Three-Dimensional Minority-Carrier Collection Channels at Shunt Locations in Silicon Solar Cells

Harvey Guthrey,^{1*} Steve Johnston,¹ Dirk N. Weiss,^{2**} Sachit Grover,^{2**} Kim Jones,^{1***}
Alain Blosse,² Mowafak Al-Jassim¹

¹National Renewable Energy Laboratory, Golden, CO 80401

²Scifiniti, San Jose, CA 95134

*Corresponding Author

**Currently at First Solar

***Currently at Whiting Petroleum Inc.

Abstract

In this contribution, we demonstrate the value of using a multiscale multi-technique characterization approach to study the performance-limiting defects in multi-crystalline silicon (mc-Si) photovoltaic devices. The combination of dark lock-in thermography (DLIT) imaging, electron beam induced current imaging, and both transmission and scanning transmission electron microscopy (TEM/STEM) on the same location revealed the nanoscale origin of the optoelectronic properties of shunts visible at the device scale. Our site-specific correlative approach identified the shunt behavior to be a result of three-dimensional inversion channels around structural defects decorated with oxide precipitates. These inversion channels facilitate enhanced minority-carrier transport that results in the increased heating observed through DLIT imaging. The definitive connection between the nanoscale structure and chemistry of the type of shunt investigated here allows photovoltaic device manufacturers to immediately address the oxygen content of their mc-Si absorber material when such features are present, instead of engaging in costly characterization.

Introduction

The dominant photovoltaic (PV) market share is still held by cells based on crystalline silicon absorbers despite recent advances in CdTe and Cu(In_{1-x}Ga_x)Se₂ solar cells. Although crystalline silicon solar cells have approached the theoretical efficiency limit (Shockley and Queisser, 1961, Tiedje, et al., 1984), these devices are made from high-purity Si material with low dislocation densities and few or no grain boundaries. Exploring different processing methods and materials has led to reduced manufacturing cost, but not without sacrificing the ability to produce champion-efficiency devices. Grain boundaries and dislocations are present in most solar cells based on crystalline silicon absorbers, and the interaction of these structural features with impurities can profoundly influence device efficiencies (Davis, et al., 1980, Hofstetter, et al., 2009, McHugo, et al., 1997, Riedel, et al., 2002, Seifert, et al., 1993, Stutzler and Queisser, 1986). Enhanced non-radiative recombination and reduced diffusion length result from impurity decoration of grain boundaries and dislocations. In addition, impurity precipitates are also known to cause diode breakdown in reverse-bias conditions (Kwapil,

et al., 2009), but the mechanisms by which this occurs is not fully understood for all cases. Significant effort has been made by the silicon PV community to understand the nature of shunts, or regions that exhibit abnormal current flow under different bias conditions (Breitenstein, et al., 2008, Breitenstein, et al., 2007, Lausch, et al., Schubert, et al.). Emerging from previous research using correlative characterization techniques is a better knowledge of shunts that includes: a classification of different types of shunts (Breitenstein, et al., 2004) and theories of how dislocations, grain boundaries, and impurities interact to result in the shunting behavior observed through thermographic imaging techniques.

Several classifications exist (Breitenstein, Rakotoniaina, Al Rifai and Werner, 2004, Langenkamp and Breitenstein, 2002) of shunts in multicrystalline silicon (mc-Si) solar cells, and they are based on the origin of each type of shunt. In this work, we consider shunt types that do not originate from gross disturbances (e.g., cracks, holes, metallization side effects) resulting from cell processing, but rather, the types associated with structural defects and impurities in the bulk of the absorber. The presence of these types of shunts is a more difficult issue to address than those associated with processing and handling of devices. Understanding the nature of shunts in a particular device is important for mitigating the presence and effects on performance for devices processed in the future. Here, we investigate shunts that exhibit an increase in current flow (heating) under forward- and reverse-bias conditions and reduced collection in plan-view electron beam induced current (EBIC) imaging.

Inversion Channels at Structural Defects

Previously, the cause of certain types of shunts has been connected to what are thought to be inversion channels around structural defects in various types of silicon PV devices. Inversion channels have been shown to exist around both dislocations (Breitenstein, et al., 2001) and grain boundaries (Breitenstein, Bauer, Kittler, Arguirov and Seifert, 2008, Breitenstein, Bauer and Rakotoniaina, 2007, Breitenstein, Rakotoniaina, Al Rifai and Werner, 2004) in ribbon-grown and cast mc-Si. In the grain-boundary studies, the inversion type around grain boundaries was connected to the presence of SiC precipitates decorating the grain boundaries acting as n-type conductors. In this case, type inversion is the result of the band offset between the SiC and the p-type silicon absorber, not the trapping of positive charges. Transmission electron microscopy (TEM) investigations of one of the boundaries found micron-sized SiC precipitates at some boundaries and long SiC filaments at others. These authors expressed some doubt regarding the ability of widely spaced precipitates to cause continuous networks of type inversion in this case. However, the small volume associated with the TEM sample may have excluded the features responsible for the observed type inversion. In another work, inversion channels were observed around dislocations and a model was constructed based on the oxygen content typical of ribbon-growth-on-substrate (RGS) silicon (Breitenstein, Langenkamp and Rakotoniaina, 2001). However, there was no direct observation of oxygen at the dislocations because there was no nanoscale analysis. It should be mentioned that the inversion channels were observed in all the previously mentioned studies via the backside EBIC technique. In backside EBIC, the back contact is partially removed; therefore, any

collection of minority carriers generated at the back of the absorber (far from the junction) is due to the presence of conduction pathways for minority carriers. In the present work, a similar analysis is performed by looking at cross sections prepared by the focused ion beam (FIB) method, where carriers generated away from the depletion region should show significantly decreased EBIC collection unless there is a minority-carrier conduction path near the excitation. In this investigation, we demonstrate both the methodology and results of a multiscale, multi-technique correlative study of shunts in mc-Si photovoltaic devices.

Material and Methods

A mc-Si solar cell about 80 μm thick was investigated in this study. The device parameters for this solar cell are: open-circuit voltage (V_{oc}) = 535 mV, short-circuit current density (J_{sc}) = 26.94 mA/cm^2 , series resistance (R_{s}) = 3.089 $\Omega \text{ cm}^2$, shunt resistance (R_{SH}) = 78.79 $\Omega \text{ cm}^2$, and efficiency measured as 7.78%. Specific conditions were used to process this device and thus these parameters are not representative of typical devices produced by this manufacturer. Dark lock-in thermography (DLIT) images were acquired with 12.6 Hz pulses at 0.9 V forward bias and 4 V reverse bias that allowed qualitative identification of the local current density-voltage (J-V) behavior of the shunts. EBIC imaging and TEM sample preparation were performed in-situ in a FEI Nova NanoLab 200 dual beam FIB. Using the EBIC stage in the FIB allows for site-specific preparation of defects, as well as serial sectioning of the entire defective regions (Yoshida, et al., 2005). EBIC images in the plan-view orientation were acquired at accelerating voltage of 15 kV and beam current of ~ 1 nA. In cross section, the EBIC images were acquired with 5 kV to enable imaging with higher spatial resolution. One shunt identified with DLIT imaging was sectioned at 90° angles to highlight the three-dimensional nature of the defect. Scanning transmission electron microscopy (STEM)/TEM imaging and energy-dispersive spectroscopy (EDS) analysis were done on an FEI Tecnai F20 field emission TEM operating at 200 kV.

Results

DLIT imaging was performed on the entire Si device to identify the locations of shunts, and the results are shown in Fig. 1. Several positions, labeled I through V, exhibited heating due to the 4-V reverse-bias pulses, as indicated in the image by the accompanying scale in arbitrary units. After DLIT imaging, marks were placed around shunt locations with a pulsed laser so that they could be easily identified for subsequent investigations. These marks are clearly visible in several of the EBIC images in Fig. 1.

Figure 1. DLIT image and corresponding plan-view EBIC images (dimensions 1.28mm x 1.1mm) for the five positions exhibiting heating in reverse bias condition. The scale of the DLIT image is in arbitrary units. The locations of the shunts in each EBIC image are in the center of the red circles and generally correspond to the intersection of multiple structural defects.

To locate the position of the shunt with greater precision than afforded by DLIT imaging, EBIC imaging was first done in the plan-view orientation inside the FIB. Images acquired from several of the shunt locations—with the locations of the shunts corresponding to the labels in the DLIT image—are shown in Fig. 1. The red circles in the EBIC images indicate where heating was observed, and it is interesting that all these regions occur at the intersection of three structural defects and exhibit dark EBIC contrast. It is well known that structural defects in silicon are preferential locations for impurity precipitation. Shunts in mc-Si have been associated with a variety of impurities, including C, N, O, and various transition metals (Breitenstein, Rakotoniaina, Al Rifai and Werner, 2004, Buonassisi, et al., 2004, Kwapil, et al., 2009). The point of intersection of multiple structural defects is likely to be decorated by impurities and impurity precipitates. Higher-magnification EBIC images revealed that the dark regions were not well defined in the shunt locations. This may be due to the electron-beam accelerating voltage used (15 kV) or perhaps the current-splitting effect described previously (Breitenstein, Bauer and Rakotoniaina, 2007, Kaminski, et al., 2004). Additionally, small precipitates (< 100 nm) have been associated with shunts and pre-breakdown sites in mc-Si; because the EBIC images were diffuse in the shunt locations, we needed a systematic approach to cross-sectional EBIC analysis to identify all the features in the region that may result in the shunt behavior. It should be noted that there are other dark spots and regions where multiple defects intersect in the EBIC images, but the features within the circles appear to be unique with respect to the DLIT signal.

All five marked positions contain features that heat in reverse-bias conditions, but only the shunt in position I exhibits heating in both forward and reverse bias, as indicated in Fig. 2. The linearity of the J-V characteristics of the shunt could not be definitively confirmed because of the different magnitude of the voltage pulse used for the forward (0.9 V) and reverse (4 V) DLIT imaging. The J-V response of shunts can reveal the possible atomic-scale origin, as has been suggested in previous work (Breitenstein, Rakotoniaina, Al Rifai and Werner, 2004).

Position 1 DLIT Images

Figure 2. Higher magnification DLIT image of position I showing heating in forward (0.9V) and reverse (4V) bias conditions.

After identifying the location of the shunted regions within several microns via EBIC imaging in-situ in the FIB, the sample was tilted so that a trench could be cut on one side of the dark feature. A final low-energy clean (5 kV) was used to reduce the effects of ion-milling damage and surface recombination on the exposed surface. After an EBIC image was acquired on tilted cross sections, additional 0.5–1 μm cuts were made, removing material in a serial fashion; after each cut, an EBIC image was acquired from the newly exposed surface. Several precipitates became visible in the SEM image near the expected center of the shunt, as shown in Fig. 3. The EBIC image on the right side of Fig. 3 was acquired at this location. The bright line across the top of the image is expected because this is the location of the p-n junction, but the other bright features below the surface are surprising. Bright EBIC contrast was observed around both precipitates visible in the SEM image, and these bright regions were connected to each other and the junction by other linear bright features. The collection profile near the junction is quite narrow, so collection of EBIC signal deeper in the device is not expected. However, if there are inversion channels in the bulk of the material that are connected to the junction, it may be possible for minority-carrier electrons to travel through the p-type bulk along the inversion channels (Breitenstein, Langenkamp and Rakotoniaina, 2001). The results of the serial EBIC analysis in the FIB suggested that the subsurface features that exhibit increased EBIC collection are three-dimensional. To illustrate this, the sample was rotated 90° and an EBIC image was taken from this face of the trench. From this image, one can understand that the linear feature in the EBIC image at the right is actually a planar feature surrounded by enhanced minority-collection channels. The convergence of multiple three-dimensional structural defects surrounded by inversion channels increases the density of pathways for current to flow. This has been observed before in RGS silicon (Breitenstein, Langenkamp and Rakotoniaina, 2001) and is a probable cause for the low R_{SH} measured for this device.

Figure 3. Higher magnification EBIC image of position I (middle) showing diffuse contrast at the location of the shunt. At the left and right are cross-sectional EBIC images of the regions indicated by the left and right red rectangles, respectively, after serial sectioning with the focused ion beam. There appear to be pathways for enhanced minority carrier collection surrounding structural defects and precipitates (bright features in the images).

Figure 4. STEM EDS map of the iron showing that the region marked by the white rectangle in Figure 3 contains iron precipitates. It is unclear from the EDS analysis whether these particles are an iron silicide species.

Following EBIC analysis of the FIB-milled cross sections, TEM specimens were prepared from the exact locations represented in the cross-sectional EBIC images. EDS analysis of the precipitates that were visible in the SEM image of Fig. 3 revealed that they contain iron, as shown in Fig. 4. The presence of these micron-sized precipitates agrees with previous synchrotron- and TEM-based studies where SiC, Si₃N₄, and metallic precipitates of similar size were found to exist at shunt locations, although the mechanism of the shunting behavior is thought to be different for the various precipitates. Diffraction analysis (not shown) on either side of the linear bright feature in the EBIC images revealed the defect to be a small-angle grain boundary. As seen in Fig. 5, this small-angle grain boundary was decorated with precipitates having diameters of less than 100 nm that were found to contain oxygen and some calcium through STEM-EDS analysis. Oxygen is a common impurity in silicon due to processing conditions and apparatus, but the precipitation of oxygen at this small-angle grain boundary suggests that this material contains a high concentration of oxygen. It has been suggested previously

that structural defects decorated with oxygen impurities may be able to trap positive charges and lead to inversion layers in the surrounding regions (Breitenstein, Langenkamp and Rakotoniaina, 2001). If there is sufficiently high oxygen content to allow for precipitates to form at the locations of the shunts investigated in this work, then it is likely that oxygen precipitates exist at other grain boundaries and dislocations. The lack of shunting behavior observed away from structural defect intersection points may be due to the lower density of inversion channels or a discontinuous network of oxide precipitates decorating the other structural defects.

Figure 5. Bright field TEM (left) and HAADF STEM (right) images of the same region where the cross-sectional EBIC image at the left of Figure 3 was taken. The feature was identified as a small angle grain boundary and is clearly decorated with precipitates with diameters of less than 100nm. The STEM image shows that the boundary is decorated along its length. The inset of the STEM image is EDS spectra acquired from the precipitates which exhibit a strong oxygen signal as well as a less intense calcium peak.

Discussion

Impurity Precipitates and Small-Angle Grain Boundaries

The authors suspect that small-angle boundaries are generally formed in the latter part of solidification after the impurities present in the melt have already become frozen in at the large-angle boundaries throughout the bulk material. As oxygen continues to diffuse from crucibles or other sources into the Si material at high temperature, it is likely that the small-angle boundaries resulting from unresolved thermal stresses in the interiors of large grains have not been decorated by other impurities; thus, they are prime locations for oxygen precipitates to form. Recent work has shown that oxygen precipitates at small-angle grain boundaries are more prevalent in slowly solidified samples than in silicon that undergoes a faster solidification based on deep-level photoluminescence (PL) and time-of-flight secondary ion spectrometry (TOF-SIMS) analysis (Katoa, et al., 2014). Other PL studies have also suggested that small-angle grain boundaries may be more likely sites for oxygen precipitates to form in mc-Si due to the presence of unresolved strain surrounding these types of boundaries (Tajima, et al., 2012). In this work, we identify oxygen-containing precipitates as well as iron precipitates at the shunt location.

Tajima et al. have shown that when both iron and oxide precipitates are located at defect clusters, the band-to-band (BB) PL emission is reduced relative to the BB PL emission from regions without significant iron content (Tajima, et al., 2010). This observation is in line with the plan-view EBIC analysis (center of Fig. 3), where dark contrast was observed at the shunt location and reduced BB emission would suggest enhanced non-radiative recombination.

Reconciliation of Light vs. Dark EBIC Signal

The plan-view EBIC image of this shunt location (center of Fig. 3) shows dark contrast; however, the cross-sectional EBIC image at the right of Fig. 3 shows the enhanced minority-carrier collection in the regions surrounding the oxide precipitates, but also around the iron precipitates, although this signal is weaker than that around the oxide precipitates. This may be a result of trapped positive charges at the silicon/iron interface that exist in lower concentrations than the trapped charge surrounding the oxide precipitates. The observation of dark EBIC signal at a current-collecting channel has been described previously. Kaminski et al. (Kaminski, Breitenstein, Boyeaux, Rakotoniaina and Laugier, 2004) interpreted the dark contrast observed at inversion channels to be due to a current-splitting effect because the channels on either side of the feature are essentially separated from one another by the charged defect. This was observed by Breitenstein et al. (Breitenstein, Bauer and Rakotoniaina, 2007) by combining back-side and front-side EBIC imaging on the same defects where dark and bright contrast were observed at the same defects, respectively. The current-splitting effect can explain why both bright and diffuse dark features are present at the same defects in EBIC images taken with different geometries; however, there is perhaps another contribution to decreased EBIC signal observed in this work. In the EBIC and SEM images shown in Fig. 3, precipitates of various sizes are clearly visible. If the electron beam is positioned on these precipitates, there will be a decrease in the e^-h^+ pair generation at that point which would reduce the number of carriers that could be collected relative to regions away from the precipitates. It is likely that the diffuse contrast at the shunt location is a convolution of the current-splitting effect and the reduced carrier generation due to the electron-beam interaction with the metallic precipitates in this case.

Potential-Induced Degradation Studies

In light of several recent studies (Lausch, et al., 2014, Naumann, et al., 2014), we need to mention potential-induced degradation (PID) observed in mc-Si modules in terms of the shunts presented in this study. The same characterization approach was used in this study where, after identifying shunts via DLIT imaging, these locations were sectioned in an FIB, imaged with EBIC, and then studied with nanoscale TEM-based chemical analysis. It should be noted that in the locations of shunts investigated in the referenced works on PID shunting, no minority-carrier collection channels were observed in the FIB/EBIC cross sections. This is likely due to the lack of an inversion layer surrounding the Na-decorated stacking faults that were identified as the cause of the observed PID. This suggests that shunts of specific character (e.g., Na-decorated stacking faults) are responsible for the PID behavior and shunts associated with three-dimensional inversion

channels are not associated with PID. The shunts in this study were associated with small-and random-angle grain boundaries, as well as with metallic and oxide precipitates; no stacking faults or increases in the sodium concentration were observed at the shunt locations.

Conclusion

In this work, we have demonstrated the benefit of combining multiple characterization techniques that vary in scale over seven orders of magnitude and provide complementary information on the origin and electronic properties of shunts in mc-Si PV devices. A shunt that exhibited heating in both forward- and reverse-bias conditions via DLIT imaging was found—using in-situ serial sectioning and cross-sectional FIB/EBIC analysis—to be associated with a three-dimensional network of enhanced minority-carrier collection channels. Furthermore, these enhanced collection channels were found to originate from nanoscale oxide precipitates by preparing STEM samples from the exact regions where the cross-section EBIC images were acquired. The results of this work highlight the importance of a site-specific correlative characterization approach, as have successfully connected device properties and device-scale defect analysis with the atomic-scale root cause. These results have the potential to reduce resource- and time-intensive analysis for mc-Si PV manufacturers because we have identified that defects exhibiting enhanced heating in both forward and reverse bias are associated with high oxygen content, which results in precipitation at these locations. The low shunt resistance also indicates the presence of such features. By addressing the high oxygen content in the absorbers of devices that contain these types of features, device manufacturers are able to mitigate the negative impact of these defects on device performance.

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