Thesis Outline:

Introduction

* Nuclear Fuel
  + Nuclear fuels have been studied extensively in order to make nuclear reactors more efficient and safe as they are running.
    - Nuclear reactors utilize chain-reaction fission events of fissile materials, such as uranium. In order for reactions to be sustainable, the amount of fissile material present must be no lower than a certain percentage.
    - UO2 is the primary fuel used in nuclear reactors.

Background

* Grain Boundaries
  + Crystalline materials are composed of ‘sub-crystals’ (or ‘crystallites’, but most commonly known as grains).
    - Grains of different orientations do not match up atomically at the boundary. This results in an increased energy (known as the grain boundary energy, discussed later).
    - Different types of grain boundaries can have different results on a material’s properties.
      * For nuclear fuels, this is important to track for efficiency and safety.
    - Grain boundaries have five degrees of freedom.
      * Misorientation
      * Inclination
* Grain Boundary Energy
  + Isotropic
    - Most researchers in the past have ignored two of the degrees of freedom available in grain boundaries, either assuming that they are of little consequence, or asserting that it is too difficult to have a five degree of freedom model.
  + Anisotropic
    - A few researchers have studied the anisotropic nature of grain boundaries, and have come up with a few ways to incorporate each degree of freedom in a model. There is not yet a universal function to describe the grain boundary energy anisotropy in every crystalline material, but some researchers have created successful models for subsets of materials.
* Bulatov’s work in 2014
  + Bulatov *et al.* developed a function that accurately interpolates any arbitrary grain boundary energy for face-centered cubic (fcc) metals.
* Results from Harbison 2015
  + Harbison used Bulatov’s results to develop a similar function that interpolates any arbitrary grain boundary energy for uranium dioxide (which has a fluorite crystal structure).

Methods

* LAMMPS
  + In creating the interpolation function, Harbison utilized grain boundary energies of uranium dioxide calculated without an anneal. This work used the same misorientation angles utilized by Harbison, but calculated the energies with an 800 K anneal.
    - Better at finding a global minimum
* An analysis of Bulatov’s paper was conducted to learn the methods used to interpolate over the 5-space.
  + Visual representations of grain boundaries can be done in many different ways, each with their own pros and cons.
    - Rodrigues
    - Axis-angle
    - Fundamental zone
    - Note that these ideas can be combined together in various ways as well.
* An analysis of Harbison’s code was conducted to learn how the fitting procedure was implemented. Bulatov’s code was analyzed as well to learn how the interpolation function worked.
* In order to better understand how well the fitted function fit the data, a method to calculate chi squared was developed.
  + Development of P and Q matrices
    - Different ways to calculated the orientation matrices.
      * Rodrigues rotation formula
      * Using same method as MARMOT
      * Bunge rotation matrix
        + Euler angles calculated a few different ways

Geometric calculation

Matlab’s MTEX package

quaternions

* + - * Tested by re-creating the 1D plots in Bulatov *et al.*’s paper.
    - Different ways to develop rotation matrices (matrix required to rotate the grain boundary normal to the [100] axis.
      * Assume same GB orientation as in MARMOT
      * Geometric calculation
  + Two methods to calculate chi squared
    - One used the P and Q matrices to test the entirety of the fit.
    - One used the 1D subsets to test the fits of the subsets, and then the full chi squared value was calculated from that.

Results and Discussion

* Results from the MD simulations show a general decrease in the grain boundary energies, allowing a trend to emerge. This results in an all around better fit.
* There are some unexpected results present.
  + <100> symmetric tilt deep cusp due to crystal realignment
    - used Harbison’s data point
  + <100> twist and <111> symmetric tilt have parts of the energy function with positive concavity.
    - Could indicate need for additional cusps
* The reduced chi squared values are much smaller than one for nearly all sets regardless of the method used to calculate chi squared.
  + Exception: <100> tilt subset using P and Q matrices is higher than it should be.
    - This is due to a scaling issue
  + Over-fitting for data
  + Indicative of need for more data points.
* A comparison of current results to a fit that uses more parameters shows a visibly better fit to the data.
  + More cusps may be needed
  + Preliminary results show a smaller reduced chi squared value
    - Good, except for the fact that it’s still smaller than 1.

Conclusion

* A better interpolation function for GB energies in UO2 has been created using improved MD simulation data.
* Further work needs to be done to calculate more data points for fitting.
  + Better chi squared value
  + Easier to identify trends.
  + This is an issue because of the huge amount of computational resources required for one data point (each data point represents about a day of calculations using the high performance computing available at Idaho National Laboratory).
* An application of this function to other polycrystalline materials with a fluorite crystal structure would help with further validation.
  + Requires a large amount of grain boundary energies for each different materials
    - Also requires a large computational expense.