Computation of Effective Diffusion Coefficient in a Polycrystal

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Motivation

- Diffusion database is key to modeling kinetics of thermally activated processes
  - *Phase transformations, grain growth, recrystallization*

- Diffusion coefficients depend on the microstructure
  - *Grain size, grain boundary misorientation distribution*

- Microstructure may not be stable at the processing temperature
  - *Varying diffusion coefficients*

- Success of Integrated Computational Materials Engineering (ICME) depends on reliable microstructure-processing models
  - *Lack of diffusion data for key lightweight materials*
Project Plan

- Experimental measurement of diffusion coefficients rely on
  - Measuring a concentration profile of a diffusing species when subjected to a diffusion anneal
  - Using analytical approaches to relate concentration gradient to an effective diffusion coefficient

- Several uncertainties exist in measurements
  - Varying contributions from bulk and high diffusivity interfaces
  - Concurrent microstructural evolution

- Effective diffusivity models at the mesoscale help to de-convolute the various contributions
Mapping algorithms used to develop realistic input microstructures

- EBSP data for annealed 6022 mapped to 3D grain structure
- Algorithm simultaneously minimizes error in texture and boundary misorientation distribution (BMD)
- Mapping code transferred to Miss. State
MD Simulations of grain boundary diffusion in Mg bicrystal

- 1010 tilt boundaries considered because of their prevalence
- The Sandia code LAMMPS code was for the simulations
- Bi-crystals generated by rotating single crystals to the required orientations and assembling them
- Initial energy minimization carried out using a low simulation anneal
- Bi-crystal then expanded uniformly in all directions using thermal expansion coefficient at the diffusion temperature and equilibrated at the highest temperature investigated (750K)
MD Methodology

• The motion of free-surface atoms constrained to a plane parallel to the grain boundary
• Mean square displacement of atoms measured in a 2 -3 nm wide region centered on the grain boundary
• Grain boundary diffusion coefficient calculated from the slope of the mean square displacement versus time
• Activation energy for grain boundary diffusion calculated from an Arrhenius plot of the diffusion coefficient versus 1/T
MD simulation of grain boundary diffusivity in Mg

- At low temperatures (600K) boundaries showed facet formation and the MSD measurements are not reliable;
- Computed activation energies for the two tilt boundaries were roughly the same;
- Simulated diffusion coefficients at 750 K three orders of magnitude higher than for single crystal magnesium at 741K (Shewman and Rhines, Trans. AIME, 1954).
Mesoscale simulation approach

- Input to the model is a realistic 3-d microstructure that matches experimental conditions
  - *Either simulated or mapped from 2-d characterization data*

- Random walkers are introduced and operate at the mesoscale

- Large 3D microstructures with periodic boundary conditions

- The properties of the random walkers obtained from lower length scale models
  - *Apparent activation energy for bulk, grain boundary or triple line diffusion*
  - *Flip attempt frequency is proportional to the activation energy*
  - *For each walker the local neighborhood defines the location type*
  - *Function of concentration*

- Diffusivity measured from simulation of mean square displacement (MSD) of walkers

- To introduce large number of non-interacting walkers large microstructure is required (need for parallel computing)
Example 1: Single Crystal

- Apparent bulk activation energy of 7000 (dimensionless)
- Temperature (dimensionless) varied from 600-1000
- Results averaged over roughly 1500 walkers in a 80 x 80 x 80 single crystal
- Smooth variation of MSD with time and recovery of bulk activation energy
Example 2: Bicrystal

- MSD curves are non-linear
- Effective diffusion coefficients obtained from long-time slope of MSD-time curves
- Simulations show the transition from GB dominated diffusion at low T to bulk dominate diffusion at high T
Bicrystal - continued

- At low T, GB jumps significantly higher than bulk jumps
  - Linear MSD vs T
- At high T, GB contribution appreciable, increases with time
  - Non-linear MSD vs T
- In real metals, GB diffusion is dominant when $D_{gb}\delta >> D_l d$
- In mesoscale simulations $\delta/d$ is large; should perform simulations with varying $d$ and extrapolate to small ratio
Example 3: Polycrystal

- MSD vs time curves are not smooth, need bigger microstructure and more walkers
- Effective diffusivity increases with decreasing grain size
- Effective diffusivity increases with temperature at a given grain size
Future Work

- Calibrate the mesoscale model for tracer diffusion in polycrystalline magnesium
  - Validate using SIMS measurements

- Extend the model for chemical diffusion in two component Mg alloy (Mg-Al)
  - Perform MD simulations to obtain GB diffusivity
  - Effect of solute segregation

- Investigate the effect of simultaneous evolution of microstructure during diffusivity measurements