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EBSD of Rough Native $CuInGaSe_2$ Thin-Films

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The polycrystalline Cu (In, Ga) Se2, or CIGS, based thin-film materials system has long been studied for use in photovoltaic technologies, where its bandgap tunability, mechanical flexibility, and relatively low production costs are all appealing. Nonetheless, significant defect populations, which serve to reduce efficiency, create performance instabilities, and increase concerns about long-term reliability, have hindered wide-scale adoption. Prior work, including application of a scanning probe based deep level trap spectroscopy (SP-DLTS) defect mapping technique and scanning transmission electron microscope (STEM) based electron energy loss spectroscopy (EELS), has shown that the most detrimental defects, with energy level near mid-gap (thus serving as a carrier recombination center), are most likely caused by CuIn/Ga antisites and tend to cluster at or around certain grain boundaries [1,2]. However, the exact nature of these particular boundaries — their structures, chemistries, or even the relative misorientation of their associated grains — and their relation to this defect clustering and/or its formation is yet unknown. As such, electron backscatter diffraction (EBSD) orientation mapping, directly correlated with defect-sensitive techniques like SP-DLTS and/or STEM-EELS, could prove critical for providing the final missing links toward understanding the mechanisms behind these defects. Indeed, recent studies using correlative electron beam induced current (EBIC) with EBSD have been able to identify boundaries, and their relative misorientations, that possess detrimental electronic properties [3]. However, because EBIC is unable to resolve the defect energy levels, many questions are left unanswered. Furthermore, this study, and others like it, employed focused ion beam (FIB) milling to flatten the natively-rough CIGS [3-5], which may run the risk of changing the nature of any near-surface defect structures.

Because of the large sample tilt angle typically used in EBSD measurements, it is highly surface sensitive, making analysis of rough surfaces particularly challenging. Additionally, the noisier EBSD patterns arising from rough surfaces lead to significantly more error following conventional Hough transform indexing routines [6]. As such, rough samples are thus usually polished or milled smooth in preparation for EBSD measurements. Conversely, native CIGS films can have peak-to-valley roughness of several 100's of nanometers (see Figure 1), but because the defective regions are often close to the surface, any kind of polishing or milling runs the risk of disturbing them. Additionally, SP-DLTS is highly sensitive to conditions at the surface, and additional atomic-scale defects (e.g. dangling bonds) created via milling/FIB flattening can render it ineffective for correlative analysis. Therefore, it would be of value to be able to perform such measurements on the natively rough surface. Prior work has shown that the dictionary indexing (DI) approach, which effectively works by matching the complete experimental pattern against a simulated master pattern, can be used to analyze low-quality (i.e. high noise) EBSD data [6]. The recently introduced spherical indexing method, which uses the detector geometrical parameters to back-project the experimental pattern onto the Kikuchi sphere, where its cross-correlation with a spherical master pattern is performed, promises similar robustness to the DI method but with improved speed and practicality [7].

Here we present the use of the spherical indexing method, as implemented within the EMSoft package [8], toward EBSD analysis of un-flattened CIGS thin-film samples. Production CIGS solar cells were procured from multiple commercial vendors. The transparent conductive oxide and CdS layers were etched away using dilute HCl to reveal the rough underlying CIGS, shown in Figure 1a. Figure 1b and c compare the EBSD orientation imaging microscopy (OIM) map produced using the conventional Hough method (Fig. 1b), yielding a poor confidence index (CI) of ~0.05, versus the spherical indexing approach using the same dataset (Fig. 1c), with a much-improved CI of ~0.5. The latter is then used to calculate the average grain misorientations, shown in Figure 2. Although optimization of the acquisition and indexing processes is still ongoing, initial results from the fully-rough samples are strongly consistent with [3], including significant populations of 60° and 70° twin boundaries, indicating reasonably good accuracy of this approach thus far.

These early results indicate that spherical indexing is likely to be an enabling process for performing EBSD grain boundary analysis on native rough CIGS samples that can be directly correlated with SP-DLTS (or other measurements) performed on the same. Nonetheless, the high roughness of the surface does still present questions regarding the reliability of the orientation analysis and whether shadowing and/or other 2D projection artifacts may be creating inaccurate results. Preliminary comparisons of EBSD OIM maps taken at different azimuthal rotations reveal a small number of discrepancies, which often correspond to the noisiest portions of the map. We are thus currently investigating digital 3D reconstruction of the surface morphology to help provide guidance for error correction procedures. At the conference, we will present up-to-date results and best practices developed in this work.

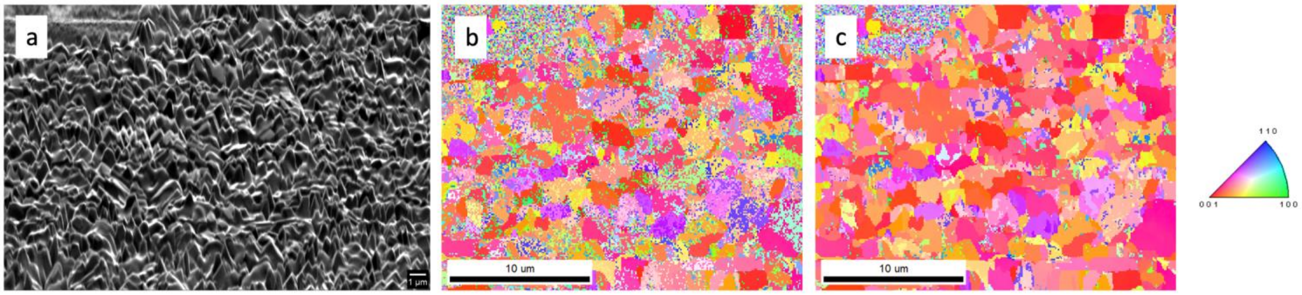

Figure 1. (a) SEM image of CIGS sample taken at 70° tilt, with tilt and focus correction applied. EBSD orientation maps of the same area using (b) Hough transform indexing and (c) spherical indexing.

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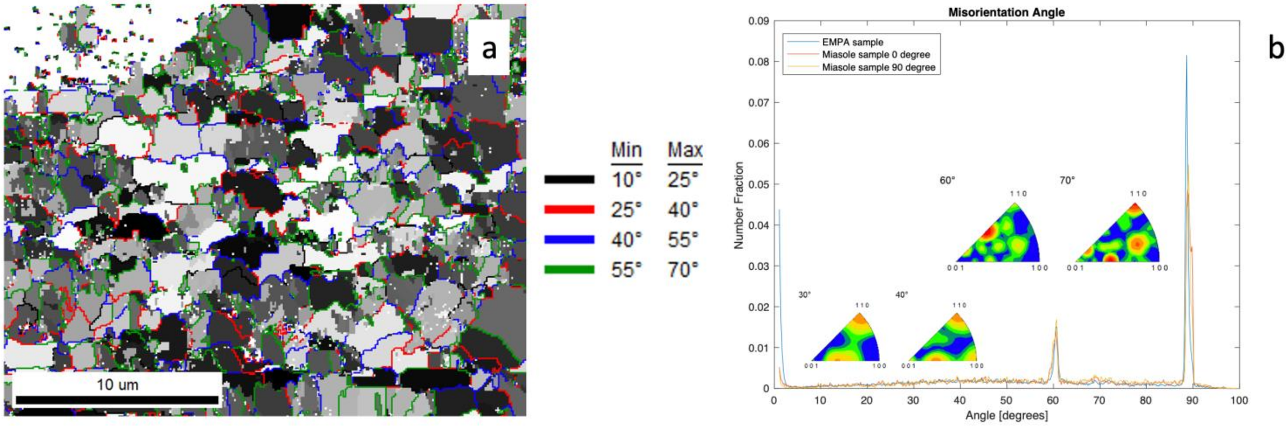

Figure 2. (a) Map of grain boundary misorientation angles using the OIM map from Fig. 1(c), and (b) histogram of the same. Note that the strong peak at 90° contains a combination of noise pixels that were indexed opposite to the surrounding grain (and thus artifacts) and some amount of real boundaries.

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