ksi [>1380 MPa]), especially at elevated temperature [45]. The strength and creep resistance of Ni-base superalloys often exceeds solid-solution strengthened Ni-alloys. High temperature stability of superalloys makes them an attractive engineering material in severe thermo-mechanical environments such as gas-turbine engine components in aerospace or power generation applications.

Ni-base superalloys are comprised of a face-centered cubic matrix, $\gamma$, with a dispersion $\text{Ni}_3(\text{Al},\text{Ti})$ ($\gamma'$) ordered (L12 structure) intermetallic precipitates. The ordered $\gamma'$ within a $\gamma$ matrix functions as an obstacle to dislocation motion. When dislocations move from the $\gamma$ matrix through a $\gamma'$ precipitate, an antiphase boundary (APB) (stacking fault) is created due to the disruption in normal atomic sequence for $\gamma'$. To overcome the disruption in atomic stacking sequence, a second dislocation is required to move with the first one. The strengthening effect of APB originates from the considerable energy required for formation [46]. Additional strengthening up to temperature of 700-750 ºC originates from the anomalous high temperature strength behavior of $\gamma'$; i.e., positive temperature dependence of the intrinsic strength with temperature. It is commonly accepted that this behavior is attributed to cross slip of dislocations from octahedral ($\{111\}$) to cube planes ($\{001\}$), which consequently form Kear-Wilsdorf locks. The higher critical resolved shear strength on the cube planes relative to octahedral results in additional stress required for deformation thereby increasing strength. As temperature increases further (past ~700-750ºC), thermally activated slip occurs on the cube plane with a corresponding decrease strength proportional to temperature. The deformation behavior as stated above, while commonly accepted, may be oversimplified and does not fully explain the mechanical behavior of superalloys [47].

Like solid-solution strengthened alloys, Ni-base superalloy properties are tailored by the modification of alloying additions. Alloying additions greatly affect the thermo-mechanical properties by strengthening the $\gamma$ matrix, altering the size, volume fraction, and misfit of $\gamma'$ precipitates. Additionally, alloying additions also alter the formation, morphology, and kinetics of formation of grain boundary and topologically close packed
TCP) phases, which are commonly regarded as deleterious superalloy mechanical properties. Figure 2.12 schematically shows the role of different alloying elements in Ni-base superalloys.

Thermomechanical processing and fabrication of Ni-base superalloys depend greatly on the volume fraction of $\gamma'$ [47]. As a general trend, superalloy elongation is inversely proportional to the $\gamma'$ volume factions. Wrought superalloys compositions contain a large fraction of matrix strengthening alloying additions. Gamma prime fractions of wrought superalloys can be as high as 45% [46]. Superalloys with higher fractions of $\gamma'$ are often produced by casting. Cast superalloys have gamma-prime fractions typically 50-60 vol% [46, 48]. Creep strength can be further increased by
tailoring the grain structure of cast superalloys using directional solidification techniques. Other routes for processing superalloys include powder metallurgy processing. Powder metallurgy processed superalloys possess even higher strength and creep resistance due to the elimination of coarse solidification microstructure characteristic of casting thereby allowing the formation of even higher fraction of γ’ precipitates.

The last category of Ni-base alloys includes specialty alloys. This category of alloys includes oxide dispersion strengthened (ODS) alloys. These alloys contain a fine dispersion of insoluble fine oxide particles such as Y₂O₃ and Al₂O₃. Such oxide particles act to pin dislocation motion at high temperatures and give ODS alloys excellent high temperature strength and stability. Although ODS Ni superalloys can be produced by chemical methods, solid state methods such as mechanical alloying via high energy ball milling are used instead to produce ODS superalloy powder with uniform distributions of dispersoids [49]. Joining of ODS Ni superalloys using conventional fusion welding techniques is difficult due to the inevitable destruction of the fine oxide distribution in the fusion zone [50]. Solid state joining and processing techniques such as FSW and FSP are potential processing routes that offer significantly benefits relative to fusion welding. Examples of ODS Ni superalloys include MA6000 and Inconel® MA754. Such alloys are used in high temperature engine component applications such as turbine blades [49, 51].

2.6 FSP/W of Ni-Base Alloys

2.6.1 Tooling Considerations for High-Tₘ Materials

Despite the numerous applications Ni-base alloys, the number of FSP and FSW investigations of Ni-base alloys is very limited, especially compared to the widespread use of FSW/P of Al-alloys. The majority of FSP/W tool material development is based on the development of high speed machining tool materials [52]. High process forces experienced during the FSP/W process combined with required used of costly, exotic tool
material leads to added complexity and capital investment. For example, ceramic and cermet tool materials such as PCBN and WC-Co have considerably lower fracture toughness compared to many metal systems. The low toughness tool materials are especially susceptible to fracture during the initial plunge portion of the process. Additionally, brittle tool materials (low fracture toughness) necessitate low spindle eccentricity during rotation (typically less than 0.0002 in. [53]) to avoid fracture. Such a demanding machine tolerance adds cost to the process and drives development and use of metallic tool material systems with higher fracture toughness.

Metallic-based tool materials are often more susceptible to thermomechanical instability compared to ceramic and cermet tool material systems [53]. Regardless of material system, instability such as creep and wear of the tool material leads to deleterious dimensional changes in tool geometry. No one definitive wear mechanism for friction stir tooling exists; however, it is generally accepted that wear (if it occurs) is a combination of adhesive, abrasive, and diffusive mechanisms. Contributions from the aforementioned mechanisms depends highly on the workpiece, tool material and process parameters [15, 52]. Additionally, unwanted chemical reaction between the workpiece and tool can also affect the process parameters by affecting heat generation and material flow [54]. Only with recent developments in high-temperature tool materials has the application of FSP/W to Ni-base alloys become a more feasible endeavor.

Despite a handful of studies demonstrating the feasibility of Ni-base FSP/W, development of stable, development of reliable tool materials remains an ongoing process. Table 2.2 is a comprehensive list of Ni-base alloy FSP/W studies listing the alloy type, tool materials utilized, and whether or not wear debris were detected in the workpiece after processing. Tool materials reported to date rely on cemented carbide or nitride cermet systems. Despite the ability of these tool materials to successfully create a FSP region or FSW, tool wear is an ever-present issue. Such a realization is evident from the summarized data in Table 2.2. Further development of tool materials combined with an understanding of the interplay between tool wear and process parameters for Ni-base alloys represents a gap in knowledge and is clearly a topic of further study.
Table 2.2. Comprehensive list of Ni-base alloys FSP/W studies

<table>
<thead>
<tr>
<th>Workpiece Material</th>
<th>Tool Material</th>
<th>Wear observed?</th>
<th>Author(s)</th>
<th>Year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inconel Alloy 600</td>
<td>WC-Co</td>
<td>not discussed</td>
<td>F. Ye et al. [55]</td>
<td>2006</td>
</tr>
<tr>
<td>Inconel Alloy 600</td>
<td>PCBN</td>
<td>yes</td>
<td>Y.S. Sato et al. [56]</td>
<td>2008</td>
</tr>
<tr>
<td>Inconel Alloy 600</td>
<td>WC-Co</td>
<td>yes</td>
<td>K.H. Song et al. [30]</td>
<td>2009</td>
</tr>
<tr>
<td>Inconel Alloy 738</td>
<td>WC-base</td>
<td>not discussed</td>
<td>S.M. Mousavizade [57]</td>
<td>2009</td>
</tr>
<tr>
<td>Inconel Alloy 738</td>
<td>PCBN-Mo</td>
<td>not discussed</td>
<td>O.M. Barabash [58]</td>
<td>2009</td>
</tr>
<tr>
<td>Inconel Alloy 600</td>
<td>WC-Co</td>
<td>not discussed</td>
<td>K.H. Song et al. [59]</td>
<td>2009</td>
</tr>
<tr>
<td>Inconel Alloy 600</td>
<td>WC-Co</td>
<td>yes</td>
<td>K.H. Song et al. [60]</td>
<td>2009</td>
</tr>
<tr>
<td>Inconel Alloy 625</td>
<td>WC-Co</td>
<td>yes</td>
<td>K.H. Song et al. [61]</td>
<td>2010</td>
</tr>
<tr>
<td>Inconel Alloy 718</td>
<td>PCBN</td>
<td>Yes</td>
<td>B.K. Jasthi et al [63]</td>
<td>2011</td>
</tr>
</tbody>
</table>

2.6.2 Friction Stir Processing/Welding of Ni-Cr-Fe Alloys

The feasibility of FSW Ni-base alloys was initially demonstrated in 2006 [55]. In this process-oriented study, FSW butt-welding of 2 mm Inconel® Alloy 600 (IN600) sheets was demonstrated using a PCBN tool of unknown grade. A peak temperature of 800°C was measured at the plate backside. From the micrographs presented in the study, a refined SZ is apparent; however, no quantitative measurements of grain size before or after FSW was reported. The authors allude to grain refinement as the primary mechanism responsible the modest SZ microhardness increase.
Since the 2006 study by Ye et al. [55], other FSW investigations of IN600 have been reported. The resulting microstructure of FSW IN600 has been studied in greater detail by Sato et al. [56]. The authors report two distinct SZ regions unique to FSW IN600 using PCBN tools. The authors named the two distinct regions as the black zone (BZ) and gray zone (GZ) based on the appearance after etching. In both the BZ and GZ, grains were refined relative to the BM; however the BZ exhibited a marked difference in grain size and grain boundary character. Within the BZ, the average grain size was 15 µm compared to the GZ (22 µm). Additionally, annealing twin fraction was significantly lower in the BZ compared to the GZ—6% and 24%, respectively. The authors concluded the ~1 µm particles within the BZ were CBN tool wear particles that influenced the microstructure and therefore the etching characteristics. The authors suggested that the small CBN wear particles contributed to the smaller grain size and lower annealing twin density by pinning mobile grain boundaries. The fine SZ microstructure qualitatively demonstrated a higher dislocation density in the TEM compared to mill-annealed BM. The FSP/W process introduces dislocations by continuous plastic deformation; however, a large fraction of these dislocations are eliminated by subgrain formation and coarsening during the dynamic recrystallization (DRX) process [25]. Comparisons of dislocation density of the SZ compared to the BM likely vary depending on the material system as well as the condition of the BM (i.e., degree of stored energy of cold work).

The relatively high SZ dislocation density observed by Ye et al. [55] could originate from incomplete recovery/recrystallization and/or residual plastic deformation by the passing tool [25]. Assuming that locally recrystallization was not complete after the passing tool or deformation (not stirring) occurring behind the moving tool, the lower stacking fault energy (SFE) of Ni-base alloys compared to Al-alloys also leads to more difficult rearrangement of dislocation structure (i.e, recovery) [41]. The increased SZ dislocation density observed for Ni-base alloys has potential to increase the number of active nucleation sites for precipitation [37, 64]—an interesting prospect for FSP of age-hardenable Ni-base alloys discussed further in this document.
Further studies of FSP IN600 have sought to increase the degree of SZ grain refinement by lowering linear heat input. It is difficult, however, to make a direct comparison of linear heat input between different studies due to differences in workpiece size, clamping conditions, tool material, tool/pin features, gas flow, and FSW machine. Song, Fujii, and Nakata [30] utilized relatively high (up to 250 mm/min, 9.8 IPM) traverse speed FSP runs with a WC-Co tool. Constant force and tool rotation speed FSP runs with varied traverse speeds between 150 - 250 mm/min (5.9 - 9.8 IPM) were explored. Defects aside, the average SZ grain size for the lowest heat input condition was significantly smaller (3.4 µm) than previous studies in which grain sizes varied from 15 to 22 µm [55, 56]. Additionally, such high tool travel speeds resulted in the highest fraction of low angle grain boundaries with the lowest fraction of annealing twins. The authors suggest that insufficient dynamic recrystallization occurred in the aforementioned lowest heat input FSP run.

It has been shown that reducing the linear heat input functions to decrease peak process temperatures and increasing cooling rates thereby retarding coarsening and increase the extent of refinement after processing. From a processing point of view, further reducing grain size in Ni-alloys has many potential benefits such as improved strength, wear resistance, ductility and corrosion resistance (in oxidizing environments or passive electrolytes) [65]. Techniques used on Al-alloys to further reduce grain size of FSP material such as multiple passes [66] or rapid cooling behind the FSP/W tool [67] has not been investigated for high Tₘ materials. Such techniques can potentially increase SZ homogeneity and remove features such as the ‘onion ring’ features.

2.6.3 Friction Stir Processing/Welding Ni-Cr-Mo Alloys

Song and Nakata [61] evaluated FSW IN625. The tool utilized was similar to the WC-Co tool used in previous studies by Song et al. on IN600 [30, 59, 60, 62]. The higher elevated-temperature flow stress of IN625 led to significantly higher tool downforce compared to IN600—42.1 kN versus 22.5 kN [30] measured on identical
workpiece dimensions. The differences in process forces likely accounts for increased observed tool wear. As with previous Alloy 600 FSP/W studies, a refined SZ microstructure was obtained. Hall-Petch strengthening for IN625 was more pronounced than for IN600; an increase of nearly 100 HVN within the SZ for grains reduced to 1-3 µm from 5-25 µm in the BM. While not regarded as a superalloy, strengthening via γ” precipitation can occur in Alloy 625. However, evidence of possible γ” formation as-FSW was not presented by the authors. Post weld heat treatment (PWHT) further increased hardness for all regions of the weld. Selected area electron diffraction identified the formation of some γ” and intergranular Cr-rich carbides. Tensile testing results of PWHT FSW samples were in agreement with the microhardness trends.

2.6.4 Friction stir Processing/Welding of Ni-base Superalloys

Only very recently have studies been published examining aspects of FSP/W of Ni-base superalloys. Work by Mousavizade and coworkers examined FSP casting modification of IN738 before fusion welding [57]. This unique study represents the only published study focusing on using FSP as a material modification process to influence the behavior of subsequent fusion welds on FSP-treated material. In that study, HAZ liquation cracking of as-cast IN738 Ni-alloy was investigated with and without prior FSP. Shallow (~1.5 mm depth) single pass FSP runs were performed using a WC-based tool. Autogenous welds were simulated using laser surface melting. The authors observed the elimination of HAZ grain boundary liquation cracking when material was ‘pre-treated’ using FSP prior to welding. The as-cast HAZ was comprised of a segregated dendritic microstructure with a high volume fraction of liquation-susceptible semi-continuous intergranular carbides and primary γ’. Following FSP, the majority of the intergranular carbides were broken up due to the stirring action—quite similar to microstructural changes during FSP for Al casting modification [68-70]. Consequently, the comminution and/or solutionization of large second phase particles via stirring action by the tool circumvented constitutional liquation and subsequent HAZ hot cracking entirely.
The microstructural evolution of FSP IN738 was detailed in a separate study by Barabash et al. [58]. Similarly, FSP was performed as a microstructural modification technique for castings. Friction stir processing runs approximately 6 mm in depth were made with a PCBN-Mo tool on 0.375 in. (9.5 mm) as-cast plate. Resulting FSP runs were analyzed using several metallographic techniques including SEM, EBSD, OM, and a proprietary polychromatic X-ray microdiffraction (PXM) technique to generate Laue diffraction patterns from FSP regions as small as ~0.3 x 0.5 µm.

The HAZ microstructure of FSP IN738 was not greatly affected by FSP. The as-cast microstructure consisting of γ and large primary γ’ with some secondary γ’ formed upon cooling persisted in the HAZ. Laue microdiffraction patterns from the HAZ demonstrated sharp diffraction spots indicative of a well-annealed material. More significant changes occurred in the TMAZ where Laue diffraction patterns from these regions indicated diffraction streaks along with spots. Such streaking of diffracted intensities corresponds to strong strain gradients and high dislocation density within the material. The streaking of diffraction spots is analogous to X-ray peak broadening resulting from non-uniform strain such as the strain gradients surrounding arrays of dislocations. Stored energy input (as dislocations) from FSP is high within this region results from an incomplete recovery and recrystallization processes. Gamma-prime morphological changes in the TMAZ varied based on peak temperature and strain, i.e., proximity to the SZ. By far, the SZ experienced the greatest extent of microstructural modification compared to the parent as-cast material. The initial dendritic microstructure was totally replaced by fine recrystallized equiaxed grains. The distribution of grain size within the SZ varied by location, with the majority of fine grains located towards the top of the SZ region. Onion ring shear band structures similar to those observed in FSP/W of aluminum alloys were also observed in the SZ. Closer examination with EBSD showed interpenetrating layers of 2-5 µm grains along with very fine grains (~1 µm). Microdiffraction from the SZ regions did not yield any indication as to a qualitative measure of the degree of stored energy. The significantly finer grains compared to other
regions surrounding the SZ led to significant diffraction peak broadening thereby making assessments as to the dislocation density ambiguous.

Song and Nakata [62] investigated FSW of wrought Ni-base Inconel 718 superalloy. Rather than γ’, heat treated IN718 is strengthened by γ” (Ni3Nb). The γ” precipitate formation kinetics are more sluggish compared to γ’ [45]. Resulting SZ microhardness was approximately 75 HVN higher compared to the parent material—attributed to the refinement of grains 5-25 µm in the base metal to grain 1-3 µm after FSW. In an earlier Song et al. study [30], IN600 SZ grains were between 5-20 µm for the same tool traverse speed. It can be suggested that fine precipitates and/or increased alloying content in IN718 inhibited coarsening of SZ grains upon cooling. Selected area electron diffraction patterns, however, were not gathered for the as-FSW condition. Therefore, a clear determination any fine precipitate presence after FSW was not made. As expected, a fine γ” distribution was observed in samples after 8 hr. PWHT at 720 °C. Microhardness in all regions of the weld increased as a result of the PWHT fine γ” formation.

A more recent study by Jasthi et al. [63] investigated FSP for casting modification of cast Inconel Alloy 718 to improve chemical homogeneity, close casting porosity, and comminute large dross inclusions. After FSP, the large solidification grains in the casting were reduced in size to less than 10 µm. Micrographs also suggest that large interconnected regions of dross near the surface of the plate were broken up after FSP. No detailed microstructural characterization was performed on the as-FSP material. Although, precipitation of strengthening precipitates (γ”) is not expected in FSP (based solely on typical durations of thermal cycles (<60 sec.)), the authors found the measured hardness of the SZ is only approximately 50 HV less than the fully heat treated casting. Although not explicitly mentioned, grain refinement within the stir zone likely results in the observed strengthening. In addition to grain refinement, fracture toughness measurements of SZ material using sub-size compact tension specimens indicate a nearly 30% improvement in apparent fracture toughness despite some large comminuted dross.
inclusions remaining. Significant wear of the grade CS4 PCBN tool was noted by the authors; however, the extent and distribution of wear was not discussed in detail.

2.7 Additive Friction Stir Processing

Friction stir processing is typically an autogenous process requiring no filler/additive material. However by incorporating additive methods local chemical composition and/or phase content can be combined with the beneficial microstructural modification of the friction stir process. Possible improvements in localized properties and performance are numerous and can include: improved strength, creep behavior, fatigue life, wear resistance, corrosion resistance, etc. Additionally, the severe plastic deformation under forging pressure of the friction stir tool also has the ability to improve the as-deposited microstructure of the original additive process via grain refinement in addition to elimination/reduction of discontinuities from the additive process[71].

One possible manifestation of the AFSP process is shown schematically in Figure 2.13. The generic AFSP process depicted in the illustration incorporates an additive process. This additive process could include fusion base additive processes such as laser engineered near net shaping (LENS), thermal spray, laser-assisted direct metal deposition, etc. Solid state additive processes such as cold spray, kinetic metallization, or selective laser sintering (SLS) could also be utilized as additive techniques. Because the microstructure will be changed drastically, the additive process utilized, parameters, and even deposition quality are not of great importance as long as the additive material can adhere to the substrate. Example materials that are susceptible to deposition cracks using LENS, such as high γ’-fraction superalloys, require deposition process parameters that are not conducive high deposition rates. It can be postulated that AFSP can enable the use of high deposition rates of additive material because any cracks or discontinuities formed during deposition will be eliminated thus increasing overall productivity.
Following deposition of material by the additive process, FSP is performed for purposes of microstructural modification of the additive material. Similar to FSP, beneficial microstructural changes including grain refinement, homogenization, particle comminution, and/or enhanced second phase formation kinetics can be expected. Post AFSP, a site-specific region with special properties remains. Properties within this region are not otherwise achievable using either the additive process or FSP alone.

While not performed in the manner as previously described, there exist several studies that address the feasibility of FSP as an additive method. In such prior studies, dissimilar materials typically in the form of unconsolidated powder were added to the parent substrate by filling machined holes or channels in the parent substrate. The majority of existing FSP studies focusing on the addition of a second phase have focused on the formation of in-situ metal matrix composites (MMC).

The solid state nature of FSP is advantageous for Al- and Mg-MMC fabrication because very limited or no reactivity occurs between the matrix and reinforcing phase during processing. Other higher temperature processing routes involving liquid phases lead to deleterious reactions between the matrix and the reinforcing phase. In its current state, the solid state nature of FSP makes it an attractive method for site-specific fabrication of near-surface light metal based MMCs. Previous work has included the addition via FSP of WC to AA7050 [72], single-wall carbon nano tubes to AA7075 [73], and SiC to AA5083 [74].
In contrast to the aforementioned investigations which incorporate dissimilar materials with the intent of no reactivity, additive FSP with the intention of compositional modification to affect microstructural evolution has been studied considerably less. In a study by Inada et al. [75], copper powder was incorporated via FSP in commercially pure AA1050 by filling an intentional gap between two Al plates in butt configuration with Cu powder. Macroscopically, the distribution of Cu particles in the Al matrix was not uniform—even after FSP by two overlapping passes (Figure 2.14). However, microhardness measurements of the double pass runs show the SZ exhibits microhardness nearly twice the base metal hardness of 45 HVN. Upon further examination of these regions using TEM, the presence of nano-scale Al$_2$Cu precipitates was observed. These precipitates led to the increased hardness within the SZ. The authors, however, did not discuss in detail the precipitation phenomena.

Figure 2.14. Brightfield TEM micrograph of Al$_2$Cu precipitate formation after two FSP passes [75].

For materials with $T_m$ significantly higher than Al-alloys, very few AFSP studies exist. Work by Mukherjee and Ghosh [71] explored the combination of laser-assisted
direct metal deposition (DMD) with FSP for corrosion hole repair in 70/30 copper-nickel alloy. This additive FSP process allows for site-specific filling and repair while eliminating deleterious features of laser DMD microstructures including porosity, large grains size, residual stress, and solute segregation. Although the AFSP process as utilized in Mukherjee and Ghosh’s work was not used to change chemistry, the additive Cu-Ni alloy region exhibited several benefits relative to DMD alone. The additive FSP region demonstrated higher yield and tensile strengths as well as reduced porosity from 3.3% to 0.35% and eliminated Cu segregation typical of the DMD solidification microstructure.

High temperature AFSP was also reported by Shamsipur et al. [76]. In this work, AFSP was used to create location-specific Ti/SiC surface composites. SiC powder was introduced into the Ti plate via powder-filled channels. Up to four overlapping passes were used to create a homogenous distribution of nano-sized SiC particles within the SZ (Figure 2.15). Surface layers with microhardness values approximately 3.3 times greater than the as-received CP Ti-alloy were successfully fabricated. The authors observe the presence of nano-size SiC within the SZ that contribute to the high hardness in two ways. The high intrinsic hardness of SiC particles within a composite microstructure increased hardness. Secondly, the nano-scale SiC particles contributed significantly to grain boundary pinning. The restriction of grain boundary mobility during dynamic recovery/recrystallization led to the development of a highly refined stir zone microstructure with an average grain size of 400 nm. Under identical FSP parameters without the addition of SiC particles, the average grain size was an order of magnitude larger. The authors believed a reaction of the Ti matrix and reinforcing SiC particles occurred as a result of AFSP; however, it was not directly observed.
Currently, additive FSP processes involving dissimilar materials or additions of materials to react with the parent matrix upon processing do not exist for high $T_m$ systems, including Ni-alloys. Incorporation of additional materials or dissimilar Ni-alloys by AFSP to locally enhance material properties/performance is a promising subject that warrants further study.

2.8 Fusion Weld Modification via Friction Stir Processing

Some applications for FSP have included crack repair or microstructural modification of fusion welds. A handful of studies have investigated using FSP as a post-arc welding technique to reduce weld discontinuities and improve the weld mechanical properties of various Al-alloys [77, 78] and NiAl bronze [79]. However, very little attention has been focused on the prospect of using FSP to modify base material microstructure such that weldability issues such as hot cracking are reduced or eliminated for subsequent fusion welds. FSP pretreatment can be used as an alternative conditioning process for the rejuvenation of microstructures with deleterious features or phases thus
facilitating subsequent welding. One possible application for FSP pretreatment includes rejuvenation of Inconel Alloy 718. The presence of large quantities of δ phase (Ni$_3$Nb, orthorhombic) present in susceptible Alloy 718 structures exposed to many thermal cycles poses weldability issues due to grain boundary liquation from δ phase dissolution that occurs during repair welding thermal cycles [80]. Location specific microstructural homogenization via FSP pretreatment can enable localized repair welding without the performing time-consuming homogenization heat treatments usually required.

Naturally, welding in the solid state via FSW is a simpler process than the combination of both FSP and fusion welding. However in some situations, using FSW entirely is not entirely practical—especially for high $T_m$ materials. By nature of the FSW process, joint geometries are limited to primarily butt and lap configurations. Other joint configurations require complex fixturing and/or complex machine control schemes [81]. Additionally, high process forces experienced during FSW of high $T_m$ materials, especially for thick sections, limits welding to only very robust FSW machines with large associated workspace footprints. Such practical limitations hinder FSW capabilities in the field. One can envision FSP pretreatment performed remotely to only modify the microstructure where necessary, e.g. a portion of the workpiece that is under high restraint, stress concentration, etc. The locally modified workpiece can then be transported to the field after FSP where final joining can be accomplished via conventional fusion welding techniques.

To date, only a single study has investigated the utility of using FSP as a pretreatment for microstructural modification of subsequent fusion welds. The unique work, by Mousavizade et al. [57], focused on the effect of FSP pretreatment prior to laser welding of cast IN738. The top ~1.5 mm of 5 mm thick IN738 plate was friction stir processed. Following FSP, the SZ region was laser surface remelted. Any cracking that occurred during laser surface remelting was the result of inherent mechanical restraint. The authors noted qualitative improvements in HAZ liquation cracking susceptibility due to increased material homogeneity and refinement of liquation-inducing microconstituents from FSP. The HAZ of non-FSP IN738 contained many cracks
transversely oriented the fusion boundary. In contrast, the FSP treated material exhibited no cracks in the near-fusion boundary HAZ after laser surface remelting. The authors suggest that constitutional liquation of coarse primary $\gamma'$ and interdendritic $\gamma-\gamma'$ eutectic in the cast base material was circumvented as a result of FSP from breakdown of interdendritic eutectic structures as well as refinement of $\gamma'$ compared to the as-cast material. While $\gamma'$ generally not regarded as a phase contributing to constitutional liquation; evidence of constitutional liquation has been observed in IN738 in a few studies [82-84].

Although not explicitly discussed by the authors, it is not likely that the liquating phases of IN738 are simply broken up as is the case with FSP of high-Si aluminum alloys with large fractions of intermetallic constituents [85]. It is more likely that large primary $\gamma'$ and $\gamma-\gamma'$ eutectic goes into solution during FSP and reprecipitates upon cooling after FSP. The reprecipitated $\gamma'$ would be considerably finer than in interdendritic regions of the cast base material. The significantly finer $\gamma'$ reduces the constitutional liquation susceptibility. Mousavizade et al. [57] used an analytical model outlined by Ojo et al. [86] to predict the dissolution of $\gamma'$ precipitates. The authors determined that refinement of $\gamma'$ due to FSP resulted in dissolution occurring nearly 80 times faster compared to the base material. The more rapid dissolution of finer $\gamma'$ precludes constitutional liquation. Other microstructural changes occurring within the HAZ of the laser weld resulting from FSP pretreatment were not discussed. It is very likely the HAZ of the FSP treated material was more refined with respect to grain size compared to the base material. Heat affected zone refinement of fusion welds resulting from FSP pre-treatment is discussed in a following section of this document.

Beneficial changes resulting from FSP pretreatment are not limited to the HAZ and may also occur in the weld metal (WM) of pretreated material due to the refinement obtained from FSP. The WM grain structure during arc welding is primarily controlled by epitaxial nucleation and preferred growth from the fusion boundary with competitive growth dominating remote from the fusion boundary [87]. Like a casting, the WM grain structure can be directly influenced by altering the extent of nucleation [88]—with
increased nuclei density leading to decreased grain size. Many studies have investigated the effects of increasing the heterogeneous nucleation site density to refine WM grains by using additions of inoculant particles. Studies have been performed in several alloy systems including ferritic steels [89], Al-Zn-Mg alloys [90], and Al-Li alloys [91]. However, a significant downside to the practice of introducing inoculant particles is the alloy-specific change in chemistry required for the base material or filler material. Additionally the thermal stability of the particles makes the inoculation effect somewhat dependent on the welding parameters. For some applications where WM refinement is sought, a change in alloy composition is not feasible, e.g., autogenous welding. Non-compositional methods such as arc manipulation techniques can also be used to change the extent of nucleation in fusion welds via several mechanisms including decreased weld pool temperature. [88]. Example methods include electromagnetic stirring [92-94], mechanical vibration [92, 95], and AC pulsed current [96, 97]. These techniques have demonstrated effectiveness in altering WM grain morphology. However, arc manipulation requires direct implementation of additional equipment, and adds complexity to the welding processes. Furthermore, additional parameters such as arc oscillation and current pulse frequency must be developed and controlled for different materials. Increasing nucleation site density by grain refinement using FSP prior to fusion welding is a promising straightforward technique that does not require compositional or fusion welding process parameter alternations.
CHAPTER 3: RESEARCH OBJECTIVES

Based on the limited number of studies described in detail in Chapter 2, the feasibility of Ni-base alloy FSP/W has been demonstrated for a range of alloys from low-strength solid solution-strengthened alloys such as IN600 to some high-strength superalloys such as IN718 and IN 738. Due to the current limitation in conventional FSP/W machinery and a lack of suitable robust tool materials, a widespread understanding and application of FSP for Ni-alloys is still in initial stages and requires further investigation. Moreover, manipulation of near-surface microstructure using AFSP has not been explored to any degree for Ni-alloy systems. The following list summarizes the salient objectives of this research.

1.) Evaluate the effects of varied FSP process parameters on measureable process outputs (e.g., process forces) and resulting microstructure. The varied FSP process parameters will be used to develop process parameter windows for subsequent FSP work.

2.) Characterize the stored energy distribution within FSP microstructures using advanced characterization techniques as a function of varied FSP process parameter combinations.

3.) Develop and optimize techniques to incorporate dissimilar material additions for AFSP of Ni-alloys.

4.) Characterize the flow, distribution, and microstructural evolution of additive Ni-base alloys the SZ.
5.) Gain an understanding of the interplay between recrystallization, AFSP process parameters, and additive material composition on the resulting SZ microstructure.

6.) Investigate grain refinement in Ni-alloys produced when FSP is used to pretreat base materials prior to fusion welding.
CHAPTER 4: EXPERIMENTAL APPROACH

4.1 Materials

A wide variety of Ni-base alloys were evaluated in this study. Inconel® Alloy 600 (IN600) and commercially pure Ni, Ni-201, were used as substrate materials. All substrate materials were in 0.25 in. plate form. IN600 was provided by Huntington Alloys Corp., Huntington, WV. The melt was produced via electric furnace melting using argon oxygen decarburization followed by electroslag remelting. Plate material was hot rolled, annealed, and pickled. Ni-201 was also provided by Huntington Alloys Corp. The melt was produced using argon induction melting followed by electroslag remelting. Material was provided in hot-rolled and annealed conditions. Table 4.1 shows the composition of the heats of IN600 and Ni-201 used in this study along with mechanical properties provided by the vendor. Figure 4.1 and Figure 4.2 show the typical base metal microstructure of IN600 and Ni-201, respectively. Both materials exhibit predominantly equiaxed recrystallized grain structure typical of hot-rolled and annealed plate material.
Figure 4.1. Brightfield optical micrograph of IN600 base material. 10% oxalic acid electrolytic etch.

Figure 4.2. Differential interference contrast optical micrograph of Ni-201 base material. Glacial acetic + HNO₃ swab etch.
Table 4.1. Measured chemical compositions and mechanical properties of substrate alloys IN600 and Ni-201.

<table>
<thead>
<tr>
<th>Element (wt%)</th>
<th>IN600</th>
<th>Ni-201</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr</td>
<td>16.69</td>
<td>---</td>
</tr>
<tr>
<td>Fe</td>
<td>9.36</td>
<td>0.02</td>
</tr>
<tr>
<td>Mn</td>
<td>0.21</td>
<td>0.23</td>
</tr>
<tr>
<td>Al</td>
<td>0.188</td>
<td>---</td>
</tr>
<tr>
<td>Ti</td>
<td>0.22</td>
<td>---</td>
</tr>
<tr>
<td>Si</td>
<td>0.12</td>
<td>0.05</td>
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<tr>
<td>C</td>
<td>0.08</td>
<td>0.01</td>
</tr>
<tr>
<td>P</td>
<td>0.005</td>
<td>---</td>
</tr>
<tr>
<td>S</td>
<td>&lt;0.001</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Cu</td>
<td>---</td>
<td>0.01</td>
</tr>
<tr>
<td>Co</td>
<td>---</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Ni</td>
<td>balance</td>
<td>balance</td>
</tr>
<tr>
<td><strong>Mechanical Properties from Mill Certification</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hardness (HRB)</td>
<td>85.6</td>
<td>61.4</td>
</tr>
<tr>
<td>0.2% Yield Strength (ksi)</td>
<td>49.0</td>
<td>33.2</td>
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<tr>
<td>Tensile Strength (ksi)</td>
<td>99.3</td>
<td>55.0</td>
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<tr>
<td>Elongation (%)</td>
<td>41.4</td>
<td>46.2</td>
</tr>
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</table>

Several other alloys were utilized in this work as additive materials. Superalloys including Haynes® Alloy 282, Alloy 214, Mar-M-247, and René 41 were utilized in various forms. Haynes Alloy 282 and Alloy 214 wire, provided by Haynes International (Kokomo, IN, USA), was used as an additive material in a cold-wire gas tungsten arc welding (GTAW) process. Both Mar-M 247 and René 41 were utilized in powder form.
(Sandvik Osprey Powder Co., Sandviken, Sweden) with a size distribution of -63+32 \( \mu \text{m} \).

Powders were incorporated as an additive material using the LENS process. Lastly, titanium was incorporated into substrate alloys directly using AFSP techniques. Gas atomized -100+325 mesh Ti-6Al-4V powder (Crucible Research Corp., Pittsburgh, PA) was utilized as the Ti source. Compositions of the various additive materials can be found in Table 4.2
Table 4.2. Compositions of additive materials utilized in this study. Asterisk denotes nominal composition

<table>
<thead>
<tr>
<th>Element (wt%)</th>
<th>Alloy 282*</th>
<th>Alloy 214</th>
<th>Mar-M247*</th>
<th>René 41*</th>
<th>Ti-6Al-4V</th>
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<tr>
<td>Al</td>
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<td>4.23</td>
<td>5.5</td>
<td>1.6</td>
<td>6.3</td>
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<tr>
<td>Cr</td>
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<tr>
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<tr>
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<tr>
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<tr>
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<td>&lt;0.1</td>
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<tr>
<td>Ta</td>
<td>---</td>
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<td>3</td>
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<tr>
<td>Ti</td>
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<td>&lt;0.1</td>
<td>1</td>
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<tr>
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<tr>
<td>Y</td>
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<td>wire (0.045”)</td>
<td>-63 +32 μm powder</td>
<td>-150 +45 μm powder</td>
<td>-100+325 mesh powder</td>
</tr>
</tbody>
</table>

51
4.2 Friction Stir Processing

All friction stir processing experiments were performed using an Accustir (General Tool Corporation, Cincinnati, OH) gantry-style FSW machine. Figure 4.3 shows a photograph of the FSW machine. Both conventional (i.e., Al- and Mg-alloys) as well as high temperature FSP/W can be performed on the Accustir machine. The high-process force capability of the Accustir machine is suitable for welding/processing of thick sections or materials with high elevated-temperature flow stress. Applied forces up to 60 kip (267 kN) can be applied in the direction normal to the workpiece. Additionally a maximum force of 15 kip (67 kN) can be applied to the tool travel direction and normal to the tool travel direction, respectively. The tool spindle motor features a high-torque motor capable of producing 1350 ft·lb (1830 N·m) of torque at the tool at spindle speeds of up to 1000 RPM. Movement of the machine in the principle directions (i.e., plunge, travel and travel normal) is repeatable to within 0.001 in. (25.4 µm) An additional tilt axis in the spindle head allows the angle of tool tilt to be varied.

Figure 4.3. General Tool Corp. Accustir gantry-style friction stir machine
High-speed force data during FSP is logged during machine use. Relevant process forces such as tool normal force, travel force, lateral force, and spindle torque are all recorded at rates up to 10 Hz. Such data is useful in the development of FSP processing parameter windows.

Tooling for FSP/W of high-temperature materials remains under continued development. For this study, tungsten-based alloy friction stir tools were chosen. Other viable high-temperature ceramic and cermet tooling such as polycrystalline cubic boron nitride require specialized tool holder and spindle run-out tolerance to use reliably. Friction stir tools were fabricated from either W-25Re or W-1wt% La$_2$O$_3$ bar stock using diamond grinding to produce tool features. All tools utilized in this study featured machined internal channels for water cooling. Cooling of the W-based tools is necessary to maintain dimensional stability of the tool during FSP of Ni-base alloys. Several tool geometry iterations were tested before selecting final feature geometries. All tools tested were based on un-threaded truncated conical pin designs with featureless, flat shoulders. Such geometries are derivatives of the Variable Penetration Tool developed by Edison Welding Institute. Friction stir processing tools were designed to create a stir-processed region that was nominally 0.125 in. (3.2 mm) in depth. Figure 4.4 shows a schematic of the W-25Re tool used to produce FSP and AFSP runs on IN600 substrates.
All FSP trials were performed on 0.25 in. thick plate material nominally 3 X 6 in. in width and length, respectively. A trailing gas-shield was used in conjunction with flowing Ar gas to prevent excessive oxidation of the plate surface during FSP. For all FSP runs, a constant tool tilt angle of 3° was utilized. To vary the heat input into the workpiece, parameters including the spindle speed and travel speed were varied. Additionally, the amount of shoulder plunge into the material was initially varied to provide sufficient consolidation of material behind FSP tool during the traverse portion of the run. Most FSP and AFSP trials used a nominal shoulder plunge depth of 0.040 in. (1.01 mm) into the surface of the plate.

4.2.1 Thermal Acquisition during FSP

The thermal history of material undergoing FSP was recorded using thermocouples embedded in the workpiece. Thermal measurements during FSP/W are often difficult to perform due to the highly dynamic flow of material under high applied loads within stir zone region.
Thermocouples (ANSI Type K) were embedded into machined holes placed in the workpiece plates at varied depths to capture the thermal history from various locations near the SZ including the SZ centerline, advancing, and retreating side. Figure 4.5 shows the locations of the thermocouple holes relative to the FSP/W tool. Figure 4.6 shows the locations of machined thermocouple holes on the backside of the plate relative to the tool travel direction.

![Figure 4.5. Locations of thermocouples placed through the plate backside relative to the FSP tool. All dimensions are shown in inches.](image)

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Figure 4.6. Locations of the three sets of RS-center-AS thermocouples shown in plan view.

The Type-K thermocouples were welded to the bottom of the machined thermocouple holes using a percussion thermocouple welder. To ensure the thermocouples remained within the holes on the plate backside, the alumina thermocouple sheaths were affixed within the holes using thermocouple cement. During an instrumented FSP run, the output of eight thermocouple channels was acquired simultaneously. Temperature data was acquired at an acquisition rate of 500 Hz.

4.2.2 Additive Friction Stir Processing Techniques

4.2.2.1 Powder Incorporation
Several different powder incorporation techniques were explored to incorporate Ti into Ni-201. Several studies investigating FSP for in-situ metal matrix composites have successfully used powder additions [72-75]. Gas atomized -100+325 mesh Ti-6Al-4V powder (Table 4.2) was utilized as the Ti source. Initial incorporation techniques involved mixing Ti-alloy powder with polymeric organic binders. The binder was added to prevent the spherical Ti-powder particles from being carried away due to high shielding gas flow rate used during FSP. The Ti-powder/binder mixture was packed into a series of 2 mm diameter holes spaced 5 mm apart located along the centerline of the plate. Binderless incorporation techniques were also explored. Ti-powder was placed into holes and a GTAW spot weld was created adjacent to the hole to consolidate the powder via melting and/or partial sintering. Figure 4.7 shows a photograph of a Ni-201 plates with Ti-powder filled holes consolidated using spot welding. After spot welding near the powder filled hole, FSP was performed over the holes.

Figure 4.7. Ni-201 plate with Ti-powder in drilled holes. GTAW spot welds are placed adjacent to holes pre-sinter Ti-powder

4.2.2.2 Cold-Wire Gas Tungsten Arc Overlays
Weld metal overlays of Haynes Alloy 282 and Alloy 214 on IN600 substrates were created using cold-wire gas tungsten arc welding (GTAW). In this process, a coiled filler metal wire is fed from a wire feeder mechanism into the leading edge of a GTAW weld pool. A programmable GTAW machine (Jetline Engineering Inc., model: TKM-72-M, Irvine, CA) was used to produce shallow overlay beads with low dilution with a welding speed, current, voltage, and wire feed speed of 1.27 mm/s (3 IPM), 105 A, 9.8 V, 6.56 mm/s (15.5 IPM), respectively. A total of four passes were placed atop a 3 x 6 x 0.25 in. IN600 plate with approximately 33% overlap. This particular overlap produced a relatively flat deposition surface that could then be subsequently friction stir processed with no post-deposition machining.

4.2.2.3 Laser-Engineering Net Near Shaping (LENS).

The lack of available filler metal wire for Ni-base superalloys, especially alloys with high Al + Ti content, necessitated alternative additive material techniques. For Mar-M247 and René 41 (Table 4.2), LENS was used to create depositions on IN600 substrates. All LENS depositions were performed on a LENS 850R (Optomec Inc., Albuquerque, NM) machine. Prior to creating LENS depositions, the IN600 substrates were cleaned ultrasonically using acetone. Table 4.3 lists pertinent process parameters used to create depositions within 1 in. wide channels machined in IN600 plate to a depth of approximately 1 mm. Prior to the first deposition pass, the surface of the IN600 plate was remelted by rastering the laser over the plate surface. After laser surface remelting, the first deposition layer was then created. The number of deposition layers required varied from 7-10, depending largely on the powder feed rate. To create each layer, the laser was rastered across the plates, thus creating multiple passes. The approximate width of each pass was 0.84 mm. An approximate pass overlap of 30% was utilized.
Table 4.3. LENS parameters used to create additive material depositions

<table>
<thead>
<tr>
<th>Deposition Depth (mm)</th>
<th>Approximately 1 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Powder Auger Speed (RPM)</td>
<td>5-10</td>
</tr>
<tr>
<td>Laser Power (W)</td>
<td>850</td>
</tr>
<tr>
<td>Laser Travel Speed (mm/min)</td>
<td>3000</td>
</tr>
<tr>
<td>Dwell between layers (sec.)</td>
<td>30</td>
</tr>
<tr>
<td>Number of passes per layer</td>
<td>30</td>
</tr>
<tr>
<td>Carrier Gas</td>
<td>Ar</td>
</tr>
</tbody>
</table>

### 4.3 Thermomechanical Simulation

The heating and cooling thermal histories measured during FSP were simulated using the Gleeble® 3800 (Dynamic Systems Inc., Poestenkill, NY) thermo-mechanical physical simulator (Figure 4.8). The Gleeble is able to replicate microstructures that result from dynamic thermal cycles (such as those experienced during FSP) that are otherwise very difficult or impossible using traditional furnace tests. The Gleeble is typically used to perform a wide variety of weldability and formability tests such as hot ductility, HAZ thermal cycle simulation, PWHT simulation, strain-to-fracture, reheat cracking, etc. Thermocouple-instrumented test specimens in the Gleeble are heated resistively. Thermocouples directly welded to the samples interface with a closed-loop thermal control system. The closed loop thermal system alloys for very precise control of dynamic heating rates as high as 10,000°C/sec. Additionally force can be applied to the sample during heating/cooling via a closed-loop hydraulic servo mechanism. Both tensile (up to 10,000 kgf) and compressive (up to 20,000 kgf) forces can be generated.
using the Gleeble 3800 with extension rates varying between 0.01 mm/sec to 2000 mm/sec. Figure 4.9 shows a schematic representation of the Gleeble® 3800 system.

Figure 4.8. Gleeble® 3800 Thermomechanical Simulator

Figure 4.9. Schematic of the Gleeble® 3500/3800 system. Image courtesy of Dynamic Systems Incorporated.
By nature of the closed feedback loop, no gauge section is required for the test specimen prior, unlike a traditional mechanical test specimen. Material in the vicinity of the control thermocouple becomes the ‘thermal’ gauge section during testing. Heating rate during testing in the Gleeble is a function of the applied current and current density. The maximum achievable cooling rate depends on several testing parameters such as the sample size, jaw grip material, cooling water flow, chamber atmosphere, and jaw free span. Lastly, the Gleeble system has additional features that can augment the cooling rate by forced gas or water quenching.

4.4 Thermodynamic Modeling

Two thermodynamic modeling software packages were used in this study: Thermo-Calc and JMatPro.

4.4.1 Thermo-Calc

Thermo-Calc is a software package originally developed by the Swedish Royal Institute of Technology [98]. Thermo-Calc is able to perform thermodynamic and phase diagram calculations for complex multi-component alloy systems using the CAalculation of PHAse Diagram (CALPHAD) method, a technique that involves global minimization of Gibbs free energy. Calculations can include simple thermodynamic data such as heat capacities, enthalpies, etc. to more complex predictions such as partitioning coefficient determination and solidification simulations using the Scheil-Gulliver model. Calculations performed are based on data contained in databases consisting of extensive compiled experimental data. For the thermodynamic calculations performed in this study, the following Thermo-Calc databases were utilized: SSOL4 (SGTE Solutions database v.4 for multicomponent solution phases; contains 78 elements) and TTN17 (Thermotech Ni-based superalloys v.7 for Thermo-Calc version R was utilized in this
work. Some alloys examined within this work (e.g., Mar-M247 and René 41) were specifically used in the validation of the TTNI7 database [99].

### 4.4.2 JMatPro

JMatPro is a Java-based software package developed by Sente Software (version 5.1, Sente Software Ltd., UK) used to calculate the behavior and properties of multicomponent systems. JMatPro differs from Thermo-Calc in that various theoretical models are used in conjunction with thermodynamic calculations performed using global minimization techniques similar to those used in Thermo-Calc. Extensive databases used for physical properties are linked to the thermodynamic calculation capabilities. One of the useful capabilities of JMatPro is the ability to incorporate kinetic calculation capabilities—specifically for the calculation of time-temperature-transformation (TTT) and continuous cooling transformation (CCT) curves [100]. Additional capabilities of JMatPro include the ability to predict behavior such as high temperature mechanical behavior (e.g., yield and ultimate tensile strength) by incorporating several strengthening models. As an example, Figure 4.10 demonstrates good agreement between calculated yield strength for a Ni-base superalloy as a function of $\gamma$’ size for various temperatures. For findings presented in this document, all calculations performed using JMatPro utilized the ‘Nickel Base Superalloy’ database.
Figure 4.10. A comparison between calculated (data markers) and experimental (lines) yield strength behavior as a function of $\gamma'$ size at various temperatures for a Ni-20Cr-1.2Co-2.4Ti-1.5Al superalloy. (Figure adapted from Saunders et al. [100])

4.5 Characterization

4.5.1 Scanning Electron Microscopy

Several different scanning electron microscopes are utilized in this work. The microscopes vary based on the type of electron source, vacuum system, installed analytical systems, and lens systems. Table 4.4 lists the utilized scanning electron microscopes along with notable features and special functions. High resolution electron microscopy is utilized heavily in this work to image at very high magnifications (>50 kX). More detailed discussion on the imaging mechanism can be found in Appendix A.
Table 4.4. Comparison of features for the three scopes utilized for analyses performed in this document

<table>
<thead>
<tr>
<th></th>
<th>FEI Quanta 200</th>
<th>Philips XL30F</th>
<th>FEI Sirion</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Microscope</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Electron Gun Type</strong></td>
<td>W-thermionic</td>
<td>W cold field emission</td>
<td>W cold field emission</td>
</tr>
<tr>
<td><strong>Accelerating Voltage Range</strong></td>
<td>200 V - 30 kV</td>
<td>200 V - 30 kV</td>
<td>200 V - 30 kV</td>
</tr>
<tr>
<td><strong>Secondary Electron Detector</strong></td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td><strong>Solid-state BSE detector</strong></td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td><strong>Electron Backscatter Diffraction</strong></td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td><strong>Through-lens Scintillation Detector</strong></td>
<td></td>
<td>X</td>
<td></td>
</tr>
<tr>
<td><strong>Energy Dispersive Spectroscopy</strong></td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td><strong>Environmental/Partial Pressure Mode</strong></td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td><strong>Gaseous Secondary Electron Detector</strong></td>
<td>X</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

4.5.2 Electron Backscatter Diffraction

Electron backscatter diffraction (EBSD) is a characterization method performed in the SEM that gathers crystallographic orientation information from diffraction patterns formed by backscatter electrons generated from the incident electron beam. Electron backscatter diffraction was used to determine the microstructural effect of FSP pre-treatment as conventional light-optical micrographs do not give sufficient information regarding near-fusion boundary grain structure. Additionally, EBSD was used to create maps of crystallographic orientation within various friction stir processed regions. Among the wide range of information gathered via EBSD, the crystallographic misorientation can be characterized. This misorientation information can be used to qualitatively determine the degree of stored energy within the SZ. Additionally, the quality of acquired diffraction patterns, which can be qualitatively determined, varies based on the amount of stored energy. Electron backscatter diffraction experiments were performed on the Philips XL30F FEG SEM on well-polished samples. Typically an
accelerating voltage of 25 kV and spot size of 5 was used to generate satisfactory Kikuchi patterns for scans using a step size of greater than ~500 nm. Higher magnification/resolution scans (<500 nm) utilized lower accelerating voltages and beam currents (20 kV; SS4) to reduce the effective interaction volume. Working distances used were between 25-30 mm and largely depended on the size of the sample and area of scan. Larger scan areas required smaller convergence angles provided by the longer working distances to prevent significant beam defocus near the edge of the analysis area. For large scan areas, scan rate is paramount. The CCD camera parameters such as exposure and binning were chosen such that camera frame rate was maximized (~45 frames/sec.). Smaller scan regions, especially where more material deformation is expected, required significantly slower scan rates associated with lower image binning (~10 frame/sec.) to capture sufficient pattern detail. The exact frame rates used depended largely on the beam parameters chosen (i.e., source brightness). Interestingly, source brightness was found to vary considerably.

Proprietary EBSD software packages were used for automated analysis of acquired diffraction pattern (TSL OIM Acquisition v.5.2, Ametek/EDAX/TSL, Draper, UT). Collected EBSD data was analyzed using TSL OIM™ Data Analysis v5.0 software (Ametek/EDAX/TSL, Draper, UT). Post-processing to remove errant data points was performed using the nearest neighbor confidence index (CI) correlation technique. Data points with a CI of less than 0.1 were extrapolated using neighboring CI values. Additionally, the grain dilation post-processing technique was also used for data points with insufficient CI.

4.5.2 Transmission Electron Microscopy and Focused Ion Beam Sample Preparation

Characteristics of AFSP such as location-specific microstructural modification and dissimilar material combination makes TEM sample preparation using conventional sectioning, grinding, and electropolishing techniques rather cumbersome. Precise site-
specific sample extraction and preparation was performed using the focused ion beam (FIB) technique by which a TEM foil is milled, extracted, and thinned from the surface of a bulk sample. A focused beam of high energy gallium ions is used to ablate and sputter away material from the surface of the bulk sample. Using specific milling patterns, a specimen is then extracted, placed onto a substrate, and thinned such that it is suitably electron transparent for TEM analysis. A Helios 600 (FEI Co., Hillsboro, OR) dual beam (i.e., instrument with electron and ion beam capabilities) FIB was used for TEM sample preparation. Final thinning was performed at an ion accelerating voltage of 5 kV. Foils were thinned to approximately 250 nm.

Thinned foils were examined using a Tecnai F20 (FEI Co., Hillsboro, OR) scanning/transmission electron microscope (S/TEM) using STEM mode in which the incident beam is converged and scanned across the region of the sample. Compared to the TEM, the STEM can image thicker samples and produces micrographs with considerably more uniform contrast by the reduction in dynamic contrast formation such as thickness fringes, bend contours, etc. Additionally, STEM is advantageous in obtaining contrast from dislocation even when far from the Bragg condition due to the large deviation parameter. This allows the operator to image the dislocation structure across multiple grains.

Brightfield (BF) imaging in STEM mode is similar to TEM mode in that the formed image is comprised from a detector that captures the direct beam and electrons that have been scattered to only very small angles. Additional imaging modes include high angle annular darkfield imaging (HAADF), an imaging mode unique to the STEM mode. In this mode, an annular detector collects electrons scattered to very high angles and therefore excludes diffracted signal. Because only electrons scattered at very high angles are collected, this mode is sensitive to atomic number and thickness. The working distance of the detector can be changed by the operator which in turn changes the collection solid angle and determines the amount of diffracted signal contributing to the image formation. Both BF and HAADF imaging modes were utilized in this work.
4.5.3 Light Optical Microscopy

Light Optical Microscopy (LOM) was performed using an Olympus GX-51 inverted stage metallographic microscope combined with a 12MP Olympus DP71 digital imaging system. The GX-51 series microscope allows for several imaging modes at magnifications from 12.5 to 1000X. Such modes include conventional brightfield, polarized light, and differential interference contrast.

4.6 Metallographic Preparation

4.6.1 Sectioning, Grinding, and Polishing

Sectioning of metallographic specimens was performed using a water-cooled abrasive chop saw (Leco CM-15) for rough cuts. Precision sectioning was performed using a Leco VC-50 diamond wafering saw. Samples intended for examination in the SEM were mounted in graphite-filled Bakelite. Mechanized grinding of metallographic specimens was performed using a Leco (Spectrum System 1000) automatic polisher/grinder. Compared to manual grinding techniques, better results were obtained via automatic grinding due to higher applied loads. The dissimilar material nature of many of the samples examined in this study created some metallographic preparation challenges. Due to the widely varied hardness for some AFSP samples, local material removal rates during polishing led to significant surface relief. Severe surface relief created ‘shadowing’ effects when samples were tilted to high angles such as during EBSD analysis. To reduce the degree of surface relief, polishing techniques were developed that maximized surface flatness. This was primarily accomplished by using non-woven polishing cloths with very low nap height. Table 4.5 shows the grinding and manual polishing procedure for preparation of all Ni-base samples examined in this work. Automated polishing was also utilized. A Buehler MiniMet 2 automatic grinder/polisher
was used to prepare samples with a high degree of repeatability. The polishing schedule used for the Buehler MiniMet is shown in Table 4.7.

Table 4.5. Grinding and polishing sequence used for manual polishing

<table>
<thead>
<tr>
<th>Step</th>
<th>RPM</th>
<th>Direction</th>
<th>Load</th>
<th>Time</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Grind</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>180 grit SiC</td>
<td>200</td>
<td>contra.</td>
<td>6</td>
<td>2</td>
</tr>
<tr>
<td>320 grit SiC</td>
<td>200</td>
<td>contra.</td>
<td>6</td>
<td>2</td>
</tr>
<tr>
<td>600 grit SiC</td>
<td>200</td>
<td>contra.</td>
<td>6</td>
<td>2</td>
</tr>
<tr>
<td>800 grit SiC</td>
<td>100</td>
<td>stationary</td>
<td>manual</td>
<td>1</td>
</tr>
<tr>
<td><strong>Polish</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6 µm polycrystalline diamond paste on</td>
<td>100</td>
<td>stationary</td>
<td>manual</td>
<td>3</td>
</tr>
<tr>
<td>3 µm polycrystalline diamond paste on</td>
<td>100</td>
<td>stationary</td>
<td>manual</td>
<td>3</td>
</tr>
<tr>
<td>1 µm diamond polycrystalline diamond</td>
<td>100</td>
<td>stationary</td>
<td>manual</td>
<td>3</td>
</tr>
<tr>
<td>Final polish: Vibratory polish with</td>
<td>n/a</td>
<td>---</td>
<td>1</td>
<td>1-12 hrs.</td>
</tr>
<tr>
<td>0.05µm Buehler MaterMet 2 colloidal SiO2 on Buehler MaterTex pad</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 4.6. Alternative automatic polishing schedule utilized in conjunction with Buehler MiniMet

<table>
<thead>
<tr>
<th>Step</th>
<th>RPM</th>
<th>Direction</th>
<th>Load (lbs)</th>
<th>Time (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polish</td>
<td>6 µm polycrystalline diamond paste on Buehler TexMet pad; Leco Microid non-aqueous lubricant</td>
<td>25</td>
<td>random</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>1 µm diamond polycrystalline paste on Buehler Microcloth; Leco Microid non-aqueous lubricant</td>
<td>25</td>
<td>random</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>Final polish: Vibratory polish with 0.05µm Buehler MaterMet 2 colloidal SiO2 on Buehler MaterTex pad</td>
<td>n/a</td>
<td>---</td>
<td>1</td>
</tr>
</tbody>
</table>

Vibratory polishing (Buehler VibroMet 2) was used to obtain a deformation free surface. The time required for vibratory polishing varied depending on the examination technique. Samples for light optical microscopy were polished for approximately one hour. Other characterization techniques that require surfaces free of deformation, such as EBSD, were polished up to 12 hours. Vibratory polishing times longer than 12 hours often led to differential polishing and surface relief.

4.6.2 Etching Techniques

4.6.2.1 General microstructure etchants

Electrolytic etching using oxalic acid was used as a general microstructure to delineate high angle grain boundaries in IN600. An applied current density 0.56 A/cm$^2$ was found to be most effective for revealing general microstructure. Long etching times
(>15 seconds) revealed additional information such as the presence of Σ 3 boundaries. Oxalic acid etching does not adequately reveal general microstructure for dissimilar material such as samples created by AFSP. Table 4.7 lists details for the electrolytic oxalic acid etchant. For some samples, a reaction product formed on the surface after etching. Light manual polishing with colloidal SiO₂ for 20-30 sec. after etching removed the reaction product and improved contrast between microstructural features.

Table 4.7. Parameters for electrolytic oxalic acid etch

<table>
<thead>
<tr>
<th>Cathode</th>
<th>Type 321 stainless steel foil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anode</td>
<td>sample</td>
</tr>
<tr>
<td>Electrolyte</td>
<td>10% Oxalic Acid</td>
</tr>
<tr>
<td>Etching Time</td>
<td>5-15 sec.</td>
</tr>
<tr>
<td>Applied Current (A/cm²)</td>
<td>0.56</td>
</tr>
</tbody>
</table>

Ni 201 samples were etched using a mixed acid etch consisting of one part concentrated nitric acid and one part glacial acetic acid. The sample surface was swabbed with an acid-soaked cotton ball until the surface became cloudy in appearance (10-15 sec.). Delineation of grain boundaries using this etch was best accomplished using light optical microscopy with differential interference contrast.

4.6.2.2 Selective Matrix Etchants

Examination of the strengthening phases in Ni-base superalloys, such as γ’, poses several challenges. Etching techniques that selectively etch away γ’ yield information useful for quantitative precipitate size measurements; however, limited morphological
information is gathered. Three-dimensional metallography via serial sections is effective in revealing morphology, but the technique is time-consuming and cost prohibitive. TEM is an alternative to SEM of serial sections; however sample preparation can be tedious and interpretation of precipitate morphology from TEM images can be difficult. Additionally it is difficult to gather information regarding the bulk distribution using TEM due to the localized nature of the technique. An alternative, γ-matrix etching procedure to study γ' distribution and morphology using high-resolution SEM was developed. Figure 4.11 shows an example of a selectively etched microstructure in IN706. The complex morphological phases developed during slow cooling are clearly visible in the micrograph.

Figure 4.11. HRSEM micrographs of selectively etched Nb-containing superalloy (IN706) cooled at 0.5°F/min. A.) low magnification showing morphology of needle-like δ phase, B.) etched sample reveals morphology variation of γ'' at high magnification

The selective matrix etch used in this work was based on a formulation provided by Haynes International. The technique and etchant formula were modified to improve resulting microstructural detail and repeatability. Table 4.8 lists the modified electrolyte
and parameters. It was determined that a reduction in applied voltage to 6 VDC resulted in longer etching times, however with more controlled dissolution of the $\gamma$ matrix. Also, additional amounts of CrO$_3$ were added to the solution until saturated. A CrO$_3$-saturated electrolyte improved the definition of phases revealed by the etching technique.

Table 4.8. Parameter for Cr(VI)-based electrolytic $\gamma$-matrix etch

<table>
<thead>
<tr>
<th>Cathode</th>
<th>Type 321 Stainless Foil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anode</td>
<td>sample</td>
</tr>
<tr>
<td>Electrolyte</td>
<td>conc. H$_2$SO$_4$ + conc. H$_3$PO$_4$ (2:15) + CrO$_3$ until saturated</td>
</tr>
<tr>
<td>Etching Time</td>
<td>30-90 sec.</td>
</tr>
<tr>
<td>Applied Voltage (VDC)</td>
<td>6</td>
</tr>
</tbody>
</table>

The Cr(VI) containing selective etch has been used successfully on a number of superalloy compositions including: Inconel Alloy 706, Inconel Alloy 718, Haynes Alloy 282, Haynes Alloy 740, René 104, Inconel Alloy 725, Mar-M247, and René 77. Haynes alloy 214, however, yields unsatisfactory results using this etch. An alternative electrolytic $\gamma$-matrix specific etch was used for Alloy 214 examined in this study. Table 4.9 lists electrolyte and etching parameters.
Table 4.9. Alternative (Cr(VI) – free) γ-matrix superalloy etch

<table>
<thead>
<tr>
<th>Cathode</th>
<th>Type 321 Stainless Foil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anode</td>
<td>sample</td>
</tr>
<tr>
<td>Electrolyte</td>
<td>20 mL CH₃OH + 100 mL glycerol + 75 mL H₃PO₄</td>
</tr>
<tr>
<td>Etching Time</td>
<td>60-120 sec.</td>
</tr>
<tr>
<td>Applied Voltage (VDC)</td>
<td>5</td>
</tr>
</tbody>
</table>

4.7 **Microhardness Testing**

Microhardness measurements were performed using both manual indenters and fully automated systems. All microhardness experiments were performed with regard to load ranges, indent spacing, and indent validity as outlined in ASTM E384. Manual microhardness testing was conducted on a Leco M-400-H1 hardness tester. A diamond pyramidal Vickers indenter was used in the load range 300-1000 g. Fully automated systems such as the Leco AMH43 were used to create a large array of microhardness indents so as to generate a microhardness map. Vickers diamond indenter was used to create arrays of indents using a 300 g load. Indent spacing was typically 200-250 μm.
CHAPTER 5: Ni FSP - PROCESS AND MICROSTRUCTURAL DEVELOPMENT

5.1 Development of Friction Stir Processing Window

Friction stir process parameters including rotation speed, travel speed, tool plunge depth, and tool geometry were varied to develop FSP process parameter windows for IN600 and Ni-201 substrates. Heat generation during FSP is dependent on process parameters, specifically the tool rotation rate and the tool travel speed. As with arc welding, heat input values serve as an important metric in the development of processing windows. For FSP/W, several equations exist that relate relevant process parameters to a heat input parameter. In this work a dimensionless heat input parameter developed by Roy et al. [101] was utilized with the following relationship for dimensionless heat input parameter, $Q^*$:

$$Q^* = \frac{f\sigma_{0.8}A\omega}{kU^2} \quad (2)$$

where $\sigma_{0.8}$ is the workpiece yield stress at 0.8·$T_m$, $A$ is the cross sectional area of the tool shoulder, $\omega$ is the tool rotational rate, $k$ is the workpiece thermal conductivity, and $U$ is the tool traverse speed. The coefficient $f$ in the above equation is a factor that represents the ratio in which heat generated at the shoulder/workpiece interface is transported between the tool and workpiece given by

$$f = \frac{\sqrt{(k\rho C_p)w}}{\sqrt{(k\rho C_p)T}} \quad (3)$$
where $\rho$ is the material density and $C_p$ is the heat capacity. Subscripts $W$ and $T$ denote workpiece and tool, respectively.

Comparing FSP conditions where the same tool and workpiece are used, simplifies to:

$$Q^* = \frac{\omega}{U^2} \quad (4)$$

The concept of a dimensionless heat input parameter has been used to relate processing parameters along with pertinent thermophysical material properties to the process temperatures during FSW. Roy et al. [101] found good correlation between the dimensionless heat input parameter (eq (2)) and the homologous process temperature for a range of workpiece materials including aluminum alloys, mild steel, and stainless steel. While not a ‘true’ equation for heat input, the value of the dimensionless heat input serves as a ranking criteria relating the relative effects of travel speed and rotation rate (with all other material parameters remaining constant) for the same material type. Other sources of heating such as that occurring from deformation are not accounted for in the above equations.

Figure 5.1 and Figure 5.2 show process parameter windows developed for IN600 and Ni-201, respectively. Factors not represented in the dimensionless heat input parameter that would also affect the heat input such as tool plunge depth, tool tilt angle, and tool cooling flow rate were held constant while varying spindle and traverse speed. It is important to note that the actual values for the dimensionless heat input parameters do not include the heat transfer and thermal properties of the base material. Therefore, values for dimensionless heat input for IN600 and Ni-201 are not directly comparable.
Figure 5.1. Process Parameter window for FSP IN600. Lines indicate constant tool travel velocity.

Figure 5.2. Process Parameter window for FSP Ni-201. Lines indicate constant tool travel velocity.
For IN600, a range of parameter combinations was created such that
dimensionless heat input was varied by approximately an order of magnitude. When
process parameters were selected that resulted in excessively high heat input (either by
too low travel speed or excessively high tool rotation rate), defects such as excessive
flash became readily apparent. Process temperatures for defect-containing runs with high
heat input parameters are expected to be higher than defect-free FSP runs. For both
IN600 and Ni-201, the material flow stress is expected to decrease as the FSP process
temperature increases. Excessive flash, which is generally regarded as a defect, arises
from low flow stress workpiece material not sufficiently constrained by the tool shoulder.
Such high-heat input process parameter combinations resulting in excessive flash defects
are indicated with red squares in Figure 5.1 and Figure 5.2. Figure 5.3 shows a
macrograph of FSP Ni-201 demonstrating excessive flash.

Figure 5.3. FSP Ni-201 demonstrating excessive flash formation on the top surface
processed at 225 RPM; 2 IPM.

The process parameter window developed for Ni-201 (Figure 5.2) used a
modified-geometry FSP tool machined from W-1wt%La\textsubscript{2}O\textsubscript{3} rather than W-25Re. The
tool geometry (and to some extent tool material) used for IN600 resulted in excessive
heat generation for Ni-201 FSP and caused excessive flash formation. Additionally the
relatively small shoulder feature of the IN600 tool relative to the pin reduced the shoulder
area constraining the workpiece material during FSP. The shoulder-to-pin ratio was changed lowered from 1.28 to 1.23 with the modified Ni201 tool. Figure 5.4 shows a schematic of the modified tool used to create ‘successful’ Ni-201 FSP runs. The overall outer diameter of the tool was also reduced from 0.90 to 0.75 in. The smaller shoulder diameter decreased the overall heat generation due to frictional heating [101]. For a constant travel speed and tool rotation rate, the heat generation by the shoulder is dependent on the tool radius by the power of three [102]. In addition to the decrease in frictional heat associated with the smaller tool shoulder profile, the amount of heat input to the workpiece was also decreased when W-1wt% La2O3 was used in lieu of W-25Re. The thermal conductivity of dispersoid-containing W-La2O3 [103, 104] is a factor of two higher compared to solute-containing W-Re [103] further decreased FSP heat input into the workpiece.

Figure 5.4. Geometry of modified W-1wt%La2O3 FSP tool used for Ni-201
Friction stir processing defects were also observed when the tool travel speed and rotation rate were such that heat input was excessively low. The most common defect occurring at low dimensionless heat input parameter combinations was a lack of material consolidation behind the tool. This defect results in large void areas exposed on the top plate surface. Figure 5.5 shows a macrograph of the lack of consolidation defect. When process parameter combinations are such that heat input is low, the process temperatures are sufficiently low that material flow beneath the rotating tool is inhibited. The higher flow stress at low temperature inhibits the filling of space produced behind the traversing pin feature of the tool. Such surface lack of consolidation defects formed can be avoided by increasing the FSP heat input (i.e., increasing rotation rate and/or decreasing traverse speed) or changing the tool geometry [52].

![Figure 5.5](image.png)

Figure 5.5. Lack of consolidation defect observed on the top surface of FSP IN600 processed at 175 RPM; 6 IPM. Arrows denote location of defects.

Non-surface breaking void/lack of consolidation defects can also arise within the SZ for certain FSP process parameters, especially when heat input is excessively low. Such voids are commonly referred to as wormhole defects. These defects were not observed with the same frequency as surface lack of consolidation defects. Figure 5.6 shows an example of a wormhole defect in FSP IN600. This wormhole defect formed on the advancing side of a FSP processed at 100 RPM/4.5 IPM with insufficient tool
shoulder engagement (i.e., insufficient tool shoulder plunge depth). The lack of tool shoulder engagement decreased the amount of frictional heating thus forming the void due to inhibition of sufficient material flow around the tool.

Figure 5.6. Two wormhole defects viewed in cross section formed on the advancing side of the FSP IN600 SZ processed at 100 RPM/4.5 IPM.

The mechanism by which these voids form is similar to that of surface-breaking lack of consolidation defects, therefore mitigation techniques are similar in nature. Additionally, tool pin features such as threads can increase the extent of material mixing near the tool and also help with wormhole defect formation. While the formation of wormhole defect is generally undesirable, the intentional formation of continuous voids within stir zone has been proposed as a novel technique to create continuous channels in plate material for heat exchanger devices [19].

5.2 Analysis of Friction Stir Process Forces

Analysis of process forces during FSP is valuable to the development of process parameters. Under ideal conditions, the FSP process enters a pseudo-steady state during
the traverse portion of FSP. It can be inferred that all material modified when operating in the pseudo-steady state regime has experienced similar thermomechanical excursions. Perturbations to the pseudo-steady state condition are detected as transients in process forces (e.g., normal force, spindle torque, tool path force). Such transients represent changes to the thermomechanical state beneath the tool and therefore affect the final microstructure. Therefore, analysis of process force data is valuable in determining the efficacy of a particular process parameter combination to create a relatively uniform degree of microstructural modification.

Figure 5.7 shows representative temporal process force output for an IN600 FSP run. Figure 5.8 is the corresponding photograph of the FSP’d IN600 plate. The force data captured during FSP represents characteristic force signatures for a 'successful' FSP run exhibiting pseudo-state state behavior. The surface of the friction stir processed region shows a characteristic lack of surface defects. While exhaustive evaluation of internal defects was not performed, multiple metallographic cross sections of FSP regions reveal no internal defects. During the plunge portion of the FSP run, the tool is driven into the workpiece material. As the tool approaches the target plunge depth, the tool normal force increases. The measured spindle torque also increases. For the run shown in Figure 5.7, a peak normal force of nearly 12 kip (53.4 kN) is achieved during the plunge portion of the process. Once the tool shoulder fully engages the workpiece, frictional heat generation increases significantly. The increase in frictional heating of the stationary tool during the plunge stage locally increases the temperature of the workpiece material and decreases the material flow stress. As the tool begins traversing (approximately 35 seconds into the process), the travel force (force resisting traverse of the tool) increases to approximately 2 kip. Simultaneously, the spindle torque increases to approximately 220 lb-ft. The process output remains relatively constant across the entire 4.5 in. traverse length. The lack of significant variations in process forces during the traverse portion is characteristic of pseudo-steady state conditions.
Figure 5.7. Typical process force output for defect-free FSP run of IN600 exhibiting pseudo-steady state behavior. Sample processed at 125 RPM/3 IPM

Figure 5.8. Photograph of surface defect-free FSP run on IN600 processed at 125 RPM/3 IPM

Process parameter combinations that did not exhibit pseudo-steady state conditions during the traverse portion of the run exhibit significant transients in process
force output. Figure 5.9 shows an example of process force output for IN600 processed at 125 RPM/4 IPM demonstrating highly dynamic process forces measured during the tool traverse—characteristic of non-pseudo-steady state conditions. For this particular FSP run, the tool plunge depth was insufficient which led to a lack of complete tool shoulder engagement with the workpiece. With the shoulder not fully engaged with the workpiece it can be envisioned that frictional heat generation is reduced leading to the formation of surface lack of consolidation defects as described previously. Defect formation during FSP is observable as transients in all measureable process forces corresponding to process instabilities. For regions of the plate containing defects, the spindle torque becomes highly oscillatory compared to values observed in pseudo-steady state conditions (Figure 5.7). Additionally, the normal force measured in the region of the traverse containing the defect increases by up to 66% due to the lack of heat generation by the non-fully engaged tool shoulder.

Figure 5.9. Process force output exhibiting highly transient process force behavior and subsequent defect formation during FSP
The process parameter combination affects the extent of heat generation during FSP and thereby the process forces measured. Figure 5.10 demonstrates the decrease in process forces observed as process parameters are altered such that FSP heat input increases. Comparing both extremes in heat input, reducing the dimensionless heat input parameter from 38 to 8 resulted in a nearly six fold increase in tool normal force. The reduction in spindle torque from extreme low heat input (8) to extreme high heat (38) was approximately 30%. The large observed variation in torque values relative to the mean value is typical in FSP/W and is attributed to the tribological interaction between the tool and the workpiece in which oscillating stick/slip conditions are manifested as ‘noisy’ spindle torque data.

Figure 5.10. Extracted IN600 pseudo-steady state traverse process forces as a function of FSP parameter. Error bars denote maximum and minimum force values measured.
The process forces, especially normal force, are quite high relative to lower strength alloy systems. The measured process forces for Ni-201, especially normal force, are markedly lower compared to those experienced during FSP of IN600. Figure 5.11 shows relevant process forces measured for FSP Ni-201 with varied parameters. Large scatter in data is present for spindle torque and travel force compared to measurements performed on IN600. Such torque measurements on Ni-201 were taken from load cells that were eventually replaced before IN600 FSP was performed.

![Figure 5.11. Extracted Ni-201 pseudo-steady state traverse process forces as a function of FSP parameter. Error bars denote maximum and minimum force values measured.](image)

Although the tool shoulder diameter is smaller than the tool used for IN600 (0.90 in vs. 1.125 in.), process forces are expected to be lower for Ni-201 compared to IN600. Since Ni-201 is a relatively pure (>99.5% Ni) alloy, strength is expected to be low due to relatively small degree of solid solution strengthening. Strength and elongation for Ni-
201 as a function of temperature is demonstrated in Figure 5.12 to temperatures up to approximately 600ºC. For temperatures higher than 600ºC, such as temperatures experienced beneath the tool during FSP, little experimental data exists—likely because Ni-201 is not suitable as a structural material for temperatures exceeding 670ºC [22]. For comparison, elevated temperature mechanical properties for IN600 are found in Figure 5.13.

![Elevated temperature mechanical properties of Ni-201](image)

Figure 5.12. Elevated temperature mechanical properties of Ni-201 (data provided by Special Metals [43])
The yield strength of Ni-201 is relatively temperature independent up to 600ºC. Inconel Alloy 600 has significantly higher yield strength relative to Ni-201 across all comparable temperatures. The yield stress for IN600 at 900ºC roughly equals Ni-201 at 600ºC. Such behavior is attributed to the higher alloy content of IN600 (27 wt% alloying addition). Therefore, differences in process forces for Ni-201 and IN600 are attributed the rather large difference in mechanical behavior of base materials.

5.3 \textit{FSP Thermal Acquisition}

Thermocouple-instrumented friction stir processing runs were performed on IN600 and Ni-201. Type K thermocouples were positioned on the backside of the plate such that temperatures could be measured on the advancing, retreating, and centerline of the stir zone (see section 4.2.1). Thermal measurements were performed on FSP IN600 for two different FSP parameter sets. Measurements were performed on the extreme ends of the IN600 processing window. The lowest heat input parameter combination (100
RPM/4.5 IPM) in the processing window (see Figure 5.1) was selected along with the highest heat input parameter set (150 RPM/2 IPM). Figure 5.14 and Figure 5.15 show the thermal output for the high and low heat input parameter sets, respectively.

Figure 5.14. Acquired thermal profiles for FSP IN600 processed using low heat input process parameters: 100 RPM/4.5 IPM.
The peak temperature measured for the high heat input parameter set was 1093 °C on the retreating side of the SZ whereas the peak temperature for the lower heat input sample was 895 °C at the center of the stir zone. The variation in temperature values, especially near peak temperatures, results from mechanical interaction between plasticized SZ material and the thermocouple junction. The faster tool traverse speed combined with lower heat generation from the lower rotation rate accounts for the approximately 200 °C decrease in peak temperature. It is interesting that highest peak temperatures were not measured on the AS—generally regarded as the region with the highest peak temperature due to the additive nature of the tool rotation and travel vectors [4, 6, 79]. Such measurements are comparable to the very limited peak temperature data for IN600 that exists in the technical literature. Findings by Song et al. [30], indicate peak temperatures for FSW IN600 vary between 800 and 900°C, depending on process parameters. Differences in the tool geometry and workpiece thickness (2 mm sheet vs.
6.3 mm plate used in this study) resulted in heating and cooling rates that were up to two times greater than observed in this study. However, the Song study does not specify the location in the SZ where the temperature measurements were taken.

Measured peak temperatures may also be lower than actual SZ temperatures and/or not agree with expected trends due to problematic extrusion of thermocouples out of the machined holes by the passing FSP tool. Although a backing plate was used to support the thermocouple sheaths during FSP, the high processes forces during processing led to deformation of material into the thermocouples thus pushing thermocouples slightly out of the region of interest. Figure 5.16 demonstrates the thermocouple junction pushed away from the FSP nugget region from material extruded into the machined hole. Slight movements of the thermocouple junction affected the measured peak temperature. In addition to extrusion of the thermocouple junction away from the process zone, the absolute accuracy of temperature measurement during FSP is affected by other factors which can include electrical noise, direct interaction (i.e., tool rubbing) of the tool with the thermocouple junction, and thermal lag associated with the size of the system [106].

Figure 5.16. ANSI Type K thermocouple junction slightly extruded away from SZ region
Thermal measurements were also performed on Ni-201 processed at 190 RPM/4.5 IPM using the same technique performed on IN600. Figure 5.17 shows the temperature of various regions of the SZ as a function of temperature. A peak temperature of 853ºC was measured in the center of the stir zone.

In Figure 5.17, both the RS and center region measurements exhibit rapid oscillations in temperature near the peak temperature with the amplitude of oscillation approximately 10-15ºC. The AS, however did not exhibit similar behavior. Examination of cross sections of the thermocouple embedded plate, the thermal output behavior near the peak temperature appears to be dependent on the proximity of the thermocouple to turbulent SZ material flow. The thermal output from the AS did not exhibit any significant oscillations as with the center region and retreating side. Moreover, because the thermocouple was located approximately 1.5 mm from the SZ (determined from
metallographic analysis), the observed heating rate was nearly 50% lower than the center and AS. For comparison, the center SZ thermocouple was consumed by plasticized material in the SZ and accounts for the loss of thermal output after reaching a maximum value of 853°C.

Although experimental values measured likely represent conservative temperature estimates of peak temperature within the SZ, such measured values are reasonable when compared to peak temperatures measured during FSP of Alloy 625. Rule [107] observed slightly higher peak temperatures of 1150°C measured near the center of the SZ. Assuming identical FSP parameters, the higher elevated strength of Alloy 625 relative to IN600 is expected to yield higher process forces to create a defect free FSP run. Higher process forces, including normal force, result in increased heat generation by the tool. The relationship between the normal force (P) and power input into the workpiece is given by the following relationship [108]:

\[ q_o = \frac{4}{3} \pi^2 \mu \frac{P}{A} \omega R^3 \quad (5) \]

where \( q_o \) is the net power (W), \( \mu \) the friction coefficient, \( P \) is the normal force, \( A \) is the projected tool area, \( \omega \) is the tool rotational speed (RPM) and \( R \) is the tool radius (m). For similar FSP parameters/tool geometries, the measured normal force during FSP of Alloy 625, for example, is 50% higher [107] than for IN600 using similar FSP parameters. The higher process forces for Alloy 625 compared to IN600 likely accounts for an increase in power input into the workpiece and therefore higher measured peak temperature. For similar reasons, the peak SZ temperatures measured for Ni-201 compared to IN600 are lower due to the lower elevated temperature strength of the extremely low-alloy Ni-201 compared to the much more highly alloyed IN600. However, it should be noted that the power input, \( q_o \), is dependent on the coefficient of friction between the workpiece and tool. Values for the dynamic coefficient of friction between the tool and different work piece materials are not known explicitly. Such high-temperature tribological measurements do not exist. Additionally the temperature
dependence of the dynamic coefficient of friction makes it difficult to incorporate into models when the FSP SZ region near the tool experiences temperature gradients that are not well-characterized [101]. While the aforementioned analysis does not take into account deformation heating, similar trends to those predicted on the basis of frictional heating are likely expected.

Using the experimental setup in this work, absolute peak temperatures cannot be accurately measured near the top surface of the plate; however, thermocouple-instrumented FSP runs yielded useful information regarding heating rates and cooling rates experienced during FSP of Ni-alloys. This information is useful when understanding FSP microstructural evolution, especially with AFSP. Measured heating and cooling rates will be used to develop thermal profiles used for physical simulation (discussed further in this document). Table 5.1 lists heating and cooling rates observed for thermocouple instrumented IN600 and Ni-201 FSP runs. Compared to the high heat input FSP parameter combination for IN600, decreasing the tool rotation rate and tool traverse rate resulted in a nearly two fold decrease in both measured heating and cooling rates.

Table 5.1. Extracted process temperature data for FSP IN600 and Ni201.

<table>
<thead>
<tr>
<th>Location</th>
<th>Cooling Rate: 0.95→0.75T_{peak} (K/sec)</th>
<th>Heating Rate: 0.75→0.95T_{peak} (K/sec)</th>
<th>Peak Temperature (ºC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni-201: 190 RPM/4.5 IPM</td>
<td>AS: 14</td>
<td>59</td>
<td>604</td>
</tr>
<tr>
<td></td>
<td>RS: 50</td>
<td>110</td>
<td>791</td>
</tr>
<tr>
<td></td>
<td>Center: ----</td>
<td>111</td>
<td>853</td>
</tr>
<tr>
<td>IN600: 100 RPM/4.5 IPM</td>
<td>AS: 34</td>
<td>120</td>
<td>834</td>
</tr>
<tr>
<td></td>
<td>RS: 15</td>
<td>108</td>
<td>635</td>
</tr>
<tr>
<td></td>
<td>Center: 36</td>
<td>144</td>
<td>895</td>
</tr>
<tr>
<td>IN600: 150 RPM/2 IPM</td>
<td>AS: 7</td>
<td>65</td>
<td>922</td>
</tr>
<tr>
<td></td>
<td>RS: 19</td>
<td>68</td>
<td>1093</td>
</tr>
<tr>
<td></td>
<td>Center: 8</td>
<td>32</td>
<td>815</td>
</tr>
</tbody>
</table>
5.3.1 Simulations of FSP Peak Temperature and Distribution

The FSP/W process is very complex from a thermomechanical standpoint. Predictive analyses of thermal evolution are not straightforward. Nonetheless, some simplistic models have been developed. A commercially available FSW/P modeling software suite, FSWS (Applied Optimization, Dayton, OH, USA), has been developed based on a modified numerical model initially developed by Colegrove and Shercliff [106]. Thermal cycle and material flow predictions have been validated by some studies in the technical literature [67]. Acquired thermal data from FSP IN600 combined with known thermophysical values for IN600 were used to calibrate the heat generation component of the model to determine peak temperatures and temperature distribution around the FSP tool. This calibration of the heat generation using data from IN600 was also applied to other Ni-base alloy systems discussed further within this document—namely Mar-Mar247.

The approach used by Colegrove and Shercliff [106] and FSWS relies on a simplistic heat generation model of a rotating tool shoulder on a stationary work piece. This simplification allows FSWS to model heat generation using a relationship originally developed for rotary friction welding [102]. Under the assumption that sliding contact between the tool shoulder and workpiece occurs, the heat flux (q) generated by the FSP tool is given by:

\[
q = \frac{2\pi}{3} \mu P \omega R_s^3
\]

where \( \mu \) is the coefficient of friction, \( P \) is the normal pressure on the tool, \( \omega \) is the tool rotation speed, and \( R_s \) is the shoulder radius. Application of this equation makes several assumptions that may not necessarily represent actual phenomena during FSW/P. The first simplification this equation assumes is that heat generation is due to the shoulder alone. For thin workpieces, heating from the pin is negligible [102]. However, for thicker workpieces (such as those used in this study) or for tool geometries with small
shoulder-to-pin ratios (such as the geometry used in this work), heating from the pin is not negligible. The second large assumption in this equation is the value to use for the friction coefficient. In practice, this value is temperature dependent and depends on several factors including the local surface roughness (contact geometry), applied force, and temperature dependent flow stress behavior of the two surfaces [109]. Because determination of the actual value for $\mu$ is not straightforward, an effective friction coefficient is used instead which is determined by calibrating the heat flux with actual experimental data. Using experimental data for other variables in the above equation, the value for $\mu$ is adjusted within a physically meaningful range (typically values from 0.2 to 0.5) until the desired heat flux and consequently predicted temperature matches experimentally derived values.

5.3.1.1 Heat Flow Model in FSWS

*Calibration of Model with IN600 Data*

Simulations performed used geometrical parameters representative of the workpiece dimensions used for thermally instrumented FSP runs. Table 5.2 lists various thermophysical properties from manufacturer data sheets [105] and tool geometry parameters used by the FSWS software. The temperature dependence of thermophysical properties listed in the table is not taken into account by the model. The simulation software has temperature dependent flow stress data for only one Ni-alloy (Inconel Alloy 718); however, the heat generation model does not take temperature-dependent flow stress behavior into account.
Table 5.2. Inconel Alloy 600 thermophysical properties used in simulations performed in FSWS. Relevant FSP tool geometry parameters are also shown.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Conductivity @ 0.5Tm</td>
<td>20 W/M-K</td>
</tr>
<tr>
<td>Density</td>
<td>8.19 g/mm³</td>
</tr>
<tr>
<td>Specific Heat</td>
<td>435 J/kg·K</td>
</tr>
<tr>
<td>Friction Coefficient (µ)</td>
<td>0.425</td>
</tr>
<tr>
<td>Tool Shoulder radius</td>
<td>11.4 mm</td>
</tr>
<tr>
<td>Tool Pin Radius</td>
<td>5.4 mm</td>
</tr>
<tr>
<td>Tool Pin Root Radius</td>
<td>8.9 mm</td>
</tr>
</tbody>
</table>

The simplified heat generation model (eq (6)) only requires knowledge of the basic tool geometry dimensions and the plunge force to determine the power input into the workpiece, assuming a value for the friction coefficient is known. Steady state plunge force data acquired for high (150 RPM; 2 IPM) and low (100 RPM; 4.5 IPM) heat input IN600 FSP runs was used as an input to simulate the temperature distribution. Because the process forces and peak temperatures within the SZ were known a priori, the value for µ was adjusted iteratively until the predicted temperatures were in agreement with experimentally determined values. Iterative FSWS simulations were performed to arrive at a friction coefficient of µ=0.425 for IN600 that gave simulation output in good agreement with measured values for both heat input conditions. Although a difference in peak temperatures of approximately 200°C exists for the high and low heat input parameters, the value for µ was assumed to be independent of temperature thus the friction coefficient used was the same for both heat input parameter conditions.

Figure 5.18 shows the simulated steady state temperature distribution for the high and low heat input parameters at depths into the workpiece that correspond to the depth of the embedded thermocouple. The geometry of the tool is superposed on the thermal distribution. Additionally, the location of the embedded thermocouple in relation to the tool along with experimentally-derived peak temperature is also shown in Figure 5.18. The simulated temperature distribution within the process region is in good agreement
with the experimentally determined peak temperatures. It should be noted that while the heat generation model assumes frictional heating proportional to tool geometry (shoulder radius), the FSWS model assumes the temperature distribution from a point heat source and is essentially independent of tool geometry. The high temperature isotherms predicted by FSWS near the center of the pin feature likely is not representative of actual behavior. Despite this shortcoming of the model, FSWS was able to replicate the observed trend in experimentally determined temperature data as a function of processing parameters. Such a trend corresponds to higher heat input parameters combinations (achieved by increasing spindle speed and/or decreasing the tool traverse velocity) resulting in higher process temperatures. As a result, the ‘calibrated’ model inputs (i.e., friction coefficient, \( \mu = 0.425 \)) were used to determine applicability to high temperature AFSP runs where reliable thermal data was not able to be collected.
Figure 5.18. Predicted temperature contour plots for A.) high (150 RPM/2 IPM) and B.) low heat input (100 RPM/4.5 IPM) process parameter combinations. Schematic cross section shows the location of experimentally-derived maximum temperature.

Applicability of FSWS Temperature Distribution Model to High-\(\gamma'\) Fraction Ni-base Superalloys

As will be discussed in following sections within this document, thermal measurements for high-fraction \(\gamma'\) superalloys such as Mar-M47 were unsuccessful due to considerably higher process forces extruding thermocouples away from process region. The applicability of the ‘calibrated’ FSWS model inputs to other Ni-base systems was determined. Thermophysical data for Mar-M247 used for model input is shown in Table 5.3. The workpiece and tool dimensions used for AFSP Mar-M247 were identical to those used for IN600 simulations.
Table 5.3. Mar-M247 thermophysical properties used in simulations performed in FSWS.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Conductivity @ 0.5Tm</td>
<td>14 W/M-K</td>
</tr>
<tr>
<td>Density</td>
<td>8.53 g/mm²</td>
</tr>
<tr>
<td>Specific Heat</td>
<td>565 J/kg-K</td>
</tr>
<tr>
<td>Friction Coefficient (µ)</td>
<td>varied</td>
</tr>
</tbody>
</table>

Using the calibrated friction coefficient determined from FSP IN600 (µ=0.425), simulations for Mar-M247 were performed for the low heat input parameter combination (100 RPM; 4.5 IPM) using pseudo-steady state normal force data. Figure 5.19 shows the resulting temperature distribution for different µ values. The higher process normal force for AFSP Mar-M247 (125 kN for Mar-M vs. 45 kN for IN600 using similar process parameters) originating from the higher elevated temperature flow stress associated with the richer alloy composition produces significantly higher power input into the workpiece. Using the same friction coefficient used in IN600 simulations (µ=0.425), the predicted peak temperatures exceed temperatures of 3800ºC. Such temperatures are unreasonably high. As the friction coefficient used in the simulation was decreased further to µ=0.15, the predicted temperature distribution approached more reasonable peak temperature near 1400ºC. While a more reasonable prediction using µ=0.15 vs. µ=0.425, peak temperatures above 1400ºC exceed the equilibrium liquidus temperature of 1361ºC. Further decreasing the friction coefficient below µ=0.15 would result in output that is sub-liquidus. However, because experimental temperature measurements to compliment the simulation values do not exist, it is not known whether a friction coefficient value of µ=0.15 (or lower) has any physical significance. It can only be concluded that frictional conditions determined for lower strength Ni-alloys such as IN600 cannot be applied to materials with significantly higher elevated-temperature strength such as Mar-M247, despite both alloys sharing the same elemental base.
Figure 5.19. Predicted temperature contour plots for Mar-M247 processed using low heat input parameters (100 RPM/4.5 IPM) as a function of varied friction coefficients. A.) $\mu=0.15$, B.) $\mu=0.25$, and C.) $\mu=0.425$.

Limitations and Improvements

The physical relevance of the heat generation model used in FSWS and the strong dependence of proper selection of the sliding friction coefficient is questionable for the high-temperature FSP/AFSP runs performed in this work. High temperature tribological studies on sliding contact between tungsten alloys and Ni superalloys do not exist. However a study by Peterson et al. explored high temperature sliding friction behavior of Ni-Cr-Mo alloys against similar alloys [110]. The authors’ findings show that sliding friction coefficients vary between 0.6-0.8 and are relatively temperature independent until 500-600°C at which point the friction coefficients drop to values between 0.2-0.4 and
remains fairly independent to temperatures of approximately 800°C. Measurements were not performed at higher temperatures, representative of FSP. Regardless, it is unlikely that values taken from datasets generated from ball-on-flat type test techniques such as that of Peterson [110] actually represent the exact friction coefficient for a complex process like FSP. Determination of actual friction conditions during FSP for the use in analytical models proves a very daunting task.

In addition to correct selection of sliding friction coefficient, several shortcomings of the heat generation estimates and the heat condition analytical methods likely lead to erroneous predictions. As previously mentioned, the equation for power input into the workpiece described by eq (6) only takes into account the tool sliding past the workpiece. Hence, the power input equation only takes into account the coefficient of sliding friction. Experimentally, however, the tribological interaction between the tool and the workpiece is more complex. The case for pure sliding friction does not always apply and workpiece material can also stick to the tool. Contact between the tool and workpiece is combination of sticking and sliding (slipping) contact that operates in an oscillatory manner [102]. Because the workpiece material has the same velocity as the tool in the sticking condition, it is expected that heat generation with this boundary condition is likely lower than the sliding condition, thus reducing power input by the tool. In addition to leaving out a heat generation term for viscous dissipation by the deformed workpiece, the heat generation model utilized by FSWS does not account for heat generation by the tool pin.

Analysis of heat conduction is the FSWS software uses the Rosenthal solution which assumes a point source. While this assumption has utility in arc welding where the heat source (arc) can be approximated as a point source at the weld pool surface. However, the heat generated during FSP/W cannot be approximated as a point source since it generated at the tool interface as well as the region surrounding the tool. Additionally, other important boundary conditions are not taken into account using the FSWS model which include heat extraction by the water-cooled FSP tool as well the backing plate and fixturing. The resulting conduction of heat away from the workpiece is
not taken into account in FSWS when determining the temperature distribution around the tool.

For the determination of process temperatures near the tool for high-temperature materials, as is desired for AFSP of Ni-alloys, the use of embedded thermocouples in the tool near the surface may result in more reliable data. Such systems exist and have been reported in a particular study by Jasthi et al. [63]. While the authors describe the system consisting of embedded thermocouples with radio telemetry, they fail to show any temperature data for FSW of IN718 generated by the system.

5.4 Microstructural Characterization of FSP Ni-201 and IN600

5.4.1 Grain Size Distribution

A characteristic feature of FSP is grain refinement produced in the stir zone. The degree of gain refinement produced is dependent on the thermomechanical history of the SZ which depends largely on the FSP process parameters. Average SZ grain size as a function of process parameter combination was determined for FSP IN600. Because FSP parameters including RPM and tool traverse speed were often varied simultaneously, the grain size as a function of dimensionless heat input was determined (Figure 5.20). Values in Figure 5.20 represent average SZ grain size which is an average of grain size from the AS, RS, and central SZ regions near the top, middle and bottom of the SZ using linear intercept method of LOM micrographs as described previously.
As the FSP process parameters were varied such that dimensionless heat input is decreased, the average grain size also decreases. Average SZ grain sizes as small as 8 µm were observed for ‘cold’ FSP runs and as large as 27 µm for ‘hot’ FSP runs. Up to a 5-fold reduction in average grain size was observed for the lowest HI parameter combination (100 RPM/4.5 IPM) when compared to the as-received base material average grain size of 55 µm. The average SZ grain size for IN600 processed twice (double pass) is approximately the same as the single pass for the same parameter combination (100 RPM/4.5 IPM, dimensionless HI=4.9). This interesting result suggests that average SZ grain size is not a function of the starting grain size, but rather a function of only the thermomechanical conditions during processing.

Work by Song et al. [60] measured average SZ grain sizes of FSP IN600 of 3 to 5 µm depending on the process parameters. Significantly smaller grains were likely
achieved because the reduced heat input via considerably smaller shoulder diameter (0.6 in. vs. to 1.125 in.) and significantly shorter pin length (0.070 in. vs. 0.125 in.)

Thermal measurements which indicated increased cooling rates for lower heat input parameter combinations likely accounts for the increase in grain refinement as dimensionless heat input is decreased by lowering spindle RPM and/or traverse speed. After the FSP tool has traversed away from the region interest, faster cooling rates of low HI FSP runs reduces the time spent in temperature regimes that allow for coarsening of the dynamically recrystallized SZ material. For IN600, rapid coarsening can occur at temperatures above 980ºC because of the solutionization of M₇C₃ and M₂₃C₆ carbides which provide stability against coarsening due to grain boundary pinning [111].

Average grain sizes measured using intercept methods on LOM do not give a clear picture of the distribution of grain sizes. As with FSP of other material, the rather heterogeneous distribution of temperature and strain within the SZ can lead heterogeneous SZ regions with respect to grain size. Figure 5.21 shows an example of gradients in grain size distributed within a region near the central portion of the SZ. Automated analysis of grain size using EBSD was used rather than LOM intercept methods to determine the distribution of grain size for particular locations in the SZ. Figure 5.22 shows the number fraction of grains as a function of equivalent equiaxed grain diameter.
Figure 5.21. Typical microstructure found in central SZ for IN600 processed at 100 RPM/4.5 IPM (dimensionless heat input=4.94)

Figure 5.22. Central SZ grain size distribution for different parameter combinations measured using EBSD
The distribution of grains for all process parameter combinations is heavily weighted for small grains less than \( \sim 10 \mu m \). EBSD is able to unambiguously distinguish grain boundaries (boundaries >5º comprise grains for this analysis) unlike conventional metallographic techniques combined with LOM. The difference in measurement techniques likely accounts for the smaller average grain size measured using EBSD.

The grain size distribution for the lowest heat input parameter combination (100 RPM/4.5 IPM, dimensionless heat input =4.9) was nearly identical for single pass compared with double pass. Multipass FSP has been applied in low-\( T_m \) FSP of Al-alloys to determine the effect on superplastic behavior [112]. Multiple overlapping passes were created on AA7475 with a slight increase in grain size (\( \sim 3.2 \) vs. \( \sim 2.2 \mu m \)) after 6 overlapping passes. The slight increase in average grain size was attributed to dissolution of some grain boundary pinning particles, namely MgZn2 and Al13Cr2Mg3. Similarly, for IN600 a second overlapping pass appears to have little effect of the microstructural evolution with respect to final SZ grain size. The slight increase in average grain size measured via EBSD for double pass vs. single pass (5.2 vs.4.8 \( \mu m \)) may be due a change in distribution/fraction of Cr-carbides from the initial pass which slightly alters the coarsening behavior of the grains in the second pass as SZ material cools away from the tool. However, the distribution/fraction of Cr-carbides was not studied in detail in this study. Higher heat input process parameter combinations (150 RPM/2 IPM, dimensionless heat input=37.5) exhibits a similar distribution of grains size, i.e., heavily weighted for small grains (i.e., \(< 10 \mu m \)). However the distribution includes significantly larger grains with diameters as large as 60 \( \mu m \).

### 5.4.2 Microhardness Measurements

The SZ microhardness of FSP IN600 under various processing conditions was measured. Figure 5.23 shows the hardness distribution along traverses along the SZ midline. As the spindle speed and traverse speed combinations are changed such that heat input is increased, a subtle decrease in hardness is observed. Comparing extremes of
the processing window, average SZ microhardness decreases from $193 \pm 7$ HV to $174 \pm 5$ HV for the lowest (100 RPM; 4.5 IPM) and highest heat input (150 RPM; 1.8 IPM), respectively. Despite the nearly two-fold difference in average grain size (measured optically) for the different FSP parameter combinations, the overall SZ hardness is not dramatically different. Furthermore, the as-received IN600 base material with an average grain size of 55 $\mu$m exhibits microhardness values very similar to FSP SZ microhardness for the range of heat inputs evaluated. For the grain sizes obtained in this work, significant Hall-Petch strengthening as evident in microhardness measurements is not expected as the Hall-Petch effect is small for FCC material such as IN600. A similarly small effect of grain size on SZ microhardness was observed for IN600 by Song et al. [60]. The authors used much thinner workpiece materials (2 mm sheet) using smaller tool shoulder diameter (15 mm) at significantly higher traverse speeds (9.8 IPM). The expected reduction in heat generation and associated increase in post-processing cooling rates reduced coarsening and resulted in fine grains approximately three times smaller than those obtained in this study using low heat input parameters. However, despite the additional refinement, the measured microhardness in the Song et al [60] study was only approximately 15% higher.
Figure 5.23. Microhardness traverses performed on FSP IN600 for various process parameter combinations

With the exception of the highest heat input condition (150 RPM; 1.8 IPM), a very slight (10-15 HV) hardness increase is observed near the periphery of the SZ and into the TMAZ. The very slight increase in strengthening in the TMAZ/near-TMAZ region may be due to an increase in work hardening via incomplete recrystallization. This effect has been observed by Steuwer et al. [39] in AA7010 FSW.

5.4.3 Characterization of Stored Energy after FSP

It is generally regarded that the resulting microstructure after FSP is the product of dynamic recovery and recrystallization processes occurring at elevated temperatures while the material is severely plastically deformed under the tool. Consequently, changes to process parameters that affect the process temperature and imposed plastic deformation will have an effect on the extent of recovery and recrystallization. The extent of residual plastic deformation, i.e., stored energy, is essential to understand kinetic enhancement occurring during AFSP discussed later in this document.

Transmission electron microscopy (TEM) can provide information regarding residual plastic deformation based from observed dislocation structure/density, high-
order Kikuchi diffraction line intersection shifts, and Kikuchi line broadening [113]. However, the analyzed area for a typical TEM specimen is rather small compared to the overall SZ. For samples prepared using focused ion beam techniques, the sampled volume is even smaller. Other techniques that can be used include peak broadening analysis using X-ray diffraction (XRD); however, typical illumination areas for conventional diffractometers, often on the order of several square millimeters, is too large to illuminate only the regions of interest within the SZ.

Local variations in crystallographic misorientation can serve as good indicators of the degree of strain, i.e., stored energy in a material [113-118]. Electron backscatter diffraction can sample misorientation information from relatively large areas of a FSP sample with misorientation sensitivities of approximately 1º [115].

5.4.4 Electron Backscatter Diffraction for Residual Plastic Strain Analysis

The presence of dislocations from a non-uniform strain in a material distorts the crystalline lattice such that a spread in Bragg scattering angle occurs. A broadening in diffracted intensity results from the spread in Bragg scattering angle and affects the ‘sharpness’ of the Kikuchi diffraction pattern. A parameter known as image quality (IQ) is used to describe the quality of Kikuchi pattern. For the analyses performed in this work, the IQ parameter is a function of the number of detected Hough peaks [119]. However, using this parameter solely to quantify the local extent of plastic deformation is unreliable. While generally decreasing with increasing dislocation density, the measured IQ also depends highly on surface preparation, grain orientation, and structure factor [120]. Additionally, instrumental variations such as beam defocus, electron accelerating voltage, and electron beam current, and EBSD camera settings also affect the measured pattern IQ [115, #358]. An example map of IQ parameter is shown in Figure 5.24 for as-received IN600 plate material. Surface preparation for this sample is satisfactory as there are no visible metallographic artifacts such as scratches or subsurface deformation that would cause significant deviations of IQ values. Regions with lower pattern IQ appear
darker in the map whereas lighter regions denote areas of the scan where pattern IQ was high. Regions of high disorder, such as grain boundaries, produce diffuse Kikuchi diffraction patterns and appear dark in the IQ map. Despite the surface preparation condition for the sample in Figure 5.24 remaining constant across the scan area, some grains appear lighter than others depending on orientation. Shortcomings of IQ mapping are apparent with the observation of significant crystallographic rotations (>5°) present without a large change in the measured IQ. A misorientation line trace is shown in Figure 5.24 from grain I to II. Despite very similar IQ values (2800-3100), large intragranular misorientation (approximately 9°) is measured from the origin. Additionally, the 36° rotation of grain II relative to grain I does not have a corresponding change in the measured IQ. Such observations highlight the inability for IQ to be used reliably to map stored energy/residual strains for polycrystalline samples.
Figure 5.24. Generated IQ map and line trace demonstrating significant crystallographic rotations can be observed despite relatively constant values for IQ.

Techniques that characterize the local misorientation for a scan region are considerably more useful than IQ distribution analysis. Kernel average misorientation and grain reference orientation deviation are used within this work to observe the extent of residual plastic strain, i.e., stored energy in FSP Ni for several different process parameter combinations.

**Kernel Average Misorientation**

An EBSD scan consists of an array of scan points analogous to pixels in an image. A scan point, or kernel defined as ‘A’, is surrounded by neighboring kernels as shown in Figure 5.25A. The central kernel, A, is surrounded by six neighboring kernels denoted 1 through 6. Each neighboring kernel possesses a misorientation difference relative to the
central kernel A. Because of the possibility of the central kernel being adjacent to a neighboring kernel with a very high misorientation (such as next to another grain), a maximum misorientation value is applied for the analysis. In this work, a maximum misorientation value of 5° was used. Misorientations larger than 5° represent kernels such as those belonging to neighboring grains with a different orientation.

![Figure 5.25. Kernel average misorientation calculation using A.) 1st order and B.) 3rd order nearest neighbor technique.](image)

The kernel average misorientation (KAM), $\Delta_{gA}$, of the central kernel, A, is determined by averaging the measured misorientation angles ($\theta$), relative to A, of the six neighboring kernels as per the following equation [121]:

$$\Delta_{gA} = \frac{1}{6} \left( \theta_{A6} + \theta_{A1} + \theta_{A5} + \theta_{A2} + \theta_{A3} + \theta_{A4} \right) \quad (7)$$

The neighboring kernels shown in Figure 5.25A represents a first-order nearest neighbor KAM configuration. The perimeter of kernels surrounding the central kernel, A, can be adjusted in the analysis as to average a larger population of local misorientations by the following relationship [122]:

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\[ \Delta g_{AK} = \frac{\sum_{j=1}^{m} k_j}{m} \quad (8) \]

where

\[ k_j = \frac{\sum_{i=1}^{n} \theta_{ij}}{n} \quad (9) \]

Figure 5.25B shows KAM determined from higher order perimeter kernels. The analyses performed in this work incorporate third-order nearest neighbors, which averages 18 kernels about the central kernel. The length associated with this third-order parameter is approximately 2.9 times the step size used in the analysis [122]. For the parameters, this characteristic distance is approximately 1.5 µm.

The KAM itself is not a direct measure of the local residual plastic strain, but rather a measure of the degree of small lattice rotations present in a material. Such small lattice rotations originate from the introduction of dislocation arrays and subgrain boundaries in a material as a result of deformation [120]. The extent of misorientation and therefore lattice rotation is proportional to the strain present in the material [123]. Generated maps of KAM show regions in a microstructure containing concentrations of local misorientations. Within these regions it is expected that residual plastic strain (i.e., regions of high stored energy) is greater than those regions with very low KAM values.

As an example of the KAM technique, the plastically deformed region around a Vickers microhardness indentation was analyzed. Image quality and inverse pole figure map near the indent are shown in Figure 5.26. Plastic deformation occurs within the vicinity of the indenter tip. Slip lines present on the sample surface appear as dark parallel lines in the IQ map (Figure 5.26A). Pixels with low intensity (e.g., black or near-black) are visible within the entirety of the indenter tip as a result of the collection of poor quality Kikuchi diffraction patterns due to a combination of very high plastic strain as well as the breakdown of the required sample/detector geometry resulting from inclined surfaces of indentation facets. Generated KAM maps from the same region (Figure 5.27A-C) show point to point misorientation distribution near the indent.
Because the analysis is point-to-point, this technique is not sensitive gradual changes in misorientation across large grains. Instead, local misorientation indicated by the KAM distribution in regions such as near grain boundaries as well as nearby the indent are expected to be location of concentrations of geometrically necessary dislocations which cause the observed misorientation. Additionally, the configuration of high misorientation kernels (>~2°) for some highly strained grains is arranged in a cellular fashion within deformation subgrains. Kernel average misorientation values are higher (1-2°) within these higher stain regions relative to unstrained material (which has near-zero misorientation).

Figure 5.26. A.) IQ and B.) inverse pole figure maps generated for regions around Vickers indent placed on the surface of IN600.
The effect of nearest neighbors order used in the analysis is also demonstrated. The analyses shown in Figure 5.27A, B, and C are generated using first, second, and third kernel neighbors, respectively. The effect of nearest neighbor order is analogous to the ‘magnification’ or ‘strength’ of measured misorientation within the maps. In general, the absolute value for the point-to-point misorientation between two points (within the same grain) is larger when the two points are separated by a greater distance. The effect of kernel neighbor order follows a similar trend to scan step size [124] in which higher neighbor order results in less structural detail with higher overall measured misorientation. When comparing the KAM maps in Figure 5.27A (1st order) to Figure 5.27C (3rd order), fine details such as the deformation substructure is significantly more resolvable. Naturally, to avoid misleading trends in misorientation, direct comparison of KAM distributions or KAM maps for different processing conditions can only be made
using identical neighbor order, scan step size, and SEM/EBSD operating parameters. Interestingly, the generated KAM maps (Figure 5.27A-C) do not indicate a concentration of strain near the regions where slip lines were detected in the IQ map (Figure 5.26A). This observation highlights the inability for this technique to clearly reveal regions of pure slip which does not directly result in a reorientation of the grain. While pure slip does not result crystalline rotations and subsequent detectable misorientations, strain will also lead to the formation of geometrically necessary dislocations that will result in detectable misorientations. The density of geometrically necessary dislocations present in the analysis area results in the measured misorientation.

**Grain-Based Misorientation Analyses**

Kernel average misorientation is a local, point-to-point misorientation analysis technique whereas techniques such grain average misorientation (GAM) and grain reference orientation deviation (GROD) are intragranular misorientation analysis techniques. In the analysis of EBSD data, grains are defined by a closed boundary consisting of a misorientation exceeding some arbitrary value. For the analyses carried out in this work, a continuous boundary with misorientation exceeding 5° is considered a grain. Intragranular misorientation can be calculated by determining the misorientation between a central orientation and each neighboring pairs of scan points (kernels) [121]. The average misorientation value for all points that comprise a single grain are then plotted to create a map. This particular analysis technique represents the GAM method. Other intragranular analysis methods including GROD are especially useful for visualizing the intragranular distribution of misorientation. Rather than determining the average misorientation between neighboring points within the grain, the misorientation of each point within a grain is compared to a central reference point within the same grain. This central reference point is assumed to be the region of lowest misorientation, i.e., strain or stored energy. For the analyses performed in this study, the reference point (or kernel) is explicitly defined as the kernel with the lowest KAM value. Grain reference orientation deviation has been used in a number of applications to observe strain
distributions around fatigue and stress corrosion cracks in 304 stainless steel [123], and effects of strain rate and deformation temperature on the accumulation of strain for P/M René 88 [125]. Work by Dziaszyk et al. [116] has shown for low-C steel better qualitative agreement between applied strain and the GROD distribution compared to KAM or TEM analyses.

Grain reference orientation deviation maps were generated for the same region examined using the KAM analysis technique. Compared to point-to-point analyses, the intragranular GROD analysis shows no substructural detail as was observed for KAM maps with low nearest neighbor order (Figure 5.27A and B). Lattice rotations over large distances (such as within large grains), however, are much more clearly observable. This results in a clear observation of the deformation field around the Vickers indent. The distribution of intragranular misorientation is most highly concentrated within close proximity to the indent as would be expected. It can be inferred from this analysis that dislocation density is highest nearest to the indent within regions with the highest misorientation. However it is clear that the distribution of intragranular plastic strain is not homogenous, due to plastic anisotropy due to the polycrystalline nature of the sample. Similar concentrations of misorientation indicating local concentrations of strain near stress corrosion cracks have been observed in 304L stainless steel [123]. While the GROD method is not as sensitive as KAM analyses to the effects of scan step size, the GROD method relies on the prudent selection of the criteria used to define the constitution of a grain boundary [116]--unlike the KAM technique.
Limitations in Misorientation Analyses

Qualitatively speaking, highly misoriented regions contain local deformation and plastic strain. However, relating the misorientations caused by geometrically necessary dislocations to an engineering strain (defined as deformation per unit length) is not a robust, straightforward methodology despite several attempts. One notable study by Lehockey et al. [113] found a linear relationship between integrated angular misorientation density function (IMDφ) and nominal strain in IN600 measured during tensile testing. The IMDφ is a summation of the distribution of intragranular misorientation angle magnitude normalized to the number distribution of misorientations. Good linear correlation of IMDφ and known nominal strain was found (R²>0.94). However, the determination of values of strain directly from EBSD misorientation data cannot be made a priori without ‘calibrating’ misorientation results for the material of interest to known nominal strain values. Quantitative determination of local strain values within FSP process zones via misorientation data is not expected to be straightforward.
and is beyond the scope of this work. One major shortcoming is the lack knowledge in the friction stir field regarding the exact distribution and magnitude of resulting plastic strain within the process zone after FSP. Without known strain values to essentially ‘calibrate’ misorientation measurements, such data can only be qualitatively comparative.

*Misorientation Analyses of Friction Stir Processed Material*

As a thermomechanical process, mechanical energy is put into the workpiece material during FSP. A large fraction of this energy is dissipated as heat, however a fraction of that energy is expended as plastic deformation, during which, additional dislocations are introduced into the material. Many of the dislocations during deformation are eliminated during dynamic recovery and recrystallization during stirring; however, depending on processing parameters, such processes may not reach completion. As a result, networks of dislocations will result in localized regions within the process zone with low-angle misorientations. Additional plastic strain resulting in measurable misorientations can result from deformation behind the moving tool or within regions of the process zone such as the TMAZ. Material surrounding the SZ that comprises the TMAZ typically contains dislocation densities as high as $10^8$ x $10^{14}$ m$^{-2}$ (compared to base material $\rho = 4.5 \times 10^{14}$ m$^{-2}$) in AA6061-T6 [39]. Such dense dislocations will result in measurable local misorientation.

A misorientation map was generated using the KAM and GROD method for the FSP process zone of IN600 processed using low heat input (100 RPM; 4.5 IPM) parameters (Figure 5.29). Grain reference orientation deviation maps indicate the TMAZ contains large unrecrystallized grains with large ($\geq 10^o$) intragranular misorientation values. Some of the misorientation values exceeding $10^o$ appear white in the generated maps. Compared to the TMAZ, the SZ and the HAZ contain grains with significantly lower GROD misorientation values ($\leq$ approximately 2$^o$). However within the SZ and HAZ, some regions are observed with locally high (indicated by yellow and red in the map) misorientation. As mentioned previously the formation of such regions can be
attributed to post-stirring deformation and/or incomplete recrystallization within such regions. However, the elongated, highly misoriented grains in the HAZ may also originate from the base metal microstructure which contains some unrecrystallized regions from residual warm work.

Figure 5.29. A.) GROD and B.) KAM maps for edge of process zone in FSP IN600 processed using low heat input process parameter combinations (100 RPM/4.5 IPM)

The generated KAM map (Figure 5.29B) shows the distribution of point-to-point misorientation. However, using this method, the interpretation of the map is slightly more ambiguous with respect to delineation between the SZ and TMAZ. Overall, the HAZ is shown to have a large fraction of scan point with near-zero misorientation values. Within this region, misorientation is observed near grain boundaries and within the substructure of the few unrecrystallized grains. In the TMAZ, a large concentration of highly misorientated scan points (>2°) are observed primarily concentrated within the
dense deformation substructure of the TMAZ. These misoriented points originate from high densities of geometrically necessary dislocations resulting from residual plastic deformation. Interestingly, the fine-grain SZ adjacent to the TMAZ contains a large fraction of scan points with KAM misorientation values >1.5º--significantly more so than the HAZ. High energy synchrotron X-ray diffraction measurements of the AA7010 FSW indicates that dislocation densities, manifested as diffraction peak broadening, were highest in the TMAZ and the SZ region adjacent to the TMAZ [39]. The authors [39] proposed that the SZ periphery and TMAZ contain some stored energy that is not decreased via recovery/recrystallization upon cooling from the process temperatures. Although the amount of stored mechanical energy for IN600 is expected to be different due to processing conditions differences, inherent differences such as the stacking fault energy difference for AA7010 compared to IN600 are expected to influence extent of energy storage. Nonetheless, the slight hardness increase observed for lower heat input parameter runs combined with the results of KAM and GROD misorientation analyses suggests that residual stored energy is present and distributed heterogeneously throughout the FSP process region.

5.4.5 Friction Stir Processing Parameter Effects on Misorientation Distribution

Friction stir processing parameters affect the resulting microstructure by altering the thermomechanical history. Utilizing misorientation analyses described previously, the effect of FSP processing parameters on the distribution of measured angular misorientations was explored. To eliminate any instrumental effects, the identical EBSD pattern acquisition parameters were used. Generated KAM maps from the central SZ region are shown in Figure 5.30 for autogenous FSP on IN600 processed using high (150 RPM; 2 IPM), low (100 RPM; 4.5 IPM), and low heat input-double pass (100 RPM; 4.5 IPM) parameters. Additionally, the as-received base material was also examined for comparison.
The KAM maps reveal a higher density of higher misoriented scan points (>1.5-2°) for low heat input FSP samples relative to the high heat input condition and base material conditions. This suggests that despite recrystallization occurring during processing, the process may not be complete and/or some other residual plastic deformation is present associated with parameters resulting in lower peak temperatures and more rapid cooling rates after processing. Examination of the KAM map for the double pass sample also reveals no significant difference from the single pass sample processed using the same conditions. Distributions of KAM as a function of misorientation angle demonstrate more clearly the differences between processing parameters. Figure 5.31 shows the distribution of misorientation for the different conditions. For all conditions, the misorientation shows log normal type distribution. As
the FSP heat input is decreased, the average KAM for the scan area increases to larger values relative to the as-received base material.

![Figure 5.31. KAM distribution for various FSP parameters from IN600](image)

Interestingly, the average KAM for the highest heat input parameter FSP run (150 RPM/2 IPM) has an average KAM value of 0.36° compared to the as-received base material of 0.45°. This result suggests that the SZ dislocation density is lower compared to the base material by means of less residual plastic strain. Likely the high peak temperatures, combined with comparatively slower cooling rates of the high heat input parameters leads to more complete dislocation annihilation via recovery and recrystallization. The high heat input FSP sample has a distribution and average KAM similar to that of the as-received IN600 plate after an annealing heat treatment at 1000°C for 1 hour followed by furnace cooling (annealed IN600 average KAM=0.36°). Via recovery and recrystallization, the annealing process eliminated sporadically distributed
unrecrystallized grains in the base material and reduced the average KAM of the scan area. Figure 5.32 shows a comparison of the as-received base material before and after annealing heat treatment. Within the as-received base material, the location of grains with significant intragranular misorientation caused by low angle misorientations from dislocation networks is clearly visible in the GROD map (Figure 5.32C and D). After annealing, both the KAM and GROD maps show very little misorientation due to the expectedly low dislocation density.

Intragranular misorientation analyses show similar trends observed with point-to-point analyses with regard to trends with FSP heat input. Generated maps are shown in Figure 5.33A-D. Overall, parameter changes made to reduce the process heat input
resulted in an increase in the proportion of grains with non-zero GROD values. Grains with near-zero GROD values represent recrystallized grains that did not experience post-recrystallization deformation. Interestingly, material processed using high heat input parameters (150 RPM/2 IPM) indicated the lowest average KAM value (approximately equal to annealed IN600 base material); however the corresponding GROD maps shows a significant fraction of grains containing kernels with intermediate GROD values (2-5°). While the high heat input sample indicates a number of grains with intermediate intragranular misorientation, high values (≥10°) were not observed within the examined region. Such high GROD values, however, were observed for the low heat input (single and double pass) as well as the as-received base material. All FSP SZ regions exhibited small clusters or islands of misoriented grains surrounded by strain-free SZ grains with near-zero GROD values. This heterogeneous distribution is most well-pronounced for the low heat input (100 RPM/4.5 IPM) sample. The observed heterogeneous GROD distributions are consistent with expected heterogeneous temperature and strain distribution that are characteristic within the FSP.
5.5 Summary

Friction stir processing trials were performed on IN600 and Ni201 to determine process parameter windows. For both materials, several defect-free processing runs were produced using a wide range of heat input values determined from the combination of spindle speed and traverse speed. Processing parameters with exceedingly high heat input resulted in excessive flash generation as the tool traversed across the workpiece. Alternatively, exceedingly low process heat input resulted in the formation of void regions either internally within the SZ or at the surface of the plate. The presence of tool wear debris was not observed for any parameter combination.
Analysis of process forces during FSP indicated that defect formation was usually evident as a lack of process force stability. The occurrence of defects within the SZ corresponded to transient responses visible from the machine output of spindle torque, normal force, and to a lesser extent the travel and tangential travel force. Pseudo-steady state process forces were determined to be characteristic of defect-free FSP-runs. Samples for microstructural examination were taken from these pseudo-steady state regions, if possible.

Additionally process forces were examined for both IN600 and Ni-201 as a function of processing parameters. It was determined that as dimensionless heat input increased, a precipitous drop in normal force was observed. A nearly 3 fold difference in normal force was observed between the extremes of the process window for IN600. The less temperature-dependent Ni201 only exhibited a decrease of 15% for the highest heat input parameter condition compared to the lowest.

Thermal measurements were performed on selected parameter combinations. Measured peak temperatures for IN600 ranged from 900-1100ºC, depending on the FSP processing parameters. Such peak temperatures are within the few reported temperature values in the technical literature. Measured heating and cooling rates indicated that both the heating and cooling rates for the processing window extremes for IN600 differ approximately by a factor of 2. Peak temperatures of approaching 850ºC were measured for Ni-201; however, the tool geometry used for FSP of Ni-201 is sufficiently different such that direct comparisons to IN600 are difficult. Temperature measurements performed on either material represent conservative estimates of SZ peak temperature due to the remote nature of the thermocouples relative to the tool shoulder.

Force and thermal data acquired was used to 'calibrate' heat generation and temperature field model predictions made with FSWS software. The 'calibrated' FSWS model was then applied to other FSP Ni-base systems that were difficult to perform temperature measurements (namely superalloy Mar-M247). The results of the simulation show that despite IN600 and Mar-M247 sharing similar elemental basis, the friction coefficient determined using IN600 was not applicable and resulted in erroneous
temperature field predictions due to shortcomings of the thermal pseudo-physical model contained within FSWS.

Microstructural characterization of FSP IN600 and Ni201 show a strong dependence the sample thermomechanical history, which depends greatly on the FSP processing parameters. The average SZ grain size for FSP IN600 varied between 8 µm and 27 µm (compared to base material d_{avg}=55 µm). Such grain size values represent processing window extremes. A direct relationship was determined to exist between the process dimensionless heat input parameter and the average SZ grain size. Microhardness measurements of the IN600 SZ reveal that despite the reduction in grain size provided by FSP, the microhardness was relatively unchanged relative to the base material, which was shown to have some residual warm work.

Electron backscatter diffraction misorientation measurements were performed to assess the effect of processing parameters on the extent of recovery and recrystallization. Both kernel average misorientation (KAM) and grain reference orientation deviation (GROD) were used for misorientation analyses. Differences in thermomechanical histories from varied FSP process parameters were shown to have an effect on misorientation distribution. Kernel average misorientation distributions were shifted towards higher angles for lower heat input processing parameters. The highest heat input parameter condition had average KAM values that were equivalent to annealed IN600 base material, suggesting that recrystallization and recovery of any post-stirring deformation occurring during the high heat input thermal cycle is quite complete for that particular parameter condition. GROD maps show that highly misoriented grains exists in cluster configurations surrounded by strain-free grains with very low (near-zero) misorientation. Such results support the hypothesis of heterogeneous SZ temperature/strain conditions.
CHAPTER 6: HIGH TEMPERATURE ADDITIVE FRICTION STIR PROCESSING

6.1 Ti-6Al-4V Powder Additions to Ni-201

Additive friction stir processing techniques for high $T_m$ alloys were explored by incorporating Ti-alloy powder additions to Ni-201. Sufficient additions of Ti to Ni promote the formation of eta ($\eta$) phase. Eta phase is a HCP D0$_{24}$ phase of the stoichiometry Ni$_3$Ti. It can exist in cellular morphology or intergranularly in acicular platelets and has no solubility for other elements [126]. The formation kinetics of $\eta$ phase is dependent, in part, on the concentration of Ti [127]. Figure 6.1 shows TTT curves for the $\eta$ phase formation. Higher Ti concentrations can reduce the $\eta$ formation times by orders of magnitude. Although the precipitation of $\eta$ phase is generally regarded as deleterious to alloy mechanical properties such as ductility [128], Ti-additions to Alloy 201, the formation of intermetallic constituents such as $\eta$ during processing demonstrates feasibility of AFSP for high temperature $T_m$ materials.
6.1.1 Processing of AFSP Ni-Ti

Additive friction stir processing runs were performed on powder-filled Ni-201. A parameter combination of 190 RPM/4 IPM (lowest dimensionless heat input parameter of Ni-201 process window) was used. Figure 6.2 shows a photograph of the FSP plate clearly demonstrating process instabilities. Large amounts of flash are produced along the FSP retreating side with numerous lack of consolidation defects spaced along the FSP run. Upon closer inspection, Figure 6.2, a clear periodicity to the instabilities exists that appears to correspond to the spacing of the powder-filled machined holes. In addition to visual inspection of the resulting FSP region, analysis of FSP force output also suggests such process instabilities. Figure 6.3 shows a periodic variation in normal force, travel force and path force. Such process force oscillations were not observed for autogenous FSP of Ni-201. Additionally, FSP Ni-201 processed using identical parameters without Ti-alloy additions exhibits a final appearance without the presence of excess flash or lack of consolidation defects. The period of the measured process force oscillations (4.9 sec)
multiplied by the traverse speed (4 IPM) corresponds to the approximate spacing, 0.33 in., of consolidation defects observed on the top plate surface. Thus suggesting the consolidation defects observed on the surface correspond to the force instabilities. The spacing of the defects also corresponds the spacing of the power filled holes drilled into the workpiece.

Figure 6.2. Left: macrograph of resulting Alloy 201 region with Ti-powder addition. Right: a periodicity associated with the process instability is clearly visible.

Figure 6.3. Process force output for high heat input FSP run (190 RPM; 4 IPM) for A.) no Ti-powder addition and B.) with Ti-powder addition
Friction stir processing instabilities can be further explained upon careful examination of the Ni-Ti binary system. Figure 6.4 shows a binary phase diagram for the Ni-Ti system. From thermocouple-instrumented FSP runs, it can be assumed that process temperatures are excess of the measured peak temperature of 850ºC. In the Ni-Ti system, liquid is expected to form for typical FSP process temperatures, especially if the concentration of Ti is increased. A Ti-rich eutectic reaction occurs at 942ºC at 78 wt%Ti in the Ni-Ti system.

Figure 6.4. Calculated Ni-Ti phase diagram.
Although the measured peak SZ temperature is lower than the eutectic temperature, it should be expected that temperatures near the top of the SZ adjacent to the tool which cannot be easily measured likely exceed the measured temperature as well as the eutectic temperature. Exceeding the eutectic temperature at the top of the SZ near the tool leads to liquid film formation. The formation of liquid films during FSP disrupts heat generation during FSP via viscous dissipation. Although FSP/W is generally regarded as a solid state process, formation of liquid films leading to a ‘tool slip’ phenomenon observed in some Al- and Mg-base alloy systems [129-131]. In the case of the Ti-6Al-4V added to Alloy 201, process power consumed by viscous dissipation decreases the power input into the weld/process region. Eutectic films were not observed in metallographic cross sections and were likely extruded away form the process region as flash. The decrease in power into the workpiece decreases the temperature consequently increasing the workpiece flow stress. Increased workpiece flow stress cause higher normal force to sustain deformation beneath the tool. Eventually this increase in process force increases the power input into the process region/weld. Finally, as process forces increase, power input correspondingly increases. Once again, liquation temperatures are reached and liquid films redevelop. Unless the liquid films are extruded from the weld or the tool traverses past the region containing liquid films, the process repeats. Process instabilities measured by force output or observed visually occur via this repetition of the FSP/W self-regulation process.

In an attempt to reduce the AFSP heat input and bypass liquid film formation, additional runs were performed with higher tool traverse speed. Traverse speed was increased to 5 IPM, which represents a 36% decrease in dimensionless heat input compared to initial tool traverse speed of 4 IPM. Despite the reduction in process heat input, the resulting AFSP region still exhibited visible FSP defects on the surface that suggest that liquid film formation was persistent. Figure 6.5 shows the resulting AFSP run performed at 5 IPM.
6.1.2 Characterization of AFSP Ni-Ti regions

Much of the Ti addition was extruded from the stir zone into the flash (Figure 6.2). Immediately noticeable are large amounts of material missing from the stir zone as the tool pushed large amounts of the Alloy 201 including the Ti-alloy addition into the flash. Within the SZ, only a few regions near the top surface indicate incorporation of Ti-powder. Figure 6.6 shows a higher-magnification LOM micrograph of a region near the top SZ that demonstrates a limited degree of intermixing between the Ni-201 and the non-etching Ti-6Al-4V. A corresponding BSE micrograph of the same region shows the added phase remains largely as unmixed Ti-6Al-4V alloy (Figure 6.7). Near the interface between the unmixed Ti-alloy region and the Ni-201, regions containing cored structures of intermetallics with varied Z-contrast exist which indicates some reaction between the Ti-alloy addition and the Ni matrix occurred during AFSP. Upon closer examination of the reaction regions (Figure 6.7B), cored structures of Ti-Ni with different stoichiometries can be observed. Additionally the solute additions from the additive material appeared to have influenced recrystallization during FSP or grain boundary migration upon cooling. Grains nearby the band structure are submicron in size whereas the surrounding SZ grains with no Ti-alloying exhibit grains 2-20 µm in diameter.
Figure 6.6. Longitudinal section demonstrating very limited incorporation of Ti-6Al-4V into Alloy 201. Arrow denotes intermetallic formation from Ti-alloy/Ni reaction.
The selective $\gamma$-matrix etching method was used to observe fine precipitates, if present, at high magnification. Unfortunately, the HR-SEM imaging techniques typically used for higher-alloy content Ni-alloys were determined to be unsuitable for Alloy 201 due to its ferromagnetic behavior which interfered with operation of the semi-immersion lens and necessitated the use of the standard Everhard-Thornley detector. Figure 6.8 shows a SEM micrograph of an etched sample near the region between the Ti-alloy and
Ni-201. No dispersion of fine Ni$_3$Ti was observed after selectively etching the SZ using conventional non-HR SEM techniques.

Figure 6.8. Ti-6Al-4V/Alloy 201 FSP sample selectively etched to reveal precipitates. Morphology of large intermetallic phases revealed; however, no distribution of fine precipitates observed near Ti-rich regions

Because of resolution limits by not operating the SEM in non-HR mode fine $\eta$ phase, if present, may not be detected. Microhardness measurements were also performed to determine the extent of $\eta$ formation. Figure 6.9 shows a hardness traverse performed in the top SZ region across the SZ and into consolidated Ti-alloy addition. Hardness values for FSP Ni-201 away from the large intermetallics at the top of the SZ exhibited hardness values of 125 HV. Stir zone hardness increased abruptly within the region containing mostly Ti-6Al-4V. The high hardness values (350-400 HV) outside the stir zone represent the intrinsic hardness of the consolidated Ti-alloy. For Ti-alloy additions to Ni, inadequate material mixing combined with metallurgical reactions leading to FSP instabilities inhibited proper incorporation of the additive material.
6.2 Additive Friction Stir Processing of Alloy 282 on IN600

6.2.1 Process Development

Haynes Alloy 282 weld metal overlay consisting of four low-dilution passes was added to the surface of IN600 and subsequently friction stir processed to create an additive friction stir processed region on the plate surface (Figure 6.10). Dilution measurements using digital image analysis indicate dilution varied between 20 and 25% between the initial and final pass. Although a number of different parameter
combinations for FSP of IN600 were used to create a processing window, conditions on either end of the processing window were chosen. This resulted in creating AFSP material using a low heat input parameter (100 RPM/ 4.5 IPM) and a high heat input parameter combinations (150 RPM/2 IPM). Selection of either extreme of the processing window allows observation of all possible microstructures formed for the parameter set previously developed. It can be assumed that microstructural development for other parameter combinations within the processing window will exhibit characteristics of both extremes.

Figure 6.10. Cross section of Haynes Alloy 282 cold-wire GTAW weld overlay

Figure 6.11 and Figure 6.12 show photographs of the surface appearance of IN600 containing Alloy 282 AFSP regions on the plate surface for both parameter combinations. For the parameter combinations explored, no visible FSP defects were present on the FSP surface created directly on the weld overlay. Material flow around the FSP tool was sufficient to enable full consolidation behind the moving tool. Some flash was created along both the AS and RS as the tool plunge depth was slightly increased (+0.005 in. vs. conventional FSP) to ensure full engagement of the tool shoulder. However, the amount of flash was not excessive as to indicate excessive heat input. Average pseudo-steady state tool normal force measured during the traverse portion of AFSP was 12.1 and 18.3 kip (53.8 and 81.4 kN) for the high and low heat input
parameter combination, respectively. It should also be noted that the tool traverse force was consistently 50% greater for AFSP Alloy 282/IN600 compared to autogenous FSP IN600 for comparable process parameters. Likely the slightly increased plunge depth (+0.005 in) combined with the incorporation of a second, more highly alloyed material accounts for the increase in process forces observed.

Figure 6.11. Photograph of AFSP Alloy 282 on IN600 with high heat input parameter combination (150 RPM/ 2 IPM)

Figure 6.12. Photograph of AFSP Alloy 282 on IN600 with low heat input parameter combination (100 RPM/ 4.5 IPM)
6.2.2 Characterization of Additive Friction Stir Processed Alloy 282 on IN600

6.2.2.1 Distribution of Additive Phase

Transverse cross sections of AFSP regions reveal the distribution of the additive Alloy 282 within the stir zone. Light optical micrographs of cross sections etched with 10% oxalic acid are shown in Figure 6.13 and Figure 6.14. Areal measurements of digital micrographs revealed 63% and 72% of the added Alloy 282 was incorporated into the SZ for the high and low heat input FSP runs, respectively. Although the amount of Alloy 282 incorporated in the SZ for both heat input conditions is comparable, the distribution of the Alloy 282 within the stir zone is markedly different due to the different in material flow characteristics for the two parameter sets. The high heat input parameter combination (Figure 6.13) resulted in a higher degree of overall mixing, especially on the AS of the SZ. Higher strain rates combined with locally higher temperatures experienced on the AS of the tool [3, 4] is one possible reason for the increased mixing. From approximately the center of the SZ moving towards the retreating side, the extent of intermixing with the IN600 substrate appears to be lower. The Alloy 282 appears to exist in the SZ in a large contiguous region on the RS. The lack of tortuosity along the Alloy 282/IN600 boundary on the RS compared to the AS suggests the degree of turbulence and material flow during processing is relatively low compared to the advancing side. The accumulation of Alloy 282 on the RS is considerably more pronounced for the high heat input condition (Figure 6.13). From the center SZ to the outer extent of the RS, the additive Alloy 282 exists in a large contiguous region with a relatively linear boundary between the IN600. However, the Alloy 282 distribution on the AS is characterized by a highly serrated boundary between the additive material and IN600. Additionally, the fraction of additive material on the AS is considerably lower compared to the RS which suggests that additive material picked up by the front of the tool is deposited behind the tool on the RS.
Compared to the IN600 substrate, Alloy 282 contains alloying elements such as cobalt (10 wt% nominal) and molybdenum (8 wt% nominal) which are not present in IN600. Additionally, IN600 contains appreciable amounts of Fe (9 wt% nominal). Such differences in alloying species facilitates observation of material mixing using analytical electron microscopy techniques such as energy dispersive spectroscopy (EDS). The highly turbulent flow of Alloy 282 on the AS of SZ region processed using high-heat input parameters was examined more closely. Quantitative line scans of the turbulent regions indicate that intermixing sufficient to cause dilution of the Alloy 282 was not observed. Figure 6.15 shows the composition along a trace in SZ AS. Regions of lighter
grey values in the BSE micrograph are the result of Z-contrast differences between the IN600 substrate and more highly alloyed Alloy 282. Transitions from IN600 to Alloy 282 along the trace result in very abrupt changes in the quantified concentrations of Co and Mo, which are not present in IN600.

Figure 6.15. EDS composition profile on AS of AFSP region.
Material flow from the AS towards the RS is indicative of straight-through flow proposed in the Nunes kinematic model [18]. However, the combined effect of straight-through flow with the maelstrom flow proposed in the Nunes model (i.e., helical flow of material around the tool pin along the thickness direction of the plate) was not observed. Likely, the lack of pin features such as threads accounts for the lack of upward (or downward) flow of additive material within the SZ. Such threadless tool pin configurations are typical for high \( T_m \) FSP/W. A FSW study by Norton [17] using embedded stainless steel wire in Armco iron plate showed evidence of straight-through flow with very little flow along the plate thickness direction (maelstrom flow). In the Norton study, tracer material that encountered the leading edge of the moving tool was transported via the rotating tool to the backside towards the RS. Very similar behavior was observed in AFSP of Alloy 282, especially using high heat input parameters.

Although both AFSP conditions do not exhibit a large degree maelstrom flow, some downward flow of Alloy 282 was observed in etched cross sections. Macrographs of the AFSP SZ regions (Figure 6.13 and Figure 6.14) show the top most layer approximately 200-250 \( \mu \text{m} \) in the SZ is comprised of light-etching IN600 rather than the additive Alloy 282, despite the addition of Alloy 282 to the topmost surface in the initial condition before FSP. Likely, the formation of a layer of substrate material on the top surface is an artifact of the particular tool geometry used.

**6.2.2.2 Microhardness of Alloy 282 Additive Friction Stir Regions**

The rather chaotic distribution of additive material combined with the small volume of additive material makes mechanical evaluation of AFSP SZs quite difficult using standard tensile and even sub-standard tensile specimens. An understanding of the distribution of mechanical properties within the AFSP SZ region was gathered via microhardness testing. Microhardness measurements are especially useful because for many metals systems including Ni-alloys, there exists a reasonably accurate correlation between measured hardness and the tensile strength of a material [132]. Microhardness
maps were generated for the portion of AFSP SZ containing Alloy 282. Resultant maps of the AFSP SZ indicate an increase in hardness within the SZ for regions that are predominantly Alloy 282. Figure 6.16 shows the microhardness distribution in the AFSP SZ regions for both parameter combinations. AFSP Alloy 282-containing regions of the microstructure exhibit hardness values of 275 HV or greater depending on location and process parameters. For comparison, the as-deposited hardness of the original Alloy 282 weld overlay layer is 205 ± 13 HV, comparable to the hardness of FSP Alloy 600 (180-200 HV) and Alloy 600 base material (186 ± 6 HV).

Figure 6.16. Microhardness maps of AFSP regions for A.) high heat input and B.) low heat input parameter combinations.

For both parameters, the more turbulent AS exhibits slightly higher hardness compared to the RS. Interestingly, the sample processed using low-heat input parameters (Figure 6.16B), contains the highest hardness regions despite lower process peak temperatures and higher cooling rates behind the tool. Some regions within the SZ of the low heat input sample exceed 300 HV. FSP measurements from thermocouples...
embedded in IN600 adjacent to the SZ indicate temperature for material processed using the low heat input parameters reached a peak of 895ºC; approximately 200ºC lower than the high heat input condition. Additionally measured cooling rates of the low heat input sample were nearly double (36 ºC/s) compared to the high heat input sample (19 ºC/s). While these measurements were performed during autogenous FSP, the very similar process forces (within 10%) of AFSP Alloy 282/IN600 to FSP IN600 suggests that the thermal history should also be similar.

6.2.2.3 Microstructure of Alloy 282 Additive Friction Stir Regions

High-resolution SEM of selectively etched AFSP samples reveals a fine recrystallized microstructure with no evidence of the solidification structure present in the original weld overlay (Figure 6.17). Also present within the Alloy 282-containing SZ microstructure is a lamellar distribution of fine precipitates that correspond to hard regions located in the hardness map. Additive FSP material processed using the low heat input process parameters contained a larger fraction of fine SZ precipitates compared to the high heat input condition. Upon closer examination (Figure 6.18), these lamellar regions contain clusters of fine precipitates ranging in size from 15 to 125 nm depending on the location in the SZ. The majority of the particles have rounded or slightly cuboidal morphology typical of γ’ formed during relatively fast cooling [133]. A number of the precipitate-containing regions also have grains as small as 1 µm or less (Figure 6.18). The formation of extremely fine grains within these regions is likely due to a modification of the grain coarsening processes via Zener pinning by precipitates or solute drag from alloying additions in Alloy 282 [47]. A similar refining phenomenon has been observed during FSW Al-Mg-Sc alloys where thermally stable Al3Sc provides grain boundary pinning [134].
Figure 6.17 Electron micrographs of selectively-etched AFSP SZ regions processed using A.) high heat input and B.) low heat input. C.) as-deposited Alloy-282.

Figure 6.18 Higher magnification of a precipitate-containing region in AFSP sample using low heat input parameters.

*Physical Simulation of Friction Stir Thermal Cycles for AFSP Superalloy Layers*

Samples of as-deposited material were thermally cycled using representative thermal histories gathered during FSP to determine the evolution of the as-deposited microstructure in the absence of deformation. A Gleeble thermomechanical simulator was used to control heating and cooling rates such that the measured FSP thermal cycles
could be replicated. The experimentally determined thermal cycles for both the high and low heat input FSP parameter combinations were successfully replicated as shown in Figure 6.19.

Figure 6.19. Comparison of thermal cycles produced by the Gleeble thermomechanical simulator compared to acquired thermal histories during FSP (black dotted lines)

The resulting microstructure of the thermally simulated as-deposited AFSP is shown in Figure 6.20. Because no deformation occurred during thermal simulation, the solidification microstructure characteristic of the as-deposited weld overlay persists. Large (> 100 µm) solidification grains are observed for both high and low heat input thermal cycles. However unlike the as-deposited, non-cycled Alloy 282, both simulation conditions exhibit the presence of bright clusters of particles present within interdendritic regions when selectively etched.
Closer inspection of the interdendritic particle clusters for the low heat input cycle sample (Figure 6.20B) indicates regions containing fine (approximately 10-20 nm) spherical particles. These fine particles are characteristic of secondary $\gamma'$ which forms despite the rapid thermal excursion of the low heat input thermal cycle. The formation of pockets of fine $\gamma'$ is the result of Ti-enrichment of the interdendritic regions during solidification. The same phenomenon, described later in this document, is also responsible for the locally heterogeneous age hardening response of the as-deposited Alloy 282.

Low magnification micrographs for the high heat input thermal cycle also show clusters of particles within dendritic interstices (Figure 6.20C). However closer
examination of particles within these clusters (Figure 6.20D) reveals a morphology that is unlike the spherical secondary γ’ found in the low heat input. The particles present are a mixture of irregularly shaped, globular particles 100-500 nm in size along with some blocky submicron particles. Such morphology is not characteristic of secondary γ’ formed upon cooling.

The measured FSP thermal profiles used to create the thermal simulation samples differ in cooling rate as described in previous section of this document (section 5.3). In addition, thermal cycles differ in peak temperature relative to the γ’ solvus temperature. For Alloy 282 an equilibrium solvus temperature of 1006°C is predicted using JMatPro, which is in good agreement with experimentally derived values of 997 °C [135]. As a result, the peak temperatures for the high (1093°C) and low (895°C) heat input cycles represents cycles that are super- and subsolvus, respectively for γ’.

In the as-deposited condition titanium partitions strongly to the liquid during solidification. Despite the resulting local interdendritic enrichment of titanium, the rapid cooling associated with the GTAW weld overlay process suppresses the formation of γ’, leaving titanium in solution. During the low heat input FSP thermal cycle, the peak temperature remains below the γ’ solvus and cools through the γ’ precipitation temperature range. Although the simulated cooling rate is rapid (~40 °C/s from 800 to 600°C), fine secondary γ’ still forms within the Ti-rich regions.

The high heat input sample, however, differs from the low heat input cycle in that the slower traversing tool results in a measured thermal cycle that remains supersolvus with respect to γ’ for approximately 12 seconds. Upon exposure to temperatures above the γ’ solvus temperature for even brief durations, titanium-enriched regions in the deposition microstructure preferentially form titanium-rich carbides and carbonitrides. The formation of these titanium rich compounds, consumes solute from the surrounding region. When the material is then cooled through the γ’ precipitation temperature range, the interdendritic regions are now considerable leaner in Ti due to carbide and carbonitride formation. Despite slower cooling rates associated with the high heat input condition, γ’ formation is suppressed due to a lack of necessary solute atoms.
The results of thermal simulations suggest that the formation of fine clusters of spheroidal precipitates observed within some regions of the as-AFSP SZ result from regions of the original weld overlay that experienced deformation as evidence by the recrystallized microstructure (see Figure 6.18). However within these regions, mechanical mixing is not sufficient such that these solute enriched regions are not homogenized leading to the distribution of lamellar precipitate-containing areas for some regions of the SZ.

6.2.2.4 Grain Size Evaluation

Microstructural observation using HRSEM indicated that the Alloy 282-containing SZ region contained some grains smaller than 1 \( \mu \text{m} \). Rather than use lineal intercept methods on optical microscopy (OM) micrographs, electron backscatter diffraction was used to determine AFSP grain size instead. Optical microscopy to deconvolute grain boundaries depends highly on the condition of the etched sample. Etching characteristics can be affected by grain boundary state which is related to the misorientation angle, distribution of grain boundary precipitates, solute contents near grain boundaries, etc. [136]. Electron backscatter diffraction is able to unambiguously determine the boundary between grains (using a threshold misorientation) without the influence of the material etching characteristics and resolution limitations of optical microscopy. As a result, the average grain size determined via EBSD is usually smaller than that determined using OM.

Using electron backscatter diffraction, the average grain size within the SZ was determined for additive Alloy 282. Figure 6.21 shows generated inverse pole figure maps for both high and low heat input parameter combinations. The examined regions represent the central SZ region. A clear difference in grain size for the two conditions is observable in the inverse pole figure maps.
For EBSD analyses, a grain was defined as a region containing misorientation angle of great than 5°. An ellipse was fit for each grain not touching the edge of the scan area. Because the morphology of the majority of SZ grains is equiaxed, an equivalent diameter derived from measured major and minor axis measurements was used. For comparison, the grain size using the same EBSD technique and parameters was determined for of IN600 processed using similar FSP parameters. Figure 6.22 shows the distribution of grain size for the samples examined. Values for the average grain sizes are shown on the x-axis. Average grain size for both FSP IN600 and AFSP Alloy 282 was smaller using low heat input process parameters compared to high heat input parameters. However, the average grain size for AFSP Alloy 282 is nearly 1.5 times smaller compared to FSP IN600. While not able to be determined explicitly, the thermomechanical conditions for Alloy 282 are likely slightly different compared to IN600 despite the same process parameters due the difference in high-temperature flow.
stress. Such differences in thermomechanical history can affect the dynamic recovery and recrystallization processes that control the recrystallized grain size within the SZ. Although some differences in the flow behavior for the Alloy 282 weld overlay and wrought IN600 base metal exists, observed forces during AFSP suggest that Alloy 282 weld overlay has very similar measured process loads (a variation of 10% or less) to that of autogenous FSP IN600. Such small variation suggests that flow behavior of both materials tested is similar.

The observed recrystallized grain size is a function of the recovery and recrystallization processes during hot deformation. Material properties such as the stacking fault energy of a material will dictate the ease by which recovery recrystallization processes will proceed. Because dislocation partials are separated by a
greater distance in materials with low SFE, the ease of recovery (i.e., rearrangement of dislocations) is more difficult for materials with lower SFE.

Computational thermodynamic software suites such as JMatPro are able to calculate the SFE of Ni-alloys using appropriate databases. A stacking fault represents local interruption in the normal stacking sequence. For a FCC material, a stacking fault can be considered a local addition or subtraction of an atomic plane to form an extrinsic and intrinsic stacking fault, respectively. Such addition or subtraction of planes results in the formation of a local region in the lattice that contains HCP rather than FCC stacking sequence [47]. Values for SFE are determined computationally by calculating the free energy difference for the $\gamma$ phase in FCC structure versus the HCP structure [137]. Because the thermodynamic database contains free energy data as a function of temperature, temperature-dependent SFE can be calculated.

Figure 6.23 shows calculated SFE values for different alloys explored in this work as a function of temperature. For all alloy compositions, the temperature dependence of SFE is a linear function. When considering the total alloy content for the investigated compositions, the stacking fault energy decreases with increasing total alloy content. Near typical FSP temperatures (900°C), the stacking fault energy of Alloy 282 is 168 mJ·m$^{-2}$ compared to 234 mJ·m$^{-2}$ for IN600. This effect of alloying addition can also be observed in Figure 6.24 for IN600 with varied iron content. A monotonic decrease in calculated SFE is observed as the iron content in increased. Although it can be shown that an increase in alloying content can decrease SFE, no clear attempts have been made to correlate the change in SFE value with the alloying species [47].
Figure 6.23. Computed temperature-dependent stacking fault energy values for alloys examined in this study. JMatPro version 5.1 used for computed values.

Figure 6.24. JMatPro stacking fault energy predictions for Ni-19Cr alloy with varied iron content.
The lower SFE of Alloy 282 compared to IN600 could play a role in the evolution of the SZ microstructure, including the final grain size. It is possible that the lower SFE of Alloy 282 compared to IN600 resulted in a higher degree of stored energy due to the increase in difficulty for dynamic recovery to occur [138]. The increased stored energy may lead to an increase in the density of nucleation sites for recrystallization after the tool has passed while the material is at a sufficiently high temperature for recrystallization to occur. The higher density of nucleation sites for Alloy 282 compared to IN600, which dynamically recovers at a higher rate, may also be a contributing factor for the difference in observed grain size.

Additionally, inherent differences in alloying additions of Alloy 282 compared to IN600 also affects grain size due to solute drag during grain coarsening. Grain coarsening can be inhibited by the presence of solute atoms decreasing the boundary mobility [21] and/or the driving force to maintain boundary velocity [139]. The extent to which these pinning solute atoms segregate to mobile grain boundaries depends largely on the location-dependent solubility of solute atom. In general the concentration at the boundary (C_B) relative to the matrix composition (C_o), defined as the grain boundary enrichment ratio, increases with decreasing solute solubility [21]. Figure 6.25 shows the grain boundary enrichment ratios for several alloy systems. Alloying additions such as Fe and Cr in Ni-base alloys have low enrichment ratios, however, interstitial elements such as B segregate strongly to grain boundaries and have high enrichment ratios therefore playing a larger role in the modification of boundary velocity compared to some substitutional alloying additions.
High heat input FSP and AFSP processing parameters exhibited larger average grain size for both additive and substrate alloys. Additive FSP Alloy 282 and IN600 processed using high heat input parameters exhibited an average grain size of $7.3 \pm 4.1$ and $9.2 \pm 3.5$ µm, respectively. Likely the higher peak processing temperatures and slower cooling time for high heat input parameters allowed more time for grain coarsening after the tool has passed.

### 6.2.2.5 Misorientation Analysis of Additive Friction Stir Processed Regions

Local variations in crystallographic misorientation can serve as indicators of the accumulation of stored dislocations within a material. The presence of dislocation arrays
within a material leads to the formation of low angle misorientations (angles < 10°), which can be characterized via EBSD [113, 115, 120]. In general, regions of the microstructure containing a high density of small angle misorientations can be assumed to have a locally high dislocation density as the result of residual strain and or incomplete recovery and recrystallization.

The kernel average misorientation (KAM) method was used to determine the point-to-point misorientation distribution for AFSP Alloy 282. Figure 6.26 shows KAM maps for Alloy 282-containing regions of the AFSP SZ for both parameter conditions. For comparison, a KAM map derived from a scan region of the as-deposited Alloy 282 weld overlay is also shown in Figure 6.26C. The examined as-deposited scan region represents the second of four weld overlay passes. In general, regions within the microstructure with very low KAM (blue regions) represent regions where recovery and recrystallization processes are complete resulting in very low point-to-point misorientation and therefore low dislocation density. Other regions in the KAM map (shaded green, and yellow) correspond to regions of the microstructure with a high density of low-angle misorientation, i.e., stored dislocations. The frequency of such regions is higher overall for the sample processed using low heat input parameters which suggests that stored energy within the AFSP SZ is higher compared to material processed with high heat input parameters. Likely the lower peak temperatures during stirring and faster cooling rates behind the tool reduced the extent of recovery/recrystallization compared to material processed at higher peak temperatures with more sluggish cooling rates (high heat input parameters). However for both AFSP processing parameter conditions, the density of higher KAM regions is higher compared to the as-deposited weld metal. This result is expected since the as-deposited weld overlay did not experience any mechanical deformation/stirring. Additionally rearrangement and annihilation of dislocations during recovery, which function to lower measured KAM, is also likely in the as-deposited Alloy 282 due to the multiple thermal cycles associated with the multi-pass weld-overlay.
Figure 6.26. Kernel average misorientation maps of AFSP SZ for A.) high heat input, B.) low heat input parameters, and C.) as deposited weld overlay. Maps generated using 3rd order nearest neighbor perimeter kernels with a maximum misorientation of 5°.

Kernel average misorientation maps reveal useful qualitative information regarding the distribution of KAM within the scan area. However, examination of the actual distribution of KAM values within the measured scan regions yields more quantitative information regarding the misorientation, i.e., stored energy, in the material. Figure 6.27 shows the KAM distribution for both AFSP process parameters and the as-
deposited Alloy 282. For comparison, the KAM distributions for FSP IN600 with identical processing parameters are also shown on the plot.

Figure 6.27. Kernel average misorientation distribution for AFSP Alloy 282 and FSP IN600. Values for distribution averages are indicated on the x-axis.

The majority of values for KAM calculated for identical-sized scan regions show the highest frequency of KAM to be less than 0.5°. However, examination of the distribution of KAM values for higher angles (>0.5°) shows trends that are consistent with the qualitative generated maps as well as observed microstructures and microhardness trends. Samples processed using low heat input parameter combinations exhibit the highest average KAM values (0.71° for Alloy 282 and 0.53° for IN600) with distributions shifted towards higher angles. As the FSP/AFSP heat input is increased,
average KAM values decrease as the distribution in misorientation is shifted towards smaller angles. Presumably, the degree of stored energy (i.e., dislocation density) is decreased using parameters that increase the peak temperature along with increasing the total thermal cycle duration. For Alloy 282 processed using high heat input parameters, distribution and average KAM is similar to the as-deposited material.

Like the grain size trends discussed previously, the alloy composition also appears to have an effect on the KAM distribution. As mentioned before, the average KAM values for Alloy 282 and IN600 differ despite the identical processing parameters, similar process forces, and presumably similar process temperatures. Most likely, inherent differences in the way each alloy accommodates dislocations introduced from deformation during stirring or behind the passing tool affects the resulting misorientation distribution. This difference in microstructural evolution due to differences in recovery and recrystallization processes can be attributed to large differences in stacking fault energy as discussed previously. Dislocations introduced during FSP of IN600, with a relatively high calculated stacking fault energy of 234 mJ·m⁻² at 900°C, are able to be much more easily rearranged and annihilated via dynamic recovery due to the rather closely spaced dislocation partials.

It is also possible that the high temperature deformation characteristics of Alloy 282 compared to IN600 result in differences in stored energy/residual strain which may also account for the measured differences in KAM distribution. Difference in high temperature flow stress can affect resulting total strain and also peak temperatures. However, the relative volume of additive material (Alloy 282) compared to the total SZ volume represents approximately one-third of the total process volume and, therefore, may not affect the overall SZ thermomechanical conditions greatly. Moreover, the FSP machine process force output does not vary significantly suggesting that the essentially solutionized Alloy 282 weld overlay has similar mechanical properties to IN600. Microhardness measurements performed are also in good agreement. It therefore can be expected that deformation behavior of the two alloys is similar and differences in stored
energy result from intrinsic differences in dislocation behavior attributed to differences in alloying content.

Analyses of intragranular misorientation were also performed. Intragranular measurements are especially useful for visualizing the distribution of misorientation (i.e., stored energy) within the grain [120, #348] and have been used to visualize distributions of strain in stainless steel welds [123]. Figure 6.28 shows grain reference orientation deviation (GROD) maps for both tested conditions. The intragranular misorientation for each point within a grain is determined from a single reference point with the lowest KAM. Generated GROD maps indicate a higher fraction of AFSP SZ grains for the sample processed using low heat input parameter combinations contains a larger fraction of grains with higher intragranular misorientations (e.g., grains shaded colors other than blue), indicating such small angle lattice rotations within the grain are the result of dislocation arrays therefore suggesting those grains contain locally higher stored energy. However both processing conditions show a large fraction of grains possessing very low intragranular misorientation (GROD values near 0º) which suggests the dislocation distribution is heterogeneous and is very low for some grains. Presumably, recovery/recrystallization processes for these grains is more complete and decreases stored energy as evidence by the near-0º GROD values.

Grain reference orientation deviation results are in good agreement with the previously discussed point-to-point KAM analyses for AFSP samples. Some large weld metal grains for the as-deposited Alloy 282 show consistent GROD values approximately 2 to 3º. However such large misorientation angles were not apparent using the KAM analysis method. Likely GROD misorientation values in the weld metal may be an artifact of the considerably larger grain size of the weld overlay compared to the AFSP SZs. The grain size between the two AFSP conditions is not strikingly different such that similar artifacts exist. Additionally, small angle misorientations (≤~2º) that result from solidification subgrain boundaries in the as-deposited material cannot be differentiated from intragranular misorientation from residual stored work using the GROD technique. Such misorientations were not observed using the KAM analysis technique. The result of
the weld overlay GROD analysis reinforces the need to use more than one misorientation analysis technique for the determination of stored energy.

Figure 6.28. Grain reference orientation deviation maps of AFSP SZ for A.) high heat input, B.) low heat input parameters, and C.) as deposited weld overlay. Intragranular orientation deviation based relative to the minimum KAM point within the grain.
Additive FSP Alloy 282 TEM specimens were extracted from central SZ regions—the same region also examined using EBSD. Samples were examined using STEM to directly observe dislocation arrays that led to the misorientation distribution characterized by EBSD. Figure 6.29 shows a low magnification view of a FIB foil extracted from Alloy 282-containing SZ processed with low heat input parameters. Equiaxed grains are clearly observable with the same morphology and range of sizes as measured by EBSD. The contrast formation observed within some grains is due to local bending and distortion of the material’s lattice due to the presence of dislocations. The distortion locally changes the diffraction condition and therefore affects the resulting image contrast formation. Larger grains visible in the TEM sample exhibit relatively uniform intragranular contrast which indicates low dislocation density. Some annealing twins are also visible within the same region indicative of locally well-annealed material.

Figure 6.29. STEM BF micrograph of AFSP Alloy 282 processed using low heat input parameters
No evidence of fine secondary precipitation was observed in TEM, which is consistent with observations with HRSEM examination of selectively etched AFSP Alloy 282/IN600 samples in the as-processed condition. Figure 6.30 shows a selected area diffraction pattern for a [001] zone axis exhibiting only reflections for the $\gamma$ matrix with no superlattice reflections for $\gamma'$. It should be noted that FIB foils were extracted near regions in the SZ microstructure that contained lamellar distributions of fine-precipitates (refer to section 6.2.2.3); however, none of the precipitates appear to be retained in the foil. Additional extracted samples may be successful in capturing the precipitates. Other notable feature observable in the FIB-prepared TEM samples include abrupt changes in the brightness of the image parallel diagonal bands. These diagonal bands are artifacts of the FIB preparation technique.

Figure 6.30. A.) Selected area electron diffraction pattern along B=[001] and B.) computed diffraction pattern for $\gamma$-Ni B=[001]. Only reflections for $\gamma$ matrix are observed.
Closer examination of SZ grains reveals arrays of dislocations. Figure 6.31A and B show higher magnification BF STEM micrographs of AFSP Alloy 282 process using high and low heat input process parameters, respectively. Regions showing more detail of the dislocation arrays are found in Figure 6.32Figure 6.31A and B. Comparing the material processed using high heat input compared to low heat input, visible dislocations are clearly visible as sporadic tangles with large defect-free regions separating the tangles. The defect structure of the high heat input sample appears similar to TEM results presented by Sato et al. [140] for IN600 which contained only a slightly higher dislocation density compared to the mill annealed base material.

Figure 6.31. STEM BF micrographs of A.) high heat input and B.) low heat input AFSP Alloy 282
The BF STEM micrographs reveal that some grains contain very few dislocations, suggesting that dislocations introduced during stirring have been eliminated by recovery and recrystallization. This is especially valid for the high heat input sample. However, the overall density of dislocations for the high heat input sample is lower compared to the low heat input sample which exhibits many more dislocations arranged in a dense network. Both samples exhibit a heterogeneous distribution of dislocations with some grains appearing to contain a much more extensive dislocation network, suggesting incomplete recovery/recrystallization or some post-recrystallization deformation has occurred. STEM BF micrographs showing the distribution of dislocations is in agreement with misorientation analyses discussed previously, the results of which suggest a locally heterogeneous distribution of stored energy. The difference in stored energy that arises from the parameters differences with different associated thermomechanical histories accounts for the observed differences in precipitate formation.
kinetics manifested as a significant difference in heat treatment response—the results of which are presented in the following section.

6.2.3 Microstructure of Heat-Treated AFSP Alloy 282

The addition of Alloy 282 using AFSP, creates an age-hardenable superalloy surface layer. To examine the heat treatment response of the hardenable surface layer, additive FSP samples were given a post-AFSP direct age heat treatment. The samples were heated to 750 °C and held for 2 hours. Following the aging heat treatment, both AFSP conditions tested exhibited an increase in hardness due to the formation of a fine distribution of $\gamma'$. The microhardness distribution within the AFSP SZ after heat treatment is shown in Figure 6.33. The average hardness of the Alloy 282 within the AFSP SZ increases to 360 ± 28 and 410 ± 31 HV for high and low heat input parameters, respectively. For comparison, the weld overlay of Alloy 282 after heat treatment exhibited a measured average microhardness of 305 ± 13 HV, 18 to 35% lower than AFSP average hardness.

As with the non-heat treated AFSP samples, the AS of the SZ contains the highest proportion of Alloy 282 with high hardness (>275 HV). Additive FSP samples processed using low heat input parameters exhibited the greatest heat treatment response despite experiencing lower peak temperatures during processing and faster cooling rates relative to the high heat input conditions. The kinetics of $\gamma'$ formation in Ni-base superalloys depend not only on time and temperature, but also on the degree of stored energy in the form of dislocations [36, 37]. An increase in stored energy in the form of dislocations can function to increase the number of heterogeneous nucleation sites for precipitates and result in an increased density of strengthening precipitates. The microhardness results of heat-treated AFSP samples suggest process parameter changes, which affect the extent of recovery and recrystallization, influence the amount of stored energy in a material after AFSP. Such results are in good agreement with the misorientation and TEM analyses discussed in previous sections.
Examination using HRSEM of selectively etched samples after heat treatment reveals the formation of fine, spherical $\gamma'$ within SZ regions with hardness values exceeding ~275 HV. Figure 6.34 shows representative micrographs of the distribution of $\gamma'$ for the two FSP parameter conditions. The sample processed using the low heat input parameter combination demonstrates enhanced heat treatment response relative to the high-heat input condition as evidence of the increase in number of $\gamma'$ precipitates present.
Samples etched with the Cr(VI)-based selective etch effectively reveal the distribution and morphology of precipitates. However such micrographs are not ideal for automated quantitative image analysis. Automated analyses of $\gamma'$ fraction estimates are performed more effectively on samples prepared such that the $\gamma'$ is attacked and the matrix material is left unaffected. The resulting holes in the surface from the missing $\gamma'$ are imaged using HRSEM using the solid-state backscatter detector. Additive FSP samples were etched to remove $\gamma'$ using a solution of (by volume) 61% Lactic acid (85%), 36% concentrated HNO$_3$, and 3% HF (49%). The sample surface was swabbed with the etchant and then ultrasonically cleaned in methanol.

Figure 6.35 shows an example micrograph etched using the aforementioned etchant. Volume fraction estimates of $\gamma'$ using digital image analysis reveal the low heat input condition contains an average 14.2 vol% $\gamma'$ prime compared to the high heat input conditions which contains on average 8.1 vol% $\gamma'$. For comparison, the equilibrium $\gamma'$ content, determined computationally using JMatPro (Ni-superalloy database), for Alloy 282 is 24 vol%. Volume fraction measurements were taken within regions of the sample
exhibited peak harness values. Regions with lower measured hardness had lower measured $\gamma'$ fractions.

Figure 6.35. Example HRSEM micrograph of Alloy 282 etched using $\gamma'$-selective etch used for automated digital image measurement. Dark regions in micrograph correspond to ‘holes’ in sample surface occupied by $\gamma'$ prior to etching

A particular disadvantage of fusion-based additive techniques for materials such as superalloys is the formation of as-deposited microstructures that exhibit locally heterogeneous heat treatment response which typically degrades the mechanical properties of the overlay. Difficulty in achieving fusion weld strength values that are comparable to the strength of wrought base material has been reported [48, 141]. Such heterogeneous heat treatment response is due to the microsegregation of $\gamma'$ promoting elements that occurs during non-equilibrium solidification. In general solute partition coefficients, $k$, less than unity combined with low Al + Ti diffusivity in the $\gamma$ matrix [45]
lead to the formation of compositionally dissimilar interdendritic regions possessing enhanced heat treatment response which surround $\gamma'$-denuded cell core regions. Solute that promotes heat treatment response can also be tied up in intermetallic compounds that form at the end of solidification or upon cooling to room temperature [142].

If it is desired to eliminate elemental microsegregation from solidification, a solutionizing heat treatment must be applied at a temperature above the $\gamma'$ solvus temperature. However solute enriched regions in the solidification microstructure can have solvus temperatures higher than expected for the bulk alloy composition. The actual solvus temperature for solute enriched regions depends on the diffusivity of the segregated species, thermal history during solidification and the partition coefficient [45]. Figure 6.36 schematically shows the $\gamma + \gamma'$ solvus curve as a function of distance between the cell core and cell boundary region. Regions near the cell boundary with solvus temperatures above the solutionizing temperature ($T_{sol}$) will not homogenize during heat treatment.
Figure 6.36. Schematic illustration of γ’ evolution in a solidified Ni-base superalloy microstructure A.) after solidification, B.) during solutionization, C.) after quenching, and D.) after aging heat treatment. Figure adapted from DuPont et al. [44].

For the weld-deposit conditions used in this work, Alloy 282 in the as-deposited condition did not exhibit signs of γ’ precipitation. Relatively low hardness measurements of 203 ± 13 HV also validate observations using HRSEM. Despite, the lack of γ’ precipitation in the as-deposited condition, microsegregation in the weld metal still occurred. Solute segregation became evident upon inspection after heat treatment via
examination of etched weld overlay using HRSEM. Figure 6.37 demonstrates the cellular distribution of $\gamma'$ in the Alloy 282 weld overlay after age heat treatment. Interdendritic regions demonstrate enhanced heat treatment response relative to the cell core regions which are denuded in $\gamma'$. Regions in the microstructure containing higher $\gamma'$ fractions appear lighter in the micrographs not due to atomic number contrast, but due to increased electron detector signal from submicron precipitates exposed on the sample surface after selective etching. Also visible within the interdendritic cell boundary regions is the presence of some irregularly shaped carbides that appear bright in the SEM micrographs.

![Figure 6.37](image)

Figure 6.37. Locally heterogeneous heat treatment response from microsegregation in as-deposited Alloy 282 demonstrating decreased fraction $\gamma'$ in A.) dendrite core relative to B.) interdendritic region.
Although the hardness was increased after aging, the Alloy 282 deposit is expected to have inferior tensile properties compared to wrought Alloy 282. The locally hard and soft regions in the weld metal microstructure that correspond to the density of $\gamma'$ particles will lead to preferential strain localization upon loading. This strain localization behavior was observed in situ on polished weld metal samples of iron-based superalloy A-286 [143]. Localized strain within the solidification structure corresponded to titanium-denuded dendritic regions. It was found that the degree of strain localization was proportional to the degree of titanium depletion in the dendrite cores.

To better understand the heterogeneous heat treatment response of the as-deposited Alloy 282, solute partitioning coefficients, $k$, were determined computationally using ThermoCalc. The nominal Alloy 282 composition was used to determine $k$ values for the different alloying additions. Figure 6.38 shows computed $k$ values for Alloy 282.
The $k$ value calculations suggest that elements such as Ni, Co, Cr, Al, and Fe with values near unity do not segregate strongly to the liquid during solidification, but rather will tend to partition slightly to the cell core. However, other alloying elements such as Mo, Ti, Si, C, and B will partition to the liquid during solidification therefore leaving interdendritic regions in the microstructure enriched in these elements immediately after solidification. Interstitial elements such as C and B that possess high diffusivity in the $\gamma$ matrix will likely diffuse and reduce the concentration gradient. However, elements such as Ti with low diffusivity in $\gamma$ will remain, enriching the cell boundaries. The composition of the liquid during solidification was determined computationally using JMatPro which considers solidification occurring under Scheil-Gulliver conditions. The result of the solidification simulation is shown in Figure 6.39. Although the nominal composition of Alloy 282 contains 2 wt % Ti, the last liquid to form is predicted to contain over three times the nominal composition due to the $k$-value for Ti less than 1. As a $\gamma'$-promoting element, regions enriched with Ti will promote the formation of $\gamma'$. 

Figure 6.38. ThermoCalc prediction of partition coefficient ($k$) values for Alloy 282.
relative to denuded regions. Kinetic/thermodynamic simulations of the time-temperature-transformation behavior of $\gamma'$ was explored as a function of Ti-content. Holding other alloying additions constant, the predicted TTT behavior for $\gamma'$ is shown in Figure 6.40 as a function of Ti content. As the amount of Ti increases beyond the nominal composition (2 wt% Ti) to the close to the predicted interdendritic composition (approximately 6 wt% Ti), the nose of the TTT curve for $\gamma'$ is shifted towards shorter durations by nearly an order of magnitude. Based on these calculations, the partitioning of Ti in Alloy 282 is likely responsible for the heterogeneous distribution of $\gamma'$ observed in the microstructure.

Figure 6.39. JMatPro prediction of elemental distribution within the liquid phase during solidification of Alloy 282.
Figure 6.40. JMatPro predicted TTT behavior for Alloy 282 as titanium concentration is varied by substituting with Ni. Aluminum level held constant at 1.4 wt%. Curves represent the formation of 1 vol% $\gamma'$.

Figure 6.41 demonstrates the typical homogenous distribution of $\gamma'$ within the SZ of a heat treated AFSP sample. Changes in the intragranular contrast in Figure 6.41 is the result of electron channeling contrast and is not due to the density of $\gamma'$ precipitates present. The AFSP SZ does not exhibit locally heterogeneous distributions of $\gamma'$ within the Alloy-282-containing SZ regions. Both heat input parameter combinations showed an elimination of the original solidification microstructure and microsegregation. Intense plastic flow from mechanical mixing of the additive material by the FSP tool at elevated temperature eliminates microsegregation developed during solidification of the weld overlay, especially Ti. The homogenization provided via AFSP circumvents the necessity for solutionizing heat treatments normally used to eliminate weld metal microsegregation. Such mechanical mixing/deformation at elevated temperatures is analogous to a forging operation. The elimination of a high-temperature homogenization
heat treatment has the potential to reduce production costs associated with energy and time spent during fabrication.

![Image](image.png)

Figure 6.41. Heat treated Alloy 282-containing SZ exhibiting no microsegregation. Intragranular contrast change in the image results from electron channeling contrast.

### 6.3 Additive Friction Stir Processing of Haynes Alloy 214

Additive friction stir processing using the same technique utilized on Alloy 282 was also performed on Haynes Alloy 214. This simple Ni-Cr-Fe system (refer to table 4.1) contains rather high aluminum content of 4.2 wt%. The high aluminum content provides high temperature oxidation resistance and also precipitation strengthening response via the formation of $\gamma'$ [144]. Using the same GTAW and FSP experimental conditions, AFSP produced Alloy 214 surface layers on IN600 plates. As with 282, both heat input extremes were chosen for examination however the low heat input and high heat input parameter combinations were slightly modified compared to AFSP Alloy
282/IN600 to produce defect-free SZs. Table 1 lists the parameter combinations used for Alloy 214 AFSP. Double pass AFSP was also explored. A first FSP pass was created on the Alloy 214 weld overlay using low heat input parameters. A second FSP pass was then performed after the workpiece had cooled down to near room temperature directly atop the first FSP pass with 100% overlap. The traverse direction for the second pass was identical to the first pass.

Table 6.1. Process parameters utilized for AFSP of Alloy 214/IN600

<table>
<thead>
<tr>
<th>Heat Input Designation</th>
<th>RPM</th>
<th>Traverse Speed (IPM)</th>
<th>Dimensionless Heat Input (ω/U*)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low</td>
<td>100</td>
<td>3.5</td>
<td>8.2</td>
<td></td>
</tr>
<tr>
<td>High</td>
<td>150</td>
<td>1.8</td>
<td>46.3</td>
<td></td>
</tr>
<tr>
<td>Low 2X</td>
<td>100</td>
<td>3.5</td>
<td>8.2</td>
<td>Double Pass - 100% overlap</td>
</tr>
</tbody>
</table>

6.3.1 Distribution and Microstructure of Additive Alloy 214

Alloy 214 was successfully incorporated into the surface of IN600 plate. Figure 6.42 shows etched cross sections showing the distribution of Alloy 214 contained in the surface. From the etched cross sections it is clear that the distribution and fraction of retained Alloy 214 depends highly on the FSP processing parameters. Using the same digital image analysis technique used on the Alloy 282/IN600 AFSP samples, an average fraction of retained Alloy 214 was determined. The AFSP SZ, on average, contains 69, 77, and 44% of the original Alloy 214 weld deposited for the low, high, and low-double pass processing parameter combination, respectively. Portions of the original weld deposit that were not incorporated during AFSP were extruded away as flash. The
second overlapping pass in low heat input double pass resulted in the loss of additional additive material compared to the single pass samples. Reductions in the amount of flash generated during process could possibly be accomplished via modification to the tool geometry/features. Concave or flat shoulder profile tools, such as the one used in this work, have a tendency to create a slight undercut in the surface of the workpiece. Additionally such tool must be run at a slightly tool tilt angle, such as 3°. The addition of a spiral cut channel, known as a scroll feature, machined into the tool shoulder increases the amount of deformation by the tool shoulder and eliminates the need for a tool tilt angle thus reducing the amount of undercut and flash produced [52]

Figure 6.42. Cross sections of AFSP Alloy 214/IN600 processed using A.) low, B.) high, and C.) low double pass heat input parameters.
Unlike the distribution of additive material observed in Alloy 282/IN600 AFSP, the advancing side (AS) of the SZ does not show the same degree of material turbulence. Additive material does not appear to be disrupted on the AS and deposited in a large continuous region on the RS. For all parameter conditions explored, the Alloy 214 appears to remain in the SZ as a single contiguous region with some additional ribbon-like distributions above and below. Some regions of the deposit are nominally the same thickness as the as the original weld overlay layer (~2 mm). The double-pass sample appears to have the most chaotic distribution of material on the AS of the SZ which is attributed to the accumulative nature of deformation from subsequent passes.

Similar behavior for Alloy 214/IN600 on the retreating side (RS) compared to Alloy 282/IN600 was observed. For all the explored parameter combinations an accumulation of additive material was observed on the RS. Again, this behavior can likely be attributed to a redistribution of additive material by the advancing tool. As the tool approaches, material on the AS of the tool is carried around the tool periphery and deposited behind the moving tool on the advancing side. Observed vertical movement of additive material largely was not observed. However, some portions of the AFSP SZ demonstrate areas where IN600 is present on the top surface (Figure 6.43). This is contrary to the initial as-deposited condition where Alloy 214 comprises approximately the top 2 mm of material on the surface. This behavior, however, it not consistent across the whole stir zone. Some regions remain which contain additive material on the top surface (Figure 6.43B). This result clearly shows that the material flow within the AFSP is rather complex and does involve locally regions that exhibit vertical, maelstrom-like flow. For practical application, tool geometry likely will have to be modified to reduce local areas of substrate material present on the surface.
The large contiguous regions of additive material with much less tortuous boundary regions compared to Alloy 282 suggests that the material flow of the additive Alloy 214 was similar to the IN600 substrate alloy. Both IN600 and Alloy 214 in the as-deposited condition do not contain precipitates. Assuming other strengthening mechanisms are similar or negligible for both alloys, the degree of solid solution strengthening will dictate the material flow behavior. Alloy 214 and IN600 are quite similar compositionally in terms of total alloy content (refer to Tables 4.1 and 4.2) therefore it can be expected that the strengthening effect from solid solution strengthening to be similar for both alloys. Figure 6.44 shows temperature-dependent yield strength from experimental data for both IN600 [145] and Alloy 214 [48]. Testing conditions for the experimental mechanical data shown for Alloy 214 were not given. However, it can be assumed that the material was given a heat treatment to form strengthening γ’. Solutionized Alloy 214, such as the weld metal of the overlay is expected to have lower yield strength values.
The temperature dependent yield strength for Alloy 600 and Alloy 214 was also calculated using JMatPro. JMatPro is able to predict the yield strength and temperature-dependent yield strength for many engineering alloys including Ni-base and Ni-base superalloys. The model utilized in the software is described in detail by Guo et al. [100]. In this model the yield strength is determined by considering strengthening contributions from solid solution strengthening and precipitate hardening at room temperature. JMatPro contains an extensive intrinsic material flow stress and Hall-Petch coefficient database. Coupled with an input of grain size for the calculation, yield stress, $\sigma_y$, can be determined by using the standard Hall-Petch equation

$$\sigma_y = \sigma_{yo} + k d_y^{0.5} \quad (10)$$
where $\sigma_{yo}$ is the intrinsic flow stress, $k$ is the Hall-Petch coefficient, and $d_g$ is the grain size. Next, the temperature dependent strength, $\sigma_T$, is related to the room temperature strength by an Ahrenniius-type exponential decay using the following equation:

$$\sigma_T = \alpha + \beta \exp \left( -\frac{Q}{RT} \right)$$  \hspace{1cm} (11)$$

where $Q$ is the deformation activation energy. The pre-exponential coefficients, $\alpha$ and $\beta$, are directly related to the room temperature yield strength, although not described explicitly by the authors. The activation energy is determined by regression analysis of a wide range of empirical data for Ni-base alloys.

If the alloy contains strengthening precipitates such as $\gamma'$, the yield strength is largely governed by the ease by which a pair of dislocations with Burgers vector $a/2<110>$ move through the ordered structure. When the $\gamma'$ particles are small, the resistance to move these dislocation pairs largely determines the yield strength behavior [100]. In this case the yield strength is determined by following relationship:

$$\sigma_{y1} = M \frac{\Gamma}{2b} \left[ A_1 \left( \frac{\Gamma'f d}{T_l} \right)^{0.5} - f \right]$$  \hspace{1cm} (12)$$

Where $M$ is the Taylor factor, $\Gamma$ is the anti-phase boundary energy, $b$ is the Burgers vector, $d$ is the average $\gamma'$ size, $T_l$ is the dislocation line tension, and $f$ is the volume fraction of $\gamma'$. The constant $A_1$ represents a morphological factor that depends on the shape of the particles. For spherical particles, such as very small $\gamma'$, this value equals 0.72. As the size of $\gamma'$ particles increases, a different yield stress model is utilized given by

$$\sigma_{y2} = 1.72M \frac{T_l \omega f^{0.5}}{2bd} \left( 1.28 \frac{\Gamma d}{\omega T_l} - 1 \right)^{0.5}$$  \hspace{1cm} (13)$$
where the variable $\omega$ represents an empirical that accounts for the repulsion of dislocations within the $\gamma'$ precipitate.

In JMatPro, the size of the $\gamma'$ precipitates can be determined by inputting both the size and distribution directly or by estimation with prior knowledge of room temperature mechanical properties. In the calculation of yield strength in superalloys, the yield strength is governed by the lower of the two equations (12) or (13).

The calculated values for yield strength of IN600 and Alloy 214 are shown in Figure 6.44. It should be noted that the temperature-dependence of experimentally-derived yield strength for both alloys is very similar and exhibits a sharp decrease around 600°C. The calculated values for both alloys exhibit the sharp decrease in strength at temperatures approximately 300°C higher than experimentally-determined values indicate. Regardless of the position of the drop in yield strength, it is apparent that both alloy near the temperatures experienced during AFSP within close proximity to the tool shoulder (>900°C) exhibit very low yield strength values. While dependent on the total strain and strain rate during FSP, the material flow stress behavior is expected to follow the predicted trends for temperature-dependent yield strength.

Striking similarities in the SZ grain size for IN600 and Alloy 214 were observed despite the rather drastic difference in initial grain size between the wrought IN600 plate and the Alloy 214 weld metal. The single phase nature of Alloy 214 allows for the fusion zone (FZ) to be comprised of very large columnar grains with some exceeding 500 $\mu$m in length (Figure 6.45).
Figure 6.45. Optical micrograph of the as-deposited Alloy 214 weld microstructure and HAZ formed within the IN600 substrate.

Figure 6.46 shows SZ microstructures for Alloy 214 as well as Alloy 600 located away from the additive material in the same sample processed using low heat input parameter conditions. Lineal intercept method on optical micrographs of the low heat input sample indicated an average grain size 13.2 ± 0.7 and 13.3 ± 1.1 µm for Alloy 214 and IN600-containing SZ regions, respectively. The very similar resulting grain size for the two alloys suggests that the thermal and deformation histories and high-temperature mechanical behavior of the two materials are sufficiently similar to produce a final dynamically recrystallized microstructure with very similar grain size. The average grain size for the higher heat input parameter combination is larger than the low heat input parameter combination as expected. However as in the case of the low heat input sample, the measured grain sizes for the substrate alloy and the additive Alloy 214 processed using high heat input parameters were strikingly similar: 19.3 ± 0.9 and 20.5 ± 0.6 µm for Alloy 214 and IN600, respectively.
Similarities in grain size was not observed in AFSP of Alloy 282/IN600 (refer to Figure 5.22 in section 6.2.2.4). The considerably higher alloying content of Alloy 282 compared to IN600 likely resulted in a different recrystallization behavior and or different local strain/thermal history which resulted in a different recrystallized grain size.

6.3.2 Microhardness Distribution in as-AFSP Alloy 214

Microhardness maps created within the AFSP region show the distribution in microhardness (Figure ). The measured hardness of as-deposited Alloy 214 weld overlay was 180 ± 9.8 HV which is comparable to the hardness of the as-received IN600 substrate, 186 ± 5 HV. After AFSP, the measured hardness within the Alloy 214-containing regions of the SZ increased moderately to average values between 225 and 250 HV. As with the Alloy282/IN600 samples discussed previously, overall higher hardness values were observed with the sample processed using the low heat input parameter combination. However, unlike the Alloy 282/IN600 samples the AS of the SZ did not exhibit the same concentration of regions of higher overall hardness. This observation was consistent all three parameter combinations. Interestingly, the additional
stirring and thermal cycle provided by the double pass did not significantly affect the hardness of the Alloy 214 containing regions.

Like the Alloy 282/IN600 samples, the addition of Alloy 214 creates a surface layer with an age hardening response. To explore the effect of AFSP on the resulting hardness and distribution of $\gamma'$ formed, samples were given direct age heat treatment at 750 °C for 2 hours followed by air-cooling. Figure 6.48 shows the distribution of microhardness after the age heat treatment. The hardness of all samples was increased overall after aging due to the formation of $\gamma'$. Overall the hardness of the AFSP Alloy 214 SZ was increased from 225-250 to 300-325 HV depending on location within the SZ. Some regions exhibit hardness values exceeding 350 HV. For comparison, the hardness of the Alloy 214 weld overlay after heat treatment was 282 ± 14.7 HV which represents a nearly 60% increase in hardness compared to the non-heat treated weld overlay. However, unlike the Alloy 282/IN600 sample examined previously, regions of high hardness after age heat treatment are located predominantly on the RS of SZ, which contains the largest accumulation of Alloy 214. While the peak hardness values obtained for all three conditions are comparable (~350 HV), the sample processed using high heat input parameters appears to have the largest fraction of high hardness regions (>275 HV) within the SZ. This is likely due simply to the largest fraction of Alloy 214 retained within the SZ after AFSP using such parameters.
Figure 6.47. Vickers hardness maps of AFSP Alloy 214 on IN600 using A.) high, B.) low and C.) double pass low heat input parameter combinations
Figure 6.48. Vickers hardness maps of heat treated AFSP Alloy 214 on IN600 using A.) high, B.) low and C.) double pass low heat input parameter combinations.
6.3.3 Microstructure of AFSP Alloy 214

6.3.3.1 As-Processed AFSP Alloy 214 Microstructure

The microstructure of AFSP Alloy 214 was examined using HRSEM on selectively etched specimens. The ‘standard’ chromic Cr(IV)-based electrolytic etchant used for all other Ni-superalloys explored in this work has been used successfully on a number of other γ’ and γ’’ strengthened superalloys such as Inconel 718, 704, 725, and René 104 to reveal strengthening precipitates. When the Cr(IV)-based etchant was used with Alloy 214 microstructures containing γ’ the dissolution rate of the γ matrix appears to be similar to that of γ’ resulting in a lack of any morphological information. All the other alloys etched with the ‘standard’ Cr(IV) etchant represent Ni-base superalloy compositions with richer chemistries. Such alloys contain alloying additions such as titanium, hafnium, tantalum, etc. that partition preferentially to the γ’. Alloy 214 does not contain any appreciable alloying additions, such as titanium, that partition to the γ’. This difference in γ’ composition likely contributed to the poor etching response using the standard Cr(IV) selective etchant. An alternative phosphoric acid-based etchant used by Barabash et al. on Inconel 738 [58] was used instead with success (Figure 6.49).
Figure 6.49. HRSEM micrograph of heat treated AFSP Alloy 214 etched using A.) Cr(VI)-based γ-matrix selective etch and B.) Cr(VI)-free γ-matrix etch.

Microstructural examination using HRSEM of etched Alloy 214-containing SZ regions reveals the lack of any resolvable precipitation. Figure 6.50 shows a representative microstructure of Alloy 214 AFSP SZ processed using double-pass low heat input parameter combination near a grain boundary within the RS of the SZ. The microstructure is essentially featureless with some void-like regions approximately 100 nm observed along the grain boundary that are likely chromium-rich carbides or carbonitrides that were removed as a result of the etch. Despite the additional mechanical deformation and additional thermal cycle provided by the double pass processing condition, the formation of any strengthening precipitates was not observed. The lack of observable precipitation is in agreement with the only moderate increase in hardness measured within the additive regions. The results obtained for the as-AFSP Alloy 214/IN600 are contrary to observations of lamellar regions of fine precipitates within regions of the SZ that corresponded to areas of high hardness (>250-275 HV). Because both material systems were processed using very similar FSP parameters, the difference in precipitate evolution is simply due to intrinsic differences in the kinetics of □’ formation of each alloy. Using JMatPro, time-temperature-transformation behavior for □’ was determined computationally. Such TTT curves for Alloy 214 and Alloy 282 are
shown in Figure 6.51. When comparing the onset of precipitation for the two alloys, it is clear that strengthening precipitates in Alloy 214 form much more sluggishly compared to Alloy 282. The nose of the TTT curve for Alloy 214 is offset such that the duration required for the onset of precipitation is over an order of magnitude longer. Alloying additions contained in Alloy 282, such as Ti, have a significant effect on increasing the rate of precipitation in Ni-base superalloys [11, 46, 48]

Figure 6.50. Selectively etched SZ microstructure of AFSP Alloy 214/IN600 along a grain boundary demonstrating no observable precipitation. Material processed using double-pass low heat input parameter combination.
Figure 6.51. JMatPro predicted TTT curves for $\gamma'$ precipitation for Alloy 214 and Alloy 282. The TTT curves represent the transformation of 1 vol% $\gamma'$.

Thermal simulations performed in the Gleeble in an identical fashion as those performed for Alloy 282 (refer to section 6.2.2.3) were also performed for Alloy 214. High heat input and double-cycle low heat input thermal cycles were chosen as they represent the extremes of the processing window. Figure 6.52 shows the resulting microstructures which are devoid of visible $\gamma'$ precipitates. The kinetics of $\gamma'$ formation for Alloy 214 are sufficiently sluggish that even for double thermal cycle conditions representative of double pass AFSP precipitation does not occur in the as-processed condition.
Figure 6.52. HRSEM micrographs of selectively etched thermally simulated Hayes Alloy 214 weld overlay. A. and B.) Regions of high heat FSP input simulated weld overlay microstructure, C. and D.) weld overlay of simulated double pass low heat FSP input, and E.) as-deposited weld overlay with no thermal simulation.

6.3.3.2 Heat Treated AFSP Alloy 214 Microstructure

After the post-AFSP age heat treatment, the distribution of the additive alloy is much more clearly observable. Figure 6.53 shows an interpenetrating layer of IN600 within an Alloy 214-containing region located on the RS of a sample processed using high heat input parameters. Lighter regions in the micrograph correspond to regions of Alloy 214 whereas the darker regions devoid of γ’ correspond to the substrate, IN600. As with the Cr(IV)-based etchant, the grain boundaries are clearly delineated using the alternative etch. Intimate material mixing and bonding between the additive Alloy 214
and IN600 substrate is evidence by grain boundaries growing across the boundaries between the two materials (Figure 6.53B).

![Figure 6.53. A.) Intercalation of IN600 within Alloy 214-containing region within the SZ. White arrows denote grains containing both IN600 and Alloy 214. B.) Interface between Alloy 214 and IN600 is sharp with respect to heat treatment response.](image)

After heat treatment, Alloy 214-containing regions were found to contain nearly monosize distributions of fine secondary $\gamma'$ approximately 20-30 nm. Figure 6.54 shows representative micrographs of the $\gamma'$ distribution near high hardness regions on the RS of the SZ. All the micrographs were taken near grain boundaries to show to distribution of
$\gamma'$ near the boundary as well as to observe any phases decorating the boundaries. Upon closer examination of the microstructures, the low heat input condition appears to have the highest fraction of $\gamma'$ precipitates compared to the low heat input conditions. Similar parameter-dependent precipitate behavior was also observed for Alloy 282. The grain boundary, however, for the high heat input parameter sample appears to have a $\gamma'$-denuded zone approximately 250 nm wide on either side of the grain boundary. These denuded zones appear to exist near grain boundary regions containing a higher fraction of grain boundary constituent phases. The formation of intergranular chromium-rich carbides or carbonitrides could possibly locally change the Cr concentration, which has a moderate direct correlation with the $\gamma'$ volume fraction [46]. It should also be noted that the microstructure of the double pass sample after heat treatment does not appear significantly distinct compared to the single pass AFSP conditions despite the additional thermomechanical cycle. The morphology and distribution of $\gamma'$ is quite similar to the single pass sample processed using the same rotation rate and tool travel speed.
Figure 6.54. HRSEM micrographs of direct aged AFSP Alloy 214 along grain boundaries for A.) low heat input, B.) high heat input, and C.) low heat input double pass process parameters.

A locally heterogeneous heat treatment response for the Alloy 214 weld overlay was not observed. The rather homogeneous distribution of γ’ within the weld metal suggests that the degree of microsegregation of γ’ formers was not as extensive, for example as compared to Alloy 282. Figure 6.55 shows a representative electron micrograph of the rather homogenous distribution of γ’ in heat treated weld overlay.
The only γ’-promoting elements in any appreciable quantity contained in Alloy 214 is aluminum. As with Alloy 282, the extent of aluminum segregation in Alloy 214 is also minimal. Figure 6.56 shows computationally derived partition coefficients for relevant elements present in Alloy 214. The predicted partition coefficient for Al is close to unity (k_{Al} = 0.88), suggesting minimal segregation during solidification of the weld overlay. Because aluminum is the only γ’-promoting element in Alloy 214, a uniform precipitate distribution similar to that observed in Figure 6.55 should be expected. Work by Cieslak et al. [142] investigated weldability of superalloy Cabot 214, which is very similar compositionally to Alloy 214. The aluminum compositional profile across a solidification grain boundary was gathered using electron microprobe analysis (EPMA). Figure 6.57 shows the concentration of aluminum across the scan region. While aluminum concentration does vary between the dendrite core and interstices, compositional variance is small (approximately 1 wt% or less) which is in good
agreement with observations in this work as well as predictions from non-equilibrium simulations.

Figure 6.56. Predicted elemental partition coefficients for Alloy 214.
6.3.4 Potential Application for AFSP Alloy 214

Applications for AFSP are not limited to applications where enhancement of surface strength or hardness is the main priority. For Ni-alloys such as Alloy 214, the most predominant application is for high temperature oxidation resistance. Such applications in high temperature systems include exhaust path components for gas turbine engines, heat exchangers, solid oxide fuel cell supports, catalyst supports, etc. [146]. Such applications rely on the formation of surficial protective oxide layers. The excellent oxidation resistance of Alloy 214 is a result of a continuous thin (1-2 μm) adherent Al₂O₃ scale layer on the surface which forms beneath a thicker Ni-Al spinel layer (NiAl₂O₄) [147]. The refined microstructure characterized by additive layers can be advantageous for other applications including oxidation resistance.

Work by Tan et al. [148] explored the oxidation resistance of a chromia-forming (Cr₂O₃) Fe-Ni-Cr alloy (Incoloy 800H). The authors noted an increase in the oxidation
resistance and oxide exfoliation resistance as the grain size near the surface decreased. Tan et al. attributed increase in oxidization resistance to the increase in diffusive pathways for chromium as fraction of grain boundaries increases with decreasing grain size. The flux of chromium atoms via grain boundaries \( j_{Cr,GB} \) was calculated with knowledge of the diffusivity of Cr along grain boundaries and within the FCC lattice as per the following equation:

\[
  j_{Cr,GB} = \frac{j_B}{(j_B + j_L)} \tag{14}
\]

where \( j_B \) and \( j_L \) are the fluxes (of chromium) within the boundary and lattice, respectively. Values for \( j_B \) and \( j_L \) are approximated by the following relationships:

\[
  j_B \approx \frac{d^2wD_B(C_B - C_1)}{(\pi D_B t)^{1/2}} \tag{15}
\]

\[
  j_L \approx \frac{d^2D_L(C_L - C_1)}{(\pi D_L t)^{1/2}} \tag{16}
\]

where \( d \) is the grain size, \( D_L \) is the lattice diffusion coefficient, \( D_B \) is the grain boundary diffusion coefficient, \( w \) is the grain boundary width, \( C_B \) is the bulk concentration, \( C_1 \) is a fixed surface concentration, and \( t \) is the time of oxidation. Using the above relationships, the relative flux of chromium atoms via grain boundaries is shown schematically is Figure 6.58. Higher diffusivity pathways increase with an increase in the atomic flux via grain boundaries as the grain size (i.e., the fraction of boundaries increase) decreases. The ratio of atomic flux via boundaries relative to the flux via the lattice approaches unity for nanoscale grains. While Alloy 214 SZ grains produced using the procedures outlined in this document do not produce nanoscale grains, SZ grains were reduced nearly two orders of magnitude compared to the large WM grains in the weld overlay. The increase
in grain boundary area along regions exposed to the environment has potential to enhance the formation and stability of protective oxide layers thus enhancing performance.

![Graph](image)

Figure 6.58. Contribution of chromium atom flux via grain boundaries as a function of grain size.

6.4 Summary

6.4.1 Ti-6Al-4V Additions to Ni-201

Titanium alloy powder was incorporated in Ni-201 using AFSP techniques as a proof of concept attempt to form η phase in a pure Ni matrix. Processing difficulties which include difficulties in obtaining a consolidated, defect-free AFSP regions were attributed to reactivity between the Ti-alloy addition and Ni-201 substrate. Reaction between the two materials resulted in localized liquid film formation and subsequent disruption of the FSP process. Analysis of process force output showed characteristic
force signatures that corresponded to defects visible on the plate surface. Microstructural examination of the AFSP SZ regions revealed that large amounts of the additive Ti-alloy was extruded as flash. Retained Ti-alloy near the surface of the SZ exhibited evidence of reactivity with the Ni-201 matrix made apparent by the cored structure of intermetallics surrounding the additive material. However, no fine distribution of $\eta$ phase was observed in the SZ when observed using HRSEM. Microhardness measurements support HRSEM observations.

6.4.2 Alloy 282

Haynes Alloy 282 was successfully incorporated as a near-surface layer in Inconel 600 using AFSP. Cold wire GTA was used to create a surficial weld overlay layer atop IN600 that was subsequently friction stir processed. AFSP process parameters were varied to tailor the heat input. Processing window extremes (high heat input: 150 RPM/2 IPM and low heat input: 100 RPM/4.5 IPM), identical to parameters used for FSP of IN600, were used for AFSP of Alloy 282/IN600. Similar amounts of Alloy 282, 60-70%, were retained for both high and low heat input conditions. The distribution of Alloy 282, however, differed greatly with AFSP process heat input. Extremely turbulent distributions of Alloy 282 were observed on the AS of the SZ for high heat input conditions. A corresponding accumulation of Alloy 282 was observed on the RS. Samples processed using low heat input parameters also exhibited turbulent flow patterns; however, no large-scale accumulation of Alloy 282 was observed on the RS. Although the Alloy 282-containing regions were macroscopically turbulent, such regions displayed no evidence of local intermixing with IN600 upon examination using EDS. For the processing parameters examined, additive material was intercalated in the IN600 SZ rather than being diluted by mechanically alloying. Overall the solidification structure of the original weld overlay was completely eliminated as a result of AFSP.

Microhardness mapping was used to evaluate the local mechanical properties of the AFSP regions. Alloy 282-containing regions within the SZ exhibited microhardness
values of 275 HV or greater—which is 70 and 90 HV higher than the as-deposited Alloy 282 and IN600 base material, respectively. Although the low heat input parameter combination resulted in lower peak temperatures and higher post-stirring cooling rate, the highest hardness values (some exceeding 300 HV) were observed in this sample. Some of the high hardness SZ regions corresponded to areas containing equiaxed, recrystallized grains with lamellar distributions of fine precipitate particles. Gleeble thermal simulations were performed to replicate the AFSP thermal cycle on as-deposited Alloy 282 weld overlay. Similar clusters of fine $\gamma'$ were observed in the weld overlay when the weld metal was thermally cycled. Such clusters correspond to Ti-rich microsegregation resulting from solidification. Simulations of the high heat input condition resulted in the formation of Ti-rich carbides or carbonitrides in lieu of $\gamma'$ due to the supersolvus nature of the high heat input thermal cycle. Thermal simulations performed in the Gleeble correspond to the observation of considerably fewer $\gamma'$-like precipitates visible in HRSEM.

Grain size of AFSP SZ regions was determined using EBSD. Similar trends observed with FSP of IN600 were also observed with Alloy 282 AFSP. As the process heat input was reduced, the average SZ grain size also decreased. It is attributed to the change in the temperature and strain conditions which affect dynamic recovery and recrystallization processes. Additionally, higher cooling rates and lower peak temperatures associated with lower heat input reduces the window for coarsening processes to be operative. On average, the grain size for AFSP Alloy 282 was 1.5 times smaller compared to FSP IN600. Differences in thermomechanical histories for IN600 and Alloy 282 as well as inherent differences in alloying behavior with respect to dislocation accommodation likely account for the differences in resultant grain size. Significant differences in alloying content (27 total wt% for IN600 vs. 43 total wt% for Alloy 282) affect the stacking fault energy as well as boundary mobility in post-recrystallization coarsening processes.

The heat treatment response of Alloy 282/IN600 was characterized by microhardness measurements and microstructural analysis. Relative to the as-deposited
weld overlay, Alloy 282-containing SZ material exhibited enhanced heat treatment response after a direct age heat treatment at 750°C for 2 hours. Additive FSP SZ hardness was measured to be in excess of 400 HV compared to an average of 305 HV for the as-deposited weld overlay. Again, the low heat input parameter combination showed the greatest heat treatment enhancement response. Electron backscatter diffraction misorientation analyses (KAM and GROD) suggest that the degree of stored energy (i.e., dislocation density) increases as the AFSP/FSP process heat input decreases. Such increase in stored energy as process heat input decreases is attributed to incomplete recovery/recrystallization as well as incomplete recovery of post-stirring deformation. Both FSP conditions examined exhibited higher overall misorientation compared to the as-deposited weld overlay. The increased stored energy suggested by EBSD analyses was verified by STEM micrographs of the Alloy 282-containing SZ. STEM BF micrographs show dense intragranular network of dislocations with some grains exhibiting low dislocation density. A similar heterogeneous distribution of stored energy was observed with GROD EBSD analyses. Material processed with high heat input parameters had considerably fewer dislocation tangles observed in the markedly larger grains. Increased stored energy resulting from AFSP along with homogenization of the deposition material due to high temperature intermixing has the potential to improve productivity and reduce process costs by reducing the time necessary to perform post-processing heat treatments.

6.4.3 Alloy 214

AFSP of Alloy 214 was successfully demonstrated using similar techniques utilized for AFSP of Alloy 282/IN600. As with other portions of the study, processing window extremes (i.e., highest and lowest heat input conditions) were selected. Additionally, double pass low heat input AFSP runs were performed to study the effect of multiple thermomechanical cycles. The distribution of additive Alloy 214 showed similar trends with heat input regarding distribution morphology to Alloy 282; however
the morphology of the Alloy 214 suggest that the degree of material turbulence is lower. Also similar to AFSP of 282, approximately 70-80% of the additive weld overlay was incorporated. However, significantly less additive material (only 44%) was retained after two passes due to significant extrusion of additive material as flash.

The resultant microstructure of AFSP Alloy 214/IN600 shows typical recrystallized microstructure replacing the solidification structure of the initial weld overlay. Interestingly, the resultant grain sizes of AFSP Alloy 214 are nearly identical to IN600 SZ material for the parameters explored. HRSEM microstructural evaluation of selectively etched as-AFSP microstructures revealed no precipitation of $\gamma'$ in the as-processed condition, which is attributed to the more sluggish precipitation kinetics of Alloy 214. The microstructures of weld overlay thermally cycled in the Gleeble to replicate the FSP thermal profile also did not show any evidence of $\gamma'$ precipitation--even for the double thermal cycle.

Microhardness measurements of AFSP Alloy 214/IN600 after direct age heat treatment (750°C for 2 hours) reveals that Alloy 214-containing SZ exhibited microhardness values approximately 50 HV higher than the weld-overlay, which represents a nearly 60% increase in microhardness compared to the as-deposited weld overlay. Peak hardness values for all three conditions were nominally the same; however, the low heat input condition (single pass) appeared to have the largest fraction of high harness (>275 HV) material. This is likely attributed to the larger fraction of retained Alloy 214 within the SZ.

From the results of microstructural examinations and microhardness measurements, significant improvement of the mechanical properties of Alloy 214 surface layers may not be realized using AFSP. However grain refinement and homogenization provided by AFSP holds the potential to improve performance in other areas--notably high temperature oxidation resistance due increased oxide stability. Increased oxide stability is expected to result due to the increase in the relative fraction of high diffusivity pathways associated with increased grain boundary area. Grain refinement of nearly two orders of magnitude compared to the as-deposited Alloy 214
holds potential to increase the formation and stability of protective oxide layers on the surface.
CHAPTER 7: ADDITIVE FRICTION STIR PROCESSING OF HIGH $\gamma'$ - FRACTION NI-BASE SUPERALLOYS

The rich chemistry of high-fraction $\gamma'$ alloys such as Mar-M247 and to some extent René 41 have characteristically high strength at high temperature combined with low ductility. Such a combination makes such alloys prone to defects during fabrication and/or joining. Additive material techniques are not exempt from intrinsic fabricability issues associated with rich superalloy compositions. This is especially true when considering fusion based additive techniques. Although no joining of workpieces is accomplished using fusion-based additive methods, many of the same weldability and metallurgical issues associated with welding still apply. Such issues include segregation during solidification, grain growth, liquation of intermetallic species, grain boundary liquation, solidification cracking, and strain age cracking. All of these issues directly lead to cracking or to degradation of mechanical properties that ultimately can lead to unexpected failure. Additive FSP that includes a fusion-based additive method, has the potential to overcome the formation of fabrication defects and/or metallurgical degradation even for high-$\gamma'$ alloys.

7.1 Additive Friction Stir Processing of Mar-M247 on IN600 Substrates

Mar-M247 is a Ni-base superalloy developed by Martin Marietta Metals as an alloy for use in equiaxed-grained gas turbine engine blades. The rich chemistry (refer to Table 4.2) of this alloy and associated high-temperature strength and low ductility makes fabricability of this alloy quite difficult. Experimental and computed temperature dependent tensile properties are shown in Figure 7.1 and Figure 7.2. Good agreement
exists between the calculated yield strength via JMatPro and data from Bor et al [149] for cast + solution heat treated + aged Mar-M247. For comparison, the strength and elongation values for IN600 are also shown in both figures. The yield strength of the IN600 substrate is considerably lower than Mar-M247; however, the ductility is significantly higher.

Figure 7.1. Temperature-dependent yield stress for Mar-M247 and IN600. Experimental data from Nathal et al. [150], Bor et al. [149], and Harris et al. [151].
Figure 7.2. Elongation to failure for Mar-M247 and IN600. Experimental data from Nathal et al. [150], Bor et al. [149], and Harris et al. [151]

Figure 7.3 shows the typical microstructure of Mar-M247 in cast + aged equiaxed-grained gas turbine blades. The microstructure reveals a very high fraction of $\gamma'$ which is attributed to the approximately 11 wt% $\gamma'$ formers (Al + Ti + Hf +Ta). An equilibrium $\gamma'$ fraction of 69 vol% was determined computationally using JMatPro, which is in good agreement with experimentally determined $\gamma'$ fraction of 62 vol% [151]. Closer examination of the intragranular regions within the casting demonstrates a bimodal distribution of $\gamma'$ (Figure 7.3B). Other microconstituents present within the microstructure are large globular carbides that are predominantly distributed intergranularly (Figure 7.3A).
7.1.1 Deposition of Mar-M247 using LENS

Because of the fabricability issues associated with using the same cold-wire GTAW process used for Alloy 282 and Alloy 214, Laser Engineered Net-Near Shaping (LENS) was used to create additive material depositions on IN600 substrates. Before FSP was performed, the LENS depositions were characterized to gain a better understanding of the initial microstructure. Figure 7.4 shows an etched cross section of the Mar-M247 LENS deposit, approximately 1-1.5 mm in thickness (depending on number of passes performed), residing a machined channel in the IN600 plate. One of the most evident features within the LENS deposit is the presence of numerous cracks parallel to the plate normal direction. Restraint of the deposition by the relatively thick substrate likely exacerbates residual stresses generated by the LENS process. For potential additive material applications of Mar-M247 such as repair operations using LENS, such cracks likely would be undesirable in application. Upon closer examination (Figure 7.4B) the cracks within the deposition are not isolated to a single deposition layer, but rather propagate through several passes. Many of the cracks within the deposition appear to be the result of grain boundary liquation. Backscatter electron
micrographs within the vicinity of the cracks (Figure 7.5) shows the presence of film-like, high-Z structures ahead of the crack tip, indicative of liquation. The morphology of fractographic features in larger cracks also suggests cracking due to liquation. Figure 7.6A shows prototypical ‘egg crate’ fracture surface features that are microscopically smooth. Higher magnification micrographs (Figure 7.6B) of the fracture surface clearly show evidence of terminal solidification products on the fracture surface.
Figure 7.4. A.) Optical micrograph of cross section Mar-M247 LENS deposit on plate surface, B.) cracks in LENS deposition oriented parallel to plate normal direction

Figure 7.5. A.) Backscatter electron micrograph of solidification crack in Mar-M247 LENS deposition, B.) Higher-magnification micrograph of same region shows evidence of liquid films present within crack
The microstructure of as-deposited Mar-M247 selectively etched and examined using HRSEM reveals a heterogeneous distribution of $\gamma'$ (Figure 7.7) corresponding to the rapidly solidified microstructure of the LENS deposit. Interdendritic regions enriched in strongly segregating $\gamma'$ formers have characteristically coarser distribution of $\gamma'$ compared the surrounding regions which exhibit a fine (~10 nm) distribution of secondary $\gamma'$. Small blocky MC-type carbides that range in size 1-2 µm or smaller are also visible throughout the fusion zone. MC-type carbides in Mar-M247 are typically Ta and W-rich [151].

Figure 7.6. A.) Interior of crack within Mar-M247 LENS deposit with characteristic egg crate morphology, B.) solidification products present on the fracture surface of deposition cracks
Figure 7.7. A.) Selectively etched microstructure of as-deposited LENS Mar-M247, B.) heterogeneous distribution of $\gamma'$ morphology showing coarser $\gamma'$ at the solidification subgrain boundaries

Figure 7.8 is a microhardness map of a portion of the Mar-M247 deposition region. The average hardness within the LENS deposition is 424 HV. There does not appear to be any additional hardening of underlying layers from the multiple thermal cycles of subsequent passes by the laser. An approximately 200 $\mu$m wide softened region within the deposition near the initial fusion boundary was observed. Although the LENS process heat input was minimized by utilizing a very high traverse speed (3 m/min), some dilution of the Mar-M247 deposition by the IN600 base material resulted. Energy dispersive spectroscopy line trace near the interface reveals a compositional transition region that corresponds to the softened area where Mar-M247 is locally diluted by IN600 thus promoting the suppression of $\gamma'$ precipitation (Figure 7.9).
Figure 7.8. Microhardness map of the Mar-M247 LENS deposition in the as-deposited condition. Dotted white line denotes original fusion boundary of initial build layer.

Figure 7.9. Energy dispersive spectroscopy line scan across the initial build layer fusion boundary. Line drawn in A.) represents the scan location depicted in B.).
7.1.2 Mar-M247 Additive Friction Stir Process Development

Mar-M247, being designed as an alloy for extremely demanding thermomechanical environments, poses a challenge for AFSP. Even in the as-deposited condition, the hardness of Mar-M247 is significantly higher and the high temperature flow stress is also expected to be significantly higher than the substrate material due largely to the very rich composition. Tabulated experimental data [149, 150] show that the yield strength of Mar-M247 near typical AFSP temperatures (970-980°C) is over an entire magnitude higher than the substrate, IN600 (refer to Figure 7.1).

Because of the resistance of Mar-M247 to high temperature deformation, the resulting AFSP processing window is limited compared to other material systems explored in this work including autogenous IN600, Alloy 282/IN600, and Alloy 214/IN600. Table 7.1 lists the parameter combinations explored for Mar-M247/IN600 as well as tabulated force output. Three different processing parameter combinations were explored. Both ‘low’ and ‘high’ heat input process parameter combinations were explored along with an intermediate parameter combination, referred to as ‘medium’ heat input. The extremes of processing parameter combination represents a nearly ten-fold difference in dimensionless heat input as per equation (4) (section 5.1).

Table 7.1. AFSP processing parameters and extracted force output for AFSP Mar-M247/IN600.

<table>
<thead>
<tr>
<th>Heat Input Designation</th>
<th>RPM</th>
<th>Traverse Speed (IPM)</th>
<th>Dimensionless Heat Input (ω/U^2)</th>
<th>Measured Normal Force (lb_x 10^3)</th>
<th>Measured Travel Force (lb_x 10^3)</th>
<th>Measured Cross Path Force (lb_x 10^3)</th>
<th>Spindle Torque (ft-lb_x 10^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low</td>
<td>100</td>
<td>4.5</td>
<td>4.9</td>
<td>28.8 ± 1.9</td>
<td>8.2 ± 1.2</td>
<td>3.0 ± 0.4</td>
<td>210 ± 11</td>
</tr>
<tr>
<td>Medium</td>
<td>125</td>
<td>3</td>
<td>13.9</td>
<td>19.8 ± 1.2</td>
<td>4.5 ± 0.3</td>
<td>1.7 ± 0.2</td>
<td>187 ± 8</td>
</tr>
<tr>
<td>High</td>
<td>125</td>
<td>2</td>
<td>31.3</td>
<td>14.8 ± 0.5</td>
<td>2.8 ± 0.3</td>
<td>1.1 ± 0.2</td>
<td>175 ± 6</td>
</tr>
</tbody>
</table>
The overall appearance of the plate surfaces after AFSP also shows evidence of limited processability. Lack of consolidation defects were present on the top surface of each ASFP SZ region, increasing in severity as the heat input is decreased. The presence of this defect is common when process temperatures are too low to sustain sufficient material flow such that the region behind the traversing tool is not completely filled [4]. Further increasing AFSP heat input may circumvent defect formation. Interestingly, the formation of lack of consolidation voids is largely beyond the sensitivity of the process force instrumentation. Figure 7.10 shows process force output for a defect-containing segment of an AFSP run. Although the process force output alone would indicate pseudo-steady process conditions, surface defects are clearly apparent after processing.

![Figure 7.10](image)

Figure 7.10. Process force output for high heat input Mar-M247 AFSP demonstrating surface defect formation within 'pseudo-steady state' process force regime.
Measured force output during AFSP shows trends consistent with FSP of IN600 discussed previously. As the AFSP heat input is increased all three measured processes forces decrease, attributable an increase in process temperature with commensurate decrease in flow stress. Spindle torque also decreases in a similar fashion with increasing process heat input.

The tool geometry utilized in this work is such that a large total volume of material is mechanically affected relative to the volume of additive material. Estimations of the additive material fraction relative to the total volume of plasticized material can be performed by considering the dimensions of the tool and additive layer. However, the actual total volume of plasticized material is larger than the projected volume of the FSP. The volume of the additive material represents 27-35% of the total process zone volume—as measured from digital images of cross sectioned AFSP samples. The proportion of additive material relative to IN600 substrate would suggest that process forces relative to autogenous FSP of IN600 should not differ significantly despite the flow stress difference. This, however, is not the case. The significant difference in flow strength for Mar-M247 compared to IN600 causes the measured process forces to be nearly two times higher for Mar-M247 compared to autogenous FSP of IN600. Higher heat input AFSP parameters result in lower measured process forces due the corresponding drop in flow stress for both materials as process temperatures are increased. Additionally, the higher heat input parameters result in a larger SZ nugget volume relative to the tool volume thus permitting the overall SZ nugget behavior during FSP to be dictated more so by the substrate (IN600).

The high process forces associated with AFSP of Mar-M247/IN600 resulted in difficulties in temperature acquisition. High normal forces up to 28.8 ± 1.8 kip (corresponding to tool pressure of up to 45.2 ksi) resulted in plastic deformation near the drilled thermocouple hole and subsequent extrusion of embedded thermocouples away from the target position. While the temperature of AFSP Mar-M247 is expected to be higher than that measured for FSP of IN600 based solely on the higher observed process
forces, the temperature measurement with embedded thermocouples as performed in this study proved to be unreliable.

7.1.2.1 Distribution of AFSP Mar-M247

As with other additive material systems explored, the distribution and retained fraction of Mar-M247 at the plate surface varied based on the AFSP parameters used. Figure 7.11 shows cross sections for all tested parameters etched electrolytically with 10% oxalic acid to distinguish the Mar-M247. It is clear that the distribution of additive Mar-M247 is not only highly dependent on location within the SZ, but also on the processing parameters used. The low heat input conditions (Figure 7.11A) shows large regions near the surface missing. These missing regions represent the lack of consolidation defects (predominantly on the RS) encountered during AFSP.

Figure 7.11. Sample cross sections electrolytically etched with 10% oxalic acid to show distribution of additive Mar-M247 for A.) low, B.) medium, and C.) high heat input parameter combinations.
Another notable feature of the low heat input sample is the lack of plastic flow of Mar-M247 located away from direct influence of the tool shoulder. The region (Figure 7.12) appears to be unaffected mechanically by the tool evident by the observation of the original cracks in the deposition layer and original deposition solidification grain structure. The boundary between the plastically deformed additive material and the remnant undeformed Mar-M247 is a sharp microstructural boundary that represents the prior position of the tool pin feature. This very sharp boundary between plasticized SZ and undeformed HAZ highlights the difficulty of friction stir applicability to high $\gamma'$ fraction Ni-base superalloys using low heat input parameters.

![Figure 7.12. Advancing side of AFSP Mar-M247/IN600 demonstrating sharp boundary between plasticized SZ and HAZ. Dotted line represents projected FSP tool geometry at the pin/shoulder interface.](image)

As the heat input is increased further (‘medium’ heat input 125 RPM ; 3 IPM), the lack of consolidation defects are not as severe nor frequent as the low heat input condition (Figure 7.11B). As was observed for other systems discussed previously, an
accumulation of additive material on the RS relative to the AS is observed. The accompanying deficient AS region shows evidence of highly turbulent flow with streaks of additive material extending to depths greater than the thickness of the original deposition.

Although large near-surface/lack of consolidation voids were reduced significantly by increasing the heat input compared to the low heat input condition, the amount of stirred additive material retained in the SZ is significantly lower for the medium heat input compared to the low heat input parameter combination. Area fraction measurements performed on digital micrographs shows that AFSP using medium heat input parameters retained approximately 45% of the original deposition compared to approximately 70% for the low heat input parameters. It was observed during processing that additive material not incorporated in the SZ was extruded away from the process region in very small discontinuous chunks rather than continuous ribbons of material.

The observation of surface voids was decreased and the degree of additive Mar-M247 material mixing was increased by further increasing the process heat input by using the high heat input parameter combination (125 RPM; 2 IPM). The distribution of Mar-M247 (Figure 7.11C) shows the highest degree of material mixing between the substrate. Consistent with observed trends, the additive material distribution is more chaotic on the AS with the majority of Mar-M247 accumulating on the RS. However, the fraction of material retained on the AS of the SZ is higher for the low compared to the medium heat input parameters. The low heat input parameters retained approximately 83% of the original deposition. The low heat input parameter combination also shows the formation of a thin layer of substrate material on the topmost surface of the center SZ. This layer has been observed in AFSP of Alloy 282 and Alloy 214, and likely results when the degree of material flow beneath the tool is sufficient to cause substrate material to travel vertically to the top of the SZ.
7.1.3 Microhardness Distribution in Additive Friction Stir Processing of MarM247/IN600

Additive friction stir processing parameter combinations not only have a significant effect on the distribution of additive material, but also on the local mechanical properties as measured by microhardness indentation. Figure 7.13 shows microhardness maps of Mar-M247/IN600 AFSP regions for the three parameter combinations used. Within Mar-M247-containing regions, the overall hardness is increased as the AFSP heat input is decreased. The low heat input sample shown in Figure 7.11A clearly shows two distinct SZ regions with different etching response. Within the top, more uniformly etching layer, very hard regions are apparent in the microhardness map. Values in this region are in excess of 750 HV across the width of the AFSP region. The hardness of the topmost AFSP surface layer is nearly double the hardness of the additive material in the as-deposited condition and is nearly four times harder than FSP IN600 produced using equivalent FSP parameters. For comparison, measured hardness values between 450 and 475 HV have been reported for as-cast Mar-M247 material that has undergone solutionization and a double age heat treatment [152]. Similar hardness values between 445 and 455 HV were measured for cast Mar-M247 cooled at 10 ºC/s [153]. The measured hardness values for AFSP Mar-M247 approach levels reported for oxide dispersion strengthened alloys such as MA6000 [51]. This high hardness region is herein referred to as region B. The hardness values observed within Region B of the SZ are comparable to nano-grain oxide-dispersion strengthened superalloy MA6000 which had a reported hardness of 645 HV [51].

The second distinct layer located predominantly in the center stir zone below the topmost region is darker-etching compared to the top layer and undergoes more mixing with the substrate material. This region is referred to as region A. Compared to the as-deposited average hardness 406 ± 14 HV, hardness values within region A are still significantly higher and vary between 500 to 600 HV.
As the AFSP heat input is increased, the overall SZ hardness also decreases. For material processed using medium heat input conditions, maximum hardness values of approximately 625 HV were measured. However the region containing material with high hardness (> 600 HV) is rather small and is located only on the SZ RS in comparison to the low heat input condition. Additionally, the deficiency in additive material on the AS results in a softened region relative to the RS. Increasing the process heat input further (high heat input sample) decreases the peak measured hardness approximately 75 HV compared to the medium heat input sample. However, the fraction of additive material is significantly greater and more uniformly distributed compared to the medium heat input sample. Figure 7.13C shows a relatively uniform AFSP layer with an average hardness of 500 ± 17 HV. The observed topmost layer of IN600 discussed previously was sufficiently thin and therefore was beyond the resolution of the indent grid spacing used. However, this thin IN600 center SZ surface layer is expected to be much softer than the surrounding AFSP Mar-M247.
Figure 7.13. Microhardness distribution for AFSP Mar-M247/IN600 processed using A.) low, B.) medium, and C.) high heat input parameter combinations.
7.1.4 Void Formation/Cracking Healing

Optical microscopy of the AFSP SZ fails to reveal general microstructure; however, the presence some void regions is apparent in optical micrographs for the sample processed using low heat input parameters (Figure 7.14). Although the frequency of cracks after AFSP is lower than the as-deposited additive material, closer examination using SEM reveals two discontinuity types within the additive SZ. Larger discontinuities, predominantly located towards the top of the SZ have fractographic features that are consistent with liquation cracking (Figure 7.14A). Because liquation is not expected during AFSP, especially using low heat input process parameters, these voids are likely un-healed solidification cracks from the original LENS deposition. The 250 - 500 µm cracks near the top of the SZ are observed only when processing material using low heat input parameters. The process parameters used for the low heat input condition, do not achieve temperatures sufficient for the additive material to flow effectively under forging pressure from the tool such that complete crack healing throughout the SZ occurs. While the cracks/voids from the deposition are not completely closed/healed, the total crack length was reduced and orientation altered depending on SZ location as a result of AFSP. Considerably smaller void-like features are also visible within specific locations in the additive SZ. Predominantly equiaxed voids approximately 1-5 µm in diameter are interspersed in the central SZ in regions where very turbulent intermixing between the additive material and substrate occurs. Due to the length scale of these voids it is difficult to ascertain whether fractographic features contained within suggest the origin of such voids is un-healed cracks from the original deposition. More likely, the voids are a consequence of an exhaustion of ductility during stirring for the process temperatures experienced using low heat input process parameters. The distribution the SZ voids appears to be along grain boundaries suggesting that strain concentrated along grain boundaries is responsible for intergranular void nucleation (Figure 7.15). The defects formed during AFSP using low heat input
parameters indicates processing at an extreme within the processing window. However, as the heat input is increased for the medium and high heat input conditions, the frequency of voids encountered in the SZ decreases significantly as the ductility of the additive material and substrate is expected to increase accordingly with the increased processing temperatures. Higher temperatures during additive material during processing for medium and high heat input parameter combinations promotes sufficient material flow to create consolidated SZ regions.

Figure 7.14. A.) Fracture surface morphology of large SZ discontinuities shares fractographic features of hot cracks observed in original LENS deposition, B.) small intergranular voids observed within the SZ. Both micrographs are from regions of a sample processed using low heat input conditions
7.1.5 Material Flow and Distribution of Mar-M247

The as-AFSP microstructure of Mar-M247/IN600 is highly dependent on AFSP parameters as well as location within the SZ. The following sections discuss the various aspects of Mar-M247 microstructural evolution resulting from AFSP.

For Mar-M247/IN600 processed using low heat input parameters, the additive material exists in two distinct regions: Region A.) a subsurface center SZ region with hardness between 500 to 600 HV and Region B.) a surface region that spans the entire SZ width with very high hardness in excess of 750 HV. Figure 7.16 shows the two distinct regions. It should be noted that the brightness differences between the two regions is a result of different etching response, which is highly dependent on the fraction of $\gamma'$. 
Examination of Region A using HRSEM reveals significant intercalation between Mar-M247 and the substrate material (Figure 7.17A and B). While contact between the additive Mar-M247 and IN600 substrate is complex as was observed for Alloy 282 and Alloy 214, complete mixing within the SZ of the two materials was not observed. The complex flow patterns observed in region A are morphologically similar to intercalation patterns and vortices observed in dissimilar FSW combinations including AA2024/AA6061 [154], AA6061/Cu [155], and AA2024/Ag [156]. In addition to dissimilar FSP, similarities in flow pattern appearance exist between the observed dissimilar material flow during AFSP and material flow in individual powder particles produced by mechanical alloying via high energy ball milling [157]. In all previously mentioned studies as well as samples examined in this work, the flow patterns are highly complex and vary significantly with location as well as FSP/W parameters, etc. Additional features visible within the SZ Region A is evidence of tool wear (Figure
7.17B), evident as bright streaks in the BSE micrograph oriented parallel to the plate surface. The majority of tool wear encountered occurred on the AS of the weld. Very little tool wear was observed on the RS.

Figure 7.17. A.) Morphology of additive material flow within turbulent SZ areas contained within Region A, B.) arrows denote regions of tool wear within the SZ of Region A.

Further examination of the bright regions using EDS indicate increased tungsten and rhenium X-ray counts relative to the surrounding alloy (Mar-M247 contains 10 wt% W nominally) as shown in Figure 7.18. Additionally, higher magnification of the tool wear regions reveals the tungsten-rich areas are comprised of discrete submicron wear particle clusters. The presence of tool wear particles for the low heat input sample is attributed to the increased process forces relative to the other processing conditions. Rule [107] has shown similar wear particle formation in the SZ using a FSP tool with identical composition and geometry. Additionally the location of tool wear on the AS observed in the Rule study is also consistent to behavior observed in this work. Presumably, the increased wear debris on the AS is attributed to locally higher temperatures combined
with higher velocity of workpiece material relative to the tool surface. Autogenous FSP experiments by Rule on solid solution-strengthened Ni-base alloys had corresponding process forces ranging from 25 to 30 kip (111 to 133 kN), depending on parameters—such force values are comparable to measured process forces for low heat input AFSP Mar-M247. Because process forces are higher than those experienced during autogenous FSP of IN600 or AFSP of Alloy 282/IN600 and Alloy 214/IN600, tool wear is not observed for FSP/AFSP in these materials. Several studies have shown a similar relationship between the process forces during FSP/W and the threshold behavior and severity of tool wear [15, 158, 159].

Figure 7.18. A.) Backscatter electron micrograph shows tool wear products comprised of individual discrete particles, B.) Corresponding EDS line scan shows increase in W relative to background levels in Mar-M247

Electron backscatter diffraction was used to examine the heterogeneous microstructural distribution within the turbulent center SZ region, Region A. Figure 7.19
shows a generated image quality (IQ) and inverse pole figure map (IPF) for a swirl feature in Region A. Two dark circular regions in the generated IQ/IPF map represent regions of diminished diffraction pattern image quality due to subsurface deformation induced by fiduciary Vickers indents. The scan area of the low heat input additive SZ reveals a unique trimodal microstructural distribution with widely varying grain size. Sub region I (Figure 7.19-I) is band of unmixed IN600 substrate material contains the coarsest microstructure relative the surrounding microstructure. The microstructure in terms of grain morphology and size is consistent with the microstructure of autogenous IN600 discussed previously. The second region, denoted as sub region II (Figure 7.19-II) represents an interpenetrating band of the additive Mar-M247 with considerably smaller grains than those found in Region I. Average grain size within this region measured from EBSD is 1.4 \( \mu \)m—considerably lower than sub region I. Etching characteristics of the fine-grained microstructure of areas like sub region II are poor using the \( \gamma' \) matrix specific etch. Rather, a \( \gamma' \)-selective etch was used to reveal the microstructure. Figure 7.20A and B shows the microstructure of sub region II in the un-etched and \( \gamma' \)-selective etched conditions, respectively. The un-etched specimen shows channeling contrast along with bright particles corresponding to fine carbides. When the sample is etched, information regarding grain size and general grain structure/morphology becomes ambiguous; however, the small pits/holes on the sample surface indicate prior sites of \( \gamma' \). It is clear that a high-fraction of \( \gamma' \) exists within this fine-grained area of the SZ, although the morphology of the \( \gamma' \) is harder to interpret using this etch. Digital image analysis of etched micrographs indicates an average \( \gamma' \) size of 25 ± 8 nm within this region.
Figure 7.19. Electron backscatter diffraction IQ/IPF map demonstrating the unique trimodal microstructure of SZ Region A. Region A is comprised of three sub regions: I, II, and III.
The EBSD scan of the trimodal Region A also revealed a third sub region, denoted as region III (Figure 7.19-III). Unlike the sharp interface between regions I and II, region III appears to be a region of intermixing between the substrate and additive material. Intermixing within this sub region will be discussed further in following sections within the document. The microstructural refinement in this sub region is between the two extremes observed in sub regions I and II. Like the fine-grained region II, γ’ formation is observed within region. Micrographs of etched specimens from this region (Figure 7.21) show the presence of a uniform distribution of spherical γ’ with an average size of 24 ± 6 nm. The formation of γ’ occurs during the AFSP thermal cycle with no additional heat treatment. Compared to the as-deposited Mar-M247, the distribution of γ’ is considerably more homogenous due to the complete elimination of solidification structure and associated microsegregation (refer to Figure 7.7). Also present within this region of the microstructure are blocky, equiaxed constituent particles,
likely MC-type carbides. The distribution of carbides within this region does not appear to be as dense as in sub region II. Interestingly, blocky carbides observed in the as-deposited condition survive AFSP with minimal change in size and morphology. Vigorous stirring of material has been shown to break down and refine constituent particle size [70], however the carbides present in this material are likely too small (approximately 1-2 µm or smaller) to be effectively further communuted.

![Image](image.png)

Figure 7.21. A.) Selectively etched (γ-matrix) SZ region typical of sub region III, B.) Characteristic uniform distribution of fine secondary γ'

The complex flow patterns observed in Region A were not observed near the top of the SZ in Region B (refer to Figure 7.16). The material within this region has characteristically very high hardness (>700 HV) and a microstructure that cannot be interpreted using a γ matrix-selective etch. Rather, a γ'-selective etch was used to attempt to reveal the microstructure. Figure 7.22 shows a representative micrograph of the SZ within Region B. The etched microstructure suggests a high fraction of γ'; however other microstructural features such as grain size are not easily distinguished.
Electron backscatter diffraction was used better understand the grain structure within Region B.

Figure 7.22. HRSEM micrograph of selectively etched ($\gamma'$ specific) sub region III demonstrating high fraction of $\gamma'$ however few other microstructural features are revealed.

Figure 7.23 shows an EBSD-generated inverse pole figure map for Region B. The map clearly shows a uniform distribution of sub-micron grains produced using low heat input AFSP parameters. The average grain size for the measured scan region is 340 nm. Other materials in this study have been processed using identical FSP/AFSP parameters (100 RPM; 4.5 IPM); however, only Mar-M247 produced nano-scale grains. The high measured SZ hardness in Region B can be attributed to Hall-Petch strengthening due to the ultra-fine grain structure. Fine, but not nano-scale, grains (1-2 $\mu$m) in the upper portion of the SZ near the surface was also observed by Barabash et al [58] in the only FSP study of a high $\gamma'$ content superalloy, IN 738 (6.9 wt% Al + Ti compared to 11% nominal Al + Ti + Hf + Ta for Mar-M247). Within the middle and
near the bottom of the SZ, the average grain size increased and was comparable in size (2-5 µm) to the grain size observed within sub region III of Region A in this work. Tool and workpiece dimensions used by Barabash were similar to those used in the current work; however, the FSP parameters used (150-200 RPM; 0.5 IPM) resulted in a considerably higher heat input compared to the low heat input condition examined here. The higher heat input used by Barabash and the inherent alloy composition-dependent differences in precipitation behavior of IN738 compared to Mar-M247 likely contributed to the coarser near-surface grain size. Following sections in this document elucidate the mechanism by which the wide range of SZ microstructures forms in AFSP Mar-M247/IN600.

Figure 7.23. Inverse pole figure map of ultra-fine grained region characteristic of the near-surface SZ microstructure comprising Region B.
Scanning transmission electron microscopy of Region B reveals submicron grains as was observed via EBSD. Similar to AFSP Alloy 282/IN600, the additive material-containing SZ regions demonstrate a heterogeneous distribution of grains with varying internal dislocation density (Figure 7.24A). Some grains (Figure 7.24B and C) show very dense networks of dislocation tangles suggesting incomplete recrystallization or unrecovered warm work post-stirring. Also visible within the microstructure is the presence of very fine (<100 nm) equiaxed particles. Such particles are high Z due to the contrast formation characteristics in HAADF STEM (Figure 7.24C). These particles are likely Ta- and W-rich carbides that are expected to be present in Mar-M247. It should be noted that γ’ was not observed in the sample, although it is expected to be present.
7.2 Evolution of Mar-M247 AFSP Stir Zone Microstructures

The evolution of unique SZ microstructures discussed previously in high-\(\gamma'\) content Ni superalloys is attributed to supersolvus deformation and recrystallization during AFSP. The following section presents a mechanistic model by which this process occurs. Figure 7.25 illustrates a schematic representation of the expected temperature,
flow stress, and microstructural evolution of the AFSP process for a Ni superalloy. Up to temperatures of ~700-750°C superalloy flow stress is high due to anomalous strengthening of γ’ therefore creating a corresponding increase in heat generation due to increased process forces. At point A in Figure 7.25, process temperatures ahead of the tool are such that the material flow (yield) stress is high. The elevated flow stress leads to an increase in heat generation and locally raises process temperatures until the material is above γ’ solvus at point B where the tool directly interacts and stirs the material. At this point, strength of the superalloy is low due to the supersolvus process temperatures. At point C, the tool has passed and the material begins to cool. Coarsening of the recrystallized microstructure formed at point B occurs while still in the single phase γ temperature range denoted on the diagram as point C. When the material cools sufficiently below the γ’ solvus temperature (point D), the SZ grain size essentially remains constant for the rest of the thermal cycle when coherent γ’ acts to pin grain growth. The following sections discuss in detail the microstructural evolution operative during AFSP of high-γ’ fraction superalloys.
7.2.1 Supersolvus Deformation During Stirring

The additive superalloy is heated ahead of the approaching tool. The superalloy additive material resists deformation at elevated temperature when below $\gamma'$ solvus temperature due to the presence of strengthening precipitates. For the case of LENS deposited Mar-M247, the deposited material already contains a fine distribution of $\gamma'$ which is attributed to the multipass nature of the LENS deposition as well as the inherent tendency for the alloy system to form $\gamma'$. As a result, the high temperature yield stress is expected to be much higher than the substrate alloy. The higher high-temperature flow stress of Mar-M247 relative to IN600 is evident in the increased process forces measured.
during AFSP (see Table 7.1 in section 7.1.2). Although successful temperature measurements during AFSP of Mar-M247 were not obtained, the temperatures during AFSP are expected to be higher than FSP of IN600, AFSP of Alloy 282, or Alloy 214. The self-regulating nature of the FSP process dictates that increased flow stress will generate correspondingly higher process temperature. The heat generated \( q \) by a rotating tool shoulder is expressed using the following equation [102]:

\[
q = \frac{2\pi}{3} k \omega R_s^3
\]  

(17)

where \( k \) is the product of temperature dependent shear yield stress and the friction coefficient, \( \omega \) is the tool rotation rate, and \( R_s \) is the tool shoulder radius. The above equation is similar to the heat generation equation (see equation (4) in section 5.1) used by FSWS, except that the term, \( k \), replaces the product of the friction coefficient \( (\mu) \) and tool downforce pressure \( (p) \). Both equations assume heat generated occurs when material interacts with the shoulder by a slipping mechanism. Although in reality material interacting with the rotating tool interacts in a combination of sticking and slipping conditions [102], the above equation can still be utilized for demonstrative purposes. The significantly higher shear yield stress (for most metals approximately 0.5 \( \cdot \sigma_{YS} \)) of Mar-M247 relative to IN600 will lead to a proportional increase in heat generation. A significant drop in shear yield stress does not occur until near or above the \( \gamma' \) solvus temperature. The self-regulating nature of the FSP process is such that a corresponding increase in heat generation will continue until the material is supersolvus and single phase at which point the yield stress is lowest. It is therefore expected that stirring and hence recrystallization occurs within this temperature regime.
7.2.2 Recrystallization and Boundary Mobility in AFSP of High-γ’ Fraction Superalloys

Dynamic recovery and recrystallization processes characteristic of FSP/W are operative while the single-phase material is being deformed. An important consequence of recrystallization while single-phase or quasi-single phase (such is the case with superalloys very near or above the γ’ solvus temperature) is relatively uninhibited grain boundary mobility. Recrystallization within the two phase region (γ + γ’) for a high-γ’ fraction Ni-base superalloy is inhibited as the presence of coherent γ’ precipitates restrict the movement of mobile grain boundaries. While the number of friction stir studies dealing with high γ’ fraction superalloys is extremely limited, such particle-inhibited recrystallization during FSP/W of superalloys has been not been documented. However, the inhibition of recrystallization has been reported during FSW of dual-phase brass alloys where relatively high fractions of incoherent β’ stable at processing temperatures retarded complete dynamic recrystallization in the SZ [160].

A mobile grain boundary, such is the case during recrystallization during stirring or grain coarsening occurring while cooling behind the FSP tool, can experience a pinning force that causes a ‘drag’ on the moving boundary. Precipitates can exhibit a pinning force, which is quantified by Zener [161]. The pinning force per unit area of grain boundary is expressed as:

\[ P = \frac{3f\gamma_{GB}}{2r} \]  

(18)

where \( f \) is the volume fraction of precipitates, \( \gamma_{GB} \) is the grain boundary interfacial energy, and \( r \) is the radius of the precipitate. As per the above equation, the pinning will increase with either a decrease in pinning particle size or increase volume fraction of particles. However, the pinning force per unit grain boundary area as predicted by eq (18) only considers a pinning particle with no coherency or solubility of the particle with the matrix (e.g. Al₂O₃ particles in an aluminum matrix) [162]. Precipitate-strengthened systems
such as Ni-superalloys rely on the formation of γ' which for most conditions maintains coherency with the matrix. The coherency of γ' within the γ matrix makes the precipitates more effective relative to incoherent particles with respect to inhibiting grain boundary motion.

Figure 7.26 shows a mobile high angle grain boundary bypassing coherent spherical particles. The loss of coherency of the particle/matrix boundary requires additional boundary energy as it changes the boundary type from a low energy (coherent boundary) to a high energy boundary (incoherent boundary). As per Doherty et al. [163], the equation for the pinning force per unit grain boundary energy for coherent particles is modified to:

\[
P_{coh} = \frac{6f(\sigma_{incoh} - \sigma_{coh})}{r}
\]

(19)

where \(\sigma_{coh}\) and \(\sigma_{incoh}\) are the particle/matrix interface energies for coherent and incoherent boundaries, respectively. However because \(\sigma_{coh} \ll \sigma_{incoh}\) the difference between these energies is then approximated to \(\sigma_{incoh}\), which is approximately the value for grain boundary energy, \(\sigma_b\). One important ramification of the above equation is that coherent precipitates such as γ' are approximately four times more effective than an incoherent particle at restricting grain boundary motion. A generally accepted ‘threshold’ for the inhibition of recrystallization via Zener pinning is if \(f/r\) is greater than 0.2 \(\mu m^{-1}\). For the case of a high fraction γ' superalloy such as Mar-M247 the fraction of γ' can be very high (\(f = 0.5\) or higher) and depending on the thermal history the γ' precipitates can be very small (20 nm). Assuming a typical value of 750 mJ·m\(^{-3}\) [164] for a random high angle grain boundary in a Ni superalloy, eq (19) predicts a value of 112.5 MN·m\(^{-2}\) (112.5 MJ·m\(^{-3}\)) pinning force. This value is significant relative to typical values (10\(^5\) J·m\(^{-3}\) [21]) for stored energy in a lightly deformed alloy.

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Although $\gamma'$ precipitates are inhibitors to grain boundary motion and subsequently recrystallization, several studies have investigated subsolvus recrystallization in several high-$\gamma'$ fraction superalloys including Nimonic PE16 [162, 164], Udimet 720 [165], and Nimonic 115 [163]. Such studies show, despite pinning by coherent $\gamma'$, boundary motion and recrystallization is possible. There are several proposed mechanisms by which a mobile grain boundary can move through a region containing $\gamma'$. These mechanisms can include:

i.) A mobile boundary encounters and subsequently dissolves the $\gamma'$ precipitate, thus removing the barrier to boundary motion. Dissolution of the $\gamma'$ precipitate leads to supersaturation of solute along the mobile grain boundary and precipitation of a coherent particle via discontinuous precipitation occurs behind the mobile boundary [166].
ii.) Coherent $\gamma'$ pins a boundary and coarsens due to solute channeling along the grain boundary. This coarsened particle functions to nucleate new grains via particle stimulated nucleation about the original grain boundary [165].

iii.) A mobile grain boundary cuts a coherent particle and causes reorientation such that coherency is maintained in the new recrystallized grain. This possible mechanism has been observed in Cu-Co systems, but has been rarely observed in Ni-base superalloy systems due to the kinetic barrier associated with renucleating $\gamma'$ in the new orientation associated with the new recrystallized grain [163].

iv.) A mobile boundary comes in contact with a coherent particle and halts motion locally. The continued movement of the boundary causes bowing near the particle and loops around the particle. This has been previously shown schematically in Figure 7.26. Once the moving boundary bypasses the particle, it retains its original orientation and is no longer coherent with respect to the new recrystallized orientation. This mechanism of particle bypass is observed commonly with incoherent particles; however it is rarely observed with coherent particles [165]

Microstructural evidence is used to elucidate plausible operative mechanisms of subsolvus recrystallization in high-$\gamma'$ fraction superalloys. For mechanism i., reprecipitation of $\gamma'$ behind the mobile boundary (known as discontinuous precipitation) leads to the formation of large, irregularly shaped $\gamma'$ particles behind the boundary (Figure 7.27A). Observations of selectively etched Mar-M247-containing SZ material does not show any morphological evidence of discontinuously precipitated $\gamma'$ along grain boundaries. As shown previously (Figure 7.21), the $\gamma'$ near grain boundaries is similar in size and spherical morphology as intragranular $\gamma'$.  

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Furthermore, the bimodal grain size distribution characteristic of mechanism ii. in which small recrystallized grains associated with the formation of a fine-grained ‘necklace’ form around the original grain boundary was not observed within the SZ. An example of a microstructure in a Ni-base superalloy resulting from this mechanism is shown in Figure 7.27B. However, this mechanism of particle-stimulated nucleation is more often reported operative for static recrystallization rather than for the case of dynamic recrystallization which is expected for AFSP/FSP [168]. For AFSP Mar-M247, grain size distributions measured using EBSD of ultra-fine grained ($d_{avg}= 0.34 \, \mu m$) Region B and the coarser-grained ($d_{avg}= 2.9 \, \mu m$) sub region III from Region A both show normal, non-bimodal distributions (Figure 7.28A and B, respectively). Grain size
analyses indicating normal distributions suggest that new, fine recrystallized grains are not nucleating from $\gamma'$ pinning events below the $\gamma'$ solvus temperature further supporting the case for supersolvus recrystallization.

Figure 7.28. Electron backscatter diffraction derived grain size distribution for A.) ultra-fine grained Region B and B.) sub region III from Region A

Based on microstructural observations, subsolvus recrystallization via mechanism iv. is also not believed to be operative during AFSP. Loss of $\gamma'$ coherency due to a
mobile boundary would lead to an intragranular distribution of γ’ in the new grain that would be incoherent. Figure 7.27C shows an inverse pole figure/image quality map for Udimet 720 [165] of a recrystallization front moving from the blue-shaded grain into the red-shaded grain. Bowing and subsequent formation of a tortuous grain boundary observed in the IQ/IPF map results from pinning by γ’. When the boundary moves past and forms a loop around the pinning particle, it becomes incoherent with respect to the new grain (blue) and retains the original grain orientation (red). High-resolution EBSD analysis of the SZ material suggests subsolvus recrystallization via this mechanism does not occur. Figure 7.29 shows an inverse pole figure for Mar-M247 SZ material using a small step size of 80 nm.

Figure 7.29. A.) inverse pole figure map of AFSP Mar-M247 microstructure (100 RPM; 4.5 IPM) typical of sub-region III. B.) corresponding confidence index map
Although the scan step size (and likely the lateral resolution of the beam [169]) is larger than much of the secondary γ’ observed in the microstructure (Figure 7.21), the corresponding confidence index map (Figure 7.29B) indicates a rather uniform distribution of intragranular CI values. The presence of incoherent γ’, even if smaller than the probe diameter, would lead to the formation of overlapping patterns from multiple orientations. For a particular scan point, overlapping diffraction patterns will lead to ambiguity in the automated indexing of the collected pattern. Such uncertainty with indexing results in correspondingly low CI values. The lack of small, local intragranular high angle misorientations combined with uniform intragranular CI values nearing unity supports the original assumption that γ’ present in the examined Mar-M247 microstructures is coherent with the γ matrix.

None of the typical mechanisms for Ni-base superalloys characteristic of moving recrystallization fronts bypassing coherent γ’ appear to be operative for any of the microstructures observed in AFSP Mar-M247. Based on this microstructural evidence, one can conclude that the recrystallization process during AFSP occurs at temperatures at which boundary mobility is not impeded by coherent γ’. Therefore it can be concluded that the dynamic recrystallization process operative during stirring occurs at supersolvus temperatures.

### 7.2.3 Post-Stirring Microstructural Evolution

Removal of mechanical energy input when the FSP tool passes leads the cessation of dynamic recrystallization and subsequent cooling of the additive material. While still at elevated temperature, driving force will exist for boundary migration and grain coarsening of the fine grains formed during dynamic recrystallization. As the additive superalloy material cools below the γ’ solvus temperature, precipitation of fine, coherent intragranular γ’ will occur depending on the local cooling rate and local concentration of γ’-promoting elements. Mobile grain boundaries associated with grain growth are pinned by the coherent γ’, essentially ‘locking-in’ grain size during further cooling.
Highly effective immobilization of mobile grain boundaries upon the formation of γ’ during cooling is a critical factor in the formation ultra-fine SZ grains observed in the near-surface SZ (Region B) of AFSP Mar-M247. Because deformation and dynamic recrystallization occurs in a relatively narrow temperature regime for rich alloys such as Mar-M247, the possible duration for very fine recrystallized grains to coarsen after the tool has passed is very limited. It is proposed that this duration is function of the AFSP processing parameters, i.e., heat input and its effect on the cooling rate. Additionally, the local composition with respect to γ’-promoting elements has a large effect on the onset of γ’ pinning and therefore the degree of SZ refinement. Regions within the SZ that undergo intermixing with the substrate essentially become diluted by mechanical alloying and the local concentration of γ’-promoting elements is decreased. This dilution has two consequences:

1.) Dilution of the high γ’-fraction additive alloy with the solid-solution strengthened substrate depresses the γ’ solvus temperature and increases the temperature range in which single-phase γ is stable

2.) Dilution reduces the relative amounts of γ’-promoting elements and retards the kinetics of γ’ formation, i.e., longer times are necessary for precipitate formation. Practically, the formation of γ’ will be suppressed for faster cooling rates.

Figure 7.30 shows thermodynamic computational prediction using JMatPro of the variation of the single phase field (γ) temperature range as additive Mar-M247 is diluted with increasing amounts of IN600. The thermodynamic prediction clearly demonstrates that as the Mar-M247 becomes diluted by IN600 the solvus temperature for γ’ is depressed relative to the solidus temperature and the temperature range for single phase γ is broadened. The extent of grain coarsening occurring post-stirring while still in the single phase γ temperature range is likely increased with larger temperature ranges.
associated with increased duration required to cool through this range allowing boundary migration to be operative until pinning by $\gamma'$ occurs.

![Figure 7.30](image.png)

**Figure 7.30.** Computational thermodynamic prediction of the recrystallization and grain growth temperature range for Mar-M247 as a function of dilution by the IN600 substrate as predicted by JMatPro.

The extent of post-recrystallization grain growth occurring in FSP/AFSP Ni materials can be observed by noting the fraction of annealing twins in the microstructure. For materials with low to intermediate stacking fault energies, post-recrystallization grain growth results in the formation of annealing twins. The idea put forth by Gleiter suggests the formation of coherent twins is by a ‘growth accident’ [170]. Figure 7.31 illustrates the atomistic model of the ledge structure at the interface of a mobile grain boundary. In this model, the model at the bottom (grain I) is growing in the x direction into grain II. This grain growth in grain I by the formation of new (111) ledges requires atoms to be
supplied from ledges \(a, b, c, d, e, f\), and \(g\) from grain II to be added to locations \(h, i, j, k, l, m\) in grain I. Eventually a new (111) layer, o-p, will be added in grain I. With regard to the stacking sequence of an fcc crystal, the new layer, o-p, has atoms that can either occupy A or C sites assuming that the preceding layer has atoms in B sites. If layer o-p atoms go to C sites, the normal fcc stacking sequence, ABCABCABCA..., will result. However if atoms in layer o-p occupy A sites, the stacking sequence will locally be ABA, which represents a coherent twin boundary due to the interruption in the normal fcc stacking sequence, i.e., a stacking fault. This model was modified slightly by more recent studies. Like the Gleiter model [170], a model proposed by Mahajan et al [171] also envisaged twin formation due to the growth accidents on \{111\} steps. However, the authors modified the Gleiter model to include the nucleation of Shockley partials during the propagation of \{111\} steps via growth accidents as is shown in Figure 7.31. These partials glide away from each other (due to repulsive forces) to form a coherent twin. This model explains why high SFE materials such as aluminum do not form annealing twins. The stability of Shockley partial loops is inversely proportional to the SFE.

Figure 7.31. Schematic of ledge structure at a grain boundary. Each ledge is formed by the termination of \{111\} planes at the boundary. Figure adapted from Gleiter [170].

The angular misorientation associated with the formation of coherent twin boundaries (\(\Sigma3\)) can be experimentally determined. Unambiguous measurement of twin
boundary fraction is straightforward via EBSD. Because post-recrystallization grain coarsening in low SFE (such as those alloys in this study) leads to the formation of annealing twins, it can be postulated that FSP/AFSP materials exhibiting high twin fractions results from significant uninhibited grain growth. For an alloy such as Haynes 282, the significantly lower fraction of γ’ formers results in no fine secondary γ’ formation upon cooling during AFSP. The lack of coherent γ’ to restrict grain growth leads to the formation of a coarser as-AFSP grain structure with significantly higher twin fraction resulting from grain growth. Figure 7.32 shows the grain boundary misorientation distribution for two regions of AFSP Mar-M247 and AFSP Haynes 282 processed using identical AFSP parameters. All these samples exhibit similar distributions of misorientation except for the misorientation angles associated with Σ3 boundaries (approximately 60º). The measured Σ3 fraction for Mar-M247 was 5% and 9% for the ultra-fine grained Region B and coarser grained sub-region III from Region A, respectively. The increased twin boundary fraction of sub-region III is associated with increased extent of grain growth post-recrystallization. In contrast, the measured twin fraction for Alloy 282 was significantly higher--nearly 33%. Presumably higher boundary mobility expected for Alloy 282 due to the lack of boundary pinning phases forming after AFSP led to the formation significantly higher twin fraction. It should also be noted that the SFE of Alloy 282 is lower than Mar-M247 (118 vs. 144 mJ·m⁻² @ 20ºC, respectively as calculated using JMatPro) therefore assuming no boundary mobility inhibition, Alloy 282 should form annealing twins more easily due to the increased stability of Shockley partial loops [171]. However, it is not known whether stacking fault energy alone would account for such a large difference in the measured twin fraction.
Figure 7.32. Grain Boundary misorientation distribution for two regions in AFSP Mar-M247 with different final microstructures processed using low heat input (100 RPM; 4.5 IPM) parameters. The distribution for FSP IN600 processed using the same parameter combinations is also shown.

### 7.2.4 Dilution and Intermixing during AFSP

As mentioned previously, the onset of the precipitation of boundary-pinning \( \gamma' \) and subsequent cessation of grain growth is also influenced by the amount of \( \gamma' \)-promoting elements. As the relative amounts of \( \gamma' \)-promoting elements is decreased with increasing levels of dilution, computationally derived CCT diagrams for \( \gamma' \) show that slower cooling rates and additional undercooling are required for the onset of precipitation (Figure 7.33). Although the absolute accuracy of these generated CCT diagrams has not been validated experimentally, the general predicted behavior of decreased tendency for rapid \( \gamma' \) formation as the Al + Ti (and other \( \gamma' \) promoting...
elements) content decreases is expected. Such response is well-known and has been studied extensively and applied within the realm of other phenomena such as the strain-age cracking [45]. The susceptibility to strain age cracking is highly dependent on the rate at which \( \gamma' \) forms. Prager and Shira [172] showed that susceptibility was highly dependent on alloying content in that alloy compositions rich in \( \gamma' \)-promoters (Ti+ Al) leading to an increase in the rate of \( \gamma' \) precipitation and thereby increase in strain age cracking susceptibility.

For very high dilution levels (>50%), the formation of \( \gamma' \) become sufficiently sluggish that precipitation formation is not expected for expected cooling rates for the
AFSP process based on aforementioned simulations. Mechanical work associated with stirring may have an effect on precipitation kinetics as discussed in previous sections in the document. However, highly diluted regions (>50%) regions are observed as γ′ denuded regions within the microstructure (Figure 7.15). As a result of the lack of γ′ formation, grain size formed is not influenced by coherent particle pinning and is dependent on the extent of coarsening after deformation by the FSP tool upon cooling.

Intermixing (i.e., dilution) of the additive alloy was not observed in other systems such as Alloy 282/IN600; however, some dilution of the Mar-M247 was measured using EDS for turbulent regions within Region A in the SZ. These regions are remote from the surface and are located within close proximity to the substrate. The significantly richer chemistry of Mar-M247 relative to IN600 allows the distribution to be observed using BSE micrographs. Figure 7.34A shows the distribution of Mar-M247 within a turbulent Region A-type area of the SZ. The gradients in image contrast are directly attributed to Z-contrast. Mar-M247-rich regions contain a relatively higher concentration of high-Z elements (e.g., W, Ta, Hf) and therefore appear brighter. Regions of intermediate contrast represent areas in which intermixing with the substrate have occurred. Energy dispersive spectroscopy line scans within the turbulent region (Region A) reveals the relative concentration of elements contained in Mar-M247 corresponds to the Z-contrast behavior observed in the BSE micrograph (Figure 7.34A). Because the alloy systems contain several different elements, only Al and Co (γ′-promoting elements in Mar-M247 [173]) are shown on the plot for simplicity. The fractions of the aforementioned elements differ significantly between the additive and substrate alloy. Figure 7.34B shows the concentration of Co and Al across a line scan in the microstructure. Regions along the line scan (approximately 0 to 50 μm) that intersect interleaved flow bands with high dilution show a concomitant decrease in the relative concentration of Al and Co. Elsewhere across the scan, the concentration of Co and Al nears the nominal values (10 and 5.5 wt%, respectively) for the Mar-M247 base material. Although this region of the scan (approximately 50-175 μm scan distance) is richer in Co and Al relative to the high dilution regions, the concentrations are overall lower than those measured within the
ultra-fine grained Region B near the surface. The EDS measurements suggest that even in Mar-M247-rich areas of the turbulent Region A, the dilution of Mar-M247 is on the order of 10-20%. Computational thermodynamic predictions indicate dilution level of only 20% Region A expands the single phase $\gamma$ stability temperature range by 106ºC. Additionally, the local decrease in $\gamma'$-promoting elements resulting from the dilution also has the effect of requiring slower cooling rates with additional undercooling for the onset of $\gamma'$ precipitation—the formation of which terminates boundary migration i.e., grain coarsening. The aforementioned effects of dilution combined with slower cooling rates expected for the interior regions of the SZ remote from the surface accounts for the overall distribution of larger grain sizes observed in Region A relative to the near-surface Region B.

Figure 7.34. A.) Backscatter electron micrograph of EDS line trace within turbulent SZ region A, B.) standardless quantification of $\gamma'$-promoting elements (Al and Co) showing leaner composition relative to the near-surface SZ of Region B.
It has been shown that stirring and dynamic recrystallization occurs within a temperature range of single phase (γ) stability. The width and duration spent within the single phase field temperature range has an effect on the final observed grain size. Under equilibrium conditions, this temperature range depends solely on the compositional dependence of the γ’ solvus temperature. Kinetic considerations with regard to the cooling rate also plays a role in the formation of γ’ upon cooling below the solvus temperature. However, other variables that play a role in microstructural evolution, namely the total strain and strain rate, are not able to be unambiguously determined. Despite the inherent heterogeneous strain and strain rate distributions within the SZ that cannot be accounted for, the compositional effects on process temperature and post recrystallization microstructural evolution have been shown to play a significant role in the final microstructure.

7.3 Additive Friction Stir Processing of René 41 on IN600 Substrates

René 41 is a precipitation-strengthened wrought Ni-superalloy developed in the 1950’s for use in high temperature applications where considerable strength around 800 °C is required [174]. An estimated 30,000 lbs. of René 41 was utilized for the manufacture of F-1 rocket engines and rocket engine turbopumps used in the first stage of the multistage Saturn V launch vehicle [174]. The amount of γ’-promoting alloying additions in René 41 (nominally 4.7 wt % Al + Ti) is considerably less than Mar-M247 (nominally 11 wt% Al + Ti + Hf + Ta) therefore it is generally accepted that fabricability/weldability of René 41 is less problematic. However, René 41 is still prone to fabrication and weldability issues namely strain age cracking, discussed in following sections [45, 172, 175, 176].

Additive FSP René 41 surface layers were created using similar techniques used for AFSP of Mar-M 247—laser depositions were created using the LENS process on IN600 substrates and subsequently friction stir processed. Unlike Mar-M247, for the deposition conditions used (refer to Table 4.3 in section 4.2.2.3), no cracking of the
deposit was observed. Figure 7.35 shows example HRSEM micrographs of selectively etched René 41 laser deposition material exhibiting solidification microstructure with very fine features characteristic of the rapid cooling rates expected for the LENS process. As with Mar-M247, a rather heterogeneous distribution of very fine $\gamma'$ is observed which is consistent with the expected microsegregation during solidification. Interspersed within the $\gamma'$-containing interdendritic regions are fine carbides (0.5-1µm) with globular or slightly script-like morphology. Such microstructures formed during rapid cooling are consistent with experimental observations by other researchers [177]. A microhardness map (Figure 7.36A) of the deposition layers indicates that hardness within the deposit is fairly uniform (average $= 285 \pm 12$ HV). Consistent with hardness trends corresponding to the $\gamma'$ former content, the hardness of as-deposited René 41 is approximately 100 HV higher than Alloy 282 and Alloy 214, but also approximately 100 lower than as-deposited Mar-M247.

![Figure 7.35. HRSEM of selectively etched laser-deposited René 41. Bright regions in (A) correspond to interdendritic regions (B) containing very fine $\gamma'$ precipitates.](image)
Figure 7.36. Microhardness map of A.) as-deposited René 41 on IN600 substrate and B.) AFSP René 41 surface layer on IN600 substrate. C.) Corresponding macrograph of AFSP René 41 cross section

Only one combination of AFSP process parameters was evaluated due to the limited availability of laser deposited René 41 substrates. In an attempt to create the highest degree of grain refinement (as with the other alloys evaluated), low heat input AFSP parameters were selected. A spindle speed of 125 RPM and tool traverse speed of 4.5 IPM was utilized. Figure 7.36C shows a photograph of the resulting René 41-containing AFSP plate. Clearly visible on the SZ top surface is continuous lack of consolidation defect that runs down the center of the SZ. Although, lower alloying content than Mar-M 247, the high temperature ductility is low and yield strength is expectedly high. Figure 7.37 shows high temperature mechanical properties gathered for
René 41. However, it should be noted that the mechanical property values presented represent material that has undergone appropriate heat treatment cycles. The lack of solutionization combined with the suppression of strengthening precipitate formation due to the rapid thermal excursions associated with the laser deposition process results in a reduction in additive material yield strength and likely an increase in elongation. The process force output for René 41 AFSP shows process forces that are more similar to autogenous FSP of IN 600 (see Figure 5.10, section 5.2) rather than the extremely high process forces of Mar-M247 (see Table 7.1, section 7.1.2). Interestingly the observation of lack of consolidation voids on the SZ top surface do not correspond to measureable process force instabilities. The size of the instability observed relative to the entire process volume likely produces a process force ‘signature’ which is below the resolution limits of the machine used in this work.

![Figure 7.37. High temperature mechanical properties for IN600 and René 41. René 41 material solution heat treated at 1066ºC, aged at 760 ºC for 16 hours followed by air cooling. Data replotted from [105] and [178].](image)
The pseudo steady state normal force of 14.2 ± 0.4 kip (62.3 ± 1.8 kN) is comparable to IN600 processed using similar parameters. Travel force and cross path force, however, are approximately 50% lower than values measured for equivalent FSP IN600 runs. Very slight differences in plunge depth between the two runs (0.040 in. for FSP IN600 vs. 0.035 for AFSP René 41) likely are responsible for the differences. It should be noted that the torque values measured for René 41 are not comparable to values obtained for IN600 or Ni-201 because of a change in torque load cell. Figure 7.38 shows process force output for AFSP René41. A consequence of the relatively low process forces is the lack of observable tool wear within the SZ which is consistent with FSP of IN600 and AFSP of Alloy 282 and 214 which exhibited similar process forces.

Figure 7.38. AFSP process force output for René41/IN600 processed at 125 RPM; 4.5 IPM.
The distribution of additive René 41 is similar to that of high heat input Mar-M247 AFSP (Figure 7.36C). As with other AFSP samples examined, the additive SZ region is characterized by a turbulent AS with an accumulation of additive material on the RS. No significant flow of material along plate normal direction was observed. Measurements of digital optical micrographs reveal a high fraction (84%) of the additive material was retained after processing, again consistent with other material systems. Additive material not incorporated was lost as flash. Microhardness mapping of the SZ regions indicates the highest hardness regions correspond to the René41 containing areas of the SZ. Overall after AFSP, microhardness was increased by an average of 100 HV. Some regions of the SZ near the top surface demonstrate microhardness values in excess of 450 HV. Within these high hardness regions the extent of grain refinement is higher than more interior portions of the SZ. René 41-containing regions near the surface have fine equiaxed grains on average 2.5 µm in diameter whereas interior SZ regions have 2-3 times larger. Figure 7.39A and B shows representative micrographs from surface and interior regions with measured hardness values of 367 HV and 443 HV, respectively. Both regions demonstrate an elimination of the solidification structure characteristic of the as-deposited condition and differ only in grain size. Fine (<15 nm) secondary γ’ is found to form within both portions of the SZ in the as-AFSP condition (Figure 7.40B). Very fine carbides from the original LENS deposit (likely titanium and molybdenum-rich MC carbides [177]) (~0.5-1 µm) appear to be unaffected in size and distribution as a result of AFSP.
While considerable refinement, especially near the top surface of the SZ, was observed for AFSP of René 41, the formation of ultra-fine (i.e., nano-scale) grains was not obtained for the processing parameter combination explored for this material combination. Compared to Mar-M247, the $\gamma'$ solvus temperature of René 41 is
considerably lower (1204°C for Mar-M247 vs. 1057°C (calculated using JMatPro)). Additionally the considerably higher γ’ former content of Mar-M247 (11 wt%) compared to René 41 (4.7 wt%) promotes the rapid formation of higher proportions of γ’. As discussed in previous sections, the formation of ultra-fine grain SZ regions in Ni-base superalloys requires supersolvus deformation and subsequent minimization of coarsening processes until the formation of coherent γ’ arrests grain growth. Minimization of coarsening is be accomplished by rapid cooling through the single-phase γ phase field (parameter effect) and/or rapid precipitate formation (inherent material effect). The larger high temperature single phase γ temperature range combined compared to Mar-M247 (Figure 7.41) along with more sluggish γ’ formation in René 41 (Figure 7.42) promotes coarsening of the recrystallized microstructure within the γ phase field immediately after stirring. Despite the lack of ultra-fine grain SZ formation forming as a result of low heat input process parameters, the SZ still exhibits microstructural refinement over the solidification microstructure of the René 41 LENS deposit.

Figure 7.41. Predicted equilibrium single-phase region temperature stability for René 41 in comparison to Mar-M247 for dilution by IN600 substrate material.
Figure 7.42. Predicted time-temperature-transformation behavior $\gamma'$ precipitation for Alloy 282, René 41, and Mar-M247 (ascending order of $\gamma'$-former content)

7.4 Potential Use of AFSP for Strain Age Cracking Susceptible Materials

Strain age cracking of Ni-base superalloys with high fractions of $\gamma'$ formers associated is associated with postweld heat treatment. A review of the technical literature shows general agreement that residual stresses associated with welding, contraction associated with aging, and thermal stress result in cracking when exposed to temperatures within the $\gamma'$ precipitation range [45, 172, 175, 176]. The use of weld repair for service cracks is used to extend the useful life of Ni superalloy components such as exhaust nozzle flaps in aerospace gas turbine engines [175]. However, repair processes for high-fraction $\gamma'$ former superalloys are prone to strain age cracking during post-repair heat treatment.

Based on the resulting microstructures presented in this document, AFSP possesses many aspects making it ideal for repair of strain age cracking-susceptible
superalloys. A hypothetical repair process using AFSP would involve grinding out the original discontinuity. Removed material would be redeposited using a fusion-based process such as LENS. While the fusion-based process would normally increase the susceptibility due to the inherent residual stresses associated with solidification, the subsequent FSP of the additive material has the potential to mitigate residual stresses. The thermal cycle associated with FSP will allow redistribution and elimination of much of the residual stress from the deposition process. While there are inherent residual stresses associated with FSP/W, it is generally accepted that such residual stresses are lower in magnitude compared to fusion based techniques. The reduction in residual stresses provided by FSP is also combined with the development of an ideal microstructure with respect to strain age cracking resistance. Because strain age cracking is an intergranular cracking phenomenon, increases in grain boundary area provided by the inherent refinement of AFSP will accommodate and redistribute residual stresses along a larger boundary area. Some studies [176, 179] have demonstrated increased strain age cracking susceptibility for coarse grain alloys relative to fine-grained material, presumably as a result of the aforementioned mechanism. An additional benefit to the use of fine-grained material with respect to strain age cracking is the larger grain boundary region’s ability to accommodate embrittling phases [45]. However, all the AFSP microstructures examined in this work do not show evidence of embrittling phases such as carbide films along grain boundaries.

7.5 Applicability of Fine-Grained Superalloy Structures

Superalloys are used in applications requiring mechanical integrity at very high homologous temperatures. For applications where creep is a concern, it is generally accepted that larger grain size is advantageous due to the mitigation of deformation via grain boundary sliding. However potential applications exist for ultra-fine grained superalloy structures still exist. Forming applications such as superplastic forming rely on grain boundary sliding to obtain very large elongations (>100%) and form complex
geometries with high precision (see Figure 7.43). Superplastic forming is regarded as one of the major advances in forging and formability within the past 50 years [48]. Several studies have investigated the superplastic formability of superalloys for aerospace applications [180-182]. Elongations of up to 560% at strain rates on the order of 10^{-3} \text{s}^{-1} have been reported for IN718 [183]. All studies utilize extremely fine grain (on the order of 1 \text{µm} or less) microstructures. The fine-grained structures formed via AFSP highlight potential application for location-specific superplastic properties in superalloys. Cost and productivity benefits can be realized by using the microstructural refinement provided by FSP to locally enhance formability only where necessary. This method also allows for a graded material with respect to grain size. Fine grain regions where formability is desired can be produced with AFSP/FSP. In other regions of the component, the grain structure can be relatively coarse to provide necessary creep resistance.

Figure 7.43. Superplastically formed IN718 exhaust gas mixer component. Photograph from Donachie [48].
Other performance benefits resulting from the fine-grained superalloy microstructures formed by AFSP include possible enhancements in fatigue performance due to enhanced fatigue crack initiation resistance. Fatigue crack initiation (Stage I) is attributed to the pile up and subsequent formation of persistent slip bands (PSB) which act as local stress risers were fatigue cracks initiate [184, 185]. Tanka and Mura [186] propose a model which predicts the number of cycles until crack initiation being inversely proportional to the grain size. Fatigue experiments in Ni (99.98%) have demonstrated that fine-grains significantly increase the fatigue life at low stress amplitudes due to the increased number of cycles until a fatigue crack initiates. However, even at high stress amplitudes, an improvement in number of cycles until initiation was increased. The authors attribute the improvement in initiation resistance to change in PSB morphology. Coarse grain Ni exhibited cracks initiating at grain boundaries where numerous PSB impinged whereas fine grain material cracks initiated at PSBs formed intragranularly.

Microstructural modification produced by AFSP/FSP is ideal for the creation of functionally graded microstructure with respect to grain size. At the surface of the component where crack initiation is expected, the grain size is significantly reduced. Below the process regions, the material is unaffected with respect to grain refinement. For superalloy components, such interior microstructures can remain coarse to provide necessary creep properties at elevated temperatures. This creation of a graded grain size component with finer grains at the surface and coarse grains within the interior is the concept utilized for superalloy disks in high performance turbine engines where fine grains are desired within the disk bore and web for high cycle fatigue considerations and coarse grains are desired in the rim for creep resistance [187]. Without the use of AFSP, the creation of graded grain size components requires dual-microstructure heat treatments which involves complex, time-consuming heat treatments. Furthermore, grain sizes only as small as 10 µm are reported within the disk bore while simultaneously grains in the disk rim are 30-80 µm for a superalloy similar to René 104 (8.6 wt % γ’ former) [187].
Low heat input AFSP/FSP can be used to create considerably finer grain sizes, especially in high-\(\gamma\)' former fraction alloys where coarsening post-stirring is severely restricted.
7.6 **Summary**

7.6.1 **Mar-M247**

Additive friction stir processing was successfully applied using Mar-M247, a superalloy with high fraction of $\gamma^\prime$ formers (nominally 11%). Laser deposition of Mar-M247 onto IN600 substrates demonstrated significant cracking within the deposition attributed to solidification cracking. Three different AFSP parameter combinations were evaluated consisting of processing window extremes along with an intermediate heat input parameter combination.

Additive FSP of Mar-M247 proved to be more difficult than lower $\gamma^\prime$-fraction alloys (e.g., Alloy 282 and Alloy 214) to process defect-free. The process force output was correspondingly high—nearly a factor of two higher than FSP of IN600. The presence of wear debris in the SZ is a consequence of the higher process forces. All processing parameter combinations demonstrated lack of consolidation voids on the SZ top surface of varying severity. The lowest heat input parameter combination exhibited the most numerous post-AFSP defects. Cross sections revealed consolidation and healing of original deposition cracks occurred as a result of AFSP. However, crack healing was incomplete in some regions of the SZ for material processed using low heat input conditions.

Microhardness mapping revealed significant increases in hardness for Mar-M247-containing regions of the SZ. As with other alloy systems examined, the extent of hardness improvement was dependent on the AFSP processing parameters. Low heat input parameter combinations produced surface hardness values exceeding 750 HV in the as-AFSP condition, an improvement of 350 HV relative to the as-deposited condition. Interior regions still showed improvement over as-deposited material by 100-200 HV. Higher heat input parameters did not exhibit extreme near-surface hardening; however, microhardness within the AFSP SZ was still higher than the as-deposited condition.

As with other alloys examined, examination of AFSP microstructures revealed elimination of the solidification microstructure for all conditions explored. All
microstructures were characterized by a near-mono-size distribution of fine secondary $\gamma'$ along with very fine (typically \(\leq 1 \mu m\)) equiaxed carbides with no post-processing heat treatment. The grain size variation in the SZ corresponded to the measured microhardness--regions of the highest microhardness were also regions with the greatest extent of grain refinement.

The low heat input condition Mar-M247 sample exhibited a unique division of microstructure characterized by a ultra fine grained near surface region \((d_{avg}= 0.34 \mu m)\) and an interior region that had highly chaotic distribution and intercalation of additive material with the IN600 substrate. Analytical electron microscopy measurements show up to approximately 20% dilution of the additive Mar-M247 by the IN600 substrate occurs during AFSP using low heat input parameters. Composition measurements in the high-hardness surface region suggest that no dilution by the substrate occurs.

A mechanism for the microstructural evolution in high $\gamma'$ fraction superalloys was proposed in this work. The model proposes that deformation during stirring occurs above the $\gamma'$ solvus temperature due to the expected drop in high temperature flow stress combined with the self-regulating nature of AFSP/FSP. Dynamic recrystallization is expected to occur supersolvus, thus establishing the microstructure observed after processing. Morphological, orientation and grain boundary character observations via HRSEM and EBSD both show evidence that the recrystallization processes operative during deformation do not occur subsolvus, a regime in which boundary mobility is inhibited. Upon cooling after the FSP tool has passed, the recrystallized material coarsens until the material has cooled sufficiently below the solvus temperature where the formation of coherent $\gamma'$ reduces boundary mobility and pins grain growth. Therefore the grain size developed after recrystallization is a function of the cooling rate through the $\gamma$ phase field as well as the kinetics of $\gamma'$ formation. It was shown via computational kinetics predictions that the precipitate formation is dependent on the extent of dilution of additive superalloy by the substrate.
7.6.2 René 41

Limited AFSP trials of laser-deposited René 41 on IN600 substrates were performed to examine the resulting microstructure of a superalloy composition intermediate relative to Alloy 282 and Mar-M247. Only one AFSP parameter combination, low heat input: 125 RPM; 4.5 IPM, was evaluated due to limited material availability. Similar to Mar-M247 AFSP the topmost SZ region of the René 41 SZ contained a lack-of-consolidation defect; however the defects formed were much less severe (in term of depth) than those observed with Mar-M247. Process forces do not show evidence of process instability despite producing a defect and demonstrate process forces comparable with similar parameter combinations for autogenous FSP of IN600. The slightly shallower plunge depth used resulted in reduced path and cross path forces compared to autogenous FSP of IN600.

As with other AFSP SZ microstructures, replacement of the original solidification microstructure with considerably refined equiaxed grains was characteristic of additive material-containing regions. AFSP SZ regions contained fine distributions (<15 nm) of secondary γ’ and very small (~0.5-1 µm) cuboidal carbides dispersed randomly within the microstructure. Grain size was on average 2.5 µm in diameter near the top surface and two to three times greater within the interior, approximately 500 µm away from the surface. The extent of grain refinement corresponded directly to microhardness measurements. On average, the hardness was increased by approximately 100 HV. The mostly highly refined SZ region near the surface had measured hardness values near 450 HV. While grains were refined as a result of AFSP, the degree of grain refinement obtained for AFSP René 41 for the chosen parameters did not result in the formation of nano-scale grains as was the case for AFSP of Mar-M247. Inherent differences in the γ phase field stability and precipitation kinetics for René 41 with respect to composition compared to high-fraction γ’ former alloys results in increased coarsening post-deformation.
CHAPTER 8: FRICTION STIR PROCESSING PRETREATMENT FOR BASE MATERIALS

Autogenous GTAW welds were performed directly on top of friction stir processed IN600 to determine the effect of FSP pretreatment on the grain size evolution of the HAZ and WM. As with other investigations discussed in this document, extremes from the IN600 processing window were selected as to represent the gamut of grain sizes obtainable via FSP. Therefore, high and low heat input parameter combinations of 150 RPM; 2 IPM and 100 RPM; 4.5 IPM were selected, respectively. Resulting IN600 SZs for high and low heat input runs exhibited average SZ grain sizes of 15 µm and 9 µm, respectively. For comparison, the unprocessed IN600 grain size was significantly larger, 54 µm.

Following FSP, autogenous GTA welds were placed atop the FSP material. Prior to arc welding, the samples were cleaned with acetone. Arc welds were placed on the top plate surface entirely within the stir zone and made in the same direction as the FSP traverse. A programmable GTA welding machine (Jetline Engineering Inc., model: TKM-72-M, Irvine, CA) was used with welding speed, current, and voltage of 1.57 mm/s (3.7 IPM), 110A, 11V, respectively.

Figure 8.1 shows a transverse cross section of the autogenous GTA weld made atop the FSP region. The size of the GTAW weld is such that the corresponding HAZ still is located within the former SZ. For both FSP heat inputs, coarsening of the prior SZ is readily apparent. Compared to the starting SZ grain size, the average grain size in the HAZ along the fusion boundary increased to the initial FSP grain size by a factor of 4 to 5, depending on the sample. The initial SZ grain differences resulting from the different parameter combinations did not have a significant effect on the resultant near-fusion
boundary HAZ grain size. While prior SZ grains coarsened as a result of the autogenous GTAW pass, near-fusion boundary grain size remained below 100 µm, unlike the untreated IN600 base material. Near fusion boundary grains for untreated base material are several hundred microns in diameter. Figure 8.2 shows EBSD inverse pole figure maps of transverse sections near the fusion boundary. The pole figure maps clearly illustrate the difference in near-fusion boundary microstructure for FSP and non-processed material with respect to grain size. Despite coarsening of near-fusion-boundary grains from GTAW, the FSP-pretreated samples exhibit grains on average three times smaller than the GTAW HAZ grains of the non-FSP pretreated BM. Clearly, FSP is a viable method for inducing HAZ grain refinement and thereby reduce the severity of grain coarsening of a base material before fusion welding.

Figure 8.1. Light optical micrograph of transverse cross section of GTAW place on top of low-heat input FSP run
The HAZ grain size reduction obtainable using FSP pre-treatment has the potential to improve fusion weldability issues such as HAZ hot cracking. A number of weldability studies have observed a relationship between near-fusion boundary HAZ grain size and liquation cracking susceptibility. In a study by Thompson et al., the HAZ liquation cracking susceptibility of Inconel Alloy 718 was shown to be linearly dependent on grain size [188]. The benefit of reduced hot cracking susceptibility with finer grain size is attributed to the increased grain boundary area associated with smaller grains. Provided the liquid wets the grain boundary, the larger grain boundary area promotes the spreading of liquid (assuming a constant volume of liquid) thereby reducing the thickness
of liquid films present on the boundary. The increased boundary area along with liquid spread across a larger boundary area reduces the strain concentration and crack susceptibility. Because IN600 is not susceptible to HAZ liquation cracking as other solid solution-strengthened alloys with richer compositions, weldability testing of FSP pre-treated IN600 was not performed.

An investigation focused on the HAZ liquation cracking susceptibility of FSP pre-treated Ni-alloys was performed by Rule [107]. Susceptibility was evaluated using the spot varestraint test. Spot varestraint is an augmented strain type test technique that utilizes a stationary rather than traversing torch to create a weld. The spot weld creates a stable HAZ thermal and microstructural gradient before the application of strain [189]. The results of this test, which includes maximum crack length and total crack length, are measured as a function of applied strain. Figure 8.3A and B show maximum and total crack length data, respectively, adapted from the Rule study [107]. For all three liquation prone alloys (Hastelloy X, Alloy 625, and Alloy 718), FSP pre-treatment resulted in a reduction in maximum crack length up to 30% for all tested levels of strain. Additionally, for all tested alloys except for Hastelloy X, the total measured crack length was reduced by as much as 25% from FSP pre-treatment.
Figure 8.3. Spot varestraint results showing A.) maximum crack length and B.) total crack length as a function of applied strain for Hastelloy X, Alloy 625, and Alloy 718. Data adapted from Rule [107].
Rule indicated that the near-fusion boundary HAZ microstructure reveals much of the original refinement obtained as a result of FSP was destroyed by coarsening due to long exposures to elevated temperatures during spot weld pool stabilization during spot varestraint testing [107]. Despite the high heat input during spot varestraint testing, improvements in MCL and TCL were observed. As a result, HAZ refinement and weldability benefits provided by FSP pre-treatment are expected to be greater in actual applications where the local heat input would not be as high as that used in spot varestraint testing. Using similar parameters used to create autogenous fusion welds atop FSP IN600, autogenous GTA welds were made directly on the SZ of a FSP-pretreated Hastelloy X base material identical to that used in the Rule study. The lower heat input of the autogenous GTA weld relative to the spot varestraint test (180 A arc for 20 sec.), does not result in the same extent of destruction of FSP grain refinement. Figure 8.4A and B shows inverse pole figure maps for FSP- and un-treated Hastelloy X base materials, respectively. As with the IN600 results discussed previously, the prior SZ grains are coarsened significantly (initial Hastelloy X FSP SZ grain size was 6 µm for material processed using 180 RPM; 2 IPM). Although the SZ grains have coarsened as a result of GTAW, they remain smaller than the untreated base material which exhibits near-fusion boundary grains approximately 5 times larger than the FSP-pretreated condition. For applications such as repair welding of coarse-grained base materials, FSP pre-treatment is a viable solution for local refinement of resulting fusion weld microstructures.
Also apparent in the orientation maps shown in Figure 8.2 and Figure 8.4, the weld metal (WM) grain size is reduced along with the HAZ grain size as a result of the FSP pre-treatment. Because the nucleation of solidification grains in the WM occurs epitaxially [189, 190], an increase in nucleation sites associated with finer grains along the fusion boundary will decrease the WM grain size. The effect of increased epitaxial nucleation resulting from finer grains is mechanistically analogous to other techniques used to increase heterogeneous nucleation in welds [190].

The effect of FSP processing parameters (i.e., the effect of starting grain size) was examined using EBSD orientation maps of WM plan sections (Figure 8.5). The extent of grain refinement within the WM for the FSP pre-treated base material is readily apparent compared to the as-received base material. Figure 8.6A shows the difference in measured WM grain size using EBSD for the low and high FSP heat input parameters compared to the as-received base material. Interestingly despite the differences in
starting grain size for the two different FSP heat input conditions prior to arc welding, the
average WM grain size was quite similar for both FSP parameter combinations. Due to
the relatively small EBSD scan area and the highly columnar nature of WM grains,
measurements of total grain boundary length using EBSD more clearly demonstrates the
extent of WM microstructural modification by FSP. Neglecting solidification subgrain
(cell and dendrite) boundaries, WM microstructures examined were almost entirely
comprised of high angle grain boundaries (> 10° misorientation). Total boundary length
measurements for the three conditions are shown in Figure 8.6B. The WM formed from
the low-heat input FSP condition demonstrated 455% higher total grain boundary length
compared to non-FSP base material. Compared to the high-heat input FSP condition, the
low-heat input FSP condition exhibited slightly higher total grain boundary length (along
with average grain size) likely due to the difference in starting SZ grain size. This
refinement of WM grains can be directly attributed to the greater extent of epitaxial
nucleation of WM grains from refined fusion boundary grains created by FSP pre-
treatment.
Figure 8.5. Inverse pole figure maps along sample normal direction (ND) for plan view sections of A.) high-heat input FSP, B.) low-heat input FSP, and C.) non-FSP IN600. Dashed line denotes weld metal fusion boundary with fusion zone to the left of the line.

Figure 8.6. A.) average WM grain size as measured by EBSD and B.) average WM grain boundary length for untreated and FSP pretreated IN600
Face-centered cubic materials such as IN600 do not exhibit strong grain size dependence for Hall-Petch strengthening [191]. Hardness measurements of the of low and high heat input FSP conditions were 73 and 70 HRB, respectively. Without FSP prior to arc welding, the measured WM hardness was 69 HRB. Although large strength improvements are not expected from the extent of weld metal refinement obtained via FSP pretreatment, other mechanical properties such as ductility are expected to improve with increased weld metal grain refinement.

The effect of FSP pretreatment on the WM microstructure has also been examined for other Ni-base alloys. Figure 8.7A and B shows resulting WM microstructure within GTA spot welds created on pretreated and untreated Alloy 625 respectively. Although the GTA spot weld heat input was rather high (180A arc for 20 sec), the effects of weld metal grain refinement are still realized.

Figure 8.7. Inverse pole figure maps of the WM microstructure of Alloy 625 GTA spot welds in the A.) FSP-pretreated and B.) untreated base material condition. Sample section lies within TD-RD plane.
As with the HAZ, refinement of WM microstructure has potential to more effectively accommodate liquid present at grain boundaries as well as reduce grain boundary stress concentrations [44]. As a result, numerous weldability issues such as weld metal liquation (for multipass welds), ductility dip cracking, and strain age or reheat cracking (in multipass welds) can potentially be minimized [35, 44] via reductions in WM grain size.

FSP pre-treatment for HAZ and WM refinement is not limited to Ni-alloy systems. Other systems that are prone to significant coarsening of HAZ and WM microstructure resulting from fusion welding are expected to have similar benefit. Titanium alloys, for example, are especially susceptible to severe coarsening of weld metal grains (prior β grain size). For applications requiring resistance to fatigue crack initiation resistance, large grains sizes are detrimental to performance [186]. To reduce coarsening, solid state joining techniques such as FSW have an advantage with respect to preventing severe grain growth. However because FSW cannot be applied universally, FSP pre-treatment before fusion welding may be advantageous. Figure 8.8 shows a plan-view section of FSP Ti-5111 (a near-α alloy) with a spot weld placed such that one half is contained within the fine-grained SZ (Ti-5111 SZ grain size 1-2 µm) the other half is contained within the HAZ which is comprised of very coarse prior β grains similar to the parent β-process microstructure. On the FSP side of the spot weld, the near-fusion boundary prior β grains are significantly coarsened; however, the grains remain considerably smaller than those on the non-FSP side. The smaller grains on the FSP side resulted in increased epitaxial nucleation and an increase in refinement compared to the adjacent side of the spot weld. Weld metal grains nucleated from the FSP-pretreated material were reduced in size by nearly an order of magnitude larger in average diameter.
Figure 8.8. A.) Optical micrograph of plan view section of GTA spot weld along FSP SZ/BM boundary in Ti-5111. B.) Higher magnification view near boundary between FSP pretreated and untreated base material. Microstructure revealed using Kroll’s etchant.

### 8.1 Summary

Friction stir processing as a viable method for the modification of fusion weld microstructures was successfully demonstrated. Refinement of both weld metal and HAZ grain size was achieved by friction stir processing of the base metal prior to fusion welding by autogenous GTAW. As a result of the HAZ thermal excursion from arc welding, fine grains of the FSP stir zone were coarsened by a factor as high as eight depending on location within the HAZ. However, HAZ grains near the fusion boundary for stir processed material still remained smaller than the base material by a factor of three. Heat affected zone grain refinement can be expected to have several practical benefits—especially related to the weldability of hot-cracking susceptible materials. Within the weld metal, grain size was also reduced as a result of FSP. Average grain size did not vary greatly with changes in FSP heat input. The smaller grains along the fusion boundary for stir processed material increased the epitaxial nucleation site density, resulting in finer weld metal grains. The total grain boundary length (area) was increased significantly (455%) for stir processed material compared to non-FSP base material. As
with the refined HAZ microstructure, weldability and mechanical properties of the weld metal are expected to be improved provided heat input is sufficiently low to prevent excessive coarsening. The applicability of FSP pretreatment was also demonstrated for other Ni-base alloys as well as a titanium alloy.
CHAPTER 9: CONCLUSIONS

The feasibility of location-specific material modification for a wide range of Ni-base alloys was demonstrated. Friction stir processing and AFSP have demonstrated potential for local property enhancement for materials that until recently have been generally regarded as very difficult to process. As a result of this work, several conclusions can be made.

9.1 Ni-Base Friction Stir Processing Development

1.) Processing windows for IN600 and Ni-201 were determined for a single FSP tool geometry. While a wide range of parameter combinations was determined, the processing window remains considerably more limited compared to low T_m alloys, such as aluminum alloys.

2.) Evidence of tool wear in the SZ of FSP IN600 and Ni-201 was not observed. Tool wear for IN600 is typically expected during FSW/P and has previously been reported in several studies [30, 56, 60].

3.) IN600 and Ni-201 demonstrate a commensurate drop in FSP process forces as the process heat input was increased. A dimensionless heat input parameter was used to rank the relative heat input for the various runs.

4.) Temperature measurements for FSP IN600 show peak temperatures near the
bottom edges of the SZ approached temperatures between 900-1100°C, depending on the process parameter combination. Along with peak temperature the heating and cooling rate was found to vary by a factor of two for either processing extreme.

5.) Shortcomings present in the heat generation model contained within existing FSW/P modeling tools provide unreliable peak and temperature distributions when applied to high temperature materials.

6.) The extent of microstructural refinement for FSP Ni is dependent in large part on the FSP process parameters. Average SZ grain size was found to increase with the dimensionless heat input parameter.

7.) Electron backscatter diffraction misorientation analyses were performed on various FSP regions. Regions of high misorientation, corresponding to an increase in stored energy, were observed within the TMAZ. Residual plastic deformation and incomplete recovery/recrystallization led to the measured misorientation.

8.) Misorientation analyses of the SZ reveal that differences arising from processing parameter-depending thermomechanical histories affect the distribution of global (KAM) and intragranular (GROD) misorientation. Overall, measured misorientation and therefore stored energy increases as the process heat input is decreased.

9.2 Additive Friction Stir Processing
The following alloys (in ascending order of γ’ formers) Alloy 282, Alloy 214, René 41, and Mar-M247 were incorporated into the surface of IN600 plate using AFSP. Also, the addition of additive Ti-6Al-4V powder into the surface of Ni-201 plate was investigated.

9.2.1 Ti-alloy Power Additions to Ni-201

1.) Ti alloy powder (Ti-6Al-4V) was incorporated into Ni-201 by filling machined holes with powder and subsequently performing FSP atop the holes. Reaction of the Ti-alloy with the substrate resulted in the formation of liquid films via a eutectic reaction that produced process instabilities.

2.) Low fractions of Ti-alloy were retained within the SZ. Regions containing Ti-alloy addition showed reaction between the addition and matrix to form a cored intermetallic annulus around the additive material. No evidence of η phase was observed.

9.2.2 Haynes Alloy 282 Additions to IN600

1.) Additive FSP techniques were developed to incorporate Alloy 282 into the surface of IN600 by performing FSP atop Alloy 282 weld overlay deposited using GTAW on IN600 substrate.

2.) The original solidification microstructure of Alloy 282 weld overlay was eliminated due to mechanical mixing at high temperature.

3.) Additive Alloy 282 was heterogeneously distributed within the top ~1mm of the plate after FSP. The distribution within the SZ was dependent on AFSP (RPM/IPM) parameter combinations.
4.) Microhardness of Alloy 282 AFSP layers showed improvement by as much as 100 HV relative to the substrate as-AFSP and 75-90 HV relative to the as-deposited Alloy 282. The highest hardness microstructures were obtained using the lowest heat input parameters.

5.) No distributions of fine secondary $\gamma'$ formed as-processed. However, some SZ regions were found to contain lamellar clusters of fine $\gamma'$ in the as-processed condition. Gleeble simulations of overlay material suggests the origins of these clusters is non-homogenized weld metal regions near the edges of the SZ.

6.) Direct age heat treated sample demonstrated enhanced heat treatment response relative to the weld overlay. Low heat input parameters resulted in higher fractions of secondary $\gamma'$ to form during age heat treatment.

7.) Thermodynamic predictions suggest the heterogeneous heat treatment response observed in the Alloy 282 weld overlay is due to microsegregation of titanium to interdendritic region. AFSP eliminated the heterogeneous heat treatment response thus eliminating the need for solutionization heat treatments.

8.) Misorientation analyses suggest the amount of stored energy (i.e., dislocation density) increases as the AFSP heat input decreases due to incomplete recovery/recrystallization as well as incomplete recovery of post-stirring deformation.

9.) EBSD misorientation analyses suggest stored energy of AFSP Alloy 282 compared to FSP IN600 processed using the same parameters is higher. Inherent alloy differences with respect to the ease of dislocation movement (i.e., stacking fault energy) results in differences stored energy.
10.) Intragranular misorientation analysis (GROD) shows a heterogeneous distribution of highly misoriented grains surrounded by regions strain-free (near-zero misorientation) grains. As process heat input is decreased, the frequency of these highly misoriented grains increases. Observations via EBSD corresponded to defect structures observed using TEM.

9.2.3 Haynes Alloy 214 Additions to IN600

1.) Similar AFSP techniques and processing parameter combinations used for Alloy 282 were used for Alloy 214/IN600

2.) No formation of $\gamma'$ was observed in the as-processed condition for any process parameter combination. Microhardness measurements support observations.

3.) Unlike Alloy 282, Alloy 214 did not exhibit a strong microhardness dependence on the AFSP processing parameter combination. A similar trend was observed after direct age heat treatment.

4.) Double pass AFSP with 100% overlap was performed using the lowest heat input parameters. No precipitation of $\gamma'$ was observed despite the additional thermomechanical cycle.

5.) Alloy 214 SZ grain refinement was identical in magnitude to IN600-containing regions for the same sample due to similar composition of the alloys.

9.2.4 Mar-M247 Additions to IN600

1.) AFSP of compositionally-rich superalloy was successfully performed using FSP to incorporate additive Mar-M247 laser deposited on IN600 using the LENS
process.

2.) Extensive hot cracking of the Mar-M247 LENS deposit was observed.

3.) Three processing parameter combinations were explored: low, medium and high heat input. The latter two resulted in extensive healing of deposition cracks whereas the low heat input condition resulted in some unconsolidated deposition cracks. Process forces nearly two times greater were measured for AFSP Mar-M247 compared to similar parameter FSP IN600.

4.) Extremely high hardness (750+ HV) was observed at the top SZ surface using low heat input parameters. Regions beneath the surface still showed improvement between 100-200 HV depending on location. Higher heat input conditions also exhibited higher hardness compared to the as-deposited Mar-M247.

5.) AFSP microstructures exhibit a fine nearly monosize distribution of spherical $\gamma'$ as-processed. Fine (1 $\mu$m or smaller) Ta and W-rich carbides present in the original deposition remain randomly distributed in the SZ with no apparent change in size.

6.) The formation of nano-structured SZ grains was observed near the surface as a result of low heat input AFSP parameter combinations. Formation of nano-scale grains in Ni-base FSP has not been reported in the technical literature.

7.) For high $\gamma'$ fraction superalloys, such as Mar-M247, the extent of SZ refinement is highly dependent on degree of intermixing/dilution by the substrate and position within the SZ. The framework for the evolution of fine-grained SZ microstructures in Ni-superalloys has been presented.
9.2.5 René 41 Additions to IN600

1.) AFSP was performed with René 41 using the same laser deposition technique used for Mar-M247.

2.) Process forces were considerably lower than Mar-M247 and were similar to autogenous FSP of IN600 processed using similar parameters.

3.) Increases in hardness relative to the laser deposit were observed due to refinement and formation of very fine (<15 nm) distribution of γ’ as-AFSP.

4.) Near-surface ultra-fine grains were not observed for AFSP of René 41 and is attributed to the larger phase field of γ stability coupled with expectedly more sluggish precipitation kinetics compared to Mar-M247 due to lower γ’-former content.

9.3 Friction Stir Processing Pretreatment

1.) The feasibility of FSP as a 'buttering' process to improve the microstructure of subsequent fusion welds was demonstrated.

2.) Grain refinement produced via FSP led to a decrease in near-fusion boundary HAZ grain size in autogenous IN600 GTA welds. Additionally reduced grain size along the fusion boundary for treated material increased epitaxial nucleation and reduced grain size. A process to simultaneously refine HAZ and WM has not previously been reported.

3.) HAZ and WM grain refinement was also characterized for other nickel alloys, namely Hastelloy X and Alloy 625. Additionally FSP pretreatment was also
demonstrated for a Ti-alloy.
CHAPTER 10: FUTURE WORK

10.1 Modifications of Tool Geometry and Materials

The tool geometry utilized in this work was originally developed for welding rather than processing applications. The primary function of the large pin feature in the design utilized is to obtain desired penetration depth. For applications where only a shallow surface layer requires modification, penetration depth is not a primary concern. Changes to the tool geometry can be performed to reduce the process volume to only include the top-most surface. The reduction in pin length and/or pin volume to optimize near-surface processing likely will reduce process forces, which are already considerably high for Ni-base alloys. Other geometrical features that can be implemented include the use of tool shoulders with scroll features. Such features will eliminate the need for tool tilt. This has two potential consequences including 1.) ability to traverse the tool easily along features with curvature and 2.) significantly reduction of flash generation which is the mechanism by which additive material is lost from the SZ during processing.

The widespread applicability of high-temperature FSP/W is still limited by the lack of inexpensive, robust tool materials. With the current state of high-temperature tool material, one naturally comes to the conclusion that additional research is necessary to reduce wear. While the W-25Re tool did not exhibit any apparent tool wear for FSP/AFSP conditions explored for IN600, Alloy 282, Alloy 214, and René 41, compositionally-rich superalloys such as Mar-M247 showed significant tool wear within the SZ. The predominant materials of interest for high-temperature FSP/W remain focused on W-alloy systems and PCBN cermet systems; however other systems such as oxynitrides (e.g., SiAlON) remain promising alternatives.
10.2 Improved Thermal Measurements

A large difficulty in the work presented in this document is the ability to acquire temperature measurements during FSP/AFSP. Thermocouples embedded from the bottom of the plate were prone to extrusion away from the process zone due to the higher process forces. This was especially apparent for Mar-M247 where process forces were extremely high. Additionally thermocouples extending into the SZ from the bottom have an inherent inability to capture process temperatures near the top of the process zone, which is the region of interest in AFSP/FSP. This is especially pertinent for the tool geometry, substrate geometry, and deposition thickness used for this work. Future work to improve the reliability of temperature characterization can be obtained by embedding thermocouples inside the tool near the surface. A very recent study by Fehrenbacher [192] describes an experimental setup where Type K thermocouples are embedded in the tool pin and shoulder. Signal from the thermocouple is acquired wirelessly using magnetic sensors in proximity to the rotating tool. The use of rotary encoders can also provide the same function. Thermocouples embedded in the workpiece can still be used to give additional spatial information regarding the AFSP/FSP temperature distribution.

10.3 Multipass FSP and AFSP

Within this work, it was shown that 2 passes with 100% overlap had very minimal effect on the SZ microstructure of FSP IN600 and AFSP Alloy 214/IN600; However the effects of multiple adjacent passes with or without partial overlap was not explored. Applications requiring AFSP may require larger regions to be modified which would require the FSP tool to create multiple passes to essentially raster across the surface. While the additional thermal cycles are expected to have some effect on the microstructure and properties, the effect of multi-pass is unknown and requires further exploration.
10.4 Mechanical and High-Temperature Testing

This study was primarily focused on AFSP process development and microstructural development in AFSP of Ni-base alloys. Because the majority of alloys examined in this study are primarily used in high temperature applications, the next phase in development of the process is to evaluate the mechanical properties of AFSP-modified structural materials at elevated temperature. Naturally, creep testing should be performed to evaluate the change in creep properties as well as investigate the effect of fine-grain surface layers on creep life as fine-grains are generally regarded as detrimental to creep performance. Additionally, as was discussed in previous sections, the elevated temperature oxidation resistance is expected to benefit from the resulting AFSP surface microstructure. However, testing should be performed to evaluate potential benefits.

The creation of a graded grain size distribution near the surface of a component has considerable potential for improving fatigue life. Subsequent testing can be performed to quantify these potential benefits. However, the development and use of substandard, miniature fatigue specimens will be required as the AFSP regions are generally smaller than standard mechanical test specimens.

10.5 Improved Thermal Modeling

The results of temperature field simulations using commercially available FSW modeling software highlights shortcomings of the utilized models for FSP/AFSP of high-temperature materials. Shortcomings in the assumptions made in the heat generation models led to the prediction of erroneous process temperatures. Because of a lack of high-temperature tribological data, the contact conditions between the tool and workpiece remain unknown. In practice these tribological conditions likely differ from the assumption of pure sliding contact during FSP/AFSP of high-temperature materials.
Future models for heat generation should incorporate stick/slip conditions as well as heat generation from deformation.

10.6 FSP Rejuvenation for Weld Repair

The mechanisms by which FSP pretreatment can improve resulting fusion welds was demonstrated. However the effect of FSP for site-specific rejuvenation for weld repair was not investigated. Materials such as IN718 in service for long periods of time form constituent phases (such as δ, Ni₃Nb) which adversely affect the ability to perform weld repairs due to cracking concerns. The repair weldability has potential to be improved only where the repair is to be performed thereby preventing the need to heat treat the entire component in preparation of repair. However, the friction stir processability of materials that have been in service is unknown and requires further exploration. The elevated temperature strength expected for service components has potential to create friction stir processing difficulties such as excessively high process forces and corresponding accelerated tool wear.
REFERENCES


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[105] Inconel® Alloy 600 - Technical Data Sheet: Special Metals (Ed.), pp. 16.


[121] TSL OIM Analysis, Ametek/EDAX/TSL, Draper, UT, USA, 2008.


[137] M.L. Gallagher, in, Ohio State University, Columbus, Ohio, 2008.


APPENDIX A: CHARACTERIZATION BACKGROUND

A.1 Scanning Electron Microscopy

Compared to optical microscopy, SEM allows for examination of microstructural details on a much finer scale. The ability to discern fine features using SEM relates to the improved resolution relative to optical microscopy. Resolution can be defined as the minimum distance between two features while still being able to be observed as two distinct features. The classic Rayleigh criterion for visible light microscopy can be used as a first approximation to demonstrate the advantage of electron microscopy. With that approximation, angular resolution, $\delta$, is defined as [131]:

$$\delta = \frac{0.61 \lambda}{\mu \sin \beta} \quad (A1)$$

Where $\lambda$ is the wavelength of incident illumination, $\mu$ is the refractive index, and $\beta$ is the collection semi-angle. For visible light microscopy, angular resolution reaches a maximum around $\sim0.5 \ \mu m$. Considerably higher resolution can be achieved using electron microscopy because of the much smaller effective wavelength of ‘illumination’. The effective wavelength of electrons accelerated in an electron microscope can be described by the following approximation which originates from the non-relativistic De Broglie relationship [A1]:

$$\lambda = \frac{1.22}{\sqrt{E}} \quad (A2)$$
where \( \lambda \) is the De Broglie wavelength in nm and \( E \) is the electron accelerating voltage in volts. For an electron source used in the SEM, the accelerating voltage can be as high as 30 kV. An accelerating potential of 30 kV translates to an effective wavelength of 0.007 nm—considerably smaller than the wavelength range of visible light (400-700 nm). The smaller effective ‘wavelength’ of electrons yields an angular resolution orders of magnitude smaller than visible light.

The maximum resolution obtainable, however, does not approach the values predicted by the Rayleigh criterion. As with optical systems, the lens systems in electron microscopy are prone to defects or aberrations that affect performance. Figure A.1 shows cross sectional schematic of a typical SEM with various electromagnetic lens systems indicated. Inherent defects to microscope components such as the objective lens, condenser lens, etc. include spherical aberrations, astigmatism, and chromatic aberration. While these aberrations can be minimized, they cannot be totally eliminated thereby leading to a deviation between actual and theoretical maximum resolution.

Figure A.1. Schematic drawing of typical electron column in an SEM [A2]
A.1.1 High Resolution Scanning Electron Microscopy

In practice, the resolution limit is dependent on several factors and operating conditions of the microscope. The characteristics of incident electron beam such as beam size, accelerating voltage, beam current, etc. all affect the extent of interaction between the beam and the specimen. Figure A.2 shows an electron interaction volume simulation for Ni as a function of beam energies with the assumption that the incident beam diameter is 10 nm. The size of the specimen electron interaction volume determines the spatial characteristics of generated secondary and backscattered electrons. Elastically (backscattered electrons [BSE]) and inelastically scattered (secondary electrons [SE]) electrons generated from locations remote from where the beam directly interacts with the specimen results in a delocalization of detected signal from the direct beam entrance (Figure A.3).
Figure A.2. Monte Carlo simulations of electron trajectories within electron interaction volume for Ni (Z=28) for an incident beam with a diameter of 10 nm. Backscatter and secondary electrons are represented by red and blue traces, respectively. Calculations performed using Casino [3].

Figure A.3. Generation of incident-beam (at point B) induced secondary electrons (SE1) and delocalized backscattered (BSE) and secondary electrons (SE2) (Figure adapted from Goldstein [A2])
A SEM image formed using the Everhart-Thornley detector is predominantly comprised from various secondary electron signals, depending on the location and nature of the secondary electron generation mechanism. Signal created by SE generated directly where the beam directly interacts with the specimen is referred to as SE1 signal. Electrons that comprise this signal are not delocalized and represent the ‘high-resolution’ signal. High energy backscattered electrons generated where the beam meets the specimen can travel away from beam/specimen impact location and cause scattering events. Such scattering from high-energy backscatter electrons generates delocalized secondary electrons. This electron signal when captured by the detector is referred to as SE2 and does not contain ‘information’ about the specimen from the impact area. Highly energetic backscattered electrons can also travel away from the specimen and interact with the SEM chamber and create scattering events which generate additional SE signal. Signal generated from SE produced by BSE colliding with the interior of the SEM comprises the SE3 signal. This signal is highly dependent on the backscatter coefficient (proportional to the effective atomic number of the specimen) of the specimen. Lastly, delocalized secondary electron signal can also be generated when the electron beam interacts with the final aperture of the scope (SE4).

The relative fraction of these different SE signals depends highly on the composition, i.e., the specimen average atomic number. Table A.1 shows the proportion of signal generated by a gold specimen. The large backscatter coefficient of gold contributes to the very large proportion of signal generated remote from locus of the beam and specimen (SE2 and SE3).
High resolution imaging in the SEM can be achieved by reducing contribution of delocalized signal, e.g. SE2, SE3, and SE4, to the final image formed by the detector. Reductions in delocalized signal are accomplished by the use of through-lens (TLD) detectors coupled with a semi-immersion objective lens. Such a system is incorporated in the FEI Sirion FEG SEM used extensively in this work. Signal captured by the TLD is comprised of only SE1 and SE2. Figure A.4 shows a schematic of the TLD lens/detector system used in the Sirion FEG SEM. Rejection of SE3 and SE4 signal thus enabling high-resolution imaging is achieved using a magnetic field from the semi-immersion lens that is projected down onto the sample. Under influence of the magnetic field, only SE within the vicinity of the electron interaction volume spiral up the lens and are collected by the through-lens detector.
Figure A.4. A.) Schematic of HR-SEM using semi-immersion lens. High resolution imaging is achieved using a magnetic field from the semi-immersion lens that extends to sample. Under influence of the magnetic field, secondary electrons spiral up the lens and are collected by the through-lens detector (Figure adapted from [A4]). B.) Cross section of snorkel (semi-immersion) lens [A5].

The signal that reaches the TLD detector is a combination of high-resolution signal (SE1) and delocalized signal (SE2). At low magnifications (<10,000X), the scan area is large (approximately 10 X 10 µm with pixel size approximately 10 nm for a 1024 x 1024 pixel image) and the field of view is many times larger than the spatial delocalization of electrons. Thus when the beam is scanned from pixel to pixel at low magnification, the SE2 signal will actually change appreciably from pixel to pixel and mask the signal from SE1 because the region where SE1 originates is significantly smaller than the pixel size. As a result, SE2 does not form a continuous signal and appears random, like noise. However, at higher magnifications (>100,000X), the scan area is very small (1 X 1 µm with one pixel representing a distance of approximately 1 nm for a 1024 x 1024 pixel image). For this case, the field of view is actually smaller than the electron interaction volume. Although still present in the image, the SE2 signal within the small scan area is relatively constant because the electron interaction volume is approximately the size of the scan area or larger. Thus, any change in detected signal can be attributed entirely to the high-resolution SE1 signal therefore enabling HRSEM [A2].
Figure A.5 shows an example of high-magnification image of a Ni-base superalloy alloy acquired on the Sirion FEG SEM using standard chamber-mounted Everhart-Thornley detector and a high-resolution mode which utilizes the semi-immersion lens and through lens detector. Images are of the same region, representing an area approximately 0.55 \( \mu \text{m}^2 \). Each pixel in the micrograph represents a distance of approximately 8 nm. The estimated width of the electron interaction volume for Ni at 12 kV beam acceleration voltage is approximately 600 nm via Monte Carlo simulation [A3]—such volume is spatially much greater than the image scan area shown in Figure A.5. Using the standard chamber-mounted ET detector with positive 250 V bias, small microstructural details are obscured at high magnifications. Extraneous SE3 signal from BSE-produced chamber SE results in reduced resolution overall resolution, and differentiation between discrete \( \gamma' \) precipitates cannot be made. When the SE3 signal is rejected from image formation as in the case of high-resolution mode (semi-immersion lens + TLD), resolution is greatly improved. Separation distances between individual gamma-prime precipitates as small as 4 nm can be distinguished using the high-resolution mode.
Advantages of HRSEM modes in high-performance FEG SEMs are readily apparent. Fine microstructural details on the scale of nanometers can be observed without the time-investment required for sample preparation of transmission electron microscopy samples.

Sample preparation and requirements for HRSEM are similar to conventional, high-vacuum SEM. However, more stringent vacuum requirements necessary for FEG sources used in HRSEM place extra emphasis on contamination mitigation. Furthermore, the magnetic field generated around the sample by the semi-immersion lens can prevent successful imaging of certain material types in HRSEM mode. Bulk ferromagnetic samples interfere with proper operation of the semi-immersion lens. This interference
can be reduced by reducing the volume of ferromagnetic specimen in the chamber. The paramagnetic nature of engineering Ni-alloys and Ni-superalloys permits imaging using HRSEM.