Generation of dislocation clusters at triple junctions of random angle grain boundaries during cast growth of silicon ingots

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Three-dimensional distribution of grain boundaries (GBs) and generation sources of dislocation clusters is examined in a cast-grown high-performance multicrystalline silicon ingot for commercial solar cells. A significant number of dislocations are generated nearby some triple junctions of random angle GBs, although it is believed that such non-coherent GBs would not induce large strain during the cast growth. This explosive generation of dislocations would take place when the triple junctions are interacted with multiple Σ3(111) GBs. A segment of the random angle GB connected with a pair of Σ3(111) GBs nearby the triple junction would act as a dislocation source.

Supplementary material for this article is available online

Multicrystalline silicon (mc-Si) ingots with a high production ratio, manufactured by the cost-effective directional solidification method, is the important material in the solar cells with the highest market share until very recently,11 even though their solar cell efficiency is lower in comparison with monocrystalline Si solar cells. One important issue for mc-Si solar cells is, therefore, to develop functional mc-Si structures with low dislocation density, since their lower efficiency is mainly due to dislocations generated from grain boundaries (GBs) during the cast growth. High performance (HP) mc-Si with a low dislocation density, in which high density of random angle GBs (RAGBs) are intentionally introduced so as to release the thermal stress during the cast growth,2 is extensively examined to be applied toward commercial solar cells.2–10 Even in HP mc-Si ingots, nevertheless, dislocation clusters are frequently generated from some GBs during the cast growth,11,12 and this would result in the unevenness of solar cell efficiency in the ingots. Quasi-mono crystalline Si ingots, manufactured by the cast growth with monocrystalline Si seeds,13 is also expected to be applied to solar cells. However, dislocation clusters are generated from the seed joints,13–16 as well as polycrystallization takes place on the crucible,17,18 and therefore the production ratio of wafers is quite reduced. In order to solve the issue, improved quasi-mono growth method named as “seed manipulation for artificially controlled defect technique (SMART)”, by which the spatial distribution of defects including dislocations are controlled via arranging seeds with proper size and orientation,19–21 is developed. It is shown that SMART Si ingots, in which a large fraction of their volume has a low dislocation density, can be manufactured via the selective introduction of the GBs acting as release sites of the thermal stress, including RAGBs, in a specific tiny part of the ingots. Therefore, in order to optimize the dislocation density in HP and SMART Si ingots, we need to control generation and propagation of dislocations during the cast growth.

Generation and propagation of dislocation clusters have been examined in various kinds of cast-grown Si ingots, and they would be traced to their origin at GBs22,23 Many of the dislocation clusters are generated from low-energy GBs characterized with a Σ number,7,11,12,17,24 that are indexed by the coincidence site lattice (CSL) theory.25 Especially, the density of dislocation clusters correlates with the density of Σ27 GBs,12 and dislocation clusters generated at junctions of Σ27 GBs connected with Σ3 and/or Σ9 GBs are reported.1,12 On the other hand, generation of dislocation clusters at RAGBs has been scarcely observed.12 Even though the density of dislocation clusters also correlate with the density of RAGBs,12 this is explained that the clusters would be terminated at the GBs.11 It is believed that RAGBs with the non-coherent nature suppress the dislocation propagation and release the thermal stresses,2 unlike CSL GBs. Actually, there is no residual stress in non-coherent GBs,26 while a residual stress exists at CSL GBs.17,26–29 It is suggested that non-coherent GBs would not have significant internal energy for defect generation.29 Those properties have been applied to design HP and SMART Si ingots with an optimum dislocation density. In the present work, however, we have found triple junctions of RAGBs acting as a dislocation source, that might cause a modification on the design guidelines. We have discussed the mechanism of explosive generation of dislocation clusters at triple junctions of RAGBs in a HP mc-Si ingot, by using photoluminescence (PL) image processing assisted by data science and microscopic techniques.

Wafers were cut sequentially from a HP mc-Si ingot for commercial solar cells, with the slice pitch of 300 μm. The size of each wafer was 156 mm × 156 mm × 180 μm, and the total number of examined wafers was 258 (No. 611 to 868). 3D distribution of dislocation clusters in the ingot was visualized and their generation sources were determined with a spatial resolution of 0.3 mm by the PL processing with the as-sliced wafers using a PL measurement system (Hamamatsu Photonics EPL-100s)30 [e.g. see Fig. 1(a)]. Reflection images of a wafer involving the generation source of a dislocation cluster were taken using white light illumination by systematically changing illumination angles.
All the reflection images were superimposed after extracting edges using the standard Canny detector [Fig. 1(b)], and a workbench for machine learning, Weka, was applied for trainable segmentation of GBs and intra-grain dark pixels to highlight GBs intersecting at a wafer surface [Fig. 1(c)]. With the highlighted data, two-dimensional distribution of dislocation clusters and GBs at intervals of about 150 μm. Atomic structure of some generation sources was examined by transmission electron microscopy with damage-free specimens prepared only by chemical mechanical polishing.

The existence probability for generation sources of dislocation clusters peaks at the distances of 0.3 mm and 1.2 mm, with the peak probabilities of 15% and 18%, respectively, from the nearest triple junction of GBs [the red bars in Fig. 1(f)]. Assuming that generation sources distribute homogeneously in the ingot, it is simulated that the probability would peak at about 1.3 mm [green bars in Fig. 1(f)]. Therefore, the data at around 1.2 mm, encircled with the dotted black curve in Fig. 1(f), would be due to the dislocation clusters generated irrespective of triple junctions of GBs. On the other hand, considering the spatial resolution of 0.3 mm for the location of generation sources, the data at around 0.3 mm, encircled with the solid blue curve in Fig. 1(f), would indicate a correlation between dislocation sources and triple junctions of GBs. Therefore, some kinds of triple junctions of GBs would act as generation sources of dislocation clusters, and about 20% of dislocation clusters would be generated at the triple junctions.

Dislocation clusters are generated at triple junctions of CSL GBs, like a triple junction of $\Sigma_3 - \Sigma_3 - \Sigma_9$ GBs (shown in Fig. S1 in the supplementary data, available online at stacks.iop.org/APEX/13/105505/mmedia), as reported. Furthermore, we have found that triple junctions of RAGBs can also act as a dislocation source during the cast growth. Figure 2 shows the generation process of a dislocation cluster nearby a triple junction of RAGBs. Those GBs expand nearly parallel to the growth direction, and each segment of the GBs is therefore observed at the similar position in each figure. Meanwhile, in the Grain 1, a sub-grain composed of multiple twins of $\Sigma_3\{111\}$-type expands toward the different direction; the expansion direction is inclined by about 50° with respect to the growth direction. It seems that dislocations spread radially in the Grain 3 from the GB segment interacted with the multiple twins, suggesting that the interacted GB segment would act as a dislocation source. Especially, when the multiple twins interact with a triple junction of RAGBs, a huge number of dislocations would be generated at the junction [Fig. 2(d)]. Similar explosive generation of dislocations is observed at another triple junction of RAGBs interacted with multiple $\Sigma_3\{111\}$ GBs (shown in Fig. S2 in the supplementary data). Thus, triple junctions of RAGBs would act as a generation source of dislocation clusters when they are interacted with multiple $\Sigma_3\{111\}$ GBs.

Microstructure of the GB segments acting as a dislocation source is examined by SEM and EBSD in a triple junction of RAGBs (Fig. 3). In Fig. 3, a segment of the RAGB named RAGB(3) is altered to a different kind of RAGB named RAGB(4) via the connection with a pair of $\Sigma_3\{111\}$ GBs. RAGB(3) and RAGB(4) are observed as slopes on the etched surface. The intersection of the slopes with the bottom surface is obviously zigzag, and each corner of the
intersection is connected with a \( \Sigma 3 \{111\} \) GB. Meanwhile, the intersection of the slopes with the top surface is rather close to straight. This conflict morphology presumably reflects an inhomogeneous distribution of residual strains around the connections, that would determine the etching rate, and the quasi-linear morphology at the top surface would be due to a small strain relaxed at the surface before etching. A number of dislocations including small-angle GBs (SAGBs) (i.e. arrays of dislocations arranged at similar intervals) would be generated nearby a junction of \( \Sigma 3 \{111\} \) GB with RAGB(4).

Since GBs induce phonon scattering depending on strains around the GBs \( ^{34} \) and on their GB characters, \( ^{35} \) due to the deterioration of crystallinity, the thermal conductivity at the GBs would be reduced. \( ^{36} \) Therefore, GBs nearby the solid-liquid interface can locally modify the temperature distribution during the cast growth process, and this modification would induce a strain around the GBs. Ouaddar has reported that, no local strain is accumulated at \( \Sigma 3 \{111\} \) GBs except at their terminations, while a local strain is accumulated at GBs with \( \Sigma > 3 \) during the crystal growth; the degree of the strain would increase with increasing the \( \Sigma \) number. \( ^{27} \) According to the CSL theory, \( ^{25} \) when a GB is connected with a pair of \( \Sigma 3 \) GBs, the \( \Sigma \)-number of the GB segment between the connected lines triples and the area nearby the segment would be distorted during the cast growth. Similarly, the vicinity of a junction of GBs would be distorted, due to their low symmetry and non-coherency, as observed in a junction of CSL GBs. \( ^{28} \) Therefore, RAGB(4), especially nearby a junction with a \( \Sigma 3 \{111\} \) GB, would be distorted in comparison with the neighboring RAGB(3)s. Indeed, the etching rate nearby the upper intersection of a RAGB(4) with a \( \Sigma 3 \{111\} \) GB in Fig. 3(b) is higher in comparison with the neighboring RAGB(3)s, presumably due to a larger strain around the intersection. Moreover, when an intersection is close to a triple junction of RAGBs with a low symmetry and non-coherency, strains would be accumulated more nearby the intersection. Such the intersection can induce a large local strain that would be responsible for the emission of dislocations, like \( \Sigma 9 \) and \( \Sigma 27 \) GBs interacted with \( \Sigma 3 \) GBs. \( ^{17,24} \) Indeed, the intersection of RAGB(4) with \( \Sigma 3 \{111\} \) GB close to a triple junction of RAGBs would generate a much larger number of dislocations in comparison with the other.
intersection. Thus, a triple junction of GBs interacted with multiple $\Sigma 3\{111\}$ GBs, at which a huge local strain would be accumulated during the crystal growth, can act as an effective generation source of dislocation clusters. According to the model, dislocations with the same Burgers vector would be generated, and most of the dislocations in Fig. 3 have actually the same Burgers vector (Fig. 4).

We have found the generation of huge number of dislocations nearby triple junctions of RAGBs, and proposed that the key of the explosive generation is the interaction of the junctions with $\Sigma 3\{111\}$ GBs. Even though multiple $\Sigma 3\{111\}$ GBs are frequently nucleated at GBs during the cast growth, they do not influence the photovoltaic properties, since they induce no deep level and have no segregation ability for impurity atoms. However, when they interact with RAGBs, as well as with CSL GBs, their terminations would introduce a number of harmful dislocations, acting as effective segregation sites for light-elements and metal impurities. Hence, even though the density of $\Sigma 3\{111\}$ GBs seems to be independent of the density of dislocation clusters, generation and distribution of $\Sigma 3\{111\}$ GBs should be considered to design cast-grown Si ingots with an optimum dislocation density.

In conclusion, we have examined 3D distribution of GBs and generation sources of dislocation clusters in a HP mc-Si ingot, and found some triple junctions of RAGBs acting as a generation source of dislocation clusters. Although RAGBs with the non-coherent nature would not induce large thermal strain that is responsible for dislocation generation, they exhibit explosive generation of dislocations when they are interacted with multiple $\Sigma 3\{111\}$ GBs. It is suggested that a segment of the RAGB connected with a pair of $\Sigma 3\{111\}$ GBs nearby the triple junction would act as an effective dislocation source.

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