CHARACTERIZATION OF STRAIN ACCUMULATION AT GRAIN BOUNDARIES OF NICKEL-BASED SUPERALLOYS

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Abstract

Elevated temperature in-situ experiments, local crystallographic mapping and digital image correlation techniques have been used to gain insight into strain localization near grain boundaries in polycrystalline René-104. Samples were heat treated to create either smooth (standard) or serrated grain boundaries. Deformation experiments concluded that strain tended to localize at triple points and special boundaries in the standard microstructure, while strain had a less-pronounced correlation in the serrated microstructure. Both materials exhibited grain boundary sliding (GBS), but the standard microstructure proved to be more susceptible. Site-specific extraction of TEM foils indicated that boundaries that exhibited strain localization experienced activation of multiple slip systems, while boundaries showing GBS showed activation of only a single slip system. Based on m' analysis, slip transmission was expected to be difficult in boundaries that exhibited strain localization while boundaries that showed sliding allowed for easy slip transmission. A volume containing a boundary that exhibited GBS was analyzed to assess boundary surface topography and indicated that the boundary was macroscopically planar with local deviations that were on the order of the secondary γ' size.

Introduction

Creep deformation in polycrystalline nickel-based superalloys is a heterogeneous process that is primarily a function of γ' distribution [1-4], but grain size and orientation also play roles [5, 6]. Previous work has explored the effects of grain boundary character on deformation mechanisms, and has concluded that increasing the fraction of special Σn boundaries with n values ranging between 3 and 29, as determined by Coincidence-Site Lattice (CSL) Theory [7], dramatically increases creep, fatigue and corrosion resistance due to a decreased propensity for crack nucleation at Σn boundaries [8-10]. Increased creep and fatigue resistance of Σn boundaries is attributed to their increased resistance to grain boundary sliding (GBS) as compared to random high angle grain boundaries (HAGB) [8, 11]. GBS occurs when creep strain is accommodated by grains displacing relative to each other, and must be accompanied by intra-granular dislocation motion [11, 12]. Preliminary findings on modern turbine disc alloys have indicated that GBS is a contributing creep deformation mechanism around 700°C [13], and intentionally creating micro-scale serrated grain boundaries increases creep resistance by limiting GBS [14]. Soula *et al.* used a full-field digital image correlation (DIC) technique to conclude that GBS was active at conventional creep strain rates [13]. These studies have not included a detailed examination of the dislocation activity and its correlation with GBS, which is necessary to understand the mechanisms of GBS and why it occurs selectively.

It is also unclear how the complex nature of polycrystalline samples dictates why some grain boundaries do or do not accumulate strain, either because of intragranular slip inactivity or easy slip transfer across boundaries. Bi-crystals experiments have shown that accumulation versus transfer of slip across grain boundaries is controlled by crystallographic misorientation [15]. For simple analysis, the alignment of slip systems in adjacent grains can be reduced to a scalar value, m' , which is equal to the product of the direction cosine between the slip directions in each grain and the direction cosine between the slip planes normals for each grain [15]. As m' approaches a maximum value of unity it implies increasing alignment of slip planes and directions and therefore promotion of easy slip transfer. In contrast, boundaries with m' values much smaller than unity would impede slip transfer and, presumably, lead to strain accumulation and localization.

The objective of this study was to utilize full-field deformation maps created through DIC to probe strain localization over a large sampling of grain boundaries and, for select boundaries, correlate the degree of strain localization with the active deformation mechanisms. Dislocation mechanisms were experimentally observed and compared with mechanism predictions from a Phase Field Dislocation Dynamics model [4]. The propensity for slip transfer versus slip blocking was assessed using the m' factor.

Experimental

Material

The material examined in this study was René-104 heat treated under two different super-solvus conditions to produce microstructures with either macroscopically straight (standard) or serrated grain boundaries, similar to those described by Danflou *et. al* [16]. Heat treatments were tailored such that both microstructures had similar γ grain size distributions. The standard heat treatment produced a bimodal γ' distribution of cuboidal secondary and tertiary particles. The serrated heat treatment produced a complex distribution with very large secondary γ' on the grain boundaries, dendritic and cuboidal shaped secondary γ' , and tertiary γ' in the grain interiors. Flat, dog-bone specimens for constant load testing were electrical discharge machined with a gage length of 1.78 cm, and a rectangular cross-section with nominal dimensions of 0.31 cm x 0.081 cm. Four hardness indents were used as reference markers so electron backscatter diffraction (EBSD) and DIC could be performed on the same area.

Characterization

Dog-bone specimens were mechanically polished to conduct EBSD to determine grain orientation and shape prior to deformation. Grain size distributions and grain boundary character

measurements were conducted, for each processing condition, on over more than 2000 grains. Large area scans were compiled using AnyStich code [17]. Identification of Σn boundaries was conducted using Brandon's criterion: which specifies that the angular tolerance for a Σn boundary is equal to $15/\sqrt{n}$ [18, 19].

Cuboidal secondary γ' size and spacing plays a critical role in determining whether dislocations prefer to stay as $a/2\langle 110 \rangle$ type or dissociate into $a/6\langle 112 \rangle$ Shockley partials [4]. The cuboidal secondary γ' size for both standard and serrated microstructures, were determined by sampling more than 450 particles following the methods of Payton et al. [20]. In this procedure, particles were measured in grains that had a $\{111\}$ plane parallel to the plane of polish. Grain orientation was determined via EBSD prior to applying a γ' etchant (61:36:6:2.4 Lactic acid, 85%: Nitric acid, 69%: hydrofluoric acid, 49%). The γ channel width between γ' particles was analyzed with two different methods. First, the centers of mass for each individual particle in a 2-D cross-section were identified and then connected using Delaunay triangulation algorithm, as shown in Figure 1. Thus, individual channel widths are defined as the segments between particles along the lines connecting the centers of mass (the pixels within the particles are excluded).

Figure 1. Identifying individual γ channels by Delaunay triangulation for 158 γ' particles and 852 identified γ channels.

Second, in order to verify the channel spacing calculated by Delaunay triangulation, Phase Field simulations based on dislocation line tension were used to determine the minimum channel width to achieve dislocation activity for this specific microstructure at the experimentally applied shear stress [4]. Phase Field simulations were set-up so that a dislocation line was placed at the right edge of the simulation cell with either a uniform particle size and spacing (benchmark) or the experimentally determined particle distributions. Shear stress, in reduced units, was gradually applied to determine at which stress level the dislocation would move forward through the experimentally determined particle distribution. Then simulations were conducted on benchmark distributions, with different channel spacing. The critical γ channel spacing was determined when the dislocation would travel through the benchmark distribution under the same applied shear stress as the simulation on the experimentally determined microstructure. This critical γ channel spacing was subsequently compared with the Delaunay calculations. The Phase Field model was then used to create a Dislocation Activity Diagram (DAD) of the active dislocation mechanisms as a function of the angle between slip system and the tensile axis. All simulations assumed intrinsic stacking fault energy of 25 mJ/m^2 , shear modulus of 67 GPa, and a Burgers vector $2.53 \text{ \AA}$.

The dominant intragranular dislocation mechanisms were studied by bright field (BF) imaging using a Phillips Tecnai F20 scanning transmission electron microscopy (STEM), at 200kV accelerating voltage. Thin foils were extracted from site-specific regions using an FEI Helios dual beam scanning electron microscope and focused ion beam (SEM-FIB). Comparison of experimentally observed dislocation mechanisms with DAD model predictions was conducted by first calculating the most probable active slip systems as a function of grain orientation with respect to the tensile axis as determined by EBSD. The most probable slip systems were determined by assuming Schmid's law, i.e. an idealized situation where the local stress state is that in which the critical resolved shear stress is determined using the maximum Schmid Factor (SF) value. The slip systems with the highest SF were compared with the slip trace measurements on STEM foils, and these systems were input into the DAD model to determine the predicted dislocation mechanisms.

The 3-D microstructure was investigated through serial sectioning using an FEI Nova 600 SEM-FIB. A platinum cap was deposited over the boundary of interest and a volume of material was extracted and placed on a grid using an OmniProbe *in-situ* micromanipulation system in a manner similar to STEM foil preparation. A custom script utilizing FEI Runscript software was created to automate the serial sectioning process, and consisted of cross-section milling, collection of ion-induced secondary electron (ISE) images, and repositioning of the stage between the two conditions [21]. The section thickness was approximately 50 nm and the ISE images had an in-plane resolution of 223 pixels/ μm . The $406 \mu\text{m}^3$ volume was reconstructed from 116 slices and contained a grain boundary exhibiting GBS.

SEM-FIB serial sectioning data was processed for automated boundary identification using the following algorithms. Following a levels adjustment for contrast enhancement, a standard deviation filter was applied where every pixel was re-labeled with the standard deviation of its 11×11 pixel neighborhood. This served to highlight pixels near the boundary, as their standard deviation (i.e. new pixel-value) would be considerably higher than pixels in the grain interiors. Next, binary image segmentation was done by applying a global threshold to highlight the grain boundaries. In some slices the boundary was artificially discontinuous, and watershed segmentation was employed to fill in most of these discontinuities. Pixel clusters not belonging to the boundary were removed by rejecting any objects below a size of 100 pixels. Any remaining pixel clusters were manually removed and remaining discontinuous boundary segments were manually connected. This algorithm was applied to each slice and resulting binary images were stacked to produce a 3-D volume of non-cubic voxels.

At this point, the grain boundary network was represented as thin lines (1-2 pixel thick) separating the grain interiors. Although the boundary was fully continuous within each slice (i.e. the imaging plane), the boundary was not continuous in the planes perpendicular to the imaging plane and therefore, the grains could not be interpreted as separate objects. To create a continuous 3-D boundary that fully separated the grains, a 3-D watershed algorithm was applied. A closed surface was constructed from the vocalized boundaries and this surface was smoothed to facilitate visualization. The individual grains and their boundaries were then visualized using the commercial software Avizo developed by Visualization Sciences Group.

In addition to the grain boundary, carbides and twins were also segmented and visualized using algorithms similar to those employed on the grain boundary network. The different segmentations were merged and visualized to create a composite dataset where all microstructural features and their interactions could be analyzed.

In-situ Mechanical Testing

Correlated Solutions VIC-2-D DIC software was used to measure the local deformation fields from *in-situ* secondary electron images (SEI) of 2048x1886 pixel size with a 150 nm/pixel resolution. VIC-2-D analysis required a random speckle pattern, and testing conditions required an elevated temperature and vacuum compatible pattern. Soula *et. al* determined that hafnium-oxide was a viable candidate for patterns for elevated temperature testing of nickel-based superalloys [13]. Therefore, a pattern of hafnium-oxide was created using electron beam lithography for this study. The pattern consisted of a random array of diamond shaped speckles, with edge lengths between 400-700 nm, and grid lines spaced 10 μm apart for GBS measurements. Specifics on specimen preparation and speckle pattern procedures are presented in Walley *et. al.* [22].

Deformation experiments were conducted at 700°C with an Ernest Fullam tensile frame, with a 4448 N load cell and molybdenum resistance heater. Constant load tests were conducted in a step-wise fashion to limit creep deformation during imaging. Samples were first loaded in displacement control to a preload condition (~300 MPa) for initial SEI acquisition. Samples were then subsequently loaded in displacement control to the test condition (900, 1000 or 1100 MPa) and held at constant load in load control for 10-30 min, depending on strain rate, before unloading to 80% of peak load for SEI acquisition. The sample was reloaded to peak load and the sequence repeated for the duration of the experiment.

Full-field deformation maps were analyzed by comparing the SEI at preload with images acquired after various hold times at peak load. VIC-2-D DIC parameters used for this study were subsets of either 60x60 or 30x30 pixels and subset spacing of 3 pixels. Optimization of DIC parameters for spatial resolution while minimizing the strain intensity signal-to-noise ratio was based on image quality and speckle pattern. GBS was measured over the same sample area as analyzed with DIC, on standard microstructures creep tested at two different levels of stress and total strain accumulation (1000 MPa-1.7% and 1100 MPa-3.67%). GBS was also analyzed on the serrated microstructure creep tested at 1100 MPa and 3.10% total strain. The contribution of GBS to the overall accumulated plastic strain was calculated by measuring discrete offsets in grid lines at grain boundaries. Since the magnitude of the individual GBS events was on the order of 100 nm, analysis of grid marker offsets was conducted on backscatter electron images with a resolution of 25 nm/pixel, following the equations expounded by Langdon [11].

$$\varepsilon_t = \varepsilon_g + \varepsilon_{GBS} \quad (1)$$

$$\varepsilon_{GBS} = 2n_l \left(\frac{w}{\tan \theta} \right)_l = n_l \left(x \tan \theta \right) \quad (2)$$

The total plastic strain, ε_t , is a function of the intragranular deformation, ε_g , and grain boundary sliding deformation, ε_{GBS} . The strain from GBS was calculated as a function of average of

discrete offsets on either longitudinal or transverse grid lines, w or x , the average number of grains per unit length of line, n_l or n_t , and θ , the angle between the tensile axis and grain boundary trace.

Results

Microstructural Analysis

Measurements of the γ phase grain size distribution of the standard and serrated microstructures confirmed that the heat treatments produced statistically equivalent microstructures. The average and standard deviation of the grain size distributions are presented in Table I. Analysis of grain boundary character from EBSD confirmed that both microstructures contained a significant number of special boundaries, with most of these being $\Sigma 3$ annealing twins. Table I lists the percentage of boundaries that are random high angle grain boundaries (HAGB), twin boundaries ($\Sigma 3$), and higher order special boundaries (Σn).

Table I. Distribution of microstructural features, grain boundary types are presented as a percentage of the total number of grain boundaries. The mode (first) and mean (second) of the γ channel width are presented.

		standard	Serrated
grain size	(μm)	22 ± 7.8	23 ± 8
HAGB	(%)	53	48
$\Sigma 3$	(%)	45	44
$\Sigma n \ n > 3$	(%)	2	7
intragranular cuboidal γ'	(nm)	114 ± 82	69 ± 50
γ channel width	(nm)	38, 74	35, 55
intergranular γ'	(μm)	~	2.3 ± 1.5

The standard and serrated microstructures had substantially different γ' distributions. The standard γ' microstructure had a bi-modal distribution of larger cuboidal secondary γ' and smaller spherical tertiary γ' particles. The serrated γ' microstructure had a quad-modal distribution consisting of (a) globular intergranular and (b) intragranular dendritic secondary γ' that formed during slow cool and lower temperature hold, also intragranular (c) cuboidal secondary, and (d) spherical tertiary γ' formed during fast quench and subsequent aging, this distribution is presented in Figure 2. The average cuboidal secondary γ' sizes for each heat treatment, and the average size of the intergranular particles in the serrated microstructure are presented in Table I. The larger inter- and intragranular γ' particles that form first during the slow cool remove some of the driving force to form additional intragranular particles during fast quench and age. Thus, a smaller secondary γ' particle size is created in the serrated microstructure as compared to the standard microstructure.

Figure 2. γ' particle distribution in the serrated microstructure, the tertiary particles are not visible at this magnification.

The Delaunay analysis of both microstructures concluded that the distribution of γ channel widths was right-skewed. As such, the mode and mean values of the distribution were different; both are presented in Table I. Intuition indicates that the mode would be more representative of the typical obstacle spacing encountered by dislocations traversing the microstructure. Phase Field simulations on experimentally determined microstructure, as seen in Figure 3, concluded that the reduced critical resolved shear stress needed for dislocation motion was $\tau^*=0.32$. Simulations on benchmark distributions concluded that for a dislocation to pass through a uniform particle field with the same critical applied stress, a minimum γ channel width of 38 nm was needed. This confirmed that the mode value determined by the Delaunay analysis was indeed the critical γ channel width for the standard microstructure.

Figure 3. Phase Field dislocation simulations through experimentally determined microstructure with gradually increasing stress in reduced units. Light gray (red in PDF on CD) indicates the sheared region.

DIC Analysis

Full field strain maps for the standard microstructure indicate that triple points and special boundaries play a pivotal role in the location of initial regions of high strain intensity. Figure 4 shows the accumulation of strain as a function of time at load, or total strain accumulation. After 0.28% average strain accumulation at 900 MPa, Figure 4a, shows that strain localizations are predominantly observed near $\Sigma 3$ boundaries and triple junctions which contain two or more Σn boundaries. The very high intensity location in Figure 4a is an artifact due to a patterning problem. In Figure 4b the total strain has progressed to 0.4%, and two observations can be made: 1) slip near previously inactive boundaries begins to accumulate and 2) the high strain regions associated with the first set of accumulated positions continue to increase in intensity. In Figure 4c, the final condition, total strain has progressed to 0.74% and the positions of the highest strain regions correlate well with “hot-spots” observed at lower strains (Figure 4a), suggesting that these regions continued to accumulate more strain. The time-lapsed information indicates that both grain

Figure 4. Strain accumulation in a standard microstructure as a function of time at stress. (a) 1.5 hrs at 900 MPa, strain hot-spots begin to form at triple junctions and $\Sigma 3$ boundaries, (b) 2.5 hrs, hot-spots link-up and new locations activate and (c) at the final time of 3 hrs the average strain is 0.74% strain. $\Sigma 3$ boundaries are black, higher order Σn boundaries are gray (red), and HAGB's are white. Color images are visible in the PDF on the CD.

boundary misorientation and boundary orientation with respect to the tensile axis contribute to the position where strain localization occurs. What is missing from this data is how the local stress state, as influenced by grain neighborhood effects, contributes to localization behavior.

Unlike the standard microstructure, a correlation between boundary character and strain localization sites was not as obvious in the serrated microstructure. Strain still localized at grain boundaries but tended to widen into the grain interiors, instead of expanding along the grain boundaries. A comparison of the degree of strain localization between the standard and serrated heat treatments in Figure 5 shows the differences between strain localization sites. Soula *et. al* utilized the standard deviation in the strain maps as an indicator of the degree of heterogeneity in the strain fields [23]. The average and standard deviation in the axial strain fields of the standard and serrated materials are very similar, $1.48 \pm 0.69\%$ and $1.6 \pm 0.62\%$ strain respectively. This indicates that the intensity distribution of the strain fields for these two materials types are equivalent, but the standard deviation does not provide insight into differences in spatial distributions. By applying a threshold to the strain field distributions so that strains greater than 3% appear as white and strains less than 3% are represented as black, Figure 5 provides a succinct representation of the differences between the spatial distributions of the strain fields of these two materials. The data indicates that the average size of the strain localizations is larger for the serrated material ($\sim 70 \mu\text{m}^2$) as compared to the standard material ($\sim 30 \mu\text{m}^2$).

Analysis of more regions is needed to conclusively determine that there is indeed a statistical difference in the size distributions.

The total strain accommodated by GBS was calculated for each material type, with values shown in Table II. In all cases the amount of calculated strain accommodated by GBS was similar even though the creep rates and total strain accumulated were different. Over the areas analyzed in these experiments, the serrated material had fewer active boundaries per unit area as compared to the standard material. This indicates that a more comprehensive analysis beyond comparing averaged behavior is required to adequately describe the differences in GBS behavior of these two material types.

Table II. Grain boundary sliding strain (ϵ_{GBS}) and the percentage of total strain accommodated by ϵ_{GBS} . Also listed is the total number of boundaries exhibiting GBS in the area of interest (# boundaries) and the number of those boundaries that are $\Sigma 3$ and higher order Σn boundaries.

	# boundaries	# $\Sigma 3$ / Σn	ϵ_{GBS} (%)	% of ϵ_t
Standard				
1100 MPa	63	11/9	0.82	22.2
1000 MPa	24	1/2	0.44	26.3
Serrated				
1100 MPa	28	5/4	0.86	28.4

Figure 5. Axial strain maps for both the standard (left) and serrated (right) microstructures tested at 1100MPa to 1.48% and 1.6% average strain respectively. Grain boundaries determined by EBSD are overlaid with the strain data in the upper pair of images, $\Sigma 3$ boundaries are white, Σn boundaries are gray, and HAGB's are black. The lower pair of images were created by applying a threshold to the data to highlight regions exhibiting the highest amount of strain localization, these show that these higher strain regions are spread over larger regions of the serrated sample, indicating less localized behavior. Color images are visible in the PDF on the CD.

Dominant Mechanism Observations

Phase field simulations allowed for the creation of a Dislocation Activity Diagram (DAD) using the critical channel width [4]. The DAD shows the influence of stress magnitude and orientation on the dislocation dissociation behavior and is illustrated in Figure 6. The light gray curve (red in PDF on CD) indicates the critical stress needed for the leading partial to pass the γ channel, while the dark gray line (blue in PDF on CD) is the critical stress needed for the trailing partial to pass the γ channel. In the region between the two lines, decorrelated motion of partials occurs and leads to the formation of extended stacking faults and nano-twins [4], as the stress magnitude increases motion of decorrelated partial dislocations leads to the movement of dissociated ones. Along the vertical axis, which is in units of resolved shear stress, the angle between the Burgers vector and loading direction is 0° and this angle can vary from $\pm 90^\circ$, for angles greater than $\pm 90^\circ$ the nature of the leading and trailing partials flips.

Figure 6. Dislocation Activity Diagram used to compare with experimental observations. The effective resolved stress is on the vertical axis, and the angular coordinate represents the angle between the full dislocation Burgers vector and the loading direction.

Select grain boundaries that exhibited above average strain accumulation or GBS were extracted for STEM analysis. In all instances the dominant mechanisms observed in the grain interiors were consistent with DAD model predictions, however in certain instances the dominant slip system observed in STEM was that with the second highest Schmid factor as opposed to the highest Schmid factor.

An example of this analysis is presented in Figure 7 for a grain boundary that exhibited significant shear strain accumulation. It can be seen that strain accumulates at the quad-point between the twins of each grain and the grains themselves. The misorientation of the grain boundary between grain 1 and grain 2, as described by an axis-angle pair in grain 1 reference frame, is $[0.64 \ 0.1 \ 0.76]38.6^\circ$. An axis-angle pair describes the rotation axis and minimum rotation required to bring two lattices into coincidence. The misorientation between the twin of grain 1 and the twin of grain 2 is characterized as a $\Sigma 9$ boundary. The STEM analysis indicated that both grain 1 and grain 2 exhibited extended stacking faults as the dominant deformation mechanism, while the twin of grain 2 exhibited non-dissociated dislocation motion as the primary mechanism. The comparison of some of the experimental measurements from STEM and DAD model predictions are shown in Table III. The discrepancy between the slip plane angles arises from the fact that STEM angles were measured from STEM images of the foils, while the model predictions were based on EBSD orientation data. Grain 1 had two active slip systems with very similar Schmid factors (SF) of 0.47 and 0.45, analysis is

shown in Table III. A comparison of both systems with the DAD model confirmed that in both cases the observed extended stacking fault mechanism was predicted as dissociated partials by DAD analysis. This indicates that the magnitude of the local stress state might be less than the macroscopic stress state. Grain 2 also had two possible active slip systems, both on the same slip plane (111), with SF of 0.41 and 0.34 for $[-110]$ and $[-101]$ respectively. The DAD model predictions indicate that $[-110]$ should exhibit decorrelated partials while the lower SF system $[-101]$ should exhibit dissociated partials. When comparing the slip systems of the twin of grain 2, DAD model predicted that this grain should exhibit non-dissociated dislocation activity (not shown in Figure 7c), which was experimentally observed. The STEM analysis indicated that there was increased dislocation content and activation of a second dislocation mechanism in grain 2 near the grain boundary, as seen in Figure 7a. Activation of a secondary mechanism was also observed at another extracted boundary that DIC indicated to exhibit above average strain accumulation.

Table III. Comparison of experimentally observed and DAD model predicted dominant slip systems for a grain boundary set that shows shear localization.

	TEM	Model Predictions
Grain 1		
Slip System 1		
System Angle	35°	36°
System	$\sim$	$(\bar{1}\bar{1}\bar{1})[0\bar{1}1]$
Schmid mechanism	$\sim$	0.47
leading+trailing	extended faults	dissociated
	$\sim$	$(\bar{1}\bar{1}\bar{1})[\bar{1}\bar{1}2] + [\bar{1}\bar{2}\bar{1}]$
Slip System 2		
System Angle	131°	132°
System	$\sim$	$(\bar{1}\bar{1}\bar{1})[011]$
Schmid mechanism	$\sim$	0.45
leading+trailing	extended faults	dissociated
	$\sim$	$(\bar{1}\bar{1}\bar{1})[\bar{1}12] + [\bar{2}\bar{1}\bar{1}]$
	TEM	Model Predictions
Grain 2		
Slip System 1		
System Angle	135°	139°
System	$\sim$	$(111)[\bar{1}10]$
Schmid mechanism	$\sim$	0.41
leading+trailing	extended faults	decorrelated
	$\sim$	$(111)[\bar{2}11] + [\bar{1}2\bar{1}]$
Slip System 2		
System Angle	135°	139°
System	$\sim$	$(111)[\bar{1}01]$
Schmid mechanism	$\sim$	0.34
leading+trailing	extended faults	decorrelated
	$\sim$	$(111)[\bar{2}11] + [\bar{1}\bar{2}\bar{2}]$

The arrows in Figure 7 indicate the superimposed direction of the Burgers vector needed to cause decorrelated/dissociated partial motion on each slip system. Without knowing the edge/screw character of the dislocations it is unclear if slip is moving towards or away from the boundary. Therefore, the m' parameter was calculated for all possible full and dissociated slip systems in each grain. Since DIC analysis indicates that strain is accumulating in grain 2 at the boundary between it and grain 1, Figure 7a, m' analysis was conducted for slip transferring from grain 2 to grain 1. In this direction, slip transfer would be difficult, since the highest m' value is only 0.5, this is for full dislocation transfer from the highest SF system in grain 2 to the highest SF system in

grain 1. If slip piles-up at the grain boundary to create a higher stress state, the DAD model predicts that slip on the highest SF system in grain 2, would occur by full dislocation motion, not partial dislocation motion. The activation of this second system can account for the increased full dislocation content near the grain boundary. If on the other hand, slip initiates in grain 1 and transfers to grain 2, the m' analysis indicates that transfer would be difficult, but would occur most easily by full dislocation slip from the second highest SF system in grain 1 to the second highest SF system in grain 2, with an m' value of 0.44. Since the second highest SF system in grain 1 is active in the dissociated sense, the partial dislocations would need to recombine before transferring to grain 2 before subsequently decorrelating. In both cases, slip transfer from grain 2 to grain 1, or grain 1 to grain 2, would be difficult because m' values are low, indicating that this boundary is likely to see strain accumulation instead of easy slip transfer. The m' analysis provides a self consistent explanation of the increased full dislocation content near the grain boundaries, but more in-depth STEM analysis is required to confirm active slip systems. The conclusion from m' analysis indicates that the difficult slip transfer from grain 2 to grain 1 might be the cause of the strain accumulation seen in the DIC analysis.

Similar analysis was conducted on a HAGB, with misorientation as described by an axis-angle pair in grain 1 reference frame as $[0.6 \ 0.5 \ 0.62]50.2^\circ$, that exhibited grain boundary sliding. From the surface seen in Figure 8a, it can be seen that the boundary trace appears to be mostly planar, with limited serrations. Experimental analysis confirmed DAD model predictions that grain 2 should exhibit decorrelated extended stacking faults on the slip systems with the first and second highest SF, both of which on the same slip system that makes a predicted 129° with the foil edge. The fourth highest SF system was also observed in STEM, with predicted angle with the foil edge of 20° . The DAD model predicted that this system should also deform by decorrelated motion, which was observed in STEM images. Grain 1 on the other hand was predicted, and exhibited non-dissociated full dislocation behavior. STEM analysis indicated that near the nodes of the grain boundary some extended stacking faults formed in grain 1, but limited activation of this secondary mechanism was observed. The analysis of the slip systems in grain 1 was complicated by the curtaining effect perpendicular to the foil edge due to FIB damage during milling.

Since SF analysis confirmed the active slip systems, slip transmission calculations with m' were first conducted assuming slip transmission from grain 1 to grain 2 because grain 1 had a larger SF than any of the systems in grain 2. Transmission could occur from $(-111)[0-1-1]$ system with SF of 0.47 or $(-111)[-1-10]$ system with SF of 0.44 to the largest SF slip systems in grain 2: $(-1-11) [1-10]$. The m' calculations indicate that easy slip transmission ($m'=0.98$) would occur from the highest SF system in grain 1 to the highest SF system grain 2 by full dislocation motion. If slip transmission goes from grain 2 to grain 1, easy slip ($m'=0.98$) would occur for the leading partial dislocation for either of the highest SF systems in grain 2 to the leading partial of the highest SF system for grain 1. Since this dislocation motion is in the reverse sense compared to slip transmission from grain 1 to grain 2, it is expected that partial-partial transmission would occur, thus explaining the formation of extended stacking faults at the nodes of the grain boundary. The m' analysis indicates that both forward and reverse slip could occur across the grain boundary, in both cases slip transmission is very easy since

$m'=0.98$. Since very few observations of partial dislocation motion exist in grain 1, it was concluded that slip is primarily transferring from grain 1 to grain 2.

Figure 7. a) Shear strain accumulation near a grain boundary after 1.46% tensile strain accumulation at 1100 MPa. The STEM foil position is indicated by the gray bar. $\Sigma 3$ boundaries are white, Σn boundaries are gray, and HAGB's are black. b) STEM BF image on grain 2 (112) zone showing the angle the dominant slip planes make with the foil edge. Activation of a second mechanism (full dislocation activity) is seen in grain 2 near the grain boundaries. c) DAD predicted dislocation mechanisms for grain 1 (black) and grain 2 (white). Colors are visible in the PDF on the CD.

Figure 8.a) Grain boundary exhibiting grain boundary sliding, as seen by the circled discrete offsets in grid markers. b) STEM BF image on grain 2 (112) zone showing that the dominant dislocation mechanism in grain 2, extended faults, shows minimal breakdown near the grain boundary. c) DAD predicted dislocation mechanisms for grain 1 (black) and grain 2 (white).

3-D Grain Boundary Morphology

A comparison of the grain boundaries shown in Figure 7 and Figure 8 indicate that there are differences in the boundary tortuosity, and subsequent analysis indicates that this is dictated

by the γ' distribution. Also evident from the STEM images was that the boundary exhibiting grain boundary sliding appears to be more macroscopically planar. Since the STEM analysis only provides limited insight into the 3-D shape of the grain boundary surface, 3-D serial sectioning experiments were conducted to quantify the grain boundary surface morphology. This study has only recently been undertaken, and only the 3-D shape of a boundary that exhibited GBS is presented here.

Figure 9a shows the surface of the grain boundary between two grains which have slid past each other. Also shown are deformation twins which emanate from intragranular carbides. Figure 9a shows that this grain boundary plane contains both macro-scale curvature, which is dictated by the grain boundary triple lines, and micro-scale tortuosity due to the γ' structure. To analyze the micro-scale tortuosity of the grain boundary, a surface was fit to the macro-scale curvature, indicating that a plane surface describes the boundary with an R^2 value of 0.98. The micro-scale topography was calculated as the difference between each data point and the surface fit. In Figure 9b the topological map is overlaid on the original grain boundary surface, and shows the degree to which the local boundary varies from an ideal surface. Most regions vary between ± 120 nm, with maximum and minimum values of +400 nm or -400 nm. Examination of Figure 9b indicates that the deepest trenches are due to the intergranular carbides (indicated by black arrows in Figure 9b and white arrows in Figure 9a), the raised region in the lower right corner of the boundary is due the boundary is being pinned by a third grain.

Figure 9.a) Grain boundary that exhibited GBS (purple) and deformation twins (yellow) emanating from intragranular carbides (white) b) boundary topography is overlaid onto the grain boundary surface. Colors are visible in PDF on CD.

Discussion

The DIC measurements indicated that strain accumulation often occurred at CSL boundaries, $3 \leq \Sigma n < 29$, or triple points. At this point it is unclear if the nature of the boundary, HAGB vs. Σn , causes localizations to occur. It could merely be that statistically most triple points contain at least one Σn boundary because they account for 47% of all boundaries in the standard microstructure. DIC measurements also indicated that only accounting for intensity variations in the strain noise provides an incomplete picture. To understand how strain is accumulating it is important to understand the spatial distributions of the localizations with respect to the microstructure. The initial findings indicate that the serrated microstructure is accommodating a more homogeneous strain field near the boundaries as compared to the standard heat treatment. In an attempt to understand why some boundaries in the standard microstructure exhibited localization while others did not, an m' analysis was conducted, but not confirmed with STEM analysis, on other boundaries that did or did not show strain localization. Though this analysis assumes an idealized scenario, where the local stress state is equivalent to the macroscopic applied stress state so that SF analysis could be conducted, initial findings are encouraging. First, for the two or three highest SF slip systems in each grain the most likely deformation mechanisms were determined. Next, m' analysis was conducted assuming both partial-partial dislocation transfer and full-full dislocation transfer. The preliminary conclusion from this analysis, is that boundaries exhibiting strain accumulation had low m' values, indicating that slip transmission was difficult, while boundaries that did not exhibit strain accumulation typically had several slip systems with high m' values. These initial observations indicate that strain accumulation at boundaries might be primarily a function of grain boundary misorientation and orientation with respect to the loading direction. The results also indicate that boundaries exhibiting GBS also had high m' values, suggesting that slip transmission may facilitate this deformation mechanism. These results are preliminary, and need to be confirmed with additional work. The present set of observations, while providing a more complete view of GBS, and how boundary crystallography dictate associated deformation substructure, are nevertheless very limited in terms of the number of boundaries analyzed. It is still unclear why only some grain boundaries with high m' values exhibit grain boundary sliding while others do not. The m' analysis is also hindered by the assumption that the stress state at the grain boundary is equal to the macroscopic applied stress state, and that the equation does not incorporate any information about the grain boundary plane orientation with respect to the tensile direction. Clearly, additional work is needed to understand how these types of boundaries are different from those analyzed in detail in the present work.

The method of calculating strain associated with GBS used an average displacement from all the sliding events. To accurately apply equation 2, all of the non-sliding boundaries must also be considered, or a new derivation is needed which accounts for all of the discrete measurements instead of an average value. The relationship in equation 3 was derived to account for discrete offsets on active boundaries, so that the strain associated with GBS is the sum of all the discrete sliding events.

$$\epsilon_{GBS} = \frac{\bar{g}}{L^2} \sum s_i n_i \quad (3)$$

Where $\bar{g}$ is the average grain boundary length determined from reconstructed grain boundaries in EBSD analysis, L^2 is the area analyzed, s_i is the sliding displacement, and n_i is the grain boundary normal each respectively projected in the tensile direction. With this equation, the number of events per area comes into play, indicating that the standard and serrated microstructures behaved differently. When strained to similar amounts, the percentage of total plastic strain accommodated by GBS in the standard and serrated microstructures are 6.3% and 0.035% respectively. These values are considerably smaller than those presented in Table II, because equation 2 assumes averaged behavior and that all boundaries are active, which is not the case in these materials. More measurements are being conducted, over larger areas, to confirm that this observed difference is real and not caused by variations in boundary morphology as a function of area analyzed. It is expected that at more traditional creep strain rates, strain accumulation by GBS should be similar or more prevalent since the active intergranular deformation is diffusion mediated.

The serial section data provided valuable insight into the surface structure of a grain boundary that exhibited GBS. An analysis of the reconstructed 3-D data set suggests that the apparent planar boundary in STEM foil was likely a planar surface in 3-D, however, more characterization is required to confirm this hypothesis. The 3-D topography measurements show that the serrations on the boundary have serration amplitude consistent with the secondary γ' particle size. Additional analysis will be conducted to determine if there is any defining serration wavelength or preferred directionality relative to the grain boundary curvature (i.e. is the boundary more or less serrated in the direction of sliding). Comparison between selected grain boundaries may reveal if there are morphological differences between grain boundaries that exhibit GBS versus grain boundaries those that show strain accumulation due to difficult slip transmission. Since macro-scale serrations have been employed to eliminate GBS as a deformation mechanism in the serrated microstructure, it is expected that grain boundary morphology should play a role in the standard microstructure.

Conclusions

DIC provided a unique opportunity to examine the development of strain distributions as a function of strain accumulation. When the tool was coupled with EBSD it allowed for correlation of strain localization sites with microstructural features. When applied to elevated temperature creep of René-104 this technique indicated that the variables dictating strain localization on the sample surface are complicated and include boundary orientation and misorientation. When DIC is coupled with STEM analysis and 3-D serial sectioning, initial observations indicate that strain localization and grain boundary sliding might also be a function of grain boundary tortuosity, but at this point it is inconclusive and needs further work.

STEM analysis was conducted to assess application of the DAD model to predict dominant dislocation mechanisms in the grain interiors. In all cases when decorrelated dislocation motion was observed in STEM the DAD model predicted decorrelated or dissociated partial motion. SF analysis as expected, because of its simplification, did not always correlate with the actual slip system observed in the grain interiors. At this time, further work is being conducted to validate the predicted mechanisms through in-depth

STEM analysis. Since comparisons between experimental observations and DAD model were proven successful, subsequent m' analysis provided a self-consistent explanation of how dislocations transfer between grains and accounted for strain accumulation observed at boundaries in the DIC analysis.

The 3-D serial sectioning was a valuable tool for exploring grain boundary surface structure. A technique was developed to characterize both macro-scale grain boundary curvature and micro-scale grain boundary tortuosity. The analysis indicated that a grain boundary that exhibited grain boundary sliding was planar on the macro-scale and had topography on the order of the secondary γ' particle size.

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References

1. B. Decamps, et al., "On the Shearing Mechanism of Gamma Prime Precipitates by a Single $\langle a/6 \rangle$ Shockley Partial in Ni-Based Superalloys," *Philos. Mag.*, 84 (2004), 91-107.
2. R. R. Unocic, et al., "Mechanisms of Creep Deformation in Polycrystalline Ni-Base Disk Superalloys," *Mat. Sci. and Eng. A*, 483-484 (2008), 25-32.
3. G. B. Viswanathan, et al., "Investigation of Creep Deformation Mechanisms at Intermediate Temperature in Rene 88dt," *Acta Mat.*, 53 (2005), 3041-3051.
4. R. R. Unocic, et al., "Dislocation Decorrelation and Relationship to Deformation Microtwins During Creep of a Gamma ' Precipitate Strengthened Ni-Based Superalloy," *Acta Mat.*, 59 (2011), 7325-39.
5. W. W. Milligan, et al., "Effects of Microstructure on the High Temperature Constitutive Behavior of In100," Superalloys 2004, ed. K. A. Green, et al., (Warrendale, PA: The Minerals, Metals and Materials Society, 2004), 331-339.
6. J. J. Schirra, et al., "Effect of Microstructure (and Heat Treatment) of the 649°C Properties of Advanced P/M Superalloy Disk Materials," Superalloys 2004, ed. K. A. Green, et al., (Warrendale, PA: The Minerals, Metals and Materials Society, 2004), 341-350.
7. H. Grimmer, et al., "Coincidence-Site Lattices and the Complete Pattern-Shift Lattices in Cubic Crystals," *Acta Cryst.*, 30 (1974), 197-207.
8. V. Thaveerungsriporn, et al., "Grain Boundary Deformation of Ni-Cr-Fe-C at 360°C," Sixth International Symposium on Environmental Degradation of Materials in Nuclear Power Systems-Water Reactors, ed. E. P. Simonen, (Warrendale, PA: The Minerals, Metals and Materials Society, 1993), 721-727.
9. U. Krupp, et al., "The Effect of Grain-Boundary-Engineering-Type Processing on Oxygen-Induced Cracking of In718," *Mat. Sci. and Eng. A*, 349 (2003), 213-217.
10. E. M. Lehockey, et al., "Improving the Weldability and Service Performance of Nickel-and Iron-Based Superalloys by Grain Boundary Engineering," *Met. Mat. Trans. A*, 29 (1998), 3069-3079.
11. T. G. Langdon, "Grain Boundary Sliding Revisited: Developments in Sliding over Four Decades," *J. Mater. Sci.*, 41 (2006), 597-609.
12. R. C. Giffkins, "Mechanisms of Intergranular Fracture at Elevated Temperature," *Fracture*, (1959), 579-627.
13. A. Soula, et al., "Analysis of High-Temperature Creep Deformation in a Polycrystalline Nickel-Base Superalloy," *Mat. Sci. Eng. A*, 510-11 (2009), 301-306.
14. H. L. Danflou, et al., "Formation of Serrated Grain Boundaries and Their Effect on the Mechanical Properties in a P/M Base Superalloy," Superalloys 1992, ed. S. D. Antolovich, et al., (Warrendale, PA: The Minerals, Metals and Materials Society, 1992), 63-72.
15. J. Luster and M. A. Morris, "Compatibility of Deformation in Two-Phase Ti-Al Alloys: Dependence on Microstructure and Orientation Relationships," *Met. Mat. Trans. A*, 26 (1995), 1745-56.
16. H. L. Danflou, et al., "Mechanisms of Formation of Serrated Grain Boundaries in Nickel Base Superalloys," Superalloys 1996, ed. R. D. Kissinger, et al., (Warrendale, PA: The Minerals, Metals and Materials Society, 1996), 119-127.
17. A. L. Pilchak, et al., "Anystitch: A Tool for Combining Electron Backscatter Diffraction Data Sets," *J. of Microscopy*, 244 (2010), 38-44.
18. H. K. Alexander and S. Shekhar, "What Does It Mean to Be Special? The Significance and Application of the Brandon Criterion," *J. Mat. Sci.*, 41 (2006), 7675-82.
19. D. G. Brandon, "The Structure of High-Angle Grain Boundaries," *Acta Metall.*, 14 (1966), 1479-84.
20. E. J. Payton, et al., "Semi-Automated Characterization of the Gamma Prime Phase in Ni-Based Superalloys Via High-Resolution Backscatter Imaging," *Mat. Sci. and Eng. A*, 527 (2010), 2684-92.
21. M. D. Uchic, et al., "3d Microstructural Characterization of Nickel Superalloys Via Serial-Sectioning Using a Dual Beam FIB-SEM," *Scripta Mat.*, 55 (2006), 23-28.
22. J. L. Walley, et al., "In-Situ Mechanical Testing for Characterizing Strain Localization During Deformation at Elevated Temperatures," *Exp. Mech.*, DOI 10.1007/s11340-011-9499-7 (2011),
23. A. Soula, et al., "Grain Boundary and Intragranular Deformations During High Temperature Creep of Pm Ni-Based Superalloys," Superalloys 2008, ed. R. C. Reed, et al., (Warrendale, PA: The Minerals, Metals and Materials Society, 2008), 387-93.