Understanding Grain Growth in 3D: Microstructural Evolution of Nickel during Annealing using Three-dimensional X-ray Microscopy

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To my favorite person in the world and my inspiration - my late grandmother Manjulika Chatterjee.
Abstract

This thesis characterizes and quantifies microstructural evolution in nickel. The orientation maps of nickel were collected using a non-destructive, synchrotron based 3D characterization technique called Near-field High Energy X-ray Diffraction Microscopy (nf-HEDM) by the Suter group at Carnegie Mellon University. Six snapshots of the nickel sample were collected after being annealed at 800°C in 30 minute intervals. The temporal changes in crystallographic orientation, grain size and misorientation in the microstructure were measured over the different anneal states. The observed microstructure was also modified to represent a more isotropic system by merging twin related domains into a single grain. Subsequently the microstructures were also meshed and smoothed to determine grain boundary properties as a function of the five grain boundary parameters.

Various morphological and topological features such as grain size, number of neighbors, difference between nearest and average of second nearest neighbors, grain curvature and integral mean grain curvature were analyzed with respect to grain growth. It was observed that a grain’s volume change is best predicted by the difference between the number of neighbors a grain has and the average number of neighbors the neighboring grains have as well as curvature. However, the correlation between neighborhood topology and curvature becomes significant only when adjacent twin related domains are merged.

A study of the influence of grain boundary curvature on grain boundary velocity revealed that they did not show a positive linear correlation. Grain boundary velocity and curvature also varied with crystallographic parameters.
An isotropic simulation used to predict grain growth of the merged twin microstructure did not reproduce the observed microstructure.

A statistical data multivariate model called Canonical Correlation Analysis was used to examine grain growth in both twinned and twin merged microstructures. The evolution of the twin merged microstructure was better predicted than the evolution of the twinned microstructure.

Input parameter sensitivity and interrelationship study revealed that grain curvature, grain radius and neighborhood topology were the most important factors that correlated to grain volume change.
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1 Introduction

In most cases, metals have polycrystalline microstructures with properties that depend on processing conditions such as temperature and prior deformation. Materials science research today is fueled by the goal of optimizing microstructures to elicit the finest performance at the lowest cost. Microstructures are made of a network of grains, the arrangement of which evolves when subjected to high temperatures in such a way that the average grain size increases with a reduction in grain boundary energy and thus area. The final distribution of interfaces strongly influences a number of critical material properties such as strength, hardness, corrosion resistance, magnetic susceptibility [2][3]. However, the characteristics of grains influencing grain growth in polycrystals are still not very well understood. This thesis analyzes experimentally measured microstructural evolution in three dimensions. Factors such as grain radius, number of neighbors, grain shape, grain neighborhood and grain curvature are evaluated to determine if they can predict grain volume change. Grain curvature is measured in two different ways and both are compared to grain volume change. The region of mismatch between two grains has less efficient atomic packing and thus excess free energy and is called a grain boundary. Grain boundaries are the longest known crystal defects and have been poorly understood until recently. Grain boundaries have been found to have a strong dependence on crystallography [4]. Studies of the movement of grain boundaries in bicrystals and simulation have shown that grain boundary velocity is directly proportional to the driving force which is given as the product of grain boundary energy per unit area and the grain boundary curvature. However, this relationship is yet to be investigated in polycrystals. This
thesis explores the structure and properties of grain boundaries especially the relationship between GB velocity and curvature as well as with crystallography. The misorientation between neighboring grains as well as the grain boundary plane orientation dictate grain boundary properties [5]. Understanding the motion of grain boundaries is key to deciphering how microstructure evolution (during recovery, recrystallization and grain growth) occurs. A complete understanding of how grain growth works ultimately will lead to being able to predict it.

1.1 Objectives

The goals of this research work are as follows:

1. Create algorithm to track crystals in successive anneal states based on their volume and crystallographic orientation.

2. Determine the effect of grain radius, number of grain faces, local grain neighborhood and integral mean grain curvature on grain volume changes during annealing.

3. Measure grain boundary velocity and curvature from three-dimensional orientation maps and determine correlations with crystallographic parameters and with each other.

4. Apply Canonical Correlation Analysis to determine if there are characteristics of the grains in the initial structure that are correlated to grain volume changes that occur during annealing.

5. Compare the predictions of isotropic grain growth simulation to the changes in grain volume that are experimentally observed.
1.2 Hypothesis

1. **The volume change of a grain during annealing depends more on the local neighborhood of the grain than its own morphology.**

   This is tested by tracking grains and their neighbors in a polycrystal during multiple annealing steps and correlating the volume change with neighborhood (the difference between the number of neighbors a grain has, $F$, and the average number of neighbors the neighboring grains have, $<F_{NN}>$) and morphological characteristics of the grain (size, the number of neighbors, $F$).

2. **Normalized integral mean curvature of a grain is better than the face-averaged curvature in predicting the grain growth.**

   There are two metrics that can be used to quantify the integral mean grain curvature: $NormCA$ (normalized area weighted grain curvature derived from triangle curvatures) and $G$ (normalized integral mean grain curvature is the summation of turning angles between adjoining triangles and the length of the edge shared between them). Both metrics are used to explain grain growth in the twinned as well as the twin merged microstructures. Correlations of these metrics to volume change is compared to test this hypothesis.

3. **Grain Boundary Velocity shows a strong dependence on crystallographic parameters such as grain boundary misorientation and grain boundary plane orientation. Grain Boundary Velocity shows a strong positive correlation with GB curvature.**

   Experiments and simulations have shown that for bicrystals, grain boundary velocity is directly proportional to curvature [6]. However, it is not known if grain boundaries in polycrystals move in the same way. In a polycrystal, the effects of constraints associated with attachments at triple junctions and the continuity of a curved boundary are currently unknown.
The characteristics (size, curvature, neighborhood) of grains in a hypothetical microstructure constructed by merging annealing twins will be better predictors of grain volume change than the characteristics of grains in the original twinned microstructure.

This hypothesis is tested in two different ways. First, a direct comparison of grain features like radius, neighbors, local neighborhood and curvature and grain volume change in both the twinned microstructure and merged microstructure. The second approach is to use Canonical Correlation Analysis to quantify the total effect of multiple factors in grain growth for twinned and merged microstructures.
2  Background

This chapter focuses on understanding the fundamentals of microstructures including the standard methods of characterizing grains and grain boundaries. It also considers the current understanding of different aspects of grain growth.

2.1  Characterization of Microstructure of Polycrystals

2.1.1  Microstructures

Most solid materials are comprised of a fine cellular substructure which is revealed when examined under a microscope as shown in Figure 2.1. In metals, these substructures form when clusters of atoms crystallize in different orientations during solidification. A region of atoms ordered at the sites of a single crystal is known as a grain (or crystal) while the interface between two impinging grains is called a grain boundary. Grain boundaries are regions of mismatch and can be considered a defect. They are preferred sites for chemical reactions and their motion dictate the final interfacial distribution in a material and thereby its properties. [7][8]
2.1.2 Crystal Structure and Orientation

The atoms or molecules within grains in a polycrystal are ordered and repeated three dimensional patterns. This spatial arrangement of atoms in a crystal is known as crystal structure. The unit cell is an arrangement of atoms that can be repeated along its unit axes to form the entire crystal. The orientation of a crystal’s lattice vectors with respect to a reference frame can be represented in several ways [9]. Some of the most common ones are orientation matrix, Euler angles and Quaternions.

The Orientation Matrix is a 3x3 orthonormal matrix that specifies the crystal coordinate system with respect to the specimen coordinate system. Each row in this matrix corresponds to the direction cosines between x, y and z of the crystal coordinate axis with each of the three specimen axes. The rows and columns are unit vectors.

**Euler angles** can also be used to represent the relative orientation of a grain with respect to a reference coordinate system. The Euler angles are a set of three rotations in a specific order.
about specific axes to rotate a crystal to bring it into coincidence with a coordinate system. These rotations are as follows and shown in Figure 2.2:

1. Positive rotation of \( \varphi_1 \) about \( Z' \) axis.
2. Rotation of \( \Phi \) about \( X' \) in its new position
3. Rotation of \( \varphi_2 \) about \( Z' \) axis in its new position.

The three-dimensional space defined by the coordinates \( \varphi_1 \), \( \Phi \) and \( \varphi_2 \) is called Euler space. Conventionally, the domain of \( \varphi_1 \) and \( \varphi_2 \) is defined between 0 and \( 2\pi \) and the domain of \( \Phi \) between 0 and \( \pi \). Each of the three rotations can be expressed mathematically as:

\[
\begin{align*}
g_{\varphi_1}^{Z'} &= \begin{bmatrix} \cos \varphi_1 & \sin \varphi_1 & 0 \\ -\sin \varphi_1 & \cos \varphi_1 & 0 \\ 0 & 0 & 1 \end{bmatrix} \\
g_{\Phi}^{X'} &= \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos \Phi & \sin \Phi \\ 0 & -\sin \Phi & \cos \Phi \end{bmatrix} \\
g_{\varphi_2}^{Z'} &= \begin{bmatrix} \cos \varphi_2 & \sin \varphi_2 & 0 \\ -\sin \varphi_2 & \cos \varphi_2 & 0 \\ 0 & 0 & 1 \end{bmatrix}
\end{align*}
\]

Multiplying these three matrices gives us the orientation matrix \( g \):

\[
g(\varphi_1, \Phi, \varphi_2) = \begin{bmatrix} c\varphi_1 c\varphi_2 - s\varphi_1 s\varphi_2 c\Phi & s\varphi_1 c\varphi_2 + c\varphi_1 s\varphi_2 c\Phi & s\varphi_2 s\Phi \\ -c\varphi_1 s\varphi_2 - s\varphi_1 c\varphi_2 c\Phi & -s\varphi_1 s\varphi_2 + c\varphi_1 c\varphi_2 c\Phi & c\varphi_2 s\Phi \\ -s\varphi_1 s\Phi & c\varphi_1 s\Phi & c\Phi \end{bmatrix}
\]

Here \( c=\cos \), \( s=\sin \) [10].
2.1.3 Misorientation

Misorientation refers to the separation between two orientations in the three-dimensional crystallographic space. If two grains have their orientation given in Euler angles, the misorientation between them is:

$$\Delta g = g_1 g_2^T$$

(3)

If either crystal is transformed by any of the permissible symmetry operators, then we obtain an indistinguishable misorientation. Accounting for the crystal symmetries, we get misorientation as:

$$\Delta g = C_{p1} g_1 (C_{p2} g_2)^T$$

(4)

($C_{p1}$ and $C_{p2}$ are the symmetry operators for each crystal). And doing the same to the other grain;

$$\Delta g = C_{p2} g_1 (C_{p1} g_2)^T$$

(5)

A common way of representing misorientations in physical metallurgy is to use an angle-axis pair. The same crystal coordinate system rotation or misorientation ($\varphi$) can be achieved by rotation around a specific axis (axis of rotation) and through a specific angle (angle of rotation).

The minimum misorientation is called the disorientation.

Using the rotation matrix, the axis and angle of rotation can be written as:

$$\cos \varphi = \frac{1}{2} (g_{11} + g_{22} + g_{33} - 1)$$

(6)

Where $g_{11}, g_{22}, g_{33}$ are the diagonal values of the orientation matrix.

The axis of rotation, [uvw], is given as:
\[ u = \frac{(g_{23} - g_{32})}{2\sin \omega}; \quad v = \frac{(g_{31} - g_{13})}{2\sin \omega}; \quad w = \frac{(g_{12} - g_{21})}{2\sin \omega}; \]

Other representations of orientation (and misorientation) like Rodrigues Frank vector and Quaternions are convenient for numerical calculations, but are not as intuitive as the representations discussed here [9].

### 2.1.4 Stereographic Projection

The stereographic projection is used to project three dimensional angular relationships like crystallographic directions and misorientations in two dimensions. It is used in generating pole figures and inverse pole figures among other things. The biggest advantage of stereographic projections is that all the angular crystallographic relationships are preserved in the projection. This is not a new problem as its application is well known in the representation of geographic features of Earth in 2D maps.

![Stereographic projection of {100} poles of a cubic crystal.](image)

*Figure 2.3. Stereographic projection of \{100\} poles of a cubic crystal. (a) shows the crystal in a unit sphere, (b) shows the projection of the \{100\} poles on equatorial plane and (c) shows the top view of the equatorial plane with the pole figure angles \(\alpha\) and \(\beta\). Figure reprinted from [11]*

Any direction vector in a 3D crystal can be represented in the sphere as shown in Figure 2.3. The figure shows the reference sphere of unit radius with the crystal positioned at the center. TD, ND and RD are Transverse Direction, Normal Direction and Rolling Direction respectively as they are the three most commonly used terms to identify orthogonal directions in a crystal.
This projection is given by $r \tan \angle OSP$, $r$ being the radius of the sphere [9]. Figure 2.4 shows a reference sphere. If we imagine a crystal at the center of the sphere at O, then the normal through a certain face of the crystal passes through the reference sphere at say P' which is called the pole of the plane. When this pole P' is projected to the southern hemisphere at S, it passes through the equatorial plane at point P. P is the stereographic projection of the pole P' [9].

While the Pole Figure (PF) represents the orientation of the crystal coordinate system in the specimen coordinate system, the Inverse Pole Figure (IPF) represents the specimen coordinate system in terms of the crystal coordinate system.

Mathematically if specimen coordinate system is denoted by $\{s_1, s_2, s_3\}$ and crystal coordinate system by $\{c_1, c_2, c_3\}$ and the coordinates of the Pole are $(X, Y, Z)$. Assuming $R$, a vector parallel to the Pole, then it can be expressed in both the crystal and specimen coordinate systems as the following:

Using the angles $\alpha$ and $\beta$ from Fig. (c), we get:

$$R = s_1 \sin \alpha \cos \beta + s_1 \sin \alpha \sin \beta + s_1 \cos \alpha$$

$$R = \frac{1}{N}(c_1X + c_2Y + c_3Z)$$
Here $N = \sqrt{X^2 + Y^2 + Z^2}$, which is used to normalize the vector R.

Scalar multiplication gives:

\[
\begin{pmatrix} \sin \alpha \cos \beta \\ \sin \alpha \sin \beta \\ \cos \alpha \end{pmatrix} = \frac{1}{N} \begin{pmatrix} g_{11} & g_{21} & g_{31} \\ g_{12} & g_{22} & g_{32} \\ g_{13} & g_{23} & g_{33} \end{pmatrix} \begin{pmatrix} X \\ Y \\ Z \end{pmatrix}
\]

This equation results in nine equivalent expressions to calculate $\alpha$ and $\beta$ for a given $(X,Y,Z)$.

$g$ is the orientation matrix [12].

Similarly, for Inverse Pole Figure, we get:

\[
\begin{pmatrix} \sin \gamma \cos \delta \\ \sin \gamma \sin \delta \\ \cos \gamma \end{pmatrix} = \begin{pmatrix} g_{11} & g_{21} & g_{31} \\ g_{12} & g_{22} & g_{32} \\ g_{13} & g_{23} & g_{33} \end{pmatrix} \begin{pmatrix} x_s \\ y_s \\ z_s \end{pmatrix}
\]

Here, $\gamma$ and $\delta$ are the angles of a vector parallel to the specimen axis in the crystal coordinate system.

### 2.2 Grain Boundaries

As described in the previous section, a grain is the region of space occupied by a continuous crystal lattice. The surface formed between two contiguous grains of different orientations is called a grain boundary. Grain boundaries (Figure 2.5) are lattice defects of only a few atomic diameters width that are created when two crystals of the same crystal structure, but different orientation come together in a polycrystalline material [3].
Grain boundaries are objects of much interest because they are different from the rest of the grain. [13]. Microstructure of any material is characterized by size, shape and crystal orientation of its constituent grains. Presence of different phases and other impurities are also important factors. All these features depend on the nature and positioning of the grain boundaries. Consequently, grain boundaries directly control the material properties. For example grain boundaries act as obstacles to dislocations and thus help strengthen a material; they are also a potential site for impurity segregation and ensuing corrosion [14][15]. Larger grains have less grain boundary area than smaller grains for a fixed volume of material. This affects the strength, toughness and ductility of a material due to the Hall-Petch relation [16][17]. Thus, understanding grain boundary structure is very important in connecting the gap between
material and property and their study makes for an important part of material science and physics.

2.2.1 Grain Boundary Parameterization

There are five independent macroscopic parameters that distinguish each grain boundary from the other. These include three parameters for describing the lattice misorientation ($\Delta g$) between the two grains sharing a boundary and two parameters for grain boundary plane orientation ($n$). [5].

We have already talked about $\Delta g$ (Section 2.1.3), now we shall understand the $n$.

The grain boundary plane space is parameterized with a unit normal or two spherical angles $\theta$ (polar) and $\phi$ (azimuthal) as shown in Figure 2.6.

![Diagram of grain boundary parameterization](image)

*Figure 2.6. Spherical angles $\theta$ and $\phi$ made by the vector $n$ at P illustrate the parameters for the grain boundary normal vector.*

For every $\Delta g$, $n$ is given as:

\[ n_{ij} = C_{p1g} n_{ij}' \text{ or } n_{ij} = C_{p2g} n_{ij}' \]

depending on how $\Delta g$ is calculated.

Since the normal can point into the first or second crystal, the total number of symmetrically
equivalent boundaries that can be formed from a single interfacial segment are $2 \times 2 \times P^2$. [5]

### 2.2.2 Bicrystal Symmetries

Even after specifying a grain boundary by its 5 degrees of freedom (DOF), it may not be unique owing to symmetry. It is seen that the distribution of grain boundary planes about a single misorientation have symmetry such that it renders looking at all possible grain boundary planes unnecessary. Grain boundary plane orientation symmetry seen with respect to a boundary interface of two crystals in three dimensions is called Bi-crystal symmetry [16]. It is found that in general, the grain boundary planes of a bicrystal for a fixed misorientation can be uniquely defined in a hemisphere [18]. When the rotational symmetry elements of the two crystals are parallel, we get more symmetry from the combining rotational symmetries with other operations that generate equivalent interfaces [19].

FCC crystals have 24 symmetrically related orientations. While, each orientation is mathematically distinct, it is physically common. Thus, to remove duplication and thus confusion, the grain boundaries are viewed in a reduced orientation space (reduced by a factor of 24) which is known as the Fundamental Zone (FZ) for cubic symmetries. [18]

The shape of FZ in Rodrigues space for cubic symmetry is shown in Figure 2.7.
It is a cube with truncated corners which form equilateral triangles and octahedra. This shape is determined by the crystal symmetry of the material [20][21].

2.2.3 Types of Grain Boundaries

Grain Boundaries are essentially lattice defects that occur between two crystals of different orientation. They are still being understood because being complex structures that require extensive analytical and macroscopic characterization.

There are several ways of defining a boundary. As shown in Section 2.1.2 the orientation relationship is a transformation to make one grain coincide with the other. It can be represented in Euler angles or by angle/axis method. The angle/axis method being more intuitive will be used to understand tilt and twist boundaries.
Imagine two grains sharing a boundary such that to bring one into coincidence with the other, one grain as to be rotated about the rotation axis which is perpendicular to the grain boundary plane as shown in Figure 2.8(a). This is called a twist boundary. Here, the grain boundary plane is fixed irrespective of the rotation angle.

If, however, the grain boundary plane is parallel to the rotation axis as shown in Figure 2.8 (b) and (c), we get a tilt boundary. Here, the grain boundary plane has infinite options while still being parallel to the rotation axis. It is called a symmetrical tilt boundary (Figure 2.8(c)) if the
two grains are mirror images of each other. Thus, annealing twins discussed in Section 2.4.2 are also symmetrical tilt boundaries. If they are not mirror images of each other then it is known as asymmetrical tilt boundary (Figure 2.8(b)). There is well documented evidence of the dependence of grain boundary energy on the tilt angle of symmetrical tilt boundaries as well [22].

Another way to classify grain boundaries is by their atomic structure. Based upon the angle of orientation between two crystallites, we can categorize the grain boundaries by their misorientation angle: Low angle grain boundaries (LAGB) and High angle grain boundaries (HAGB). Figure 2.9 illustrates the misorientation between two crystals at the grain boundary [23][24][25][26]. The region between the two grains in Figure 2.9(a) is a region of distorted lattice. The angle between grains at Low angle grain boundaries is typically lower than 10°-15°. Their properties depend on the misorientation angle and the structure can be created by an array of dislocations.
Symmetric tilt boundaries consist of parallel edge dislocations and the energy of these boundaries is easily determined. It is seen from Fig 19(b) that the dislocation spacing $d$ decreases with tilt angle $\varphi$ and their relation is shown as:

$$\frac{b}{d} = 2 \sin \frac{\varphi}{2} \approx \varphi$$

Where $b$ is the Burgers vector and $d$ is spacing between the dislocations. Burger vector $b$ is a function of lattice parameter. Thus, lattice misorientation determines the set of geometrically necessary dislocations required to construct a low angle grain boundary (LAGB).

As misorientation angle $\varphi$ increases the dislocations get closer and closer until they lose their identity as they overlap. The dislocation model falls short at higher angles and the equation above is not valid anymore. These boundaries are called HAGB (Figure 2.10). Grain boundary energy for these boundaries is seen to be independent of rotation angle.
2.2.4 Coincident Site Lattice theory (CSL)

Unlike low angle grain boundaries, high angle grain boundary structures aren’t very well known. Although the atomic structure of a HAGB is more complex than a LAGB, ways have been found to simplify their understanding and the Coincident Site Lattice theory (CSL) does just that. The development of the CSL theory is a milestone in the study of grain boundaries in the last few decades.

This model was first proposed in 1926. The idea is that if two grains were superimposed on each other on all possible combinations of angle of misorientation about the rotation axis, it was found that for some specific angles, there was good coincidence between the lattice positions for both crystals. This idea becomes physically real at grain boundaries. Thus, at specific angles there is a high density of coincidence between grain boundary lattice points [27][28][29][30]. In Figure 2.10.
2.11 we see a rotation of the second lattice <100> axis by 36.87° called Σ5 brings into partial coincidence many sites (triangles within circles) of either lattices (denoted each lattice by triangles and circles) to create a superlattice (CSL) of its own. This is a 2D example, however, in real it is a three-dimensional lattice.

Each CSL boundary is defined by the degree of coincidence and expressed in units of Σ. Σ is defined as the normal lattices per coincidence lattice site:

\[ Σ = \frac{Volume \ of \ elementary \ cell \ of \ CSL}{Volume \ of \ elementary \ cell \ of \ crystal \ lattice} \]

One of the most common CSL boundaries are Σ3 boundaries which means every third lattice is coincident. These boundaries are also known as “special boundaries” in polycrystalline materials. They include both coherent and incoherent annealing twins as well as other boundaries with a Σ3 misorientation which is 60° [31] [9] [32] [33] [11].

Another low CSL boundary of interest is the Σ7 boundary. These boundaries have misorientation of 38.2°. The Σ7 tilt boundary has been observed to have the highest mobility [6].
This good fit sometimes translates to improved properties in a material such as energy and mobility. Thus, this new superlattice formed from the coincident sites of the two disoriented lattices is called the Coincident Site Lattice. Aust and Rutter et al. noted that some grain boundaries of high coincidence in Pb-Sn alloys migrated much faster than the rest [34]. The most important take-away is that CSL theory applies to real grain boundary behavior. Since grain boundaries are effectively defects, they have excess energy owing to their non-equilibrium structure. Brandon et al found that “the reciprocal density of common lattice points in a boundary is not a measure of the actual degree of fit”. This density only refers to the 2D surface of the boundary. They extended the dislocation model of Read and Shockley and the coincidence site lattice model of Kronberg and Wilson to note that the density of coincidence site at the boundary...
limits the number of dislocations that can be introduced at the boundary without breaking the coincidence. They concluded that their model has a sub boundary in the coincidence site lattice when superimposed with a dislocation network. The boundary was found to be stepped and non-planar and anisotropic with regions of disorder interspersed with regions of coincidence [35][36].

Hasson et al measured interfacial free energies of [001] and [011] tilt boundaries of high purity aluminum. They found that interfacial energies drop sharply called deep energy cusps corresponding to $\Sigma 3$ on 111 planes (at 70°) seen in Figure 2.12. Other studies revealed shallow cusps at $\Sigma 9$ and $\Sigma 27$. However, the grain boundary at 109° is also a $\Sigma 3$ but does not have low energy. This has led to the phenomenon of grain boundary engineering where thermomechanical processing is implemented to increase the population of special boundaries in order to improve properties of polycrystalline materials [37][28]. However, Goodhew et al showed that CSL theory for energy of boundaries could not predict the same for tilt boundaries. It was also concluded later by other studies as well that $\Sigma$ value may not be a good predictor of grain boundary energy [38][39][40].
2.2.5 Topology of grains and grain boundaries

A very interesting thing about microstructure is that arrangement of grains in a polycrystalline material is not random but a result of topological constraints. Since grain boundaries are defects, they are thermodynamically unstable. Thus the low and high angle boundaries are rearranged to fit the topological requirements of space-filling and surface tensions[14][41]. The microstructure consisting of vertices, edges, faces and grains obeys the topological rule of Euler characteristic (Figure 2.13). According to this formula:

\[
1 = V - E + F \quad \text{(for 2D plane)} \quad (7)
\]

\[
1 = V - E + F - C \quad \text{(for 3D space)} \quad (8)
\]

Each is number of: vertices(V), faces(F), edges(E), cells(C)

For example, in a cube: \( V = 8; F = 6; E = 12, C = 1 \), thus \( 8 - 12 + 6 - 1 = 1 \)

According to C.S. Smith, “Normal grain growth results from the interaction between the topological requirements of space-filling and the geometrical needs of surface tension equilibrium” [42].

\[\text{Figure 2.13. Schematic of 2D section of a microstructure depicting the faces, vertices and edges of grains. Reprinted from [3]}\]
2.2.6 The Grain Boundary Character Distribution (GBCD)

While grain boundaries can be uniquely defined in 5D space [31], viewing them in 5D space is a challenge and thus we normally look at an assortment of projections of these distributions. These are called the Grain Boundary Character Distributions. It shows the relative areas of grain boundaries as a function of their lattice misorientation and GB plane orientation. Some of these distributions are shown in Figure 2.14 for pure Nickel. Figure 2.14 (a) shows the distribution of the grain boundary disorientation angles. The actual distribution is shown by the blue circles while the red squares represent a random distribution. There is a distinct peak at $60^\circ$ and then at $39^\circ$. The significance of the random disorientation distribution is that in 1958 Mackenzie showed that if a large number of orientations are randomly sampled in an orientation space, the disorientation angle would show a peak at $45^\circ$. This is also called the Mackenzie plot [43].

GBCD computation requires parameterization of the five-dimensional space of grain boundary types. In 2002, Rohrer et al. used Euler angles of the disorientation and the two spherical angles for grain boundary planes for this parameterization. They could segment the 5D space into equal volume parts with an average angular resolution of $10^\circ$ for each parameter. The area of each grain boundary plane was added to its corresponding discrete configuration followed by normalization with total area to obtain the values in MRD [44]. Another method of calculating the GBCD was given by Morawiec et al. in 2014. In this method, they applied a kernel density estimation method which calculates the distribution at points by summing the areas of boundaries that fall within a certain distance of that point. This method eliminates certain artifacts borne by the partition method and shows sharper peaks and better smoothness[45].
Figure 2.14 (b) shows the grain boundary plane distribution (GBPD). This is a distribution of the grain boundary area as a function of the GB plane orientation irrespective of disorientation information. They are represented in a standard stereographic triangle in a crystal reference frame. The distributions are plotted in units of Multiples of Random (MRD) which denotes the relative areas in a normalized distribution. The understanding is that MRD value greater than 1 signifies observation of grain boundary types that are greater than expected. Figure 2.14(b) shows that the grain boundary area peaks at the (111) position, meaning that there is a preference for (111) planes.

Figure 2.14. Holistic view of grain boundary character distribution. (a) shows the disorientation distribution and the break at 60° shows a huge peak. (b), (c), and (d) are stereographic projections of relative boundary areas in units of Multiples of Random (MRD). These distributions tell us that there is a peak at 39° and 60° in the disorientation of grain boundary, and
there is a strong peak at (111) around 60° disorientation. These are the coherent Σ3 twin boundaries commonly found in fcc materials [46].

Figure 2.14 (c) shows the distribution of grain boundaries planes with a disorientation of 60°.

The most commonly occurring boundaries are (111) meaning most boundaries that have a 60° disorientation around [111] [47].

In Figure 2.14 (d) we look at the distribution of all grain boundaries with a misorientation of 60° around [111]. There is a strong peak at the (111) position which corresponds to the coherent twin position common among fcc metals.

2.2.7 Grain Boundary energy distribution

Grain boundaries are essentially non-equilibrium defects. They possess excess free energies. It is due to this variation in energy that causes grain boundary motion. Grain boundary energies have been known to vary with crystallography [48]. Thus measure boundary energy is of paramount importance to being able to understand microstructural evolution.

The energy of LAGBs can be calculated using the misorientation angle since it is essentially an array of dislocations. Then energy of this boundary is given by the following equation [25]

\[
\gamma_s = \gamma_0 \varphi (A - \ln \varphi)
\]  

(9)

Here \(\gamma_0\) is the energy of one dislocation = Gb/4\(\pi(1-v)\) and A = 1+ln(b/2\(\pi r_0\)), \(r_0\) being the radius of dislocation core. From equation 2 we can see that energy of low angle grain boundaries increases with increase in misorientation or decrease in \(h\) [49].

Suppose the energy required to create two new surfaces is \(2\gamma_s\) and the bonding energy between the two surfaces created is \(B\), then the grain boundary energy can be written as:
\[ \gamma_{gb} = 2\gamma_s - B \quad (10) \]

Mullins approximated \( \gamma_{gb} \) to be in the range of 0.5-1.0J/m\(^2\) [50]. The driving force for grain growth is given by this excess energy [51]. Grain boundary energy is also anisotropic as argued by Smith from the measured dihedral angles [14]. This anisotropy is due to the difference in the microscopic structure of boundaries and can be because of either \( \gamma_s \) or \( B \). Grain boundary energies were measured using Herring’s rule for equilibrium of interfacial forces at a triple line [52]:

\[ \gamma_i t_i + \frac{\partial \gamma_i}{\partial \beta} n_i = 0 \quad (11) \]

\( \gamma_i \) is the energy of the \( i \)th interface, \( t_i \) and \( n_i \) are the tangential and normal forces, \( \beta \) is the rotation angle (see Figure 2.15).

![Figure 2.15. Interfacial energies, tangential and normal forces at equilibrium at a triple line.](image)

The interface with higher energy will tend to pull the triple line along itself, thereby destroying itself and replacing with lower energy interfaces in the process. The differential of the energy
with respect to the rotation angle arises due to anisotropy and causes a torque to rotate the triple line in a lower energy direction. The Herring equation can be simplified to the Young’s equation as follows [53]:

\[
\frac{Y_1}{\sin \theta_{2,3}} = \frac{Y_2}{\sin \theta_{1,3}} = \frac{Y_3}{\sin \theta_{1,2}}
\]

However, to accurately map the anisotropy, Herring’s equation in its complete form needs to be used. Several studies measuring the variation of relative grain boundary energy with grain boundary misorientation or grain boundary plane orientation indicate the existence of anisotropy in grain boundary energy. Using automated methods for determining grain boundary energies for all grain boundaries it has been found that anisotropy is greater with variation in grain boundary plane orientation than lattice misorientation [44][54][55][56][57]. It is also found that there is an inverse correlation between grain boundary energy and grain boundary population. A detailed comparison between calculated and observed grain boundary reveals strong correlation between the two [58][59][60][61].

2.3 Grain Boundary Geometry-Boundary Curvature

Grain Boundary geometry is believed to have a profound effect on grain boundary movement.

One of the most important aspects of geometry is curvature. Curvature can be defined as a quantity that defines the amount by which a curve deviates from a straight line or a surface deviates from a plane. To derive curvature, we first need to define an arc, arc length and how we are parameterizing the curvature by arc length.

Let us define the arc length with respect to a parameter \( t \):

\[
r(t) = x(t)\hat{i} + y(t)\hat{j} + z(t)\hat{k}
\]

If \( r \) is moving from \( a \) to \( b \) and is differentiable, arc length \( s \) is given by the following equation:
\[
\begin{align*}
    s &= \int_a^b \sqrt{(\frac{dx}{dt})^2 + (\frac{dy}{dt})^2 + (\frac{dz}{dt})^2} \, dt = \int_a^b |\mathbf{v}(t)| \, dt \\
    \text{Thus, we can define the arc length function } s(t) \text{ as } \\
    s(t) &= \int_0^t ||\mathbf{v}(u)|| \, du
\end{align*}
\]

Before we get to curvature, we first look at the tangent to a curve. Since the tangent is barely touching the curve at a point, the slope of a tangent gives the instantaneous slope of a curve at that point. This is shown as follows:

\[
    T = \frac{dr}{dt}
\]

And unit tangent vector is given as:

\[
    \hat{T} = \frac{T}{|T|} = \frac{dr/dt}{|dr/dt|}
\]

Now, \(dr/dt\) is velocity of the moving point in time and thus we can say that \(|dr/dt|\) is the speed, or the magnitude velocity. Thus \(\hat{T} = \frac{velocity}{speed} = \frac{dr/dt}{ds/dt}\)

Also, if the arc length \(s\) is parameterized in terms of \(s\) instead of \(t\), the tangent vector becomes:

\[
    T = \frac{dr}{ds}
\]

And

\[
    \hat{T} = \frac{T}{|T|} = \frac{dr/ds}{|ds/ds|} = \frac{dr}{ds}
\]

Curvature is the measure of the degree of deviation of a curve from a straight line. In other words, it tells us how the tangent at a point is changing as the point travels along the curve. Thus, it is the rate of change of the tangent:
$$k = \left| \frac{d^2r}{ds^2} \right| = \left| \frac{d\hat{T}}{ds} \right|$$

We can also write it in terms of the parameter $t$ (using chain rule):

$$k = \left| \frac{d\hat{T}}{dt} \cdot \frac{dt}{ds} \right|$$

$$= \frac{1}{|ds/dt|} \left| \frac{d\hat{T}}{dt} \right|$$

$$= \frac{1}{|v|} \left| \frac{d\hat{T}}{dt} \right|$$

If $T(t)$ is defined as the unit tangent vector, then

$$k = \left| \frac{d}{ds} (T(t)) \right| = ||r''(s)||$$

Here, $k$ is defined as the rate of change of tangent $T$ with respect to time $t$ and by corollary arc length $s$.

With the help of chain rule we can write it in terms of $t$:

$$\left| \frac{d}{ds} (T(t)) \right| = \left| T'(t) \frac{dt}{ds} \right|$$

$$\frac{||T'(t)||}{||ds/dt||} = \frac{||T'(t)||}{||r'(t)||}$$

This gives $K(t)$ as:

$$K(t) = \frac{||r'(t) \times r''(t)||}{||r'(t)||^3}$$

For a plane curve, if we assume that the curve is in the $xy$-plane and defined as $y=f(t)$, such that

$$r(t) = ti + f(t)j$$

Then curvature is given as:
\[ K(t) = \frac{|f''(t)|}{\left[1 + (f'(t))^2\right]^{3/2}} \]

There are several methods of measuring curvature of grains and grain faces. In this thesis, we have used two methods namely Face-averaged curvature \( (M_S) \) [4] and normalized integral mean curvature \( (\mathcal{G}) \) [62]. Their derivation is discussed in more detail in Chapter 4.

### 2.4 Recrystallization, Recovery and Grain Growth

Annealing is a heat treatment that involves heating a material above its recrystallization temperature for a suitable amount of time and then slow cooling it. The three stages of annealing are classified as recovery, recrystallization and grain growth. Recovery occurs in a deformed microstructure and it marked by reduction in stored energy and annihilation of defects. It is followed by the nucleation of stress free grains which is called recrystallization. The stress free grains grow until the stressed grains have been consumed. When upon further heating, these new stress free grains keep growing, it is called grain growth. This results in further reduction of grain boundary energy and reduction in total grain boundary area. These processes improve the properties of a material by making it more ductile and tough. In the current study recovery and recrystallization are irrelevant so we will move on to grain growth directly.

Annealing a polycrystalline material at high temperature leads to grain growth, during which some grains shrink and are eliminated and the average grain size increases [41][63][64]. Grain growth can be classified as normal grain growth and abnormal grain growth (AGG). In AGG, a few grains grow abnormally large consuming every small grain on its way. This leads to a bimodal grain size distribution for the AGG.

Several factors may affect grain growth such as: Temperature: the higher the temperature, higher is the mobility of grain boundaries resulting in significant grain growth. Presence of
impurities/solutes also adversely affect grain growth. Precipitate particles tend to pin grain boundaries and slow down grain growth. The rate of grain growth is also diminished when the grain size is bigger than the thickness of the sheet sample. The higher the grain size, the lesser is the driving force. Texture also plays a part in influencing grain growth. A highly textured microstructure consists a high concentration of low angle boundaries, which reduces the driving force for grain growth [3].

2.4.1 Classical Theories of Grain Growth

The earliest studies that focused on individual grains were quasi two-dimensional and used soap froths or polycrystalline succinonitrile as proxies for inorganic microstructures because their transparency made it possible to make dynamic observations. Early studies of grain growth were also usually limited to one or two-dimensional cases.

C.S. Smith examined the growth of cells in a two-dimensional soap froth formed in a small flat glass cell and remarked upon the positive correlation between a cell’s size and its number of near neighbors [14][65]. It was realized that the process occurred to eliminate excess energy while also balancing topological requirements for space-filling and local interfacial equilibrium at triple junctions [42][66].

In 1965 Hillert and Feltham came up with a statistical grain growth model, commonly known as the drift model, which predicts a value of a critical radius ($R_{\text{crit}}$) below which a grain should shrink and above which it should grow (equation 18). Hillert showed that the mean grain size was equal to this critical size owing to topological constraints. He also obtained grain size distributions and concluded that a distribution with grain size greater than $1.8\bar{R}$ would result in abnormal grain growth, which was later found to be incorrect [67][68].
Burke and Turnbull worked on the kinetics of grain growth and deduced a parabolic growth law for both 2D and 3D systems. They found the following equation:

\[
\frac{dR}{dt} = cM\gamma_b \left( \frac{1}{R_{\text{crit}}} - \frac{1}{R} \right)
\]

Here \( n \) is called the grain growth exponent, and ordinarily equals 2. They considered an isotropic model and assumed grain growth is driven only by boundary curvature. They also considered the radius of curvature to be proportional to the mean radius \((\bar{R})\) [69]. Other extensive studies have revealed that \( n \) may vary in the range 2 to 4.

In 1970, Aboav [70] recognized empirically that grains in 2D sections were arranged such that the number of neighbors of a grain \((F)\) is related to the average number of neighbors of the neighboring grains, \(\langle F_{NN} \rangle = 5 + 8/F\). This eventually resulted, after some theoretical advancements, in what is now known as the Aboav-Weaire (AW) [71][72] law:

\[
\langle F_{NN} \rangle = (\bar{F} - 1) + \frac{(F + \mu_2)}{F}
\]

where \( \bar{F} \) is the average number of neighbors and \( \mu_2 \) is second moment of the distribution of grain nearest neighbors (faces). Noting that 3D microstructure studies find a strong positive correlation between \( F \) and the grain size [73][62][74][75], one can interpret the AW law as simply saying that large grains (those with many neighbors) are on average surrounded small grains (those with fewer neighbors) and vice versa. In the context of grain growth, the AW law emphasizes that the size and shape of a grain is related to its neighbors and it has successfully described different kinds of cellular patterns [72] in steady state and transient structures. [76]
Von Neumann deduced a theorem that shows how evolution of a cellular structure is governed by topological changes. It is famously called the Mullins’ “n-6” rule and is given as:

$$\frac{dA}{dt} = k \frac{\pi}{3} (n - 6) \quad (16)$$

This equation implies that in 2D grains with more than 6 sides will grow and fewer than 6 sides will shrink; growth rate depending on the number of sides [51].

This system tends to minimize its energy by shortening the edge lengths between any two vertices. Assuming isotropic properties of grain boundary energy, integral mean curvature dictates the growth of a grain.

Equation 18 was extended to three dimensions by MacPherson et al. Assuming isotropic properties again, they derived the following equation:

$$\frac{dV}{dt} = -2 \pi M \gamma \left( L(D) - \frac{1}{6} (\sum_{i=1}^{n} e_i(D)) \right) \quad (17)$$

The quantity $L(D)$ is called the mean width of domain $D$, $e_i$ is the length of triple lines, and it is being summed over all the n triples lines [77].

The first extensive observations of three-dimensional (3D) grain growth dynamics were produced by computer simulations, which assumed isotropic grain boundary properties. 3D grain growth simulations using the Monte Carlo Potts model [78], boundary tracking methods [79], the vertex method [79], and the phase field method [80] make it possible to study isotropic grain boundary motion in three dimensions. These studies verified the idea that the distribution of grain sizes normalized by mean grain size was time invariant during normal grain growth.

Those studies that compared $\langle F_{NN} \rangle$ and $F$ for the simulated 3D structures found a functional relationship consistent with the AW law [78][79]. While computer models have been used to simulate 3D microstructural evolution during grain growth, experiments have been more
difficult. It was found that grains with number of faces less than 15.5 were expected to shrink and ones more than 15.5 to grow. Analytical theories suggest that the minimum integral mean curvature should occur for a grain with 13.4 neighbors [62]. The difference was explained on the basis of consideration of the grains’ nearest neighbors, thereby focusing on the substantial role of nearest-neighbors within a grain network required for grain stability.

Palmer et al. [81] verified the Mullins-Von Neumann "N-6" rule [51] by observing grain growth in succinonitrile. McKenna et al. compared the experimentally measured morphology of grains of β-Ti after a single anneal period, to the simulated one using a phase field model assuming isotropic grain boundary properties and predicted that for isotropic grain boundary properties this model will provide an accurate prediction of the evolution of the polycrystalline system. They reported good agreement between the two in many areas of the polycrystalline structure [82].

There has also been a recent report of matching grains between microstructures reconstructed by two different techniques [24]. The two data sets shared a common volume and had different orientation reference frames. The sampling grids were aligned so that the reconstructed grains coincided. Grains that did not coincide were eliminated. An algorithm was then employed to match the data sets on a grain-by-grain basis. The difference in orientations between matched grain pairs was used to better align the sampling grid which allowed for accurate voxel to voxel comparison thus providing a means to evaluate the accuracy of each technique.

Another study non-destructively measured 3D grain growth (using 3DXRD) and comparing between two anneal states, explored correlation between grain orientation, growth of grains and neighboring relationships [25].

Grain boundaries when annealed in a recrystallized system, migrate towards the center of their curvature [83]. Grain boundaries follow the rule of motion of plain curves which posits that the
boundary moves towards its center of curvature at a speed proportional to the curvature in an ideal case of a pure metal [51]. The driving force for grain growth is a function of grain boundary curvature and grain boundary energy. In this work, we shall explore in some detail the relationship between grain boundary curvature and grain boundary velocity in experimentally measured three-dimensional nickel polycrystal microstructure data.

Initially C. S. Smith studied bubble growth in an aggregate of soap froth and showed that diffusion through a bubble wall is proportional to the mean curvature of the wall [14]. According to Von Neumann and Mullins, volume growth rate is proportional to curvature by the following equation:

\[
\frac{dV}{dt} = -M \gamma \mathcal{H}
\]  

\[\mathcal{H} = \int_{\text{faces}} \left( \frac{1}{R_1} + \frac{1}{R_2} \right) dS \]

M is the mobility constant; \(\gamma\) is the interfacial energy; and \(\mathcal{H}\) is the integral mean curvature of the grain faces [51][84][62]. Assuming M and \(\gamma\) to be constants, the volume growth rate is directly proportional to the negative curvature, i.e. grains with negative curvature will have a positive growth rate and grains with positive curvature will shrink. If equation 20 is divided by the equivalent radius, we get:

\[
\frac{dV}{dt} \frac{1}{V^{1/3}} = -M \gamma \mathcal{G}
\]

Where \(\mathcal{G}\) is the normalized integral mean curvature of the grain faces.

There have been extensive studies to understand the workings of complex microstructures. Recent developments in three dimensional data characterization has allowed us to understand grain boundaries by measuring grain data such as grain size, topology, orientation, grain boundary populations in 3D [62][44][1]. Zhong et al. found that the mean grain boundary
curvature varies with grain boundary crystallography and is more sensitive to grain boundary plan orientation than disorientation [4].

2.4.2 Annealing Twins

Understanding annealing twins is important for this work because the nickel polycrystal being investigated in this study has a highly twinned microstructure. A twin occurs when some portion of the boundary of the grain forms a mirror image of itself across the plane of twinning. Twins are distinguished based on how they are generated. If the twin orientation is generated with the help of external deformation, it is called a deformation twin while it is called an annealing twin if it forms during recrystallization. Thus while deformation twin formation is accompanied by the need to minimize strain energy, annealing twins are formed in order to minimize interfacial energy.

Twin boundaries tend to be flat and have a faceted appearance. First observed by Carpenter and Tamura [85], these twin boundaries have a special symmetrical relationship which translates to a 60° rotation about <111> in F.C.C crystal structures. This is also called Σ3 in the CSL lattice system. If the boundary plane is parallel to {111} it is known as a coherent twin boundary and if it is not, then it is known as incoherent twin boundary.

There have been numerous studies aimed at understanding the mechanism of formation of annealing twins [86][87][88][89] and is still a disputed topic. In 1951, Fisher et al. showed that annealing twins would form if there was a possibility of lowering the overall interfacial free energy upon their formation. They made direct observations of the formation of twins and correlated twin boundary spacing to grain size as well as commented on the small mean dihedral angle in twinned corners. It was also shown by that a higher boundary velocity has a higher
probability in causing these growth accidents thus more stacking errors causing the occurrence of annealing twins.

One of the most widely believed theories is the “growth accident” theory which states that a twin is formed due to stacking fault error. The stacking sequence of \{111\} planes on either side of the twin boundary is ABCABC…and thus across the boundary it is ABCABACBA….Thus a low stacking fault energy assists in twin formation and has been found to be related to twin density by the following equation:

\[
\rho = \frac{b}{d} \log \frac{d}{d_0}
\]

Here, \(\rho\) is the twin density, \(d\) is grain size and \(d_0\) is the grain size at which \(\rho\) is zero, while \(b\) is a constant [90].

2.5 Near-field High energy diffraction x-ray microscopy (nf-HEDM)

High energy diffraction x-ray microscopy is a non-destructive imaging technique.

Monochromatic X-rays of energy between 30KeV to 100KeV are generated by accelerating electrons. The high energy of the beams ensures a larger penetration volume and is able to map crystallographic orientation fields of polycrystalline materials. High brilliance ensures reasonable time frames for conducting the experiment. Monochromicity allows the beam to focus to a micron width which gives greater special resolution. The beam is like a sheet, about 5\(\mu\)m in vertical dimension and 1mm in the horizontal to achieve enough narrowness so that each point contains a single orientation. The setup consists of CCD detector, and a single crystal polished tungsten attenuator (Figure 2.16). In this rotating sample experiment, the distance between the rotation axis of the sample and the imaging plane is maintained between 4-7mm which gives it the name near-field. The sample is centered on the rotation axis and rotated. As the rotation
invariably bring the sample into Bragg’s condition, the imaging is done in intervals of 1° rotation for contiguous intervals ranging from 100° to 180°. After capturing all the diffraction patterns for one slice, the sample is shifted along the z-axis to capture the next slice and so on so forth. In this method the diffraction spots are predicted using simulation and optimized to produce the best possible match for the same planes. The orientation information thus extracted for 2D slices are reconstructed into 3D microstructure using Dream.3D which will be explained in detail later [91][92][93][94].

![Diagram of nf-HEDM setup](image)

*Figure 2.16. Schematic of nf-HEDM setup. High energy beam is incident upon cross-section of sample (in green). 2-3 Detectors are placed to capture the diffracted rays from the sample that obey Bragg's law. Reprinted from [77]*

The recent development of near-field High Energy X-ray diffraction microscopy (nf-HEDM) [91] and diffraction contrast tomography [95] make it possible to track the evolution of three-dimensional microstructures while they respond to an external stimulus. For example, the response of a high purity Al polycrystal to annealing was measured by nf-HEDM [96]. The nondestructive nature of the orientation mapping made it possible to observe the growth of new grains and verify the salient features of microstructure recovery and recrystallization. The effect of the tensile strain on the microstructural evolution of polycrystalline copper was also mapped
using nf-HEDM [97]. Reconstructed voxels were used to study the misorientations and populations of certain dislocations and these were compared against computational models. Although considerable work has begun on the three-dimensional orientation mapping and algorithm development, little work has been done for direct grain-to-grain comparisons at different points in and the evolution of the 3D grain size distribution and number of grain faces [98][99].

2.6 Statistical tools and techniques

- **DREAM3D, Paraview**: Open source software that uses data analysis tools to create customized workflows for reconstruction of three-dimensional nickel microstructure using an array of thresholding, cleaning, segmentation and coloring, meshing and smoothing algorithms. Paraview is used for visualizing the reconstructed microstructures. [100] [101]

- **MATLAB, MTEX package**: Used for scripting algorithms, data analysis and data visualization [102][103]. They are used to track grains across anneal states, generate correlations between several datasets. MTEX is used for calculating disorientation between grains using crystallography.

- **FORTRAN**: Used for grain tracking, GBCD calculation and generating stereographic projections [104]. Also used for visualizing these stereographic projections as a function of 5 parameter grain boundary distributions.

- **R, CCA package**: Used for conducting Canonical Correlation Analysis on grain growth data [105]. Used on grain level data to understand which parameters influence the growth of grains as well as their interrelationships.
• **Analysis of Variance (ANOVA):** Used for identifying variance between grain boundary velocity and curvature distributions with the help of JMP [106].

• **Canonical Correlation Analysis (CCA):** Statistical model for multivariate analysis to define interrelationships and parameter sensitivity between input and output [107] [108].
3 Grain Tracking

3.1 Abstract

The orientations, locations, and sizes of approximately 2500 grains in a Ni polycrystal were measured at six points in time during an interrupted annealing experiment by synchrotron x-ray based, near field high-energy diffraction microscopy. The volume changes were compared to the geometric characteristics of the grains in both the original microstructures and microstructures in which adjacent twin related domains were merged. Neither the size of a grain nor the number of its nearest neighbors correlates strongly to a grain's volume change over the time scale of this experiment. However, the difference between the number of neighbors a grain has, $F$, and the average number of neighbors of the neighboring grains have, $<F_{NN}>$, is correlated to the volume change. A grain with more (fewer) neighbors than $<F_{NN}>$ usually grows (shrinks). The correlation between the volume change and $F - <F_{NN}>$ is obvious only if adjacent twin related domains are merged. The correct sign of the volume change is predicted by $F - <F_{NN}>$ about two thirds of the time. The results show that the volume change for a given grain is better predicted by comparing a grain's characteristics to its neighbor's characteristics than to the characteristics of the entire ensemble of grains.

3.2 Introduction

The goal of this work was to determine which characteristics of grains in an initial state best predict the sign of the volume change after some grain growth has occurred. To do this, an algorithm was developed to track each grain across all the anneal states. The volume changes of the grains were compared to factors such as initial volume, the number of contacting neighbors
(F), and the average number of neighbors of the neighboring grains, \(<F_{NN}>\). The results lead to the conclusions that while the size and number of neighbors are not well correlated to the volume change, the quantity \(F - <F_{NN}>\) is correlated to the volume change of the grains, but with two important stipulations. The first is that the correlation is strongest when adjacent twin related domains were merged and treated as a single grain [109]. The second is that the volume change and \(F - <F_{NN}>\) are correlated only on average, while some individual grains behave differently. For more information on grain growth please refer to Section 2.4.1.

### 3.3 Experimental Method

High purity Nickel (99.999% pure) was obtained from Alfa Aesar in the form of 5mm diameter x 50mm rod. Using Electric Discharge Machine (EDM) a cylindrical sample of 1mm diameter and 2cm length was cut. To identify the sample alignment for subsequent anneals, a fiducial (square cut notch), made using EDM, was marked as a reference, at 1mm above the measured location. The sample was initially annealed at 750°C for about 2 hours in order to fully recrystallize it and achieve a desired grain size (grain size was of the order of a few microns in the as-received nickel microstructure, deemed too small for imaging). Post this the sample was annealed sequentially in a reducing gas atmosphere (97 % \(\text{N}_2\), 3 % \(\text{H}_2\)) for approximately 30min, five different times, as shown in Table 1. After each anneal stage, the sample was cooled to room temperature and the 3D microstructure was measured.

After each annealing interval, near-field High Energy Diffraction Microscopy (for more details refer to section 2.5) was used to create 3D volumetric orientation maps of the microstructure using the 1-ID beam line at Argonne National Laboratory’s Advanced Photon Source. This setup used a monochromatic X-ray beam of operating energy 64.5KeV. The detector-to-rotation-axis
distances were set at 4.8, 6.8 and 8.8mm. The 3D orientation map was created by scanning the sample one layer at a time, each of 4μm spacing.

Thanks to the non-destructive nature of the microscopy, the same region of the sample was mapped six times. The volumes of the six annealing states were in the order of 280μm x 1mm (initial state referred to as Anneal 0, Anneal 1), 336 μm x 1mm (Anneal 2, Anneal 3, Anneal 4) and 400 μm x 1mm (Anneal 5). The increasing mapped volume was deliberate in order to capture the growing grains as well as any protruding grains outside of the imaging volume. The details of this data collection have been reported previously [110] and preliminary analyses are contained in two Ph.D. theses [93][94]. Of importance to the current work, the volume of materials characterized contained approximately 2500 grains, the x-y plane was discretized into 600 x 600 voxels with lateral dimensions of 2.4 μm, the vertical separation of the x-y planes is 4 μm, and each volume contains at least 70 x-y planes.

<table>
<thead>
<tr>
<th>Anneal State</th>
<th>Number of layers mapped</th>
<th>Duration (min)</th>
<th>Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>70</td>
<td>120</td>
<td>750</td>
</tr>
<tr>
<td>1</td>
<td>71</td>
<td>23</td>
<td>800</td>
</tr>
<tr>
<td>2</td>
<td>87</td>
<td>30</td>
<td>800</td>
</tr>
<tr>
<td>3</td>
<td>86</td>
<td>25</td>
<td>800</td>
</tr>
<tr>
<td>4</td>
<td>101</td>
<td>35</td>
<td>800</td>
</tr>
<tr>
<td>5</td>
<td>104</td>
<td>25</td>
<td>800</td>
</tr>
</tbody>
</table>

*Table 1 Summary of Annealing Parameters*
3.4 3D Reconstruction and grain segmentation

The three-dimensional Ni microstructure was reconstructed from nf-HEDM data using the IceNine implementation of the Forward Modeling Reconstruction Method (FRM) [92]. The result is a file that discretizes the microstructure into parallel 2D layers made up of equilateral triangles. For each triangle, the spatial coordinates, the Euler angles in the Bunge convention, and a confidence index, C, reflecting the reliability of the orientation determination are specified. The resolution limit was less than 0.1°. An in-house program was used to convert this information to 2 µm x 2 µm square pixels in each layer that could be directly read by DREAM.3D, an open source software consisting of data analysis tools using customized workflows (Pipelines) for analyzing data [100].

The sequence of steps for creating a 3D grain structure in DREAM.3D was similar to the default pipelines used to generate 3D orientation maps from focused ion beam serial sectioning experiments [74]. Briefly, the procedure begins by applying a threshold on C to generate a Boolean mask that distinguishes high and low confidence voxels. The individual sections are then laterally aligned by minimizing the layer-to-layer misorientation using only the high confidence data. The lateral shifts required for alignment ranged from 0 to a few voxels. After alignment, the microstructure is represented by three-dimensional voxels with dimensions of 2.8 µm x 2.8 µm x 4 µm. So-called clean-up procedures are then used to assign orientations to low confidence voxels using information from surrounding high confidence voxels. The data are then segmented into grains by grouping sets of contiguous voxels with small relative misorientations. In this procedure, there were two threshold parameters that influenced the results. One was the minimum number of voxels that are considered to reliably reflect the volume of a grain. A test of the sensitivity of the results to this parameter is summarized in Figure 3.1. Based on this, the
minimum accepted grain size was set to be $3^3$ or 27 voxels because this was the first integer cubed beyond which the average grain size and number of grains changed only slowly. The second threshold parameter was the minimum disorientation for segmenting voxels into grains. This was set at $2^\circ$ because the typical orientation difference between the same grain in two successive anneal states was less than $2^\circ$.

![Graph](image)

*Figure 3.1 Changes in the average grain size and number of grains in the reconstruction depends on the minimum size cut off during grain segmentation. The data are from anneal state 5 and are characteristic of other anneal states.*

After the grain segmentation, each anneal state had approximately 2500 grains, two-thirds of which are bulk grains and the others are surface grains (grains that contact the exterior volume of sample or the vertical limits of the field of view are referred to as surface grains, the remainder are referred to as bulk). The average equivalent spherical grain radius in the first anneal state (denoted 0) is 19 $\mu$m. This increased at each anneal state to 23 $\mu$m in the final anneal state (denoted 5). There are numerous challenges in identifying the same grains in successive anneal states. First, the volume imaged in each anneal is not exactly the same (see Fig. S1). The intent was to slightly increase the volume measured so as to minimize the loss of internal grains as growth took place; however beamtime limitations prevented a uniform increase. Therefore, there
are grains in the field of view after one anneal state that might not have been in the previous field or the next one. Second, a grain that is classified as a bulk grain in one state may impinge with a surface in another state and have a different volume and centroid location. Third, some grains will shrink and disappear in the course of the experiment, so it will not be possible to track them throughout the entire annealing sequence.

3.5 Grain Matching Algorithm

An algorithm was developed to adapt to these challenges. From each of the reconstructed microstructures, a list of all grains was constructed which included the grain’s centroid location (coordinates) and the orientation (Euler angles). As a first step, grains in successive anneal states with similar orientations and locations were identified and the distribution of their misorientations and separations were determined (see Figure 3.2 a and b). Note in Figure 3.2b that the distribution of separations reaches maximum at about 20 μm. This is assumed to be a rigid translation associated with aligning the samples after the heat treatment. The three components of the translation vector were obtained separately (see Fig. S2) and this vector was subtracted from the coordinates of the voxels in the second state to improve the alignment. After this procedure, the average difference in the locations of the centroids was nearly zero. Similar rigid translations were applied to all anneal states.
A kernel approach was used to identify the same grain in sequential anneal states. For each grain in the first anneal state, the distances to all grains in the next anneal state were calculated. Next, the disorientation between each grain from the first anneal state and every grain in the next anneal state whose centroid was less than five times the grain radius from the centroid of the grain in the first anneal state was calculated. These values were stored in a matrix that contained the disorientation and centroid distance of every grain in first state with respect to a neighborhood kernel of grains in the second state. To identify the same grain in sequential anneal states, we assumed that grains with similar locations and orientations were the same. To perform this match quantitatively, we defined a confidence index ($CI$) to measure the likelihood that two grains match:

$$CI_{ij} = C_1 \Delta g_{i,j} + C_2 \Delta L_{i,j}$$

where $\Delta g_{i,j}$ is the disorientation between grains $i$ and $j$, $\Delta L_{i,j}$ is the distance between the centroids of grains $i$ and $j$, and $C_1$ and $C_2$ are constants to scale the parameters so that disorientation and position had similar weights in determining $CI_{ij}$. Typical values were $C_1 = 0.2 (°)^{-1}$ and $C_2 = 0.005 \mu m^{-1}$.
A grain in the first anneal state is then matched with the grain in the second anneal state that has the lowest value of $CI$. The algorithm was tested by comparing duplicate microstructures and in these cases, 100% of the grains were matched appropriately. The algorithm was further tested by using the initial structure as the starting point for a 3D grain growth simulation. The phase field simulation was run until the average volume change was 15%; the matching algorithm found all of the pairs except for the grains that disappeared. When the algorithm is used on real data, multiple grains in the first anneal state were occasionally matched to the same grain in the second anneal state. In these cases, the grain pair with the lowest $CI$ was selected as the true match and the others were classified as unmatched. This problem is related to multiple twinning, which creates grains with the same orientation in close proximity. Pseudo-code for the matching algorithm is provided in Fig. S3. A schematic illustration of the process is illustrated in Figure 3.3. For grain 92 in the first state, $CI$ is computed for all grains whose centroids are separated from grain 92's centroid by less than five times the radius. Because grain 2743 in the second state had the lowest $CI$, it is assumed to be grain 92 in the second state. A visual comparison of Figure 3.3 (c) and (d) confirms that the two grains are the same.

When matching grains in different anneal states with real data, it is not possible to match all observed grains. One of the most significant problems is that the field of view at each time step is not exactly the same and if a grain is not included in both 3D images, it is not possible to match. For example, the image in Figure 3.4 shows anneal state 4, where the unmatched grains are opaque and the matched grains are transparent. In addition to some small bulk grains (these are grains that shrunk and disappeared), there is a large aggregate of grains at the lower boundary of the sample that were not matched because they were outside the field of view of the next set of data. All six fields of view are shown for comparison in Fig. S1. The next problem is that
small grains can disappear (or shrink to smaller than the segmentation threshold) and cannot be matched in the next time step. For real data, the matching efficiency (matched grains/total grains) was 88%, which accounted for an average of 97% of the volume of the microstructure (see Figure 3.5a). The matching was implemented both with time (states 0 to 5) and in reverse time (states 5 to 0) and the results were comparable.

![Figure 3.3. Schematic illustration of the grain tracking algorithm. (a) Grain 92 in the initial state is considered, all other grains are displayed as semi-transparent. (b) In the next state, all grains within five grain radii of grain 92’s centroid are considered candidates and displayed as opaque. (c & d). After computing the CI for all candidate grains, grain 2743 in the next state has the minimum CI and is matched to grain 92 in the initial state.](image)

![Figure 3.4. The sample after anneal state 4. Grains that matched are displayed semi-transparent and unmatched grains opaque. The unmatched grains at the bottom did not overlap with the field of view of the next state and could not be matched.](image)
The tracking program was also run with the condition that only bulk grains were considered. In this case, the average matching efficiency falls to 81%. The reduced efficiency is because some grains within the bulk in one state may, in the next state, impinge on a surface either because they grew and intersected the surface or because of a change in the field of view.

From the images of the microstructure in Fig. S1, it is obvious the microstructure is twinned, as expected for Ni. In fact, in the final anneal state, grain boundaries with the twin misorientation (60° misorientation about [111]) make up 29% of all grain boundary area. Because twins have odd shapes [75] and twin boundaries have unusually low mobilities [111], it is not clear how they influence the grain growth process. To minimize the influence of twins, we transformed the twinned microstructure to a more equiaxed microstructure by combining into a single grain all grains that share a grain boundary with the twin misorientation. This was implemented using the "merge twins" filter in DREAM.3D [100][112]. After merging the twins, there were fewer grains. For example, in the initial state, 2972 grains were merged to 926 grains. Examples of the
microstructure before and after the twins were merged are illustrated in Fig. S4. The grain matching algorithm had to be modified to match the twin related domains. Specifically, it was necessary to preserve the characteristics (orientation and centroid location) of all the grains merged into a single twin related domain in each state, and compare these groups to match the merged grains.

3.6 Grain Tracking Results

3.6.1 Visualization of matched grains

The initial and final reconstructed volumes are illustrated in Figure 3.6. Note that grain visualizations shown in this paper show the sets of voxels (as described above) with no smoothing applied. The results discussed here do not require smoothing. Comparing the two, the average grain size in the final anneal state is noticeably larger than in the initial state. Note that there is a difference in the field of view; a larger volume was mapped in the final anneal state than in the initial anneal state. While significant changes occurred during annealing, it is still possible to identify some common grains on the periphery of the sample, such as the one marked with a white X; all six reconstructed volumes are illustrated in Fig. S1. A comparison of three matched internal grains in the initial and final anneal states (Figure 3.6 c and d) shows that they have the same orientation (color) and similar shapes. The three individual grains that are illustrated all increased in volume. Two are relatively equiaxed but the other has a ‘U’ shape in the initial state and has a tunnel through it in the final state. Because of extensive twinning in the microstructure, this shape is not unexpected. The grain within the tunnel is a twin.
Figure 3.6. Visualizations of the microstructures in the (a) initial and (b) final anneal states. The grains are colored by orientation with respect to the cylinder axis, according to the inset color key. One example of a matching surface grain in the two states is marked with a white "X". Three selected grains in the initial anneal state are illustrated in (c) and the grains they are matched to in the final anneal state are shown in (d).

To demonstrate the effectiveness of the grain tracking algorithm, many individual grains were visualized in each anneal state. Figure 3.7 illustrates a grain that started small, but grew during annealing. The growth rate was clearly different during different annealing periods. For example, it grew dramatically between (b) and (c). Similarly, Figure 3.8 is a visualization of a grain that shrank during annealing. In the final state (Anneal 5), the grain could not be detected, suggesting that it shrank below the 27 voxel threshold used in the grain segmentation.
1800 grains were identified that matched in all five anneal states. While this is less than the total number of grains, it is limited by the condition that the grain had to appear in all five anneal states. Grains that shrank below the minimum detectable grain size (such as the one illustrated in Figure 3.8) are not counted. The greatest percentage fractional change in grain radius amongst these grains (between the initial and final anneal states) was 965%. It should be noted that the surface grains at the top and bottom of the measured volume do not provide reliable measures of the volume change; a small change in the field of view can dramatically change the measured
volume of a grain. Concentrating on only those grains that did not touch the surface of the volume in any state, and that matched in all states, eliminates this artifact, but creates other artifacts. For example, this selection criteria is biased against the largest grains (those that intersect the surface in some state) and smallest grains (those that disappear during the experiment). Accepting these limitations, there are 701 bulk grains that appeared in all anneal states; 330 grains grew while the remainder (371) shrank.

3.7 Grain size statistics

The distribution shown in Figure 3.9 illustrates that almost 80% of the grains have small volume changes during the experiment (less than ±20%).

As expected, the average grain size increases during annealing. Figure 3.10 shows the average grain radius in each anneal state, using grains selected by various conditions. When all grains in the reconstructed microstructure are considered, the average spherical equivalent grain radius increases from 19 µm to 23 µm. The surface and bulk grains behave the same way, but the
surface grains are larger than average and the bulk grains are smaller than average. This is because the probability that a grain intersects the surface is proportional to its size, so the surface grains are larger on average. Those grains that do not contact the surface and are tracked in each anneal state have a roughly constant grain size. These are the grains that were not small enough to shrink and disappear during the experiment nor were they so large that they impinged with the surface as a result of growth or shift in the field of view. The normalized grain size distributions before and after merging twins are illustrated in Fig. S5 and the distributions of grain faces before and after merging twins \((F)\) are illustrated in Fig. S6. In each case, the distributions appear self-similar throughout the experiment.

![Graph showing grain radius vs. anneal states](image)

*Figure 3.10. Average grain radius in each state for all grains (circles) and for selected subsets.*

The changes illustrated in Figure 3.10 are the sum of contributions from hundreds of grains. Whether a grain is growing or shrinking depends on the point in time when it is observed. If the end point of grain growth is a single crystal, then all but one grain must, at some point in time,
shrink. The behaviors of individual grains are not as regular. To illustrate examples of how individual grains change size over time, the spherical equivalent grain radii of six grains in each anneal state are illustrated in Figure 3.11. Grain 1 initially increased in size, and then shrank, from 43 µm to 33 µm. Grain 2 increases, decreases, and increases again. Other grains changed continuously, such as Grain 6, which grew from a diameter of 25 µm to 38 µm.

3.7.1 Grain growth and Aboav Weaire law

The results were compared to the AW law to determine if the number of neighbors that a grain has is correlated to characteristics of the neighboring grains. In Figure 3.12 Aboav’s empirical formula (for ascertaining whether grains in a polycrystal are arranged in a quantifiable manner) was used to plot AN (average number of neighbor of neighbors) vs N (number of nearest neighbors). In this plot, the number of nearest neighbors of the nearest neighbors of grains were counted and the average was calculated and noted for every bulk grain in the initial Anneal State.

![Figure 3.11. Grain radii at six anneal states for six representative grains.](image-url)
which had about 2200 grains. A best fit model was applied to calculate the correlation coefficients, which gave us the rule:

\[ AN = 16 + \frac{25}{N} \]  

(22)

This seems to follow the general rule developed by Aboav initially \( (m = 5 + 8/n) \), thus corroborating the idea that the arrangement of grains in a polycrystalline system can be quantified by a rule. A random distribution, on the other hand, could be visualized as a horizontal line in the plot of Figure 3.12.

![Figure 3.12. Variation of N with AN.](image)

Rearranging Eq. (15), we find a relation between \( \langle F_{NN} \rangle F \) as a function of \( F \):

\[ \langle F_{NN} \rangle F = (\bar{F} - 1)F + (\bar{F} + \mu_2) \]  

(23)

The data in Figure 3.13, which includes all grains in the twinned microstructure, shows good agreement with Eq. 33, where \( \bar{F} = 16 \) and \( \mu_2 = 21 \). The actual values of \( \bar{F} \) and \( \mu_2 \) measured from the data are \( \bar{F} = 12 \) and \( \mu_2 = 78 \). While the fit to Eq. 33 is good, the resulting parameters (\( \bar{F} \) and \( \mu_2 \)) do not correspond to the measurements. We do not currently have an explanation for this discrepancy, but it does bring into question the application of the AW law to three dimensional data.
Figure 3.13. The product of the number of near neighbors (F) and the average number of nearest neighbors of the neighbors (\(F_{NN}\)) plotted versus the number of neighbors (F). The line is a fit to the AW law.

3.7.2 Does grain volume change with change in neighbors?

The results in Figure 3.14 show how the change in the number of neighbors is related to the change in the volume. Each line corresponds to one matched pair of bulk grains. There is a distinct trend that those grains with a positive volume change mostly gained neighbors and those with a negative volume change mostly lost neighbors. In the central region, where the volume change was small, the change in the numbers of neighbors was also small. While the trend in the data is clear, there are also examples of grains that behave in the opposite way. The results in Figure 3.14 (a) are for two consecutive anneal states. We can make a similar plot for changes from the initial to final state. In Figure 3.14 (b) the change in the number of neighbors in the initial and final state are correlated to the change in the grain volume. Grains that shrink in size mostly lose neighbors, however grains that increase in size seem to gain as well as lose neighbors. However, grains that gain between 1 and 5 neighbors have similar chances of growing or shrinking.
In Figure 3.14 (a) and (b) positive change in grain radius is seen at the highest positive differences in number of neighbors. This implies that as the grains grow, their number of neighbors also increases. While, Figure 3.14 (a) plots the same for a shorter time step (30 minutes), Figure 3.14 (b) plots for the initial and final time step (150 minutes). However, upon closer inspection of both plots, it can be said that although the general trend is followed in Figure 3.14 (a) but not so much in (b). Certain grains do not confirm to this even in Figure 3.14 (a). For example, in Figure 3.14 (a) there are certain cases of grain growth accompanied by a reduction in their number of nearest neighbors. We shall take a closer look at such cases (circled in red) and attempt to explain the anomalies. It can also be speculated that when we consider only the matched bulk grain pairs, we are severely limiting the sample space of grains to be able to comment conclusively on any trend definitively. This is because, upon considering only bulk matched grains, we are including about 500 grains while the approximate number of grains in any anneal state is 2500.
Figure 3.15. Study of a grain whose size increases but number of neighbors decreases from (a) initial anneal state to (b) final anneal state.

(a) No. of neighbors: 118
Radius: 33 μm

(b) No. of neighbors: 57
Radius: 37 μm
In Figure 3.15 we look at a specific grain (one that was encircled in Figure 3.14(a)) to investigate why its change in volume is positive and yet the change in number of nearest neighbors is negative using Paraview. Figure 3.16(a) and (b) show us the neighbors of grain ID 1, which has grown from a grain radius of 33.62 µm in Anneal 0 to 37.46 µm in Anneal 5. Its number of neighbors, however, have decreased from 118 to 57. This is then visualized in Figure 3.16(c). The number of neighbors of this grain have decreased because the neighbor grains have grown. Figure 3.16(c) plots the average of the neighbor grain size (grain diameter) at each anneal state. This value increases monotonically from 24 µm to 43 µm. This will reduce the number of grains that can surround the grain in question.

3.7.3 Predicting volume changes using the effect of local neighborhood

In Hillert's [67] classical mean field theory of grain growth, it is assumed that there is a critical grain size above which grains (on average) grow and below which they shrink. However, when the volume change of a grain is considered as a function of the initial grain radius, the two
quantities do not appear to be strongly correlated. The data are shown in Figure 3.17, where grains were binned according to their radius and the mean (square) and standard deviations (bars) are plotted for each grain size. Volume changes in the twinned microstructure (Figure 3.17a) appear independent of grain size. In the microstructure with merged twins (Figure 3.17b), the three largest positive volume changes occur for the three largest grain size categories (approximately twice the mean size), but at smaller sizes the volume change is independent of size.

Some grain growth theories have suggested that there is a critical number of neighbors above which grains grow and below which they shrink [113][114]. In fact, a grain's size and number of neighbors are strongly correlated such that larger (smaller) grains have more (fewer) neighbors, as shown in Fig. S7. In Figure 3.18, the grains were grouped by their number of faces and the mean (squares) and standard deviation (bars) of the volume change in each category is plotted. For the twinned microstructure (Figure 3.18a), $F$ is a poor predictor of whether a grain will shrink or grow. For the microstructure after the twins have been merged (Figure 3.18b), there is some evidence that grains with more sides are more likely to grow. Grains with more than 18
sides have positive mean volume changes, except for four categories among the five with the largest number of sides.

![Figure 3.18. Change in volume versus initial number of grain neighbors for (a) the twinned microstructure and (b) after merging twins. For each class of F, the marker is the mean value and the bar shows one standard deviation of the distribution.](image)

It has been shown by Von Neumann and Mullins [115][51] that when local mean curvature drives grain boundary motion, the total volumetric growth rate \((dV/dt)\) is given by Eq. 34:

\[
\frac{dV}{dt} = V^{1/3}M\gamma \mathcal{g}
\]

where \(M\) is the grain boundary mobility, \(\gamma\) is the excess interfacial energy per unit area, and \(\mathcal{g}\) is the normalized integral mean curvature of the grain faces. \(\mathcal{g}\) is given by:

\[
\mathcal{g} = -\frac{1}{V^{1/3}} \int_{\text{Faces}} \left(\frac{1}{R_1} + \frac{1}{R_2}\right) dS
\]

Here \(R_1, R_2\) are the radii of curvature. \(\mathcal{g}\) is obtained by integrating the curvature of a infinitesimal area over all the faces of a grain. Integrating Eq. 35, we get:

\[
\Delta V^{2/3} = \frac{2}{3} \int M\gamma \mathcal{g} \, dt
\]
It has been observed experimentally that $\mathcal{g}$, the normalized integral mean curvature, is linearly proportional to $F - \langle F_{NN} \rangle$ [62][4]. Using this relation, we approximate Eq. 36 in the following way:

$$\Delta V^{2/3} = V_f^{2/3} - V_i^{2/3} \propto \int (F - \langle F_{NN} \rangle) \, dt \approx (F - \langle F_{NN} \rangle) \Delta t \tag{27}$$

where $V_f$ and $V_i$ are the final and initial volumes, respectively. This suggests that in a given period of time ($\Delta t$), grains that have more (fewer) faces than the average of their neighbors should grow (shrink).

The data in Figure 3.19 show the relationship between $\Delta V^{2/3}$ and $F - \langle F_{NN} \rangle$ for all grains (a) and after merging the twins (b). In each plot, the marker shows the mean value in each class and the bar indicates the standard deviation. Volume changes in the twinned microstructure (Figure 3.19a) appear independent of $F - \langle F_{NN} \rangle$. When the twins are merged, the correlation between $\Delta V^{2/3}$ and $F - \langle F_{NN} \rangle$ is much stronger. In this case, all positive mean values of $F - \langle F_{NN} \rangle$ are associated with positive values of $\Delta V^{2/3}$. The average volume changes for grains with negative $F - \langle F_{NN} \rangle$ are generally negative, but not strongly negative. This is likely to be due, in part, to the fact that when small grains shrink below the detection limit, they are no longer tracked and these negative volume changes are not included. However, it is difficult to discern if a grain is not tracked because it actually disappeared for if it is no longer in the field of view. It should be noted that while the mean values follow the expected trend, there is considerable scatter.

Therefore, a positive or negative value of $F - \langle F_{NN} \rangle$ is not an absolute predictor of the sign of the volume change. However, the sign of $F - \langle F_{NN} \rangle$ predicts the sign of the volume change in 65% of the grains.
Figure 3.19. Change in volume versus the difference between the number of neighbors and the average number of neighbors of the neighbors in (a) the twinned microstructure and (b) after merging twins. For each class of $F - \langle F_{NN} \rangle$, the marker is the mean value and the bar shows one standard deviation of the distribution.

Note that all grains, including surface grains, were included in this analysis. This clearly introduces errors, but some of the errors compensate. For example, a surface grain does not have its full complement of near neighbors, so $F$ is reduced from the true value. However, most of the neighboring grains face a similar restriction, so $\langle F_{NN} \rangle$ is similarly reduced. In an ideal situation, these grains could be excluded or included according to the criterion introduced by Rowenhorst [62]. He argued that grains whose centers of mass are less than $2\langle R \rangle$ from the boundary of the volume should be eliminated, where $\langle R \rangle$ is the mean grain radius. Unfortunately, when this criterion is applied to our data, there are so few grains remaining that there is insufficient data for the analysis, especially for the sample where the twins were merged. If we relax Rowenhorst's criterion and remove grains whose centers of mass are less than $\langle R \rangle$ from the boundary of the volume, more than half of all grains are removed, but 300 to 400 grains remain in each anneal state. The correlation between the volume change and $F - \langle F_{NN} \rangle$ for this more selective data is illustrated in Fig. S8. While the result is certainly noisier than Figure 3.19, because there are fewer grains, the results are consistent with the trends in Figure 3.19. In other words, the
comparison of Figure 3.19 and Fig. S8 suggests that eliminating grains near the surface does not change the conclusion that grains with positive mean values of $F - \langle F_{NN} \rangle$ are associated with positive values of $\Delta V^{2/3}$.

### 3.8 Discussion

Synchrotron x-ray based, near field High Energy Diffraction Microscopy (HEDM) makes it possible to measure the shapes and volumes of grains within a solid at different time intervals during interrupted annealing. Using these data, it is possible to determine which grains increased in volume and which grains decreased in volume and compare the volume changes to the characteristics of the grains in the initial state. The results do not support the idea that there is critical size, or a critical number of near neighbors, above which a grain grows and below which a grain shrinks. Instead, mean values of the volume change are correlated to $F - \langle F_{NN} \rangle$. It should be noted that the grain size, $R$, and its number of nearest neighbors, $F$, are strongly correlated with one another (see Fig. S7). Therefore, it should also be true that volume changes are correlated to $R - \langle R_{NN} \rangle$, where $\langle R_{NN} \rangle$ is the average radius of the nearest neighbor grains. The correlation between $R - \langle R_{NN} \rangle$ and the volume change is illustrated in Fig. S9 and, while having more noise, shows the same trend as Fig. 30. It should also be mentioned that a recent study of $\alpha$-Fe also found a correlation between the volume change and $F - \langle F_{NN} \rangle$ [116].

In retrospect, it is naïve to think that the size of a grain, compared the ensemble average grain size, determines whether it grows or shrinks. A specific grain grows or shrinks by exchanging mass with its neighbors. During a small increment of time, a specific grain has no information about ensemble averages and is only influenced only by its nearest neighbors. So, it is a comparison between the characteristics $(F, R)$ of the grain of interest with the characteristics of its immediate neighborhood $(\langle F_{NN} \rangle, \langle R_{NN} \rangle)$ that determines whether it grows or shrinks. In other
words, for a grain to grow (shrink), it is less important that a grain be larger (smaller) than the average grain size than it is for a grain to larger (smaller) than its neighbors.

While the correlation between the mean values of the volume change and $F - \langle F_{NN} \rangle$ illustrated in Figure 3.19 is strong, there are many exceptions which are apparent in the distributions about the mean values. On a grain-by-grain basis, $\Delta V$ and $F - \langle F_{NN} \rangle$ have the same sign 65% of the time. Among the other 35 %, some are certainly linked to limitations in the data. One source of error arises when the volume change is very small. As illustrated by Figure 3.9, most grains have small volume changes and small differences in the reconstruction may change the sign of the volume change. Another source of error is that grains that disappear are not included in the statistics and this artificially reduces the mean volume change of shrinking grains. The free surfaces of the volume are also obvious sources of error. When some of the largest outliers in the distribution were visualized, they were almost always caused by erroneous volume changes in grains at the upper and lower limits of the field of view. Errors associated with the free surfaces could be minimized by having data sets with more grains and we hope this will be possible in the future. Finally, there are rare events where a grain is divided in two by another (usually a twin). When divided, the grain experiences an apparent large negative volume change that is not the result of the normal grain shrinkage process.

The time interval over which the experiment occurs also plays an important role in our ability predict whether a grain grows or shrinks. Consider, for example, the idealized case where an ensemble of grains continues to grow without stagnation. The average grain size increases continuously until the end point is reached when the entire sample is made up of one grain. The so-called solid state crystal conversion process is a real world approximation of this ideal situation [117][118]. At any point in time while the average grain size increases, some fraction
of all grains can be classified as growing. However, before the endpoint is reached, all grains other than the one left at the end must eventually shrink and disappear. In other words, while all grains can be classified as shrinking or growing at any instant in time, some growing grains must become shrinking grains given sufficient time. In the current experiment, the total volume changes were, on average, 15% for each anneal state. Changes in the local neighborhood of the grains that influence both \( F \) and \( \langle F_{NN} \rangle \) during the time intervals used here are one source of uncertainty in the results. While this uncertainty might be reduced by using smaller time intervals, uncertainties in the accuracy of the volume measurement will at some point become influential.

Dake et al. [119] recently reported grain rotation during liquid phase sintering. While we observed no such systematic rotations, it must also be mentioned that our method of matching the grains in different time steps relied on the similarity of the orientations in the two states. Therefore, rotations of several degrees would have led to an unmatched grain pair and it would not have been detected. The fact we are able to match 88% of the grains, and the unmatched grains are well correlated with the non-overlapping fields of view and the disappearance of small grains (see Figure 3.4), suggests that any large rotations, if they occur, are rare. Also, the average disorientation between matched grains in two time steps is on the order of 0.05° (see Figure 3.2a). This is a measure of the uncertainty in the orientation measurements, so rotations below this limit would not be detected in our experiment. In an earlier analysis of these data, rare changes in nearest neighbor disorientations were explained by singular events rather than grain rotation. [93]

From Figure 3.13 we can say that the arrangement of grain distribution in a polycrystalline microstructure is systematic to a certain degree, such that large grains tend to be surrounded by
small ones and vice-versa [12]. Since the correlation developed by Aboav et al. [12,13] was derived from the geometry and topology of two dimensions, it could not be conclusively said for three-dimensional topology as well. However, using the polycrystalline Ni 3D data, plots of similar nature have been observed for all anneal states. It is thus concluded that there exists a certain relationship between a grain and its nearest neighbors (and perhaps even the second nearest neighbors, (chain reaction or domino effect). This lack of randomness has been attributed to have occurred out of a need to maintain the averages of either the number of faces or the ensemble average area in two or three dimensions. Any grain which is large (or has many neighbors) will naturally need to be surrounded by smaller ones. The relations derived above allow such arrangements to occur and can be thus validated. [29]

The analysis of a twinned microstructure clearly presents challenges. When twins are considered as separate grains, the relationship between $\Delta V$ and $F - \langle F_{NN} \rangle$ is not obvious. There are several reasons that twins can complicate the analysis. Considering the large fraction of coherent twins, almost every grain will have at least one twin boundary and these boundaries are likely to be far less mobile than the other boundaries. Another difficulty is that plate shaped twins within a grain can have as few as two neighbors; this likely leads to a negative value of $F - \langle F_{NN} \rangle$. However, if the incoherent segments at the edges of the plate are expanding within the grain, its volume will increase. As an example, Lin et al. [110] showed, using a subset of the data considered here, that when twins nucleate at triple lines to lower the grain boundary energy, they expand in size, even though they have only three neighbors and a negative value of $F - \langle F_{NN} \rangle$. Also, because of the very anisotropic shapes of the twins in 3D, the typical relation between grain size and number of neighbors is not expected to be as regular. For example, in Fig S7, the standard
deviation in each grain size class is much larger in the twinned microstructure compared to the microstructure without twins.

Finally, we should question whether or not the microstructure is in a steady state during the experiment. If we base this only on the distribution of grain sizes for faces, one might conclude that they are self-similar (see Figs. S5 and S6). Furthermore, the distribution of grain boundary disorientations is also nearly constant. The only measurable change is an approximately 10% increase in the relative area of grain boundaries with a 60° disorientation (see Fig. S10). While this is consistent with the idea that low energy grain boundaries increase in area with respect to higher energy boundaries, they are eventually expected to reach a steady state. [104] Although the increase in the fraction of twins suggests that the distribution of grain boundaries has not reached a steady state, it should be noted that these boundaries have no influence on the microstructure from which twins have been eliminated and this is the one whose volume changes are best correlated to $F - \langle F_{NN} \rangle$.

3.9 Conclusion

The shapes and sizes of 2500 grains in a Ni polycrystal were measured at six points in time separated by 30 min anneals at 800 °C. During the experiment, the average grain volume increased by 77%. For the individual time intervals, neither the grain size nor the number of neighbors were useful predictors for the sign of a grain's volume change. However, in microstructures where adjacent twin related domains were merged, the difference between the number of neighbors and the average number of neighbors of the neighboring grains is correlated to the sign of the volume change. The correct sign of the volume change is predicted by this metric 65% of the time. These results show that a grain's local neighborhood is more important in determining its volume change than how it compares to the average characteristics of the
entire sample. Volume changes in the twinned microstructures were not well correlated with any of the metrics and this is most likely a result of the high concentration of twins (more than 27% of grain boundary area) and their distinct properties.
4 Grain growth and curvature

The three-dimensional evolution of nearly 2500 grains in pure polycrystalline nickel was studied in an interrupted annealing experiment using synchrotron based near-field high energy diffraction x-ray microscopy. The distribution of grain boundary curvature is measured and compared to grain volume changes. This correlation is also studied when the adjacent twin related domains are merged in the microstructure.

4.1 Procedure

4.1.1 Meshing and smoothing the microstructure

To calculate grain boundary curvature, face curvature, grain boundary planes we need to smooth the boundaries and triple lines. Using Dream.3D, we first mesh the surface of the segmented grains in the sample [100]. This creates a geometry that can be represented in discrete triangles consisting of vertices, edges and faces. The meshing algorithm demarcates the voxel faces into triangles. This means the face between any two features (grains in this case) in 3D is divided into connecting triangles. This is done by identifying all voxels (building blocks of grain) whose neighboring voxel is part of a different grain and defining its face by a pair of triangles. The surface mesh is characterized by no free edges that are shared between no more than two triangles. The edge lengths of these triangles vary from 0.02µm to 7.5µm, with an average of 2.9µm and a mode of 2.8125µm (this is on order of the dimension of the voxels). The information required to describe this meshed structure is given in the form of several lists:

- SharedTriList (lists the vertex Ids of every triangle),
- SharedEdgeList (lists the vertices connecting every edge),
• **SharedVertexList** (lists the x, y and z coordinates of each vertex Id).

Each triangle is defined by the feature Ids on either side, the triangle normal, the triangle area, centroids and eventually curvature. The meshed structure of a grain can be viewed in Figure 4.1. Meshing creates a certain stair stepped pattern on the surface. To reduce this chunkiness, a smoothing procedure is followed. An effective technique for smoothing these meshes is a polyhedral surface smoothing called Laplacian smoothing. Unlike other triangulation methods, Laplacian smoothing improves triangulation by moving point locations. As described by Belyaev [120], a triangulated surface is considered for a vertex P to define an umbrella operator:

\[
U(P) = \frac{1}{\sum_i W_i} \sum_i W_i Q_i - P
\]  

Here, \(W_i\) are positive weights and defined as the inverse distance between P and the connecting vertices, \(Q_i\) describes the neighbors. The summation is done over all the neighbors of P (as shown in Figure 4.1).

![Figure 4.1. Schematic of \(U(P)\), the umbrella operator. Picture reprinted from [101]](image)

The vertex P is updates by the following rule:

\[
P_{\text{new}} = P_{\text{old}} + \lambda U(P)
\]  

The factor \(\lambda\), a small positive number usually in the range (0,1) controls the degree of smoothness obtained. Eq. 39 is iterated several times until the required smoothness is obtained.
\( \lambda \) is too small, it may require more iterations to achieve smoothness, slowing down the smoothing process, and if \( \lambda \) is too large, the process is unstable. A value of 0.25 was used for \( \lambda \) in our smoothing process [120][121]. Figure 4.2 shows a smooth grain after using the Laplacian Smoothing technique.

![Figure 4.2. Image of a grain (a) before and (b) after meshing and smoothing using Dream.3D.](image)

### 4.1.2 Grain boundary curvature calculation

#### 4.1.2.1 Normalized integral mean grain curvature (NormCA) from triangle curvatures

The driving force for grain growth is given by the product of curvature and energy making grain boundary curvature one of the most important properties of microstructure. For calculating the principal direction vectors and the curvature of grains, Dream.3D uses the Cubic Order Algorithm [122]. The principal directions and principal curvatures are computed by considering the eigenvalues and associated unit eigenvectors of the Weingarten Curvature Matrix. The surface normal at each vertex is calculated and the Weingarten curvature matrix is defined in a local orthonormal coordinate system. After this, the Adjacent Normal Cubic Approximation method is performed. In this method, a least squares fit is done to the surface adjacent to the vertex data which is in the form of triangles (Figure 4.1). Solving the Weingarten matrix, the eigenvalues are the principal curvatures and the eigenvectors are the principal directions. Each triangle has a mean curvature which is the mean of the two principal curvatures.
To investigate the effect of grain boundary curvature on grain growth, we need to calculate the integral mean grain curvature. This being the triangle curvatures integrated over grain faces and then entire grains. This is achieved by adding the mean curvature of all triangles across all the faces of that grain weighted by the corresponding triangle areas, as shown in the Eq. 32. Let’s call this curvature $CA$.

$$CA = \sum_{j}^{n_{f}} \left( \sum_{i}^{n_{t}} H_{ij} \times S_{ij} \right)$$

Here, $CA$ is the integral mean curvature of the grain faces, $H$ is mean curvature of the $i$th triangle on the $j$th face, while $S$ is its area. The integral mean curvature was calculated for both the as-received twinned microstructure as well as the fabricated merged twin microstructure. The twinned microstructure had an average of 2500 grains while there were approximately 900 merged grains obtained in each state.

While the integral mean curvature is the property of the whole grain, we also consider individual face curvatures which is the property of the face of the grain. The face curvature, $C$, is related to the integral mean curvature by the following equation:

$$C = \frac{\sum_{i=1}^{n} f_{i}A_{i}}{\sum_{i=1}^{n} A_{i}} = \frac{CA}{\sum_{i=1}^{n} A_{i}} \quad \text{(31)}$$

$f_{i}$ is the curvature of the $i$th face, $A_{i}$ is the area of the $i$th face and $n$ is the total number of faces in a grain, thus the integral mean curvature is the weighted average of the face curvatures.

The normalized integral mean curvature of grain faces is then given by:

$$NormCA = \frac{CA}{R}$$
Here, CA is the integral mean curvature obtained from before and R is the spherical equivalent radius of the grain.

4.1.2.2 Normalized integral mean grain curvature (G) from Mean Width $\mathcal{L}(D)$

Another method to calculate integral mean width has been applied which was given by Srolovitz and MacPherson [77]. They defined volumetric grain growth rate in 3D by:

$$\frac{dV}{dt} = -2\pi M \gamma \left( \mathcal{L}(D) - \frac{1}{6} \sum_{i=1}^{n} e_i(D) \right)$$

Here, $\mathcal{L}(D)$ is the mean width of a domain D, $\sum_{i=1}^{n} e_i(D)$ is the summation of triple line lengths $e_i$ over all n triple lines, M is the mobility, $\gamma$ is the surface tension of the domain wall.

The integral mean grain curvature $G'$ is thus defined by:

$$G' = \int k dA = 2\pi \left( \mathcal{L}(D) - \frac{1}{6} \sum_{i=1}^{n} e_i(D) \right)$$

The mean width of an arbitrary three-dimensional domain was defined as the following:

$$\mathcal{L} = \frac{1}{2\pi} \sum_{i=1}^{n} e_i \beta_i$$

Here $e_i$ is the length of the edge shared by two triangles and $\beta_i$ is the exterior turning angle between the two triangles. The mean width can also be written as the sum of two quantities-triangles at the faces and triangles at the triple lines as shown by Rowenhorst et al. [98].

$$\mathcal{L}_{face} = \frac{1}{2\pi} \left( \sum_{i=1}^{m^F} e_i^F \beta_i^F + \sum_{i=1}^{m^TJ} e_i^{TJ} \beta_i^{TJ} \right)$$

If one assumes that the turning angles of all triple lines are 60°, then the second summation of the above equation exactly equals the triple line summation in the equation for $G$ and they cancel each other. Then we get the normalized integral mean grain curvature as:
\[ G = \frac{1}{V^3} \sum_{i=1}^{m_F} e^F_i \beta^F_i \]

The curvature has been normalized by spherical equivalent grain radius. This assumption obviates the need to consider triple lines and use only those triangles that are shared between two faces. This is useful in this case since it was found that the dream3d smoothing and meshing can be problematic for triple lines and render inaccuracies.

It must be noted that in this study, the calculation of \( G \) yields only positive values because the turning angle was taken as the smallest positive angle between the normal of the adjoining triangles. The measurement of signed angle is currently a work in progress.

### 4.2 Results

#### 4.2.1 Curvature estimation

![Image of a twinned grain after smoothing colored by its faces.](image-url)

*Figure 4.3. Visual of a twinned grain after smoothing colored by its faces.*
Figure 4.3 shows a grain after meshing and smoothing using the Laplacian Smoothing method, after which the principal curvatures and directions were generated using Cubic Order Algorithm [122]. This grain has 41 faces and a mean curvature of $-0.01\mu\text{m}^{-1}$. This grain is colored by its face IDs, bringing into view how each face looks after smoothing. Figure 4.4 show the same grain from Figure 4.3, where the grain is colored by the mean curvature across the triangles making the grain. Figure 4.4 shows that twinned grains have odd shapes and affect grain growth differently as twin boundaries usually have low mobilities. [123] It has been illustrated from Figure 4.4 that there are areas in the proximity of the twins which have extreme curvature values. These local fluctuations in curvature could be artifacts from the Dream.3D meshing and smoothing algorithm.

Upon merging the twins, these local fluctuations are avoided thereby increasing the confidence in the mean curvature values. Therefore, we transform the twin microstructure into a slightly more equiaxed structure by merging all the adjacent twin related domains. Henceforth, all our
observations include both the as received twinned microstructure and the microstructure obtained after merging twins. Surface grains as well as bulk grains have been considered in this study.

The histogram in Figure 4.5 shows the distribution of average grain curvature of both the as received and twin merged datasets (for all six anneal states compiled together). The distribution shows more than 99% of the data points while the 1% outliers cannot be seen in the given x-range. Merging the twins changed the total number of grains in the microstructure from 2972 grains to 920 grains after merging (for initial anneal state). Both distributions have more than 70% of the curvature values between the range -0.05 μm⁻¹ to 0.05 μm⁻¹. This is an important observation with respect to Figure 4.4. This is because due to the scale the mean curvature in the heart of the faces looks like it is very close to zero, but is within ± 0.01 μm⁻¹ where most of the curvature values are concentrated. It can be observed that merged microstructure has a higher number of grains with lower curvature values. This is because during the process of merging, a lot of twins are combined to form larger grains thereby eliminating higher curvatures and small
faces that decreases the average grain curvature. The curvature distribution is right skewed. During grain growth, larger grains tend to grow at the expense of the smaller grains surrounding them until only one grain remains, thereby having more grains shrinking at any point in time. Consequently, number of grains that have positive mean curvature values is greater than those with negative values because according to Eq. 23, grains with positive (negative) curvature tend to shrink (grow). Thus, at any point in time, more grains will have positive mean curvatures than negative. After calculating integral curvature of grain faces (CA), we plot it as a function of number of grain faces for the twinned data in Figure 4.6. The x-axis was restricted to F=40 so that the graph shows the clear majority of data and is not dominated by outliers; the plot capturing the entire range can be visualized in Appendix. In the figure, we see that the mean curvature crosses over the x-axis at roughly F=15, which means that, on average, a grain with more than 15 faces has negative mean curvature while a grain less than 15 faces has positive mean curvature. This result is in agreement with previously established zero growth rates by multiple researchers [62][4][124]. Several researchers have put this value down at 13.4 faces [62], however it is safe to say that declaring an exact value for zero grain growth is not effective as it has been found that it is the range that makes more sense [98]. Also, the growth of a grain is more affected by local topological changes in the neighborhood than the entire ensemble of grains [62][1]. Based on our findings, it is believed that a grain with more (less) than 15 faces will have negative (positive) G. This trend is predicted correctly 80% of the time on the entire dataset as shown in Appendix (Fig. S11). Grains with very small faces and large faces seem to have to lowest standard deviation while the grains in the middle (20 < F < 30) have higher standard of deviation; this result is mainly driven by the number of grains in each bin. At very
small and very large number of faces, the bins have very few data points and hence less standard deviation.

4.2.2 Grain growth and curvature

Rowenhorst et al. showed a strong correlation between the normalized integral mean curvature of grain faces in a $\beta$ titanium alloy and the difference between the number of faces of a grain and the average number of faces of the grain’s nearest neighbors ($F - <F_{NN}>$). The correlation plot was linear and passed through the origin, implying that the grains which have more (less) faces than the average number of faces of their neighbors tend to grow (shrink). The difference in grain volume raised to the power two-thirds has also been shown in recent studies that shows the same correlation to $F - <F_{NN}>$ [116][1].

Figure 4.7(a) and (b) show the integral mean curvature (C) vs $F - <F_{NN}>$ for two scenarios considering all the twinned grains and after merging the twins. Also, it should be mentioned that in all these figures (Figure 4.6 Figure 4.7 Figure 4.8 Figure 4.9), surface as well as bulk grains are considered. This is because given the sample dataset, removing the surface grains would greatly
reduce the number of grains for statistical significance. And also because when surface grains are considered, say for example, in the F- \(<F_{NN}\)> plots, both the F and \(<F_{NN}\)> parts get adequately compensated due to the lack of neighbors.

However, including surface grains leads to the possibility of errors. For different fields of view between the anneal pair, the change in volume of surface grains is inaccurate. In Figure 4.7 the curvature is linearly related to F- \(<F_{NN}\)> and passes through zero. The current data confirms the trend highlighted by prior studies [62][4]. Since curvature is strongly correlated to F- \(<F_{NN}\)> and it has been shown that \(V_F^{2/3} - V_I^{2/3}\) (\(V_F\) – final volume, \(V_I\) – initial volume) also strongly correlates to F- \(<F_{NN}\)> [1][116], it is expected that average integral mean curvature should correlate to change in volume in some form.
In Figure 4.8(a) and (b) we plot the normalized integral mean grain curvature against the volume change \((V_F^{2/3} - V_I^{2/3})\) of the grains that have been tracked for the twinned microstructure as well as after merging adjacent twin grains. In Figure 4.8(b) the mean values pass through the x-axis very close to \(x=0\) which does not happen in Figure 4.8(a) i.e. the trend is clearer and prominent after the twin related domains have been merged. Correlation coefficients for Figure 4.8(a) is \(-0.05\) and for (b) is \(-0.34\). Grain volume change \((V_F^{2/3} - V_I^{2/3})\) is also plotted as a function of the normalized integral mean curvature \((G)\) derived from mean width (4.1.2.2) in Figure 4.9. The correlation coefficients are 0.4 for twin merged (b) and 0.03 for twinned microstructure(a).
Figure 4.9 Normalized Integral mean curvature of grains (G) plotted versus change in volume of the grain raised to power two-thirds in (a) twinned microstructure as a binned plot and (b) after merging twins between Anneal1-Anneal2.

Face curvatures of a grain have been derived in the same way as C. The number of positive and negative face curvatures were noted for every grain. Correlation coefficients for Figure 4.10 for the twin merged microstructure (b) is -0.13 and for the twinned microstructure (a) is -0.005.

Figure 4.10. Ratio of positive to negative face curvature of grain faces plotted versus change in volume of the grain for (a) twinned microstructure and (b) twin merged microstructure.
Thus, according to this plot, excess positive face curvature has a weak correlation with fractional volume change.

Figure 4.11. Meshed structure of grain shown has 1 positive and 4 negative faces curvature values with positive integral mean curvature.

Figure 4.12. Meshed structure of grain shown has 6 positive and 1 negative face curvature values with negative integral mean curvature.

Figure 4.11 shows a grain with all but one face with positive curvature and Figure 4.12 shows a grain with all but one face with negative curvature. In Figure 4.11, 4 faces have negative
curvature and 1 face has positive, i.e. 4 faces are concave in geometry with a negative mean curvature. This grain is more likely to grow than the grain in Figure 4.12 which has 6 positive face curvatures and 1 negative. It is seen that the grain in Figure 4.11 does grow by 18%.

The above figures (Figure 4.13) show a specific grain that grew by approximately 40% upon annealing. It has 18 faces in Anneal 0 and it gained one face in the next anneal state. Looking at
the evolution of a specific face of this grain, we see that the face has grown wider but shorter.

Above figure (Figure 4.14) shows the face in the former anneal state (left) and in the subsequent anneal state (right).

![Figure 4.14. Visualization of grain face evolution of a grain between two consecutive anneal states. The different facets of the grain are colored by its faceID.](image)

In Figure 4.15 we look at the face evolution of a specific grain between two consecutive anneals. The spherical equivalent grain radius changes from 94 µm to 100.3 µm in Figure 4.15 (a) and (b) respectively. In Figure 4.15 it is seen that there are some distinct changes at several faces of the grain. Some faces have grown larger and more concave (marked ‘x’), while a face grew a lot (marked ‘Y’) and the face labelled ‘b’ has shrunk. Face b shrank indicating that the neighboring grain most likely shrank too.

It has been shown through simulation work that coherent Σ3 twins are essentially immobile [123]. Olmsted et al. showed that while incoherent Σ3 boundaries have very high mobilities, coherent Σ3 boundaries are immobile within the resolution of the simulation using synthetic driving force molecular dynamics method. They examined 388 boundaries of nickel and found
that boundary mobility is independent of boundary energy, disorientation, $\Sigma$ value, thermodynamic quantities etc. [123][4].

Figure 4.16. Fraction of sigma3 boundaries plotted vs the fractional change in volume as (a) a binned plot and (b) scatter plot for all states.
Figure 4.16 shows the fraction of $\Sigma 3$ boundaries of a grain vs the volume change of the grain. There is a weak correlation and thus fraction of $\Sigma 3$ twins of a grain cannot predict grain volume change.

Figure 4.17 shows the difference of the absolute values of mean curvature from the absolute value of the average of the mean curvatures of the nearest neighbors. This result is plotted for the twinned microstructure. According to the plot, if $C - <C_{NN}>$ is positive, there is negative fractional change in volume and when $C$ is equal to $<C_{NN}>$, then fractional change in volume is also zero. However, when $C - <C_{NN}>$ is negative, the fractional change in volume seems to be initially positive at larger negative values and then comes closer to zero but due to lack of significant data points is inconclusive.

![Figure 4.17](image.png)
4.3 Discussion.

Figure 4.7 shows that NormCA has a strong negative linear correlation with F-<F<NN> for both the twinned and twin merged microstructures. This was also observed in previous studies [62][4]. We also saw in Chapter 3 that the grain volume change is well correlated with F-<F<NN> for the twinned microstructure. Thus it can be expected that grain volume change will also be correlated to grain curvature. This is examined in Figure 4.8 where we look at volume change as a function of the normalized integral grain curvature (NormCA). It is seen that the NormCA of the twin merged microstructure is better correlated to volume change than the twinned microstructure. We also determine grain curvature by summation of the turning angles of adjoining triangles with shared triangle edges (G) and determine its correlation to grain volume change in Figure 4.9. Again, the twin merged microstructure shows better correlation than the twinned microstructure. This might be due to the elimination of twin boundaries in the twin merged microstructure which are known for being highly anisotropic as already discussed in Chapter 3. In the twinned microstructure, the twin plates are flat and thus have low curvatures, while the incoherent segments at the edges of twins will have higher curvature, affecting the curvature of the grain as a whole. Thus grain curvature of the twinned microstructure may not be reflective of the actual volume change of that grain. Another reason could be inaccuracies in curvature estimation. Among the triangle mean curvature values generated from smoothing procedure of Dream.3D, especially the triangle curvatures at the periphery of a grain face is seen to have much higher magnitudes as compared to the heart of the face (as seen in Figure 4.4). While the magnitude of face curvature mostly varies between 0.04 μm⁻¹ to 0.05 μm⁻¹, the periphery has curvatures as high as ± 0.1 μm⁻¹. The high values at the triple lines could affect the overall curvature of the grain and is a source of error. This can be improved by using a different
smoothing procedure (future work). There are also several artifacts generated in Dream.3D such as weird grain shapes generated of very tiny grains with high mean curvatures.

Comparing the correlation for the merged microstructure in Figure 4.8 (b) and Figure 4.9 (b) we see that G shows slightly better correlation than NormCA. In the twinned microstructure, the correlation coefficients in Figure 4.8(a) and Figure 4.9(a) are too small to make any meaning comparisons. Measuring curvature by including the turning angles of the triangles leads to a better approximation of volume change than area weighted grain curvature only in the twin merged microstructure but not in the twinned microstructure.

The relationship between volumetric growth rate and integral mean curvature (G) of individual grains has been shown as a perfect linear correlation in another work on Iron based on experiment and simulation [125]. Considering that G takes into account turning angles it can capture the irregular shaped grains better than a simple area weighted average which is NormCA.

In Figure 4.15 evolution of faces could be the result of consuming a neighboring grain wholly or in part. This shows that evolution of faces of a grain is a dynamic process. In other words, although the grain is growing, not every boundary is growing in area. Some boundaries are growing while some are shrinking. Changes in the face depends on the growth behavior of both grains around it. Therefore, boundary evolution is directed by both grains and may not always be predictable from the point of view of only one grain.

Fractional change in grain volume is plotted as a function of excess positive face curvatures, fraction of Σ3 boundaries and C - \(<C_{NN}\) in Figures Figure 4.10Figure 4.16Figure 4.17 for the twinned microstructure. All three plots show a weak correlation between the fractional volume change and the respective x-axes. In Figure 4.10, the correlation has been shown to slightly
improve from -0.04 to -0.3 after the twin grains have been merged but still a weakly negative correlation at that. Figure 4.17(a and b) reveal more about how the magnitude of curvature affects grain growth. When a grain has higher magnitude of curvature than the average of its nearest neighbors, it has a higher tendency to shrink than the neighbors. High magnitude of mean curvature equates to small grains. However, there is no growth/shrinkage reported when the mean curvature of the grain is equal to or less than the average mean curvature of its nearest neighbors. However, it is the boundaries (the grain faces) of a topological network that essentially do the moving. Hence, looking at individual face curvatures of grains might lead to a new understanding to understand why a grain grows/shrinks. This idea shall be further investigated in Section 5.4.4.

4.4 Conclusion

- Normalized integral mean curvatures ($G$ and $NormCA$) have stronger correlations with grain volume change in the twin merged microstructure than the twinned microstructure.
- The normalized integral curvature obtained from mean width $G$ has a slightly stronger correlation with volume change than the normalized integral curvature obtained from triangle curvatures ($NormCA$) in the twin merged microstructure.
- Excess positive face curvatures of each grain can weakly predict whether a grain will grow or shrink.
- Depending on grain size of nearest neighbors and curvature of the faces, a growing grain can have both shrinking and growing faces.
5 Grain Boundary Velocity

5.1 Abstract

A high purity nickel microstructure was mapped using nf-HEDM in an interrupted anneal experiment at several points in time. The translation between any two consecutive states was adjusted by minimizing the average disorientation between voxels of the two volumes. For each anneal state, the average velocity for all grain boundaries was calculated using a method that tracks voxels whose grain IDs change from one grain to another. Grain Boundary (GB) velocity and GB curvature are a function of the five grain boundary parameters. \( \Sigma 3 \) twins contribute 29% of grain boundary area fraction and GB velocity was found to be directly proportional to the curvature in these boundaries. By contrast, \( \Sigma 7 \) boundaries show a negative correlation between velocity and curvature. For other boundaries examined, there is minimal correlation between velocity and curvature. The uniformity of GB curvature likely affects the driving force for grain boundary motion.

5.2 Introduction

Grain boundaries are regions of mismatch between any two adjoining grains. They form a three-dimensional interfacial network that connects all grains in the microstructure. Grain boundary motion (GBM) is the phenomenon by which these boundaries migrate to lower their energy. When one grain grows at the expense of another, the grain boundary gets displaced. GBM is one of the most investigated and not very well understood phenomenon in the realm of grain growth. Understanding GBM is extremely relevant because the final grain boundary configuration influences some physical, chemical and mechanical properties of a material. This process which
involves motion of atoms across boundaries occurs mostly during recrystallization, recovery and eventually grain growth. Previous studies show that GB velocity is proportional to a driving force \[126\] \[127\]

\[ v = MP \]

Here \( v \) is the Gb velocity, \( P \) is the driving force and \( M \) is called mobility which is the proportionality constant. In this regard, the velocity is also written as:

\[ v = Myk \]

Here, \( \gamma \) represents GB energy per unit area and \( k \) is the integral mean grain curvature \[128\][129\]. GB mobility determines how fast the boundaries move and is usually assumed to vary with temperature in an Arrhenius type equation as follows \[130\]:

\[ M = A_0e^{-Q/RT} \]

\( A_0 \) is the pre-exponential factor, \( Q \) is the activation energy (enthalpy) for migration, \( R \) is the Boltzmann constant and \( T \) is the temperature.

A variety of factors can affect GBM such as impurities or precipitates (solute drag), chemical composition as well as temperature \[131\][132]. Several studies have shown that mobility, grain boundary energy and curvature vary with grain boundary crystallography. However, the effect of crystallography on grain boundary velocity has not been directly established, especially in three-dimensional microstructures \[133\][134][135][136\]. Recent studies have attempted to quantify the dependence of grain boundary mobility to five macroscopic degrees of freedom with contrasting results. Zhang et al. found no correlation with the boundary’s five macroscopic degrees of freedom implying grain growth is more complicated than perceived and has other factors driving it \[137\].
GB velocity in three-dimensions is more complex possibly because of all the constraints in a polycrystal and the requirement of having equilibrium at triple junctions. This work attempts to deconvolute the interrelationships between GB velocity, GB curvature and crystallography.

5.3 Methods

5.3.1 Alignment of 3D Volumes

It has been previously shown that the 3D microstructures for each of the Anneal States have different mapped volumes and have different field of views [138]. To calculate grain boundary velocity accurately, it is imperative that the data be in the same spatial reference frame. The following procedure is used to find the translations that will place the data from each time step into the same reference frame.

At each time step, there is a three-dimensional array of cubic voxels. Each voxel, indexed i, j, k, contains an Euler angle triplet that characterizes the orientation of that voxel. The sequential volumes do not have the same number of voxels, do not cover the same field of view, and are not aligned so that we do not know the correspondence between the voxels. In addition, because the grains in the latter volume have evolved during annealing, they are similar, but not identical.

- It is assumed that there is some rigid translation such that a voxel in the first volume (i₁, j₁, k₁) corresponds to the same location as a voxel in the second volume (i₂, j₂, k₂).

- It is assumed that at this translation, the average disorientation between voxels with the same index in the two volumes will be a minimum.

A program was written to find the translation that minimizes the average disorientation between voxels in the two volumes:
\[ \overline{\Delta g} = \frac{\sum_{k=1}^{Z} \sum_{j=1}^{Y} \sum_{i=1}^{X} g_{ijk, t=1} g_{ijk, t=2}}{N} \]

where \( X, Y, \) and \( Z \) are the numbers of voxels in the three perpendicular directions, \( g \) is an orientation, and \( N \) is the number of locations where position \( i, j, k \) at both \( t=1 \) and \( t=2 \) contain non-zero data.

The program computes the average disorientation at 729 different translations from -4 voxels to +4 in the three perpendicular directions and selects the minimum. It is therefore necessary to have a preliminary approximate alignment within ± 4 voxels in each direction. In practice, the first volume is shifted to this approximate alignment before the calculation of the average disorientations at each position within the 9 x 9 x 9 range of translations. The approximate alignment can be found by randomly sampling the space of translations or by aligning the centroids of the largest grains.

In all the cases examined, the minimum disorientation was a unique and global minimum for all translations. For example, for time steps four to five, the average disorientation in a plane of constant \( k \) (the plane containing the point of minimum disorientation) is illustrated in Figure 5.1.
5.3.2 Grain Boundary Velocity Calculation

At this point it is pointed out to the reader that over the course of this chapter the words- “face” and “grain boundary” have been used interchangeably.

Grain Boundary Velocity has been quantified based on the number of voxels that change their orientation from one grain to another. This method was originally proposed by Prof. Carl Krill. As per the 3D reconstruction, each grain is made of voxels. An array of these voxels constitutes the 3D object that is the microstructure. Each voxel can be identified by a unique id assigned to it by dream3d. It is assumed that this id stays invariant over every anneal state. The mean velocity of a boundary separating two grains is the number of voxels whose orientation changes from one grain to another, divided by the grain boundary area and annealing time. To understand this visually we look at the schematic in Figure 5.2(a) shows grains 1(blue) and 2 (red) in Anneal 0 and (b) in Anneal 1 in 2D. These grains share a boundary AA` which has changed from the solid line to the dotted line from (a) to (b). The blue region between the two boundaries is denoted as “C” and the red region “D”. Grain 1 has lost region C voxels to Grain 2 while gaining voxels in region D from Grain 2. The volume exchanged can thus be specified as:

\[ Vol \text{ exchanged} = \{\text{vox} \in (C)\} \pm \{\text{vox} \in (D)\} \]
We identify the voxels that were a part of Grain1 in Anneal 0 but now are in Grain2 in Anneal 1 as voxels “lost” by Grain 1. Similarly, voxels that used to be in Grain2 in Anneal 0 but are in Grain 1 in Anneal 1 qualify as voxels “gained” by Grain 1. It is to be noted that only voxels gained and lost between Grain 1 and Grain 2 will be considered with respect to the boundary shared by them. This voxel trade-off can be quantified and converted to volume (by multiplying the volume of one voxel- $21\mu m^3$ to the number of voxels). Volume divided by average grain boundary area and annealing time would give us a quantity with the units of velocity. This is the grain boundary velocity. It can be achieved in two ways –by adding or subtracting the grain and lost voxels:

$$ Vel_{Face_{AA'}} = \frac{\{vox \in (C)\} \pm \{vox \in (D)\}}{Area_{AA'} * \Delta t_{An01}} $$

The algorithm for velocity calculation was validated by running it on fabricated microstructures with “before “and “after” states with known velocity.
We have calculated velocity in two ways- by addition and subtraction of exchanged voxels and found that their trends with GB curvature and crystallography were same, however the addition method gives larger magnitudes in velocity. The subtraction method may falsely give zero velocity when the number of voxels gained equals number of voxels lost by a grain so despite the boundary having moved, it registers as a zero-velocity boundary. Thus, the velocity results shown henceforth use the addition of exchanged voxels.

Figure 5.3 and Figure 5.4 show actual grain boundary movement based on reconstructed nf-HEDM data. In Figure 5.3 the boundary between grains A and B has clearly moved leftwards in Anneal 2 (bottom) as grain B consumes parts of grain A in Anneal 2 to become grain B’ and grain A’ respectively. Similarly, in Figure 5.4, the boundary between the large blue grain and the small purple grain has been visualized. This boundary has moved towards right, the bigger grain having consumed parts of the small grain in the process.

*Figure 5.3. Visualization of Grain Boundary movement from reconstructed HEDM data. The figure shows two grains A and B separated by a purple boundary in initial (top) and final (below) states. The boundary seems to have moved towards the left leading to the Grain B consuming part of Grain A.*
5.3.3 Outliers in Grain Boundary Velocity

An outlier in a dataset can be defined as a value that lies “outside” the distribution of the data. In other words, a value deemed as abnormally large or low with respect to rest of the values. They can arise from segmentation artifacts or could be telling something potentially of scientific interest. These outliers increase the variability of the data, affect the mean of the data and thereby decrease statistical power.

In that respect, we look at two extremes in this data – zero velocities and high positive velocities. Zero velocities are the lowest velocities obtained from velocity estimation. This means when we get zero velocity, there were no voxels gained or lost between the two grains sharing a boundary. An example of this is shown in Figure 5.5.
In Figure 5.5 the grain boundary in Anneal 1 has grown by a significant quantity in Anneal 2. This however qualified as zero grain velocity. This is because the voxels gained by either grain in the next state were not a part of each other but other neighboring grains (that are not shown in Figure 5.5). Every time the boundary grows vertically but does not exchange voxels laterally across the boundary, our algorithm yields zero velocity, which according to our definition of velocity is the correct result. This however accounts for less than 5% of the boundaries considered and thus is not considered to have any significant effect on the velocity calculation. Another outlier is the very high value of velocity. One such case is shown in Figure 5.6.
This figure (above) shows that the boundary initially connected by a single voxel (or triangle in the meshed image) enlarges as both grains inch closer to each other sharing significant area between them in the next state (right). The high value of velocity obviously comes from low grain boundary area in the initial state.

There are several challenges in tracking boundaries across states. For example, in Figure 5.7 we see that the boundary between the two grains in (a) does not exist in (b).

This happened because the lower part of the multicolored grain is not associated with the same in the final state. This could happen if the average orientation of the voxels comprising the lower part of the grain changed in such a way that it overcame the misorientation tolerance set for grain segmentation. Since the boundaries are matched by Ids, this boundary is lost. Figure 5.8 shows another such example. Note that these anomalous event are either missed (Figure 5.7) or make a minority contribution to the data (Figure 5.5); the majority of the data reflect or conventional migration events such as that illustrated in Figure 5.4.
Figure 5.8. Another example of boundary not being trackable since the part of the grain that was sharing the boundary is a different grain now.

5.4 Results

5.4.1 Characterization of Grain Boundary Area

Nickel as a fcc crystal has a large fraction of annealing twins which accounts for almost 29% of all boundaries in the microstructure [138]. Figure 5.9 shows the distribution of Grain Boundary area for all boundaries with a 60° misorientation about a common <111> axis. While the peak position remains unchanged going from the initial anneal state (Anneal 0) to the final anneal state (Anneal 5), the intensity of the peak increases monotonically going from 140 MRD in Anneal 0 to almost 300 MRD in Anneal 5. The position of the peak depicts the crystallographic orientation of the GB plane and the intensity of the peak captures the relative area of GBs with that specific orientation (∑3 twin boundaries in this case).
Figure 5.9. Distribution of grain boundary planes at a disorientation of 60° by [111] axes for all anneal states.

Figure 5.10 shows the total GB area within twin boundaries and random (non-twin) boundaries. Twin boundaries are observed to have higher area than random boundaries. More than 99% of twin face areas are within $1.2 \times 10^{-8}$ m$^2$ while more than 99% of the random boundaries are within $0.5 \times 10^{-8}$ m$^2$. In fact, the largest face in the entire microstructure is a twin face (Fig.5.11).
Figure 5.10. Cumulative Probability plots of Grain Boundary area for $\Sigma 3$ twins, general boundaries and both together. CDF plot has been magnified to show the difference clearly.

Boundaries with areas less than 20 $\mu$m$^2$ have been classified as outliers and eliminated from calculations. These small GB areas are visualized as the encircled regions in Figure 5.12(a) and (b). Note that the area of one voxel face is 10 $\mu$m$^2$.

As observed here, these are likely to be data processing artifacts arising from Dream3d meshing procedures.
5.4.2 Characterization of Grain Boundary Velocity

Figure 5.13 shows the distribution of GB velocity of all boundaries in matched pairs. The histogram shows a half-normal distribution with a right skew. The mean velocity is 0.09 μm/min with a standard deviation of 0.1 μm/min. The mode of this distribution is close to 0.05 μm/min.

![Histogram of Grain Boundary Velocity](image)

*Figure 5.13. Grain Boundary velocity (μm/min) distribution of all faces for all anneal states combined*

Figure 5.14 shows CDF plots of twin and non-twin boundary velocities. Twin boundaries in general show lower velocity than other boundaries.
Figure 5.14. Cumulative probability velocity distribution for twin boundaries, general boundaries and all boundaries. The plot is magnified to show differences clearly.

5.4.3 Grain Boundary Curvature

Grain Boundary curvature is calculated as the area weighted average of the mean curvatures of all the meshed triangles belonging to that face. The calculation is described in Section 5.3.2.

Figure 5.15. Absolute grain boundary curvature (1/μm) distribution of all faces in all states
Figure 5.15 shows the distribution of the absolute face curvatures of all states together, the mean curvature is 0.09 $\mu m^{-1}$ and standard deviation around 0.1 $\mu m^{-1}$. The distribution fits a half-normal shape and is right skewed.

The one-way analysis of variance (ANOVA) was used to determine statistical differences between GB velocities for every anneal state. It is a statistical method used to test the null hypothesis. The null hypothesis states that all groups are independent. In other words, it assumes at least one mean is statistically different. This assessment is done with the p-value. P-value is the probability that an observed difference in means could have occurred by chance i.e. it assumes that the null hypothesis is true [139]. Figure 5.16 shows the box plots for boundary curvatures for all states. The labels C0, C1, C2, C3 and C4 indicate the anneal state and C denotes curvature. Thus, C01 denotes the absolute GB curvature of Anneal 0 for boundaries that matched between Anneal 0 and Anneal 1. One way ANOVA was performed for these curvatures. The p-value was 0.37 meaning that there is no significant difference between the grain curvatures of each state. It can be noted from the box plots that the means of each state are around 0.025 $\mu m^{-1}$ and do not seem to vary significantly or monotonically from each other.
This goes on to show that the short annealing intervals did not provide enough time to cause drastic changes in curvature. The similarity of the curvatures in each state indicates that the driving force was constant (within experimental uncertainty), so we can aggregate the velocity and curvature data from all the anneal states and analyze it as one set of data.

5.4.4 Correlations between parameters

Figure 5.17 looks at the relationship between GB curvature and GB area as a binned and a scatter plot. Boundary curvature is inversely related to boundary area with a correlation coefficient of -0.63 of the binned means. In plot (a) the error bars represent the first and third quartiles of the data in each bin and (b) shows the scatter plot with the red markers as bin means from (a).
Figure 5.17 (a) and (b) show the face curvature (1/\( \mu m \)) as a function of face area (\( \mu m^2 \)) as binned and scatter plots respectively.

Figure 5.18 plots the face curvature as a function of fractional change in face area. The mean values show that the lowest curvature occurs at zero fractional change in area. This implies that the boundaries with the lowest curvature do not change significantly going from one anneal state to the next. Figure 5.19 shows the grain boundary velocity as a function of the magnitude of boundary curvature for all boundaries. However, grain boundary velocity shows a poor correlation with curvature (r = -0.02).
Figure 5.18. (a) Binned plot and (b) scatter plot of Absolute GB curvature (1/μm) as a function of fractional change in boundary area. The black and red markers in either plot denotes the bin means and the bars in (a) denote the standard deviation.

Figure 5.19. (a) Binned plot of GB velocity (μm/min) as a function of GB curvature (1/μm) and (b) scatter plot of the same. The black markers in both the plots are the bin means.

5.4.5 Anisotropy of Grain Boundary Motion

Figure 5.20 shows the variation of GB velocity as a function of grain disorientation without any misorientation axis information. The disorientation angle is binned into 5° intervals. Figure 5.20 (a) plots the average Gb velocities for each bin. There is a peak in mean velocity at around 35°
disorientation and reaches a local minimum at 60°. The interquartile ranges from Figure 5.20 (b) shows that there is significant variation within boundaries with similar disorientation angles.

We also look at velocity of selected twist or symmetric tilt CSL boundaries namely \( \Sigma 3, \Sigma 5, \Sigma 7 \) and \( \Sigma 9 \). From Fig.5.21 (a) it can be seen that \( \Sigma 7 \) CSL boundaries about the [111] axes have the highest mean velocity. \( \Sigma 7 \) also has the widest range in velocity as seen from the box plots in (b). It can also be noted that \( \Sigma 3 \) boundaries around <111> axis have the lowest velocity closely followed by \( \Sigma 9 \) symmetric tilt boundaries.

![Figure 5.20](image1.png)

**Figure 5.20.** (a) Binned mean GB velocity (\( \mu \)m/min) plotted as a function of disorientation angle in degrees and (b) box plots of GB velocity as a function of disorientation angle in degrees.

![Figure 5.21](image2.png)

**Figure 5.21.** (a) Mean and (b) box plots for GB velocity (\( \mu \)m/min) for low CSL grain boundaries such as \( \Sigma 3, \Sigma 5, \Sigma 7, \Sigma 9 \) and \( \Sigma 11 \).
Next, we examine the distributions of GB velocities, GB curvature and GB population at fixed misorientations of $\Sigma 3$, $\Sigma 5$, $\Sigma 7$ and $\Sigma 9$. These are shown from Figure 5.22 to Figure 5.25. Figure 5.22(a) plots the distribution of all grain boundary velocities with a misorientation of 60° about [111] on a stereographic projection and Figure 5.22(b) plots the grain boundary curvatures for the same and (c) plots the grain boundary populations. It is noted that curvature is minimum at the coherent twin position. The minimum values of GB velocity are also at the same position as curvature. This is the twist boundary bounded by (111) planes on both sides and thus is flat. However, it can be noted that the maximum curvature and velocity positions are not the same. While the curvature is highest at the $(1\bar{1}0)$, $(0\bar{1}1)$, $(\bar{1}01)$ and $(\bar{1}10)$ boundary planes, the velocity is highest at $(1\bar{2}1)$, $(\bar{1}\bar{1}2)$ and $(\bar{2}11)$ tilt grain boundary positions. $\Sigma 3$ twins have the highest fraction and lowest peak velocity at 150 MRD and 0.09 $\mu$m/min respectively. The lowest curvature and velocity corresponds to the population maxima. 

The velocity and curvature for $\Sigma 5$ and $\Sigma 9$ does not have any strong correlation (Fig. 5.23 and Fig.5.25). The velocity in $\Sigma 5$ varies from 0.05 $\mu$m/min to 0.13 $\mu$m/min. The minimum curvature position at the (100) twist position is the minimum velocity. Also, the GB curvature distribution of $\Sigma 5$ looks approximately inverse of the grain boundary plane distribution i.e. highest curvature is found at the population minima and lowest curvature at the peak population. For $\Sigma 5$ boundaries the GB population is highest around $(\bar{2}10)$ and $(2\bar{1}1)$ planes on the left and $(210)$ to $(2\bar{1}0)$ on the right. The GB curvature is lowest at these positions and highest at $(012)$ and $(0\bar{2}1)$. $\Sigma 5$ and $\Sigma 9$ do not show any strong correlations between velocity and curvature. Fig. 5.24 (a) and (b) show a negative correlation between velocity and curvature for $\Sigma 7$ boundaries. The highest velocity reaches 0.22 $\mu$m/min at (111) twist grain boundary position.
The boundary population for $\Sigma 7$ is at its peak at (111) position which is also the position of highest velocity and lowest curvature.

Figure 5.22. (a) Velocity ($\mu$m/min), (b) Curvature (1/$\mu$m) and (c) population distribution (MRD) for all grain boundaries with a $\Sigma 3$ (60°[111]) disorientation.

Figure 5.23. (a) Velocity ($\mu$m/min), (b) Curvature (1/$\mu$m) and (c) population distribution (MRD) for all grain boundaries with a $\Sigma 5$ (36.86°[100]) disorientation.
5.5 Discussion

The lack of a positive linear correlation between GB velocity and curvature (Figure 5.19) brings into question our understanding of curvature driven boundary movement in 3D polycrystals and whether there are other factors that aren’t being considered. An assumption in the Gibbs
Thomson equation is that for curvature driven boundary motion, the grain boundary curvature is uniform. In other words, the boundary for any grain would fit any part of a solid sphere. That however is not true for the nickel sample which has irregularly shaped grains. Figure 5.26 shows that the triangle curvatures in a grain boundary may not necessarily be uniform.

![Figure 5.26. Visualization of two grain boundary in the twinned microstructure with plane normal situated at each meshed triangle. (a) shows a ragged boundary while (b) shows a relatively uniform boundary.](image)

This means that the triangles making up a face do not have their centers of curvature in the same side of the boundary, and consequently both concave and convex triangles are present in one.

This might lead to complexities in grain boundary motion thereby making the linear relationship curvature and velocity too simplistic.

Another possible reason for the lack of correlation could be grain boundary stiffness. There has been a recent study on the role of interfacial stiffness on GBM [140]. In this study, they showed that GB stiffness greatly influences curvature driven interfacial motion as well as boundary faceting. Using atomistic simulations they showed that GB stiffness can be more anisotropic than GB energy and create structural instabilities. For boundaries with negative stiffness, there were instabilities related to distortions such as faceting and re-construction processes. They also found
that the most stable Σ3 boundaries were involved with {111}, {112} and {110} planes with respect to stiffness. However, it is not possible to determine the occurrence of faceting in our data due to the limitations of resolution and 3D reconstruction.

Another aspect of GB motion that has not been addressed in this work is the role of triple junctions (TJ). It has been shown previously that when grain growth is governed by triple junction motion, the Neumann-Mullins relation is not valid anymore. More specifically the smaller the TJ mobility, the less it follows the N-M rule [141]. Triple junctions have also been shown to have a considerable effect on grain growth in nanocrystalline materials [142][143]. These studies showed that low TJ mobilities exert a drag effect on boundary motion. It is however, beyond the scope of this study to assess TJ mobility since the triple lines generated in the reconstruction follow poor smoothing procedures and hence are deemed unreliable and inaccurate.

Because of the crystal interchange symmetry at every boundary, it is not possible for curvature or velocity to have a sign. It must also be acknowledged that there are several approximations in the velocity calculation. The velocity calculated can be considered as average velocity as it counts all the voxels that changed orientation between two grains sharing a boundary. This method does not offer point by point grain boundary movement information for each voxel/triangle but a general idea of total voxel movement across a boundary. However, curvature can be calculated on a point by point basis and that could cause a discrepancy when comparing these two quantities at the triangle level.

Figure 5.20 shows that GB velocity changes with disorientation. Fig. 5.22 to Fig. 5.25 also show that for special sigma boundaries, Gb velocity and curvature vary with the grain boundary plane orientation. Thus these observations provide clear evidence that mean grain boundary velocity
depends on the crystallographic parameters. For example in Σ3 boundaries, the velocity varies from 0.055 μm/min at the (111) twist boundary to 0.095 μm/min at the (211) type boundary planes. We also see significant variations in velocity of Σ5, Σ7 and Σ9 misorientations. Fig. 5.21 (a) shows that Σ7 boundaries have the highest and Σ3 have the lowest velocity. This agrees with previous studies where it was found that boundaries with a misorientation of 38° around <111> have the highest mobilities and Σ3 twins owing to energy cusps have been recorded to have low mobilities [28][33][123].

Grain boundaries statistically do not show a strong correlation with curvature except for Σ3 and Σ7 boundaries. There is a direct positive correlation between the Σ3 twin boundaries with a correlation coefficient of 0.9 and a direct negative correlation between velocity and curvature of Σ7 type boundaries with a correlation coefficient of -0.4 as shown in Figure 5.27 and Figure 5.28. These figures plot the same data points as seen in their respective stereographic projections (Fig. 5.22 and Fig. 5.24).

This means for Σ3 boundaries, GB velocity is directly proportional to magnitude of curvature. In other words, lower curvature boundaries have low velocities.
Grain boundary energy is inversely proportional to grain boundary population [48]. Owing to the lower energy of twin boundaries they generally have higher boundary area than other boundaries as shown in Figure 5.9. The GB velocity calculation involves normalizing the displaced volume by grain boundary area. This could be yet another factor why Σ3 boundaries have low velocities. However for Σ7 boundaries, a clear correspondence between boundary area and GB velocity could not be established based on the current data.

5.6 Conclusion

Grain boundary velocity and curvature of high purity nickel annealed at 800°C for six time intervals was calculated and analyzed with respect to crystallographic parameters. The following conclusions were drawn from this work:

- Generally grain boundaries do not show any clear correlation of GB curvature with GB velocity. This contradicts a part of Hypothesis 3.
- Grain Boundary velocity and curvature are anisotropic and vary with grain misorientation and grain boundary plane orientation. This supports a part of Hypothesis 3.
- Σ3 twin boundary velocities have a positive linear correlation with GB curvature with a correlation coefficient of 0.9. This means that the flatter the Σ3 twin boundary, slower is the boundary movement.
- Σ7 boundaries have shown negative correlation of GB velocity with GB curvature (correlation coefficient = -0.4). Σ7 boundaries also have the highest mean GB velocity.
6 Comparison of Experiment with Simulation

6.1 Abstract

An isotropic simulation model based on threshold dynamics was used to predict grain growth with the Ni experimental microstructure as an initial state for every two consecutive anneal states. The simulations were compared with the final experimental anneal states. Grain volumes, number of neighbors and boundary curvatures of the predicted and observed results were compared. The differences between the simulated and measured grain volumes increased with the number of topological changes that were not correctly predicted by the simulation. The simulation also produced boundaries that have reduced curvatures compared to experimental observations. Both factors likely contribute to the inaccurate prediction of volume changes.

6.2 Introduction

It has already been established that Nickel is a highly anisotropic metal [144]. This means there is variation in properties including the grain boundary energy and mobility with crystallographic direction/orientation. This fcc crystal also has an abundance of $\Sigma 3$ twin boundaries (~29% boundary fraction) that have properties that are very different from other grain boundaries. Thus, to better interpret anisotropic microstructural evolution, we compare it with isotropic grain evolution and investigate all points of difference such as volume and topological changes and attempt to uncover the origin of these differences.

This chapter seeks to compare grain growth of Nickel with an isotropic simulation model. This model uses 3D Threshold Dynamics [145] for simulating the motion of interfaces. This simulation uses each reconstructed experimental microstructure as the initial state and attempts
to predict the next anneal state. The Threshold Dynamics model simulates microstructural evolution by applying curvature driven grain boundary motion. This simulation was conducted by our collaborators in the Civil Engineering Department namely Prof. Kaushik Dayal and Xiaoyao Peng. For more information on the simulation one may refer to [146]. My contribution was to compare the outcome of this simulation to the experiment.

There have been numerous efforts in the recent years to understand grain growth with the help of simulation. There have also been several methods starting with Monte Carlo simulations known as Potts model [147] [148] [149] [150] cellular automata models [151] [152], molecular dynamics [153][154], vertex simulations [155] [156] and Phase field models [80] [157][158]. Recent studies of 3D grain growth using simulation have attempted quantify and understand topological, volumetric growth rate, grain boundary energy evolution in isotropic as well as anisotropic grain growth [157][159][160][99].

There have been several attempts in the past to compare the results of grain growth simulations with experiments on a grain-by-grain basis. For example, in 2003 Demirel et al. [161] validated anisotropic grain growth simulation in 2D using GB curvature as a driving force as it explained 50% of the experimental grain growth. They also showed that isotropic simulation with 17% explaining power has very poor matching with experimental evolution. McKenna et al. [82] compared evolution of individual grains between experiment and 3D isotropic phase field simulation in polycrystalline Ti-β-2S. Statistical analysis revealed good agreement between experiment and simulation for grain growth kinetics. However, direct comparison of individual grains revealed a poor match in grain shapes and grain boundary widths because the simulation was unable to capture local anisotropy in grain boundary energy and mobility.
Another study by Zhang et al. [137] compared phase field simulation of grain growth with 3D experimental data to determine reduced mobilities of grain boundaries. They conducted a detailed analysis of these reduced mobilities with respect to GB crystallography and found that mobility is independent of crystallography and determined that it can be affected by time and topological events instead.

There are six anneal states in the experiment as described in [138] where high purity Nickel sample was annealed for regular time intervals. To match the isotropic simulation, each experimentally mapped microstructure was modified to merge all grains sharing a twin boundary. This was done to reduce the anisotropy from the odd shaped grains. Each experimental state was used as the initial state for simulation to predict the next state. The comparisons between simulation of the final state and experimental final state for each pair was made. The point of this chapter is to compare the measured distribution of grain characteristics with that predicted by the simulation. By comparing the predicted and observed geometric characteristics of individual grains, it is possible to say they are not the same, but it is not possible to understand the source of the differences. Here, we compare measured and predicted distributions of grains sizes, neighbors, curvatures and volume changes. There are systematic differences between the simulation and experiment in the volume changes of grains that grew and shrank the most.

6.3 Results

All anneal states have been translated to a common spatial reference frame by minimizing voxel misorientation as explained in Section 5.3.1. Next, all states (experiment and simulation) were cropped to include the same volume between the beginning and end of each anneal state.
Figure 6.1. Normalized grain size distribution in Anneal 0 and Anneal 1 of experiment and Anneal 1 of simulation

Figure 6.1 displays the grain size distribution for experimental states An0-exp and An1-exp and simulated An1-sim. An0-exp was used to simulate An1-sim. The histogram plots grain radius normalized by average grain radius. The distribution is self-similar for both cases of experiment as well as simulation.

Figure 6.2(a) shows the experimentally measured microstructure of an initial anneal state of 921 grains, (b) the experimental final state microstructure with 756 grains and (c) the simulated microstructure of the final anneal state with 601 grains. The average grain size in (a) is 32.1 µm, in (b) is 32.7 µm and in (c) is 34.7 µm. Between the grains that matched, the reconstructed initial experimental state has an average of 11.12 faces per grain, while the final state has 10.99 faces per grain and the simulation state has 9.6 faces per grain. Comparing the top surface between the two, the interfacial network of the simulated grains is smoother than the experiment. This will be addressed in detail later.
Figure 6.2. (a) Experimentally measured microstructure of the initial anneal state of 921 grains and (b) experimental final state with 756 grains and (c) simulated microstructure of the final anneal state with 601 grains.

Figure 6.3 takes a closer look at the faces of a grain for the initial and final experimental state (a), (b) and the final simulation state (c). Differences in grain face evolution are revealed upon comparison of the grain faces between the two. Complex topological changes in the experiment have not been well captured in the simulation.

While, there are six anneal states, each state was used as an initial state to simulate the next one. A bulk grain has been visualized at all states as it evolves from Anneal State 0.
to Anneal State 5 in Figure 6.4. This grain is within the bulk of the microstructure and shrinks with annealing in the experiment as well as simulation.

![Figure 6.4. Visualization of a bulk grain across all Anneal States in experiment and comparison with simulation of the same grain. The top row and bottom row shows experimental and simulation grains starting from Anneal 0 to Anneal 5 respectively.](image)

The experimental and simulated changes in grain radius for the grain visualized in Figure 6.4 has been plotted in Figure 6.5 as a function of the anneal states. Starting at radius of 42.2 µm for both, the experimental plot shows the grain grew till An2 followed by a drop to 40.8 µm while the simulated grain also shrunk in the final state (39 µm) it did not follow the same behavior as can be seen in all the states. For example, while both started from the same radius, the simulated grain shrunk between An0-An1 but in experiment it grew. Ultimately though, the grain shrunk and the simulation could capture that.
Figure 6.5. The actual and simulated evolution of the grain size of the bulk grain in Figure 4 at every anneal state.

Figure 6.6 plots the histogram of fractional change in volume for experimental and simulated grains. The data includes the fraction of grains that matched from the initial anneal state to the final anneal state. It is seen that a higher fraction of simulated grains (17%) have shrunk to zero than they did in the experiment (10%). In both simulation and experiment, the maximum of the distribution is for grains that have small volume changes near zero. Also, there are more grains in the experiment that increase in size as shown by the right tail of the distribution.

To understand Figure 6.6 in more detail we look at Figure 6.7, which plots the fractional change in volume as a function of grain radius for both simulation and experiment. The simulation follows isotropic normal grain growth which tends to shrink the small grains and enlarge the bigger grains as the orange plot points have a steep fall in the lower grain radius bracket (5-25 μm) while in experiment there are comparable number of data points at both sides of the y-axis for the same size range. This means that while almost all small grains are indiscriminately being shrunk in simulation, a good portion of them showed significant growth in experiment. There are
fewer grains that have a high magnitude of fractional change (>0.6) in volume as grain size increases. When correlated with Fig. 6 it means that the lower extreme of fractional change in volume (<-0.6) corresponds to grain size less than 35 µm.

In Figure 6.8 the change in volume between the simulated and initial experiment grains is plotted as a function of the volume change of the same grains in experiment. The markers in the plot are the bin means and the bars are the standard deviation of these bins.

![Histogram of fractional change in volume for final state experimental and simulated grains. The markers show the bin centers and the lines connect bin centers to make the plot.](image)
The fractional change in volume as a function of grain size in experimental and simulated grains.

The negative extreme of the plot is much flatter; this is because some of the grains in experiment disappeared and thus did not find a match with the initial state. Therefore, the sum of the increased and decreased volumes in not zero. In other words, the volume is not conserved because of the over prediction of grain shrinkage. The plot shows a positive direct relationship between the experimentally observed and the simulated change in volume.

When the experimental volume change is greater than zero, the simulated volume change also is positive, and we see a positive monotonic trend between the two. The slope of the trend is lower than the ideal 45° line, meaning simulation under-predicts the grain growth. For grains with a negative change in volume in experiment, the simulation did not do a good job of predicting as most of them have reported a positive change in volume. It was
found that for all matched grains between each anneal state pair, considered together, the simulation has correctly predicted the sign of volume change 62% of the times.

The microstructure in both simulation and experiment is meshed and smoothed with the help of Dream.3D (Section 4.1.1). Figure 6.9 shows a comparison between the unsigned triangle curvatures for all matched grain boundaries of the experiment and simulation. The y-axis displays the fraction of triangles in simulation and experiment. The simulation has more triangles with lower curvature and fewer triangles with higher curvature. The lower curvatures have been overpredicted by the simulation while it is underpredicting higher values of curvature. In other words, the simulation produces smaller curvatures of grain boundaries. Experimental curvatures have a higher fraction of triangles for curvatures higher than 0.0055 µm⁻¹ while simulation triangles have a higher fraction for lower curvature values (14%) than experiment (12%).
Another approach for determining the degree of robustness of the simulation was to compare the volume prediction error (VPE) with the topological error (TE) for individual grains. VPE and TE are defined as follows:

\[ \Delta N_s = N_{sim} - N_{exp\ (initial)} \]
\[ \Delta N_e = N_{exp\ (final)} - N_{exp\ (initial)} \]

**Topological Error (TE)** = \( \Delta N_s - \Delta N_e \)

**Volume Prediction Error (VPE)** = \( \frac{Vol_s - Vol_e}{Vol_e} \)

\( VPE \) is the fractional difference in volume predicted by simulation of final anneal state and experimental final state. \( TE \) is the difference in \( \Delta N \) for each grain between simulation and experiment. In other words, \( TE \) is the error in predicting topological evolution by the simulation. Figure 6.10 plots the volume prediction error as a function of topological error.

*Figure 6.9. Absolute triangle curvatures for all boundaries in the simulation and final experiment state.*
A low VPE indicates small difference between the final volume predicted and actual final volume of the grain. A high TE value means there is a large error in predicting the topological change in grains. This plot is approximately linear and monotonically increasing. When TE is close to zero, VPE is also close to zero. As TE increases, the error in the predicted volume also increases, in both positive and negative directions.

Figure 6.10. The volume prediction error (VPE) as a function of topological error (TE). VPE is the fractional difference in predicted and observed grain volume. TE is the difference in grain face evolution between simulation and experiment.

6.4 Discussion

The grains that disappeared in the simulation of Anneal 1 from Anneal 0 are visualized in Figure 6.11. When the grains that the simulation shrank are examined in experiment, it is seen that 33% of them grew. The average radius of simulated shrinking grains was less than 60% of the average radius. Basically, the simulation shrunk smaller grains and grew larger ones which agrees with conventional thinking [67]. Figure 6.4 showed that the simulation approximately maintains the shape and size of this grain. The volume change of
this bulk grain is also predicted reasonably. The fractional change in volume plots of Figure 6.6 and Figure 6.7 show that simulation shrinks more grains than experiment. It is observed that maximum change in volume occurs for smaller grain sizes. Thus, it is at the lower grain size that the simulation fails the most since small grains have been known to grow in experiment (Figure 6.7).

At the grain boundary level comparing triangle curvatures of simulation and experiment, one may reason that the simulation predicted different volume changes because the evolution of curvature is different. Higher number of triangles have lower curvature in simulation than experiment which confirms that simulated curvatures are smoother (Figure 6.9).

![Visualization of the 121 grains in Anneal State 4 that shrunk to zero in Anneal 5 in simulation. These grains are colored by IPF coloring and are opaque while the rest of the microstructure is transparent.](image)

Smother curvatures would mean a lesser driving force for grain boundaries. This would lead to lesser change in volume or underprediction of grain volume increases as observed in Figure 6.6. However, it should be acknowledged that we do not know if the rougher
interfaces in the experimental data are realistic or an artifact of uncertainties in the measurement and reconstruction.

We also compare a single face in the final experiment and simulation as shown in Figure 6.12. Visualization of the face shows that it is more curved in the experiment than in simulation. The cumulative distribution function plot of triangle curvatures of these faces also indicates that simulation has lower curvature than experiment in general.

![Figure 6.12](image.png)

**Figure 6.12.** Comparison of triangle curvatures of a single face as a CDF plot (right) and visualization of the same grain face in experiment (An5-exp) and simulation (An5-sim) on the left.

### 6.5 Conclusions

High purity nickel microstructure data mapped from an interrupted annealing experiment was used as an initial state for an isotropic simulation of grain growth. The measured microstructure was modified by combining all neighboring grains with a twin misorientation into a single untwinned grain. The topological and volume changes were compared for both individual grains as well as an ensemble. Each state from Anneal 0 up till Anneal 4 was used as an initial state for the simulation and the consecutive state as final state was compared with the simulation. In
general, it was observed that the simulation shrank more grains than experiment. Also, smaller grains shrunk more than larger grains grew. Although the simulation used interfacial mean curvature as a force for driving grain growth, we see that it was unable to capture perfectly the morphological and topological changes in the microstructure. On this basis, we have the following conclusions:

- While grain radius histograms show good prediction by simulation, on average the simulation predicts the correct sign of volume change only 62% of the times. Based on the fractional change in volumes, it is seen that grain shrinkage is overpredicted while grain growth is underpredicted.

- The simulation has systematically lower grain boundary curvatures than experimental observations. This could lead to lower driving force for grain boundary motion and hence explain the errors in prediction of grain volume changes.

- Volume prediction errors are correlated to errors in predicting topological changes. When the simulation captures the topological changes correctly, it can predict the volume change accurately as well.
7 Canonical Correlation Analysis

7.1 Abstract

Canonical Correlation Analysis is performed on a multivariate input and output dataset. It is used to analyze the as-received experimental twinned microstructure of Nickel and a modified microstructure of the same obtained after merging twin related domains. Grain characteristics such as grain radius, grain curvature and local grain neighborhood predict grain volume changes better in the twin merged microstructure than the twinned microstructure.

7.2 Introduction

Better understanding of the relative importance of different factors modulating grain growth for different systems helps with better control of the microstructural evolution. A better understanding of processing-structure-property relationships in materials with the help of predictive models will not only help reduce experimental trials but also guide in designing them. As our ability to control the evolution of microstructure improves, so does our ability to control its dynamic mechanical properties.

Canonical Correlation Analysis (CCA) is a statistical tool for multivariate analysis that explores the interrelationships between multiple input variables and multiple output variables [162][108]. The data is divided into two sets – an independent (typically input) set and a dependent (typically output) set. CCA endeavors to find linear combinations from both sets to maximize the correlation between the two. CCA has been used in several fields such as sensitivity studies, image analysis, economics among other things [163][164]. CCA allows for multiple independent
and multiple dependent variables, dimensionality reduction as well as interrelationships and that is why this is preferred over other statistical techniques. [165][166].

Grain growth is a complex phenomenon guided by several factors. For years, there have been studies conducted to determine the key parameters driving grain growth. Studies of critical grain size [67] number of neighbors [40,59,60,61,62] grain boundary curvature [69] etc. have been known to influence volumetric grain growth rate in 2D and 3D [6][125]. For more information on grain growth theories one is directed to read Section 2.4.1.

The data generated through the nf-HEDM experiment yields a lot of information about individual grains including grain size, grain volume, grain orientation, grain shape, number of neighbors of the grain, and curvature. We want to be able to predict or find the governing factors in the growth of a grain using information related to every grain before and after growing/shrinking. There are several different data analysis techniques available such as Principal Component Analysis (no interrelationships, dimensionality reduction), simple linear regression (univariate), Multiple linear regression (several independent variables but one dependent variable), Machine Learning (has hidden layers, complex mechanism, difficult to understand inner workings of model, works best with huge amount of data) [167][168][169].

In this study grain characteristics based on previous studies and present work have been chosen to illustrate the CCA methodology for quantifying parameter sensitivity and interrelationships.

The input parameters include grain radius, number of grain faces, F-<F>, grain curvature and normalized integral grain curvature (with and without triple lines) as discussed in Chapters 3 and 4. Metrics such as misorientation between the same grain in two time states, misorientation within a grain, grain shape metrics such as Surface area to Volume fraction and moment invariant (Omega3) have also been considered. For the output, the change in volume (DelV) and
difference in volume raised to the power of two-thirds (DV23) have been used. The idea is to
determine which of the input parameters are most strongly correlated to observed volume
change.

One to one parameter correlations assume that each parameter is independent of the other.
However, for complex phenomena like grain growth, this may not be true. This necessitates the
need for a holistic multivariate approach to analyzing parameter sensitivities as well as their
relationships. The main objective of this study is to determine the most relevant input variables.
Also, as seen in Chapter 3, grain growth is better predicted when the microstructure is modified
by merging adjacent twin related domains. Thus, canonical correlation analysis for grain growth
will be conducted for both twinned microstructure as well as the twin merged microstructure.
The objective is to see what percent of variance in the data can be captured by the input variables
and to determine which of the two microstructures is better correlated to the input parameters.

7.3 Methods

CCA is based on the principle of maximizing correlation between two sets of parameters by
comparing linear combinations of different parameters in both sets. As shown in the schematic,
the input and output parameters are linearly combined to form canonical variates X and Y
respectively. The first canonical set tries to maximize the correlation between the two canonical
variates. The residual variance left from the first set is further explained by a second set of
canonical variates, which again attempts to maximize the correlation. This approach is repeated
till the number of canonical sets reach the number of variables in the smaller set between input
and output. If the residual variance is fully explained before this limit, then further canonical sets
are obviously not needed. In this way, several canonical pairs are generated until the entire
dataset has been explained.
Canonical variates are synthetic variables created by linearly combining observed variables.

Depending on the dimensions of the output set, there can be as many canonical functions. They are given as follows:

\[ U_x = A_x X \]
\[ V_y = A_y Y \]

\( A_x, A_y \) are datasets, \( x, y \) are directions, \( U_x \) and \( V_y \) are the canonical variates.

The canonical correlation (\( \rho \)) between the canonical variates is given by:

\[ \rho = \frac{V_y' \cdot U_x}{\sqrt{V_y' \cdot V_y \cdot U_x' \cdot U_x}} \]

Some of the terms used in this model are described below:

- The coefficients used in the linear combinations are called “Weights” which indicates the individual contribution of each variable towards the overall set of variables. Each
coefficient tells us about the contribution of that variable towards being able to explain the set of variables.

• The bivariate correlation between each variable and its corresponding synthetic variable is called “Loadings” or “Structure Coefficients”. This is an index of what proportion of each variable is reproducible from the total canonical results.

• The sum of all squared loadings called the “Communality Coefficient” tells what proportion of variable’s variance is reproducible from the overall canonical output.

• “Canonical variate adequacy coefficient” is the average of the structure coefficients with respect to one function. This represents how adequately a canonical variate set can explain the variance of the raw data.

• Lastly, “Redundancy coefficient” is generated by averaging the cross-loadings across all the canonical functions. It explains the average proportion of variance in one set that is reproducible from the other set [165].

In this study, CCA was conducted on all grains of Anneal 1 and compared with grain growth results with respect to Anneal 2. There was a total of 14 parameters used as input set and two parameters as output set as listed below:

• **Misor**- misorientation between the same grain in two different time steps

• **S1**- Boolean array for surface grain indicator, 1 is a surface grain

• **Rad1**- Initial grain radius of Anneal 1

• **<F>** - Average number of faces of nearest grain neighbors

• **F-<F>-** Difference between number of sides and average number of faces of nearest grain neighbors

• **C1**- Area weighted grain curvature of Anneal 1
• **C1*Area1** - Grain curvature multiplied by grain surface area

• **G** – Normalized integral mean grain curvature excluding triple lines (for more information on how it is calculated please refer to Section 4.1.2.2)

• **Omega3** - Moment invariant of grains in 3D which describes the shape of the grains such that a spherical grain would have the highest Omega3 [170].

• **FAM** - Feature Averaged Misorientation (average of the misorientation angle (in degrees) between all cells’ orientation that belong to a grain and the average orientation of the grain)

• **SAV** - Surface Area to Volume ratio. For a given shape, SAV decreases with increase in size. Also, for a given volume, a sphere has the lower surface area. Thus, SAV ratio is low for rounder shapes, and decreases with increasing volume. A high SAV would mean larger grain boundary area and thus more grain boundary free energy.

Output:

• **DelV23** - Volume$_{\text{final}}^{2/3}$ – Volume$_{\text{initial}}^{2/3}$

• **DelV** - Change in volume

### 7.4 Results

Statistical models like direct pairwise correlation and Canonical Correlation Analysis have been used to compare correlations and interrelationships between variables for grain growth between twinned and twin merged microstructures.

#### 7.4.1 Pairwise Correlation

First, we look at the pairwise correlation coefficients between the 11 input parameters and the 2 output parameters described in Section 1.3. This has been carried out for both the twinned and
twin merged microstructures with the objective of comparing the relative strength of correlations. Table 1 shows the correlation coefficients amongst all possible pairs of input and output variables. The same information can be visually depicted in the form of a heat map to provide a qualitative and intuitive summary as shown in Figure 7.2.

It is evident from the comparison of twinned (2a) and twin merged (2b) matrices that there are significant differences in the pairwise correlations. Some of the input parameters which shows the most contrasting behavior are Omega3 (shape descriptor), C1*Area, F-<F>, Rad1 and G. Omega3 shows a significant positive correlation with volume change (r = 0.394) in 2(a) but not in 2(b) (r = 0.025). Similarly, C1*Area, F-<F>, Rad1 and G shows greater correlation in 2(b) than 2(a). However, it should be noted that the pairwise correlations do not consider interrelationships amongst each other.

![Figure 7.2. XY Correlation plot showing direct correlations between all input parameters (X) and output parameters (Y) for (a) twinned microstructure and (b) twin merged microstructure. The correlation matrix is colored according to the strength of the correlation. A perfect positive correlation of 1 is dark red while a perfect negative correlation of -1 is dark blue. A value of 0 signifies no correlation which is green.](image-url)
7.4.2 Canonical Correlation Analysis (CCA)

Like the pairwise study, separate analyses have been conducted for the twinned and twin merged microstructures. The relative importance of different input parameters, the interactions between the parameters and overall variance explained by the whole set have been compared.

Redundancy of a canonical function quantifies the average proportion of variance in one set that is reproducible from the other set. An aggregate or total redundancy sums the redundancies of individual canonical functions. Figure 7.3 shows both the individual and aggregate redundancies for twinned (Figure 7.3 a) and twin merged ((Figure 7.3 b) microstructures.

![Figure 7.3 Canonical Variate Redundancy plots showing Y given X and X given Y for (a) twinned microstructure and (b) twin merged microstructure.](image)

In both 3(a) and 3(b), Y given X shows greater redundancies than X given Y. This implies that given the microstructural parameters, the volume change can be predicted with a higher confidence than the other way around. In the twinned microstructure, input parameters can capture around 17% of the variance in volume change while in the twin merged microstructure it increases to 42%.
Table 2. Lists the direct pairwise correlations between all pairs of input and output parameters for both (a) twin merged and (b) twinned microstructures.

Figure 7.4 shows the canonical variate plots with the correlations ($R_c$) and variance explained ($R_c^2$) by each CV. It is seen that the first canonical variate (CV1) of twinned microstructure (4a) captures 29% of the total variance in the data followed by CV2 (4b) which captures 12% of the residual variance. The equivalent R-squared number in the twin merged microstructure (4c) is much higher- 46% for CV1. According to the Bartlett’s Chi-Squared Test which denotes statistical significance of the canonical variates, it is found that both canonical variates are important for the case of twinned microstructures but only the first one is statistically significant for the twin merged microstructure.
Figure 7.4. Canonical Variate Plots between each canonical X and canonical Y which are the synthetic input and output variables. There are two canonical variates needed to fully explain the variance in the data referred to as CV1 and CV2 for each twinned microstructure (a) and twin merged microstructure (b).

The structural correlations of input variables with respect to output variables in the form of circular helio plots are shown in Figure 7.5. In Figure 7.5 structural correlations are plotted in the form of radial bars that are demarcated by a vertical line. The bars on the left denote the input set and the bars on the right the output set. The radially outward bars represent positive correlation and radially inwards negative correlation. From the first canonical variate plot (CV1) of the twinned microstructure (5a) it is seen that RAD1, F-<F>, G and Omega3 have strong positive correlation with each other and with DelV. Meanwhile, C1*Area show a strong negative correlation with DelV. For CV1 of the twin merged microstructure (5c), it is seen that both
DelV23 and DelV have a stronger correlation with input parameters like Rad1, F-<F>, C1*area and G.

In CV2 of the twinned microstructure (5b), S1 and misor have a medium negative impact in canonical X and are positively correlated to DelV23 and DelV in canonical Y while CV2 of the twin merged microstructure (5d) will not be considered since it captures a very low variance in the data.

Figure 7.5. Structural Correlations plots for the two canonical variates show interrelationships between input and output (left and right side of the vertical line) for the merged twin microstructure. The radial bars pointing in the same direction indicate...
Next, we look at Communality coefficients which determine the fraction of total variance explained by each of the input variables over all the canonical functions (Figure 7.6). The most dominant variables in the twin microstructure (6a) are Omega3, Misor, C1*area and Rad1 and for the twin merged microstructure (6b) are C1*area, F-<F>, Rad1 and G. It is seen that while Omega3 is the most important parameter in twinned microstructure, it features quite low in the twinned microstructure. C1*area and Rad1 are the two common important input variables.

![Figure 7.6](image)

**Figure 7.6.** Communality coefficients of the input parameters in decreasing order. They indicate the overall importance of a variable across all canonical functions. (a) represents twinned microstructure and (b) twin merged microstructure.

### 7.5 Discussion

Results of pairwise correlations from Section 7.4.1 show that several input parameters show much stronger correlations to the grain volume change terms in the output. Aggregate redundancy coefficients from Figure 7.3 also show this trend. The microstructural characteristics of the merged twin dataset show better capability in predicting grain growth as compared to twin
dataset. The improvement in aggregate redundancy due to merging of the twins is almost 150%.
This is also seen in Figure 7.4 as the canonical variates plots have a much higher percentage of
variance explained in twin merged microstructures. This supports the hypothesis 4 that when the
twinned microstructure is modified by merging the twins, the resulting microstructure shows
better correlation of grain growth with different microstructural features.
The input parameter Omega3 stands out for the case of twinned dataset. Omega3 shows a
correlation of 0.394 with volume change for the twinned microstructure and only 0.025 for the
merged twin counterpart. Communality coefficients from Figure 7.6 highlights this discrepancy
as Omega3 shows the highest communality for the twinned dataset but not nearly as significant
for the merged twin one. Omega3 is the moment invariant descriptor of grain shape. A perfect
sphere has the highest Omega3 value. Omega3 shows a strong positive correlation with grain
radius in the twinned microstructure as evident from the pairwise correlation in Figure 7.2 and
helio plot for CV in Figure 7.5(a). This implies that large grains are likely to be more regular
shaped and thereby have a high value of Omega3. In contrast irregular shaped grains (lower
Omega3) are smaller in size and more likely to shrink [170]. Consequently, it’s not surprising
that Omega3 shows a positive correlation to change in volume. In twin merged microstructure,
this correlation between Omega3 and grain volume change is not significant. Due to merging of
all grains that share a twin boundary, grain shape irregularity is largely diminished causing all
merged grains to be more polygonal. Another descriptor of shape is the surface area to volume
ratio (SAV). SAV is lowest for a perfect sphere, and hence shows a negative correlation with
Omega3.
C1*Area is the most important input parameter in the twin merged microstructure as seen from
Figure 7.6. This may seem counterintuitive because the individual components C1 and Area (or
radius) are not as strong. C1 or the face-averaged curvature is calculated as the triangle area weighted sum of all triangle curvatures in the grain normalized by the total area of the grain. C1*Area thereby refers to just the weighted sum of all triangle curvatures. This factor can predict grain volume change much better than other factors established in this study like local grain neighborhood (F-<F>) and even normalized integral grain curvature (G). This dependence can be further explored across multiple anneal states and other material systems.

F-<F> (difference between the number of neighbors a grain has, F and the average number of neighbors of the neighboring grains have <F>) is the second most important input parameter for predicting grain growth in the twin merged microstructure but not very important in the twinned microstructure. This result is not surprising and was already established in Chapter 3. This is also consistent with Hypothesis 1 that the volume change of a grain during annealing depends more on the local neighborhood of the grain that its own morphology, for the merged twin microstructure.

Normalized integral mean grain curvature G shows better predictive power than Face averaged grain curvature C1 in both twinned and twin merged microstructure as seen in Figure 7.2 and Figure 7.6. This result has also been established in Chapter 4. This is consistent with Hypothesis 2.

Another interesting point to note is that G is inversely proportional to C1. This happens due to the inherent definition of these two curvature quantities. While C1 is the area weighted grain curvature, G is the summation of the product of turning angles at adjoining triangles and triangle edge lengths at grain faces. As seen in Figure 7.2 grain radius and C1 are inversely proportional (because curvature ~ 1/radius), but G is directly proportional because larger grains have more triangles that contribute to the sum that determines G.
S1 is a Boolean array that stores either 1 (surface grains) or 0 (bulk grains). A decrease in the value of S1 (or the presence of more bulk grains) show a decrease in grain volume change. However, S1 does not feature among the most important factors in both twinned and merged microstructures. It does feature in the helio plots corresponding to CV2 of the twinned microstructures, where it shows a mild positive correlation to grain volume change. This is consistent with the observation in Chapter 3 that the average surface grain radius is larger than the average bulk grain radius. Lower constraints at the surface of the grains allow for more growth of these surface grains. But, it should be noted that CV2 of the twinned dataset only explains 12% of residual variance. A separate analysis was conducted for surface grains as well as bulk grains. Here bulk grains are defined as grains at the top and bottom surface since they are not true surface grains and excludes only curved surface grains where were around 200 grains. When the CCA analysis was repeated using only the bulk grains in the twinned microstructure, the results did not differ in any significant way.

7.6 Conclusion

The following conclusions can be made from this analysis:

1. Grain characteristics of the twin merged microstructure are better predictors of grain volume changes and the grain characteristics of the twinned microstructure.

2. Grain growth is best predicted by normalized integral mean grain curvature (G), face averaged curvature (C1*area), grain neighborhood metric (F-<F>) and grain radius (Rad1). This is applicable only for the twin merged microstructure.

3. For the twinned microstructure, change in volume is not predicted strongly by any of the aforementioned input variables. It does however show a strong dependence on grain shape (omega3).
4. Normalized integral mean grain curvature $G$ shows better predictive power than Face averaged grain curvature $C_1$ in both the twinned and twin merged microstructure.
8 Hypothesis Revisited

1. A grain in a polycrystal does not exist in isolation. The hypothesis concerning the change in volume with $F < F_{NN}$ was largely supported (Hypothesis 1). $F < F_{NN}$ can predict grain growth correctly 65% of the times. Additionally, this correlation was not found in the twinned microstructure but in the twin merged microstructure. This correlation shows that a grain’s local neighborhood is instrumental in determining the changes in the said grain’s size.

2. Next, the hypothesis on curvature and its effect on grain volume change (Hypothesis 2). Normalized integral mean grain curvature ($G$) derived from mean width is seen to have a poor correlation to grain volume change in twinned microstructures ($r = 0.03$) but has a stronger correlation ($r = 0.4$) for the twin merged dataset. The correlation of grain volume change is slightly weaker with integral mean grain curvature derived from triangle curvatures (NormCA) with a correlation coefficient of -0.34 for the twinned microstructure ($r=-0.38$). The correlation coefficients for both $G$ and NormCA with respect to grain volume change for the twinned microstructure are too small to be statistically significant. This result supports the hypothesis only for the twin merged microstructure.

3. Grain Boundary Velocity varies with misorientation and grain boundary plane orientation. This part of the hypothesis 3 is supported. However, grain boundary velocity shows no correlation to grain boundary curvature except for a certain subsection consisting of $\Sigma 3$ twins and $\Sigma 7$ boundaries. Thus these results are not consistent with the other part of the hypothesis.
4. Canonical Correlation Analysis was applied to the mesoscale data to understand important features affecting grain growth. The variance in grain volume change captured by input parameters in twin merged microstructures is much greater than the real microstructure. Also, factors like grain radius, F-\langle F\rangle and normalized integral mean curvature in twin merged microstructures have a significant contribution in explaining grain growth. The same results were also observed upon direct correlation analysis in Chapter 3 and Chapter 4. These results are consistent with the hypothesis 4.
9 Conclusions

Three-dimensional characterization of high purity fully recrystallized nickel was conducted using a synchrotron based non-destructive technique called nf-HEDM in an interrupted annealing experiment. Grain orientation, location, size, shape was measured for each of the six consecutive anneal states. This highly twinned microstructure was also studied after merging twin related domains to reduce anisotropy and enable understanding of grain growth. The overarching theme of this work was to investigate and understand the phenomenon of grain growth and as a corollary grain boundary motion. Following this, several grain growth theories (2.4.1) and models were investigated with respect to experimental data.

The big picture here is that the conventional theory for grain growth does not predict grain volume change on a case by case basis and that is shown from a variety of points of view. A voxel based analysis of the initial and final anneal states of experimental microstructure showed that grain growth is not predicted by factors like grain radius, number of neighbors and local neighborhood of grains (Chapter 3). It is known that the integral mean grain curvature has a vital role to play in driving grain growth. Thus, we meshed and smoothed boundaries of grains and measured grain curvature from two different methods (Section 4.1.2). Even this metric was unsuccessful in predicting grain volume change (Chapter 4). The morphological and topological characteristics were also used as input parameters in an advanced statistical analysis model (Canonical Correlation Analysis) to predict grain growth and even that was unable to predict grain evolution (Chapter 7). Only when the twin boundaries in the microstructure are eliminated and the grains are made more regular in shape, then it does a better job at predicting grain growth. But the merging takes away orientation dependence and makes everything more uniform. The most important factors influencing grain growth in the twin merged microstructure
were grain curvature (C1*Area), F-<F>, grain radius as well as normalized integral mean grain curvature which agrees with conventional grain growth theories.

At the grain boundary level, curvature is known to drive grain boundary motion as seen in 2D studies and simulations. Velocity and curvature are supposed to be positively linearly proportional to each other [6]. The biggest find is that in 3D microstructural data of polycrystalline nickel Grain boundary velocity did not show a linear positive correlation with grain boundary curvature (Chapter 5). We also see that GB velocities have a strong crystallographic dependence (5.4.5). We also used a model that implemented interface curvature to drive grain growth to predict the twin merged microstructural evolution of nickel. Upon comparing the model prediction with the experiment, we get results that show its different by curvature and doesn’t predict what happens in the microstructure. They don’t match the reality and the reality doesn’t follow the proportionality with curvature but the model does.

Ultimately, conventional theory is bad for predicting whether a grain will grow or shrink. This may be because grain growth and GBM is a more complicated process in a polycrystal. Grains do not exist in isolation and neither do grain boundaries. Also, a grain boundary can be thought of as being made of several grain boundary plane orientations, each of which would possess different propensities to the ability to move owing to velocity’s dependence on crystallography. In addition, a grain boundary is connected to other boundaries that may or may not be mobile and thus affecting the motion of the original boundary. One must also consider the triple junctions and surface tension equilibria and GB stiffness [171] [140]. It is also noted that each of the velocity factors like GB energy, GB mobility and GB curvature also share a strong correlation with GB crystallography. Thus, GBM is affected by many factors that need to be
considered to be able to predict interfacial evolution perfectly in single phase polycrystalline materials.
10 Future Work

10.1 Effect of triple junctions on grain growth

The effect of triple junctions on the evolution of interfacial networks in a polycrystalline material is well documented [142][172].

However, in this study it was not possible to study this effect due to the smoothing algorithms that were used for smoothing the grains. The dream3d smoothing procedures can’t smooth the triple lines accurately because of which they were mostly left out from any analysis.

It is possible to smooth and mesh these grains with the help of another smoothing techniques [173] to obtain better and more realistic three dimensional triple lines and quadruple junctions. Subsequent work in this project will focus on understanding the motion of triple junctions and their effect on grain boundary motion. According to Neumann-Mullins equation, triple junctions do not affect grain boundary motion. It has also been found that triple junctions are temperature sensitive and beyond a certain temperature, they affect grain boundary network development [174]. Recently it has been shown in simulations that for sufficiently small grains triple junctions drag controls grain growth rather than curvature [175]. These results will be evaluated with respect to the nickel dataset for both twinned and twin merged microstructures.

10.2 Comparison of anisotropic simulation with experiment

Application of anisotropic grain growth simulation to predict grain growth using the twinned microstructure as the initial state. This will be then compared with experimental final state to understand and measure the degree of simulation prediction. Grain volume changes will be
measured and their correlations will be evaluated. Topological changes will be compared as well as grain crystallography shall be investigated in the simulation and compared with experiment. Currently, an anisotropic model is being developed by our collaborators – Kaushik Dayal, Kiana and Xiaoyao from the Civil Engineering Dept in CMU. Figure 10.1 shows a preliminary anisotropic model compared with the original twin grain (a), the isotropic model grain (b) and the anisotropic model grain (c).

![Fig 10.1](image-url)  
*Figure 10.1. The figure shows an experimental twin grain on the left (An4) and the isotropic (middle) and anisotropic (right) evolution of the grain using simulation.*

10.3 CCA on grain boundary level to understand grain boundary movement

Apply CCA on grain boundary data to understand parameter sensitivity and interrelationships between grain boundary characteristics and correlate it with grain boundary velocity. Also include grain boundary crystallography such as misorientation and grain boundary plan orientation to classify the relationship between GB velocity and GB curvature for different boundary types.
10.4 Application of Neural Networks and other ML models

Neural Networks (NN) or Artificial Neural Networks (ANN) are computational models in the domain of machine learning that have revolutionized everyday life. First conceived in the 1940s by two mathematicians Warren McCulloch and Walter Pitts, these groundbreaking models have taken us to the next level in Artificial Intelligence (AI). They essentially mimic neurons in the human brain to be able to decision-make in the face of large and complex data. They are being used in every real-world application today be it computer science for speech and handwriting detection, aerospace, automotive, manufacturing, robotics, medical, banking and defense.

ANNs can also be called non-linear statistical datamining tools used for detecting patterns and trends. Just like in a human cell, neurons which have nucleus, dendrites, axon and synapses to transmit electrochemical signals, ANNs also consist of multiple layers- input layer, hidden layer(s) and output layer to transmit weighted inputs and self-learn [176].

ANNs can use this structure to train large numbers of data and perform critical tasks such as:

- Classification – Based on Supervised learning the neural networks can organize datasets into predefined classes
- Prediction- They can give an output based on a set of inputs
- Clustering- Based on Unsupervised learning, they can identify features in a data and classify them without prior knowledge
- Associating- Neural networks can be trained to “remember” patterns which can be reverted to when analyzing unfamiliar version of the pattern.

Thus, neural networks can be used to find patterns in complex data or even predict outputs. In this context, it can be applied for multiscale modeling in the nickel dataset in the following ways:
• Grain Growth Prediction: Use grain information like grain size, orientation, neighbors, volume, curvature and mean width as input and change in volume as output. Since we have six anneal states, the model can be trained on a few and then tested on the rest.

• Grain Boundary Movement: Use grain boundary crystallography, curvature and other metric to understand GB velocity.
11 Appendix

Figure S1. The reconstructed volume in all six anneal states. (a) 0, (b) 1, (c) 2, (d) 3, (e) 4, (f) 6. The cylinder is 1 mm in diameter and a roughly equivalent position is marked by a white “x” in each anneal state. Note that while the fields of view overlap, each the volumes are not identical.
Figure S2. Distributions of (a) x component, (b) y component, and (c) z component of the centroid separations between matched grain pairs. The translation vector used to align the different anneal states was the average of each distribution.

Algorithm GrainTrack

input grain statistics_{Anneal_i}, grain statistics_{Anneal_j}, C_1, C_2
for each grain ID_k ∈ Anneal_i
  for each grain ID_l ∈ Anneal_j
    if distance(centroid_k, centroid_l) < 5*radius_k
      ΔG ← disorientation(orientation_k, orientation_l)
      ΔL ← distance(centroid_k, centroid_l)
      CI_{kl} ← C_1(ΔG) + C_2(ΔL)
    else
      CI_{kl} ← 999 (implies field of view mismatch)
  end for
end for
min ← 1000
for each grain ID_k ∈ Anneal_i
  for each grain ID_l ∈ Anneal_j
    if CI_{kl} < min
      min ← CI_{kl}
      store index min that gives minimum CI
  end for
match grain ID_k (Anneal_i) to grain ID_{min_j}(Anneal_j)
end for

Figure S3. Algorithm used to match grains in different anneal states.
Figure S4. Reconstructed microstructure in anneal state 0 (a) before twins are merged and (c) after twins are merged. (b) a cluster of twin related grains in the initial structure that gets merged to a single grain (d) in the final structure.
**Figure S5.** Normalized distribution of grain sizes in the six anneal states (a) before twins are merged and (b) after twins are merged. Because there are fewer grains after the twins are merger, there is more scatter.
**Figure S6.** Distribution of grain neighbors in the six anneal states (a) before twins are merged and (b) after twins are merged. Because there are fewer grains after the twins are merger, there is more scatter.

**Figure S7.** The correlation between grain radius and the number of neighbors in the initial anneal state for the twinned microstructure (squares) and the microstructure after merging twins (circles). For each grain size category, the marker is the mean value and the bar shows one standard deviation of the distribution.
Figure S8. Change in volume versus the difference between the number of neighbors and the average number of neighbors of the neighbors after removing grains whose centers of mass are less than $<R>$ from the boundary of the volume in (a) the twinned microstructure and (b) after merging twins.

Figure S9. Change in volume versus the difference between the grain radius and the average of the radii of the neighbors in (a) the twinned microstructure and (b) after merging twins. For
each class of $R^{-<R_{NN}>}$, the marker is the mean value and the bar shows one standard deviation of the distribution.

**Figure S10.** Disorientation distributions in (a) the initial anneal state and (b) the final anneal state.

**Figure S11.** Integral mean curvature of grain faces plotted against number of grain faces ($F$) in the twinned Anneal 1 state for the entire range captured.
Figure S12. Visualization of a grain matched in all the anneal states. Location of the grain shows that the fields of view do not match.
12 References

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