Final report: Constructing comprehensive models of grain boundaries using high-throughput experiments

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Project Scope

This is the final report on project DE-SC0008926**. The goal of this project was to create capabilities for constructing, analyzing, and modeling experimental databases of the crystallographic characters and physical properties of thousands of individual grain boundaries (GBs) in polycrystalline metals. This project focused on gallium permeation through aluminum (Al) GBs and hydrogen uptake into nickel (Ni) GBs as model problems.** This report summarizes the work done within the duration of this project (including the original three-year award and the subsequent one-year renewal), i.e. from August 1, 2012 until April 30, 2016.

Technical accomplishments

Novel optical methods for microstructure characterization

A major part of our work focused on developing methods for conducting high-throughput experiments on GBs in engineered microstructures. We concentrated on specially processed foils with through-thickness grains, such as those shown in Fig 1B and 1C, and developed a technique to measure the complete crystallography of all the GBs in them. Describing the crystallography of a GB requires five parameters. Electron backscatter diffraction (EBSD) measures four of these parameters for each of GB on the surface of the sample: three for the misorientation between the neighboring grains and one for the trace of the GB plane on the surface. However, EBSD does not determine the inclination of the GB plane with respect to the surface.

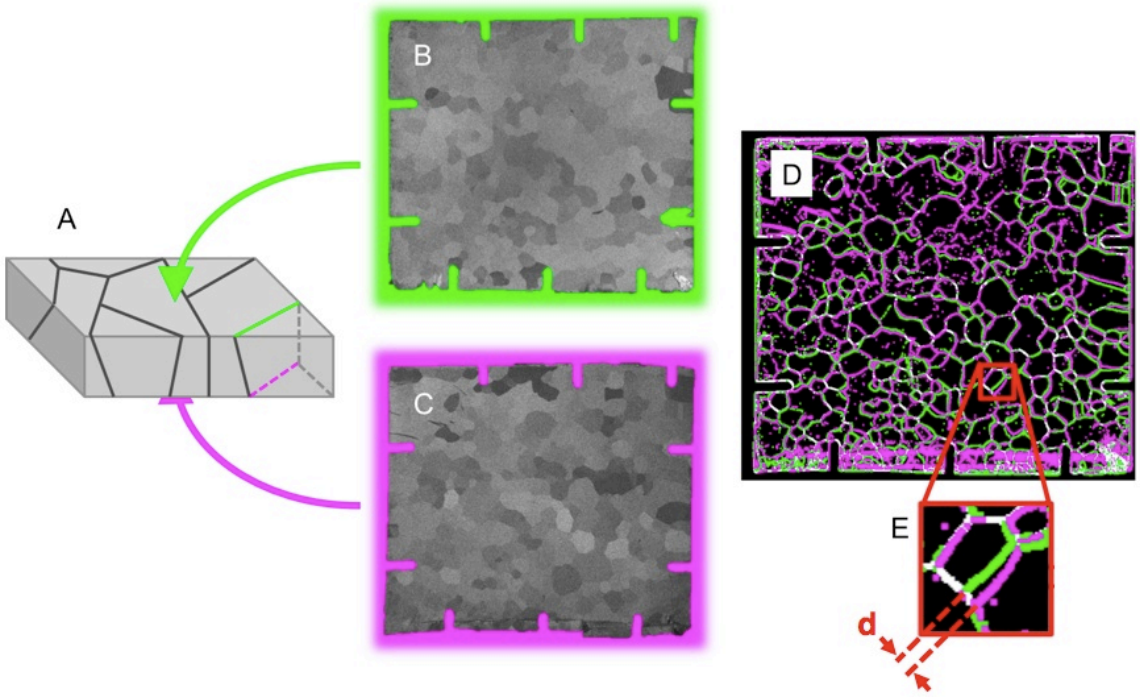

Fig. 1: Measuring GB plane inclinations: (A) schematic representation of a sample with through-thickness grains; (B) and (C) are optical stereo-micrographs from both surfaces of the sample showing the grain structure; (D)

overlapped images of GB traces from the top- and back-surfaces; (E) detail of the overlapped GB traces showing a clear shift between top- and back-traces of the same GB.

To determine GB plane inclinations, we imaged both sides of samples with through-thickness grains using an optical stereo-microscope under different illumination conditions (Fig. 1B). A chemical bath in NaOH was performed to highlight GB traces on the surface. The collected images were post-processed in MATLAB, first to locate GB traces and then to register the images taken from both sides of the sample (Fig. 1D). The registration of the top- and back-side images was performed using pre-made notches on the sample edges (visible along the edges of Fig. 1B and 1C). The inclination of each GB plane was computed from the average distance between top-surface and back-surface traces (Fig. 1E) and the sample thickness. The samples in Fig. 1 are Al sheets produced by the group of Dr. J. Erlebacher at Johns Hopkins University.

To validate our determination of GB plane orientations, we exposed the Al foil in Fig. 1 to liquid Ga, as illustrated in Fig. 2. The Ga rapidly wetted many of the GBs in the sample, allowing us to disassemble the microstructure grain-by-grain and directly image the shapes of a few representative grains, as shown in Fig. 2B. By comparing GB plane inclinations obtained from the disassembled grains with those determined by the analysis method described above, we were able to confirm the accuracy of our newly developed technique.

By merging the GB plane inclinations obtained using our surface imaging technique with misorientations found using conventional EBSD, we obtain the complete crystallography of each GB in the sample. This method is fast, does not require large-scale computing resources (all analysis is done on a laptop), and non-destructive. A description of this method was published in Nature Computational Materials (NCM) [1]. This publication was highlighted by NCM, MIT news (http://news.mit.edu/2016/metals-crystal-structure-0706), and the DOE Office of Science homepage. Further development of the optical surface scanning methods used for this technique led us to an unanticipated invention that may make these sorts of analyses rapid and inexpensive enough for large-scale industrial applications. An invention disclosure describing our findings has been filed at MIT.

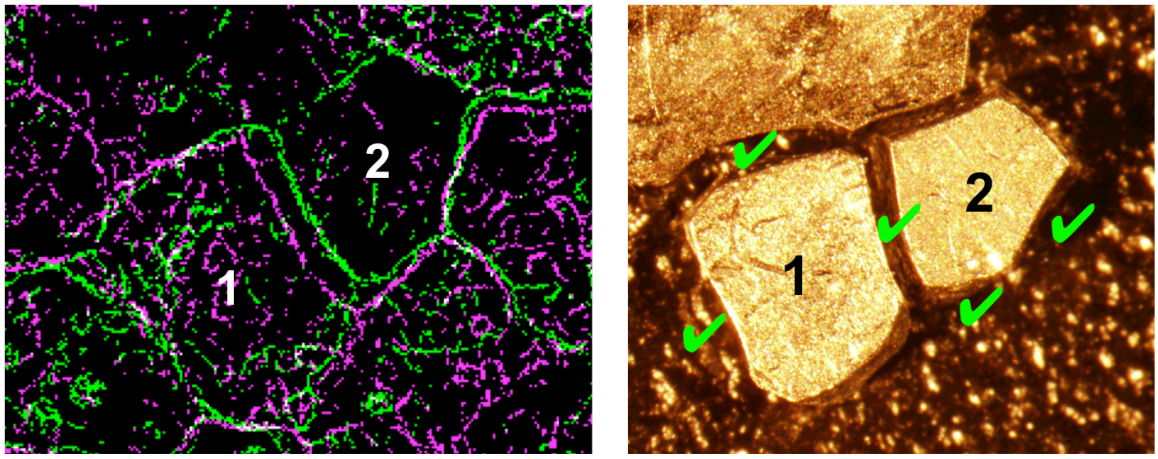

Fig. 2: Left: inclination of the GB plane between grains 1 and 2 determined by the analysis method described above. Right: validation of our method: after being wetted by liquid Ga, grains 1 and 2 were separated and their plane inclinations imaged directly in an optical microscope. The green check marks denote agreement with GB inclinations determined using our analysis method.

Building on this initial success, we have continued to develop quantitative approaches for microstructure characterization using optical microscopy methods. Recently, we have found that by measuring directional reflectance profiles for individual grains, we are able to collect partial information on grain crystallographic orientations. For example, Fig. 3 compares the distribution of grains with <110>-type out-of-plane orientations determined using optical reflectance with an analogous measurement carried out with EBSD on the same pure Ni sample, demonstrating excellent agreement between these approaches. The optical reflectance method, however, is much faster and less resource intensive than the EBSD-based measurement. Moreover, unlike EBSD, the optical reflectance method does not require vacuum and may be carried out in air. We believe that our optical

reflectance method has the potential to make microstructure analysis of metals routine in industrial settings. A manuscript describing our findings is currently in preparation [2].

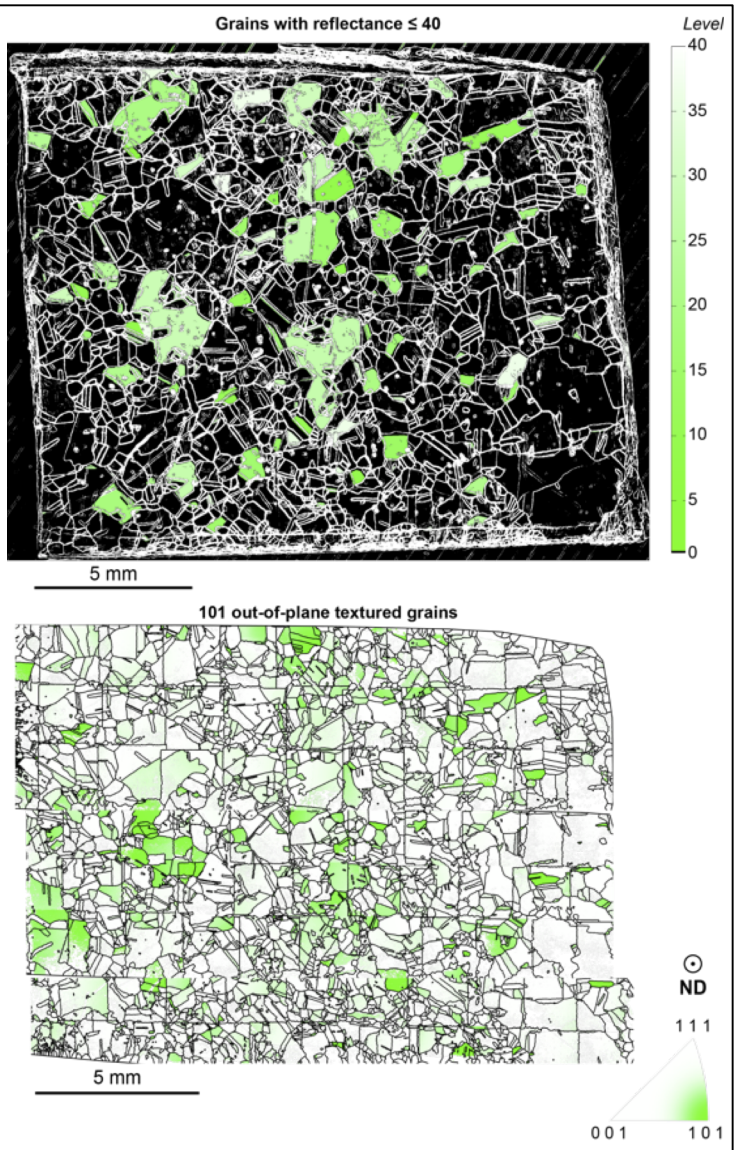

Fig. 3: Comparison between the distribution of grains with <110> out-of-plane texture in a polycrsytaline Ni sample measured using our optical reflectance method (top) and EBSD (bottom).

Localization of GB constitutive laws from measurements on GB networks

Our initial focus was to determine whether GB constitutive behavior could be inferred from measurements of effective properties of GB networks in polycrystalline solids. Specifically: given numerous polycrystals with qualitatively different GB networks (e.g. due to differences in crystallographic texture), how much can we infer about the properties of the individual GBs in them? Deconvoluting local properties of a heterogeneous system from homogenized global measurements is known as *localization*. It is the reverse of homogenization.

We studied localization of GB diffusivities from the effective GB diffusivity D_{eff} of a polycrystalline aggregate. We found that, while inferring the diffusivites of individual GBs from the behavior of GB networks may not be feasible, it is nevertheless possible to infer a small number, n, of parameters that enter into analytical constitutive laws for GB diffusion. If the character of the GB networks used for inference is chosen judiciously, then the number of measurements needed to determine these parameters scales with n, rather than exponentially with the dimensionality of the GB crystallography space [3]. The manuscript we published based on this work was the December nominee for the 2015 Robert W. Cahn prize at the Journal of Materials Science. Future extensions of this work may address more detailed descriptions of GB diffusion and other properties related to GB energy landscapes [4].

This advance is underpinned by an improved ability to represent GB networks in polycrystals [5-7]. Spectral representations of triple junction distribution functions in GB networks provide a mathematical connection between crystallographic texture and the structure of grain boundary networks. We formulated a mathematical framework to define the triple junction hull, which describes all physically realizable triple junction distribution functions [7]. These results are crucial to improving the localization schemes developed so far.

Neutron radiography measurements of hydrogen in metals

One of our research goals was to investigate hydrogen interactions with GBs in Ni. The method that we proposed to use for this purpose is neutron radiography (NR). We have applied for and received an allocation for conducting NR measurements at the BT2 thermal imaging instrument, which is operated as a user facility by the Radiation Physics Division of the Physical Measurement Laboratory at NIST. In our first set of experiments, we collected baseline neutron radiography data on commercially pure Ni samples provided by I. McCue and J. Erlebacher from John's Hopkins University. These samples were thermomechanically processed to contain large, through-thickness grains. We characterized all the GBs in this sample using the optical microscopy methods, which we developed in this this project [1]. The results from this measurement are illustrated in Fig. 4.

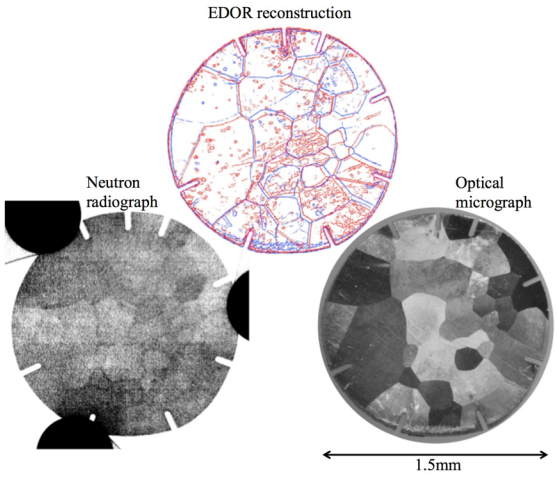

Fig. 4: Microstructure of a pure Ni sample with through thickness grains (bottom right); EDOR [1] reconstruction of the GB network in the sample (top); Neutron radiograph showing distinct signals from individual GBs and grains (bottom left).

Our initial investigations validate our approach for constructing databases of GB properties using high-throughput experiments. In particular, they demonstrate that we are able to fully characterize the complete crystallographic character of all the GBs in our samples using EDOR. This method integrates GB misorientations, determined using EBSD, with GB plane orientations, measured by correlating the locations of GB traces on opposite faces of samples with through thickness grains. Our investigations also demonstrate that our NR measurements are able to distinguish signals from individual GBs as well as from individual grains. We are currently preparing for our next round of neutron reflectometry measurements, which are aimed at detecting H in pure Ni samples.

Bayesian methods for inference of GB properties and optimal experimental design

We have explored Bayesian methods for inferring micro-scale mechanisms of GB behavior from macro-scale observables. Our goal is to apply these methods to Ga permeation through Al GBs. To that end, we have developed a phase field model for Ga penetrating GBs and are currently a database of Al GB structures for the inference process. Meanwhile, to explore general features of Bayesian inference in the context of surface-governed phenomena, we have formulated and solved a model inference problem: the determination of the properties of a hidden substrate based on the behavior of a film deposited on it [8]. Our work highlighted the need for surrogate models to perform inferences efficiently.

We have also investigated Bayesian optimal experimental design (OED) as a means to efficient extraction of GB properties using limited experimental resources. OED aims to maximize the value of experiments and the data they produce by ensuring efficient allocation of limited resources, especially when numerous repeated experiments cannot be per- formed. We have applied fully Bayesian and decision theoretic approaches to OED—accounting for uncertainties in models, model parameters, and experimental outcomes, and allowing optimality to be defined according to a range of possible experimental goals [9]. We demonstrated this approach on two illustrative problems in materials research. The first example is a parameter inference problem. Its goal is to determine a substrate property from the behavior of a film deposited thereon. We designed experiments to yield maximal information about the substrate property using only two measurements. The second example is a model selection problem. We designed an experiment that optimally distinguishes between two models for helium trapping at interfaces. In both instances, we provide model-based justifications for why the selected experiments are optimal. Moreover, both examples illustrate the utility of reduced-order or surrogate models in optimal experimental design.

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