Investigating the mechanisms of grain boundary migration during recrystallization using molecular dynamics

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Abstract. Resolving the mechanisms associated with grain boundary migration is a difficult task. This work is focused on detailing techniques to utilize atomistic simulations to better understand the energy, structure and mobility of grain boundaries (GBs). Various techniques are detailed on how to construct different simulation cells, select boundary plane normals, and measure simulated mobility. A limited number of GBs, inspired by experimental observations, are selected for the present work. The GBs studied indicate that the structure of the theoretical set of GBs has a structure consistent with an overall minimization of energy. The mobility simulations indicate that shear coupling is the preferred migration mechanism. However, when the same GBs are constrained, as might occur in polycrystalline networks, the relative mobilities of the set of GBs are significantly altered. Finally, the influence of deformation structures, as they pertain to GB migration during recrystallization, are discussed.

1. Introduction
Thermomechanical processes are essential in the production and manufacturing of a large portion of the world’s materials. These thermomechanical treatments subject the microstructure within the material to deformation, recovery, recrystallization, or grain growth [1]. Each of these steps is critical to achieving a material with a given defect density, texture, grain size and structure, which in turn play an important role in defining a material’s strength, ductility, and property anisotropy. For example, defect density correlates with strength and ductility, grain size affects both creep resistance and strength (as in the Hall-Petch relationship [2,3]) and texture is the basis for property anisotropy.

The three stages in process annealing are: (1) recovery, which is marked by the annihilation of and rearrangement of dislocations; (2) recrystallization, which marks the nucleation of new defect-free crystals that grow to consume a previously deformed material; and (3) grain growth, which involves the subsequent growth of these grains and overall reduction of energy by the reduction of grain boundary area. The latter two processes both involve the migration of GBs, although the driving force for migration is different for the two processes. In addition, recent work has demonstrated that recovery can even involve the migration of grain boundary triple junctions [4]. As a result, understanding the mechanisms and influences of boundary migration is essential to fully resolving the different annealing processes.

As noted above, GB migration during the different stages of annealing is influenced by various factors. Complete decoupling of these factors, such as the inherent mobility of a GB from the driving force facilitating its motion, the defect structure being consumed, and the GB network constraining that motion, is at present an impractical procedure. One can, however, gain insight into the relative
importance of the various factors affecting migration by examining the factors individually and comparing their effect with those directly observed.

GB migration during recrystallization, in particular, is primarily influenced by the stored strain energy due to prior strain hardening [1]. While this stored energy provides the primary driving force for migration, recent work has shown that the alignment of dislocation boundaries along particular crystal planes can lead to faster grain boundary migration [5]. This leads to the question of whether the structure and orientation of dislocations that build up during deformation play a more important role on the migration of a given GB than its inherent mobility. This is especially pertinent since it is known that deformation prior to recrystallization can have a significant influence on the resulting texture following recrystallization [1]. Finally, it is known that GB plane normal plays an important role in defining the properties and structure of different GBs, even when the GBs have the same misorientation.

The general aim of this research is to detail methods that could be used to examine the various factors influencing the migration of recrystallizing GBs in deformed aluminum. Recrystallization GB migration in aluminum is selected because of the availability of data from nondestructive techniques such as 3D X-ray Diffraction (3DXRD) [6,7] and 3D X-ray crystal microscopy (3DXRM) [8]. These synchrotron techniques provide a full 3-D voxelized map of the microstructure. The orientation is recorded for each 3D voxel, with a spatial resolution in the micron to submicron scale. By using X-ray techniques with the highest spatial resolution and comparing the orientations in these voxels, one can even characterize the local deformation microstructure.

These works often examine GBs that are nominally of a single misorientation with the surrounding deformed structure. In the experiments of interest, the recrystallized sample is examined at various times during the recrystallization. In the region examined in these snapshots, a recrystallizing GB is advancing to consume the deformed aluminum. The GBs exhibit interesting migration patterns: stop and go motion, regions that are fairly planar that migrate slowly, curved regions that migrate at higher rates, etc. [6,7]. The planar and curved regions show a particularly strong dependence on GB plane normal.

Unfortunately, the 3DXRD and 3DXRM methods do not provide mobility measurements and cannot resolve the nature of the boundary migration or the reasons that some portions of the GB remain stagnant while other portions migrate relatively quickly. One could analyze individual boundaries and their migration by more standard experimental techniques [9-14], though we seek instead to simulate these behaviors at an atomic level.

The present work utilizes atomistic simulations to investigate the structure, energy and mobility of various GB plane orientations in a high-angle GB. The nature of the study utilizes methods similar to a previous survey of 388 GBs [15-19], but the focus is on creating GBs to match conditions in experiments rather than the construction of random GBs. While the conditions cannot be identical to those in experiments, the approach does allow one to study various influencing factors and gain insight into their relative importance as they might influence experimental observations.

2. Methodology

2.1. Bicrystallography to match experimental and atomistic GBs
The GB of interest would separate a recrystallized grain from a deformed microstructure, and therefore have a range of misorientations. As a result, one must either choose to study a range of misorientations and boundary plane orientations or one can study the average misorientation, as we choose to do here. We select a high-angle GB about an arbitrary axis, not one of the commonly studied 100, 110 or 111 axes. We identify the coincidence site lattice (CSL) Σ45c GB, which is given by 53.1301° rotation about the <221> axis. The Σ45c CSL GB can be studied for various boundary plane orientations to gain an understanding of how different regions of a GB, curved or flat, might respond during migration. To eventually provide optimal comparison between experiments and the simulated
atomistic GBs, the crystallography obtained from experimental work must be examined, however here we have chosen an example GB which are inspired by the experimental observations.

The boundary plane orientations for the various GBs studied in this work are selected from a hypothetical distribution of plane normals, as defined in the crystal frame of the recrystallized grain. These normals are separated into two populations: one population of normals will have a diffuse distribution over a large range of normals, corresponding to a ‘curved’ region of a GB, and the second population will be a sharp distribution over a small range of normals, corresponding to a ‘flat’ region of the same GB. These two distributions of normals are presented in Figures 1a and 1b. The two populations overlap slightly but the curved GBs are obviously more diffuse. The normal for the ‘flat’ portion is identified as \([0.05 \ 0.55 \ 0.83]\). Since the ‘curved’ distribution of normals is diffuse, we select two normals in the region, one with a normal equal to \([-0.39 \ 0.83 \ 0.41]\), and the other to be selected for reasons described later. A total of 3 GBs will be studied, one representing the ‘flat’ GB and two representing the ‘curved’ portions of the same GB.

To obtain the atomic structures for these GBs, an adaptation of the methodology used by Olmsted et al. is used [15]. These techniques employ CSL lattice theory to determine boundary structures so that periodicity in the boundary plane can be employed to avoid the effects of free surfaces. In the Olmsted methodology, one seeks CSL bicrystals that fit within a given set of orthogonal boundary plane dimensions, thereby limiting what boundary plane normals can be studied. Since we are instead interested in creating bicrystals for specific normals, we adapt Olmsted’s methodology in the following way. First, we define the CSL lattice for the misorientation using Grimmer’s method [20]. This CSL lattice is defined in the crystal frame of one of the two bicrystals. A small portion of the CSL lattice for the \(\Sigma 45c\) misorientation is plotted in Figure 2a, where each CSL lattice site is given by an ‘x’. Using a set of CSL lattice points, one can define a set of vectors that begin at the origin and end at each of the defined lattice points, as shown in Figure 2a. Vectors that have the same direction as other vectors are discarded, and for efficiency, the smallest magnitude vector for each direction is retained. This results in a set of vectors which all have unique directions.

Given this set of vectors with unique direction, one can begin to define planes by the combination of any two vectors. Since these two vectors correspond to CSL lattice points, the boundary plane they define will capture the natural periodicity of both crystals (one on each side of the GB), thus ensuring that periodicity for the GB is maintained so free surface effects can be avoided. These two vectors become the basis vectors, \(\vec{a}_1\) and \(\vec{a}_2\), and form the boundary repeat unit which is typically a parallelogram as shown in Figure 2b, since most combinations of \(\vec{a}_1\) and \(\vec{a}_2\) are not orthogonal. The normal, \(\hat{n}\), for each boundary plane is defined by the normalized cross product of \(\vec{a}_1\) and \(\vec{a}_2\). While
the boundary plane repeat unit may not be orthogonal, this method has the advantage of producing significantly smaller GBs than if the constraint of orthogonality was enforced as in the work of Olmsted et al. [15].

In order to match the boundary plane normal to those desired (cf. Fig 1), as we seek to do here, we must find a boundary plane that is sufficiently close to the normal of interest. Using the CSL points and their corresponding vectors, one can obtain a family of possible normals from the set of selected CSL lattice points. For example, using a set of unique vectors defined by $\pm 3$ CSL lattice points away from the origin, one can obtain all boundary plane normals defined in Figure 2c. Since there may be multiple combinations of vectors that define the same plane, the combination that defines the smallest repeat unit area is kept. It is noted that in Figure 2c, certain regions have a high density of boundary plane normals and other regions have a low density of boundary plane normals.

One can select the boundary plane which is nearest to the desired normal, or if there is no boundary plane sufficiently close, one can use larger blocks of CSL lattice points ($\pm 4$ or more CSL lattice points away from the origin) to improve the overall density of boundary planes near a desired normal. Finally, it is noted that for the $\Sigma 45c$ misorientation, a full hemisphere of unique boundary plane normals exists, or in other words, the $\Sigma 45c$ boundary plane orientation fundamental zone is a full hemisphere [21].

In the present work, once the planar boundary repeat unit is determined, a third vector, $\vec{a}_3$, is identified to determine the periodicity of the CSL lattice in a direction out of the boundary plane. In the present work, the selection of this third vector is another CSL lattice point based on a combination of orthogonality to the boundary plane and magnitude of the vector. This results in a periodic triclinic simulation cell [22], which can be easily accommodated by software such as LAMMPS. The balance between the two factors is critical for efficient computations, since orthogonality maximizes parallel computing efficiency and a small magnitude minimizes the number of atoms contained in each CSL unit.

If one simulates a fully periodic bicrystal (containing 2 GBs), the basis vectors for the simulation cell must follow all three vectors identified above to maintain periodicity. On the other hand, it was recently determined that if one simulates a bicrystal with only 1 GB, this constraint of using a non-orthogonal $\vec{a}_1$ is not necessary. Since periodicity will not be enforced in this dimension, this does
enable the possibility of defining a basis vector that is orthogonal to the boundary plane, resulting in a monoclinic simulation cell that can have improved computational efficiency. The magnitude of this orthogonal basis vector would simply be the vertical distance of the next CSL lattice point that is still contained within the dimensions of the two basis vectors defining the boundary plane. Despite the possibility to define the third basis vector of the simulation cell orthogonal to the first two, the present simulation utilizes the triclinic non-orthogonal basis vectors that lie at CSL lattice points.

With the three basis vectors defined, one can evaluate the microscopic degrees of freedom that enable a particular GB to achieve its minimum energy structure. These microscopic degrees of freedom are identified as: three degrees of freedom defining the relative shift vector between the two crystals, one degree of freedom defining the placement of the actual GB plane relative the boundary plane defined by the basis vectors, one degree of freedom for the allowed proximity between neighboring atoms, and one degree of freedom determining which overlapping atoms should be discarded [15]. These microscopic degrees of freedom are essential to defining a GB because they affect how the atoms line up and coordinate at the interface of the two crystals. The shift vector adjusts the position of the two crystals relative to each other, the GB plane placement determines which two planes of the each of the crystals will line up next to each other, proximity cutoff determines how close atoms can get to one another before they must be removed, and the selection of which atoms to be deleted determines where a given atom may be placed.

The present work uses the methods established by Olmsted et al. to identify the unique configurations and combinations of these variables [15]. Unique combinations of the relative shift vector between the two crystals are determined from the displacement shift complete (DSC) lattice. Since these represent continuous degrees of freedom, which would be difficult to fully evaluate, fractions of the three basis vectors of the DSC lattice are typically considered. In the present work, each vector is divided into two increments for a total of 8 relative shift vectors. The placement of the GB plane must be somewhere between the original boundary plane and the minimum distance, \(d\), to the next CSL lattice point contained within the simulation cell (illustrated in Figure 2c), since this CSL lattice point reflects the next periodic image of the same atomic configuration. Since the lattice spacing of atoms in the two crystals is discrete, one can identify the exact placement of this plane that will result in unique configurations. Finally, following Olmsted, we consider three cases of discarding overlapping atoms that are within the proximity cutoff: deleting overlapping atoms from the first crystal, deleting overlapping atoms from the second crystal, and removing overlapping atoms from both crystals and replacing the both with an atom at a location halfway between the two removed atoms. The proximity cutoff is evaluated from 1/3 to 85% of the nearest neighbor distance evaluated in 0.1 Å steps. These combinations can result in 100s to 10,000s of configurations that may give the minimum energy configuration.

The construction of the full simulation cells also follows the method of Olmsted et al. [15]. The bicrystals are periodic in the two dimensions within the GB plane and non-periodic in the direction normal to the GB, resulting in a single GB within the simulation cell. The size of the simulation cell is set to a minimum of \(17a_0/2\), where \(a_0\) is the lattice parameter, or at least two repeats of the CSL lattice for both directions in the plane of the GB, whichever is larger. In the direction normal to the plane, each grain on either side of the GB is set to a minimum of \(40a_0/2\) or at least two repeats of the CSL lattice, which ever is larger. An additional CSL unit cell is added in between the two grains for the purposes of the GB construction and additional CSL unit cells are added to each end of the bicrystal, which are constrained to move as a block to ensure that free surface effects do not influence the GB energy calculation during minimization. The aluminum embedded-atom method (EAM) interatomic potential developed by Mishin et al. is utilized for atomistic simulations analyzed here [23].

The minimum energy structure for a given GB is obtained by energy minimization of each of the possible configurations over the microscopic degrees of freedom. In the present work, the conjugate gradient minimization technique in LAMMPS is used. We use tolerance cutoff values of \(10^{-10}\) eV/Å for the force and a relative error of \(10^{-4}\) % for the energy. These values balance achieving
minimization convergence of sufficient accuracy with the time required to achieve convergence since the evaluation of up to 10,000s of configurations can be time consuming.

All these steps can be automated for the creation of GBs to match those desired for different experiments. Access to high performance computing resources makes these calculations tractable. In an effort to make these approaches accessible to the research community, another group recently published a similar algorithm to this present work. While the algorithms are not the same, their algorithm is an open source Python utility called GBpy, which performs many of the same functions as those discussed here to determine the minimum energy structure for a GB of interest [24].

2.2. Multi-normal GBs.

One minor drawback to studying GBs solely in bicrystal form is that one can miss the effects of other boundary normals or network constraints. We do not study the effects of network constraints here because the experiments of interest frequently only examine a single GB [6,7]. However, the GBs are typically curved and therefore have a range of normals as illustrated in Figure 1. To better understand how neighboring regions of a different normal affect the overall GB motion, we also create a multi-normal GB. This is similar to a faceted GB, but the facets are large compared to the atomic level facets that may exist in a GB of a single normal. The GBs here will be composed of multiple ‘flat’ segments but will be connected to other ‘flat’ segments with different boundary plane orientations. The periodic boundary conditions utilized to create the GBs requires that all facets also obey the periodic boundary conditions. Thus, along the two periodic dimensions of the plane, \( \vec{a}_1 \) and \( \vec{a}_2 \), an additional facet must either be non-existent or more than one facet must exist to allow the boundary to return to its original position before it repeats. Thus facets that will be repeated across a periodic boundary can only exist for a circle of directions, all of which must be normal to either \( \vec{a}_1 \) and \( \vec{a}_2 \), an example of which is shown in Figure 3a for circle of normal about the \( \vec{a}_1 \) axis. By selecting facets of these normals, one can see in Figure 3b how a boundary of multiple facets can return a GB to the original normal before repeating along the \( \vec{a}_2 \) direction. These facets are continuous along the \( \vec{a}_2 \) direction, thereby preserving the periodicity of the simulation cell. In this manner, the influence of multiple GB normals on each other can be studied.

2.3. Mobility.

The mobility of the GBs is examined using the synthetic driving force method [25]. In short, the method utilizes an order parameter to distinguish between the atoms in two grains of differing orientation. Additional potential energy is added to atoms of one grain selected to be the unfavored orientation, while atoms in the other grain have no added energy, making it the favored orientation.
The gradient in energy across the GB, leads to a driving force, \( f \), inducing GB migration and growth of the grain with the favored orientation. Using the magnitude of the applied synthetic driving force, \( f \), and the measured velocity of the GB migration, \( v \), one can calculate the mobility of the GB, \( M \), according to \( v = M \cdot f \) [9].

While the synthetic driving force does not have a physical equivalent method to add energy to just one orientation of crystals, GB mobility measurements by this method have been demonstrated to be independent of the type of driving force [26]. Furthermore, this method has been compared to other boundary migration techniques and found to provide nominally identical results [27]. More recently, an improved synthetic driving force method was introduced, called the energy conserving orientational driving force [28]. While the newer method does overcome minor issues of the original method, the two give nominally similar results, indicating that the results contained herein are valid [28].

Similar to Olmsted et al. [15], the GBs studied in this work are tested under three driving forces in the set of (0.005, 0.010, 0.025) eV/atom. To mimic temperatures near those indicated in the experimental work, the mobility measurements have been carried out at 600 K using an NVT ensemble; additional simulations were conducted for a few boundaries at 800 K for purposes that will be explained below. The simulation cell is equilibrated over 50 picoseconds prior to the application of the synthetic driving force. The GB position is then tracked over a period of time and the velocity is extracted from the slope of the GB position-time data. The end groups of atoms are set to be free in some cases, enabling shear coupling to occur if the GB motion selects that, or constrained in other cases, to prevent shear coupled GB motion in cases it is desirable to be avoided.

3. Results

3.1. Energy and structure of bicrystal GBs

Using the techniques described above, it was determined that the \((5\ 47\ 76)\) crystal plane coincides well with the ‘flat’ boundary plane normal; the angular deviation is 1.6°. For the ‘curved’ boundary plane normal noted above, the \((\bar{1}\ 2\ 1)\) crystal plane was found to be within 1.4° degrees of the desired normal. Finally, a second migrating boundary plane normal, with a \((\bar{5}\ 5\ 113\ 109)\)crystal plane, was selected so it could be tested independently and in conjunction with a \((5\ 47\ 76)\) plane as part of a multi-normal GB. The \((5\ 47\ 76)\) and \((\bar{5}\ 5\ 113\ 109)\) bicrystals each have triclinic simulation cells while the higher symmetry of the \((\bar{1}\ 2\ 1)\) bicrystal basis vectors enables a monoclinic simulation cell.

The \((5\ 47\ 76),\ (\bar{1}\ 2\ 1)\), and \((\bar{5}\ 5\ 113\ 109)\) bicrystals had 32,310, 4,275, and 585 microscopic degrees of freedom, respectively, of which each configuration was minimized. A plot of the GB energy distributions for the configurations is provided in Figure 4a. The minimum GB energy for the \((5\ 47\ 76),\ (\bar{1}\ 2\ 1),\) and \((\bar{5}\ 5\ 113\ 109)\) GBs are 397 mJ/m², 466 mJ/m², and 445 mJ/m², respectively. These are higher than the textbook “high angle” GB energy listed of 324 mJ/m² [1], though the ‘flat’ boundary normal has energy lower than the ‘curved’ boundary normals and thus closer to the textbook value.

In Figure 4a it can be seen that minimum energy structures are far more sensitive to the starting configuration than those shown by Olmsted et al. [15]. The structures for these three GBs are shown in Figure 4b-d, where only those atoms with a centrosymmetry parameter greater than 1 are shown. Thus only those atoms in the GB that are not ideally coordinated are shown and the ideally coordinated surrounding atoms are not included in the image.

For the \((5\ 47\ 76)\) GB shown in Figure 4b, it can be seen that there are very clear facets to the GB. For example, the overall plane normals are \([5\ 47\ 76]\) and \([47\ 5\ 76]\) in the lower and upper grain respectively. The long facet in the GB is close to a \((3\ 8\ 10)\) and a \((8\ 3\ 10)\) plane in the lower and upper crystals respectively, while the short facet is close to a \((\bar{4}\ 5\ 14)\) and \((\bar{5}\ 4\ 14)\) plane in the lower
and upper crystals respectively. Since the GB configuration was tested in so many other configurations (Figure 4b is the low energy configuration), these facets clearly play a role in achieving this low-energy structure.

In contrast, the \((\overline{1} 2 1)\) GB shown in Figure 4c appears to have facets in the upper grain as indicated by the lower side view, but the lower grain only has slight serrations that differ by an atomic plane or so. Furthermore, the upper grain also shows some minor facets in the left side view. Upon 3D inspection, an array of peaks and valleys in the atoms of the upper crystal can be seen, while the lower crystal is more or less flat. This difference in the two grains likely results from the lack of symmetry in GB plane normal, where the lower crystal has normal of \([\overline{1} 2 1]\) and the upper crystal has a normal of \([\overline{1} 2 7]\).

Finally, the \((\overline{5} 47 76)\) GB shown in Figure 4d is relatively flat in the upper grain but the lower grain has some minor peaks and valleys as indicated in the side view, though these are minor. This GB is also asymmetric in that the lower and upper crystals have normals of \([\overline{5} 47 109]\) and \([15 15 165]\), respectively.

3.2. GB Mobility

The three GBs were each subjected to the synthetic driving force at various magnitudes of the applied driving force. Unexpectedly, all three GBs demonstrated shear coupled GB motion [29,30]. The mobility values and shear coupling factors are listed in Table 1. The \((5 47 76)\) GB had a higher mobility than the \((\overline{1} 2 1)\) and \((\overline{5} 47 109)\) GBs by almost an order of magnitude, except at the highest driving force where it is still higher but by a smaller magnitude. It is noted that the mobility of

\begin{figure}
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\includegraphics[width=\textwidth]{figure4}
\caption{Energy and structure for the three GBs examined in this work. (a) Distribution of GB energy for all possible configurations examined to find the minimum energy structure. GB structures plotted using atoms whose centrosymmetry parameter is greater than 1 for the (b) \((5 47 76)\), (c) \((\overline{1} 2 1)\), and (d) \((\overline{5} 47 109)\) boundary plane GBs.}
\end{figure}
these GBs depends on the magnitude of the applied driving force. As demonstrated by Olmsted et al. [27], this phenomenon appears to be associated with the roughening transition temperature. Below the GB roughening transition temperature, boundaries exhibit a larger dependence on the driving force, and above the temperature, boundaries show very little dependence on the magnitude of the driving force [27]. This decrease in dependence with increasing temperature is demonstrated in Figure 5, where it can be seen that the mobility of the (5 47 76) GB shows a smaller dependence on the driving force magnitude at 800 K than it does at 600 K. Since the melting point of Al is 933K, it is likely that this boundary will not roughen before it melts.

As shear coupled GB motion requires lateral motion of the surrounding grains or deformed matrix material, it is possible that this type of motion would be constrained under experimental circumstances. As a result, all three GBs were examined for their mobility under constraints that inhibited shear coupling. To do this, blocks of atoms at the two ends of the bicrystal were constrained to not move. The resulting mobilities are presented in Table 2. In this case it can be seen that all three GBs have similar magnitudes of mobility at the highest driving force but at lower driving forces, the (T̅2 1) GB has the highest mobility.

| Driving Force [eV/atom] | Nominal | Actual [GPa] | Mobility [m/(s GPa)] | Shear coupling factor \( v_\parallel / v_\perp \) |
|-------------------------|---------|--------------|-----------------------|-----------------------------------------------|
|                         | (5 47 76) | (T̅2 1) | (55 113 109) | (5 47 76) | (T̅2 1) | (55 113 109) |
| 0.005                   | 0.044    | 242         | 37         | 21     | 0.31  | 0.28  | 0.33  |
| 0.010                   | 0.089    | 347         | 44         | 22     | 0.31  | 0.24  | 0.26  |
| 0.025                   | 0.223    | 440         | 78         | 117    | 0.31  | 0.33  | 0.26  |

| Driving Force [eV/atom] | Nominal | Actual [GPa] | Shear Constrained Mobility [m/(s GPa)] |
|-------------------------|---------|--------------|------------------------------------------|
|                         | (5 47 76) | (T̅2 1) | (55 113 109) |
| 0.005                   | 0.044    | 34         | 67         | 17     |
| 0.01                    | 0.089    | 45         | 66         | 30     |
| 0.025                   | 0.223    | 87         | 77         | 71     |

**Table 1.** Mobility and shear coupling factor for three Σ45c GBs under different driving forces.

**Table 2.** Mobility for three Σ45c GBs under shear constrained conditions

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**Figure 5.** Driving force (pressure) dependence of mobility for the (5 47 76) GB at two different temperatures.
3.3. Multi-normal bicrystals

To investigate the effect of a GB with multiple normals, as is the case with curved boundaries observed in many experiments, a bicrystal with multiple normals was created. The creation of this bicrystal follows the methodology discussed in section 2.2. The simulation cell for this multi-normal bicrystal starts with the (5 47 76) GB, since it is associated with a ‘flat’ GB configuration. The (5 47 76) simulation cell is enlarged by several times and then the boundary plane is allowed deviate according to normals that follow the outline of section 2.2. For the multi-normal bicrystal shown in Figure 6a, it can be seen that there are three normals present in the simulation cell. The ‘flat’ (5 47 76) crystal plane is allowed to exist on both a lower and higher plane. One additional plane is selected as the (5 47 76) crystal plane because it is contained within the distribution of ‘curved’ normals. To maintain periodicity, an additional normal is required; this additional normal is selected to be the mirror image of (5 47 76) crystal plane and results in a (425 257 853) plane, which is not studied individually in this work. The multi-normal bicrystal is triclinic, has volumetric dimensions of 128 x 566 x 295 Å³ and contains 1.28 million atoms.

Figures 6b, 6c, and 6d illustrate the structure of the multi-normal GB. It can be seen that the (5 47 76) regions contain the structure characteristic for this boundary plane normal, as illustrated in Figure 4b. However, it is difficult to achieve exactly the same structure in the (5 47 76) region as is illustrated in Figure 4d because the lattice shifts between the crystals for the minimum energy structures are not identical between the (5 47 76) and (5 47 76) GBs. As a result, the bicrystal was constructed using the microscopic degrees of freedom that enabled the minimization of the (5 47 76) structure. It can be seen that the (5 47 76) region has a grid of minor facets while the (425 257 853) region has facets that are continuous with the facets of the (5 47 76) structure. The GB energy of this structure is 485 mJ/m², which is higher than three GBs studied individually in this work and much higher than textbook values.

Figure 6. Multi-normal GB created for this work. (a) Entire simulation cell showing the two different crystals. (b) Atomic structure at the GB plotted using the centrosymmetry parameter. (c-d) Two different views of an Ovito surface construction [31] showing the faceting of the (5 47 76) GB and its influence in faceting on the adjacent regions with different normals.
When subjected to the synthetic driving force, the lower (5 47 76) regions grow the quickest of all the different normal regions, as shown in Figure 7. This includes faster growth than the upper (5 47 76) region. It is reasonable to expect that the curvature driving force has an effect here. Though it is noted that these results are consistent with the individual examinations of (5 47 76) and (55 113 109), which would suggest that whether shear coupling is constrained or not, the (5 47 76) GB should move more quickly. It is noted that this GB, even with its multiple normals does demonstrate some magnitude of shear coupling, though the magnitude is difficult to quantify since the GB does not move uniformly.

4. Discussion

4.1. GB Energy and Structure

The three individual GBs and one multi-normal GB indicate very interesting correlations between energy and structure. From these simulations it appears that the lowest energy structures for a given boundary plane are most frequently associated with a faceted structure. This is in line with previous work and theory [32-35] that indicate that the lowest energy structure can often be comprised of two or more crystal planes with differing orientation and energy that give the same net boundary plane but with lower energy. This is very apparent in the case of the (5 47 76) boundary plane, where the facets are the same in both crystals due to the higher symmetry across the GB. In contrast, the (1 2 1) and (55 113 109) boundary planes show more complex faceting, which faceting is different depending upon which crystal is examined. As a result, it is not as surprising that these GBs have higher energy.

It is also possible that other boundary plane could have lower energy than the (5 47 76) crystal plane. For example, since the (5 47 76) plane is dominated by the long facet with a (3 8 10) crystal plane, it is likely that this boundary plane would have even lower energy. Nevertheless, this low energy crystal plane is not in the distribution of ‘curved’ boundary normals shown in Figure 1a. Thus, from the set of GBs examined here, a theoretical structure comprised only of the examined GBs would be consistent with a GB that has been minimized for energy.

4.2. GB Mobility

The mobility values of the GBs examined in this work indicate a number of possibilities for how the actual migration in a recrystallization experiment could occur. While experimental conditions cannot be replicated exactly in atomistic simulations, there is information that can be gained about how different factors may be influencing the actual GB migration in the experiment.

First, it is surprising that of the 3 GBs examined, all exhibit shear coupling when not constrained. In Olmsted’s set of 388 GBs, six Σ45c CSL nickel GBs were examined, of which four exhibited shear coupling [16,30]. In other words, the Σ45c GBs examined here show a high propensity for shear coupling. This is significant because shear coupling has been speculated as a possible mechanism underlying stress-driven grain growth [36]. Whether or not shear coupling actually occurs may be subject to various factors. A curved boundary with a distribution of boundary planes, as is often seen...
in experiments, may or may not enable shear coupling. Though the multi-normal GB examined here did exhibit some magnitude of shear coupling. One complicating factor with this shear-coupling scenario though is that if ‘flat’ GBs typically migrate slowly, this is inconsistent since the (5 47 76) crystal planes have the highest mobility under shear-coupled conditions.

Second, when the three GBs are constrained in a manner to inhibit their shear coupling, the mobility values change significantly. The ‘flat’ (5 47 76) GB that had been the fastest when not shear constrained now becomes slower than the ‘curved’ (12 1) GB at low driving forces. However, the ‘curved’ (5 5113 109) GB has a mobility lower than the ‘flat’ (5 47 76) GB. Thus, one ‘curved’ GB is faster than the ‘flat’ GB while the other ‘curved’ GB is slower. One would then need to resolve the distribution of different boundary plane orientations with the relative mobility of each to determine whether the ‘curved’ GB has higher inherent mobility than the ‘flat’ GB.

Third, in further examination of the shear-constrained motion of the (5 47 76) GB, it can be seen that the (4 5 14) facet is critical to the boundary migration. Even in this shear-constrained state these small facets migrate laterally as the GB migrates in its normal direction, as indicated in Figure 8. While most of the movement is lateral, this does not entirely account for the vertical motion of the GB. The simulation indicates that for every angstrom the short facet moves laterally, the long facet moves half an angstrom vertically. Since the (4 5 14) facet falls in the middle of the distribution of ‘curved’ normals in Figure 1a, its high mobility may be further indicative of high mobility among the ‘curved’ boundary normals.

Fourth, it is noted once again though, that despite the fact that the (5 47 76) GB has a slower mobility than the (12 1) GB or the (4 5 14) facet within the (5 47 76) GB, the (5 47 76) GB’s mobility is non-zero, indicating that under the proper driving force it could move, and in experiments, ‘flat’ portions of the recrystallized GB remain fairly stagnant or migrate very slowly.

Finally, the present work does not take into account the inhomogeneous distribution of stored energy in the deformed microstructure, nor to any possible preference for migration along specific dislocation boundaries [5]. Also, the presence of medium to high angle dislocation boundaries in the microstructure (e.g. [37]) causes sufficient lattice curvature that there is not a single orientation within the deformed material. As a result, the misorientation across the entire experimental GB is not constant. Furthermore, since dislocation structures can impact GB migration, as has been indicated by Godiksen et al. [38], it is possible that the mobilities simulated in this work would be significantly altered by the presence of dislocation structures and even enable or constrain shear coupled GB migration.

In the end, any one of these various factors influencing the migration of recrystallized GBs in experiments could dominate over the others. It is difficult to know exactly which effect is then controlling or exerting the largest influence on the actual migration. Nevertheless, based on the data and discussion above, it is reasonable to deduce a number of facts about what may likely happen. First, it is unlikely that many materials experience sustained shear coupling in a single direction over large

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**Figure 8.** Examination of the boundary migration of the (5 47 76) GB at two different snapshots (a-b). Not the lateral motion of the small facet dominating the migration, though it is noted that the boundary as a whole simultaneously migrates as the facets move.
distances. Depending on the distance migrated by the GB, this would produce significant offsets; based on shear coupling factors in Table 1, offsets would be equal to ~1/3 the distance migrated by the GB. Furthermore, the boundaries do not always migrate in a uniform fashion, so varying degrees of shear coupling would have to be accommodated and this could be difficult. In addition, not all GB plane orientations shear couple in the same direction or by the same magnitude, further complicating shear coupling over a large area. Second, the fact that the shear constrained mobilities of the GBs show some level of correlation between high/low mobility and ‘curved’/‘flat’ boundary planes, means that the mobility may be a good indicator of when a given boundary will migrate even in the presence of additional factors. Third, there may be some factor that would cause a ‘flat’ (5 47 76) GB to be less mobile, despite the fact that the atomistic simulations predict finite mobility values. Perhaps the deformation structure somehow facilitates faster motion of the ‘curved’ portions of the boundary and also manages to decrease the mobility or completely immobilize boundaries that might otherwise exhibit significant migration.

5. Conclusions
The present work seeks to provide insight into possible factors affecting the migration of recrystallizing GBs, by comparing to atomistic simulations of GB energy, structure, and mobility. The work details methods for selecting specific CSL bicrystals that can be chosen to match experimental GBs. Additionally, methods for constructing multi-normal bicrystals are presented.

As a case study, three GBs are selected to match a theoretical GB with both ‘curved’ and ‘flat’ boundary plane orientations. The structure and energy of the three GBs indicate that ‘flat’ portions of the recrystallization GB have lower energy than the ‘curved’ portions of the GB. This structure is consistent with the principle of a minimum energy GB structure, meaning that a GB will maximize the area of low energy boundary plane orientations while minimizing the area of high energy boundary plane orientations that may be required to accommodate geometry.

The three selected GBs prefers to migrate by shear coupled mechanisms when not constrained. However, since many polycrystalline materials would likely restrict shear coupled GB motion, these same three GBs are also studied using boundary conditions that restrict lateral migration of the GBs. Under these boundary conditions, the GBs are still mobile, but the ‘flat’ boundary is faster than one ‘curved’ GB, while slower than the other. As a result, one can reasonably conclude that inherent GB mobility is an important factor in the motion of recrystallization boundary migration. However, this fact alone is not sufficient to accurately describe or predict recrystallization GB migration in experiments. The deformation microstructure in the non-recrystallized regions also likely plays a roll due to the fact that the simulated ‘flat’ GB is mobile while experiments typically show slow or negligible migration of ‘flat’ GBs over long periods of time. The GB migration mechanisms (moving facets in the case of the ‘flat’ GB) may also interact with deformation microstructures in such a way as to accelerate or decelerate the inherent mobility of a GB.

Together, the atomistic methods and case study presented here are intended to demonstrate how atomistic simulations provide valuable insight into the physical factors of GB migration that are not easy to resolve using experimental techniques alone.

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