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Crystal growth mechanisms of silicon during melt growth processes

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1. Introduction

The expectations for solar cells have been increasing yearly toward solving energy and environmental problems worldwide. The multicrystalline Si (mc-Si) is one of the most important materials along with the Si single crystal (sc-Si) for the substrate of solar cells in the future, although other materials are being developed. The crystal structure of an mc-Si ingot obtained by casting is markedly different from that of sc-Si, as illustrated by the formation of grain boundaries and the distribution of crystallographic orientations. Various types of defect included in an mc-Si ingot, such as grain boundaries, dislocations, sub-grain boundaries, and metallic impurities, affect the properties of solar cells. Therefore, there is increasing importance to control the macro- and micro-structures of mc-Si ingots. In this study, the melt growth mechanisms of Si will be discussed, because fundamental understanding of crystal growth mechanisms is crucial to developing a technology for growing high-quality mc-Si ingot. The crystal growth phenomena during melt growth processes including the faceted dendrite growth and morphological transformation of crystal-melt interfaces will be discussed.

2. Faceted dendrite growth

Dendrite of faceted materials, so-called ‘faceted dendrite’, were discovered in the 1950s [1]. Si-faceted dendrites have unique crystal structures. The surface of the faceted dendrite is bounded by {111} habit planes, and at least two parallel twins exist at the center of the faceted dendrite. It is also known that the preferential growth direction of faceted dendrites is <112> or <110>. Such features can be applied in technologies for growing mc- Si ingots for solar cells, as illustrated in Fig. 1 [2].

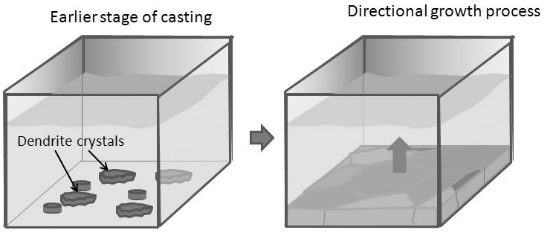

Fig. 1 Dendritic casting method.

We succeeded in directly observing the growth processes of Si-faceted dendrites by using an *in situ* observation system, which consisted of a furnace and a microscope [3].

Figure 2 shows a <110> and a <112> dendrite growing from a part of a faceted crystal-melt interface [4]. The shape of the tip of the growing dendrite is markedly different between the <110> dendrite and <112> dendrite. While the tip of the <112> dendrite becomes wider during growth, that of the <110> dendrite remains narrow during growth.

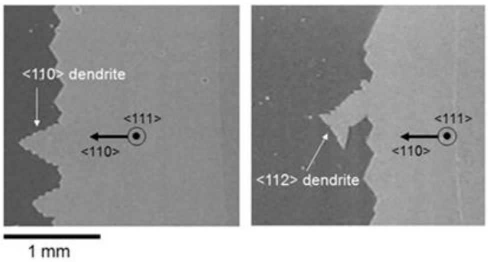

Fig. 2 Shape of the Si <110> and <112> dendrites.

We presented a growth model of <112> and <110> dendrites based on our experimental evidence [3,4], and have developed equations for the theoretical growth velocities of a Si <112> and <110> faceted dendrites on the basis of the growth model [5]. The theoretical growth velocities are described as

$$V_{<112>} = \frac{h}{2\,(h/V_1 + d/V_2)} + V_2 \qquad (1)$$

$$V_{<110>} = \frac{h}{\sqrt{3}\,(h/V_1 + d/V_2)} + V_2 \qquad (2)$$

where h is the height of the triangular corner, d is the twin spacing, V_1 is the growth velocity at the reentrant corner, and V_2 is the growth velocity on {111} plane. It is found that the growth velocity of the dendrite is inversely proportional to the twin spacing. We also investigated the dependence of the growth velocity of faceted dendrite on undercooling [6]. The growth velocity increased linearly as the increasing undercooling. We found that the relationship between the growth velocity and undercooling is most sensitive to the twin spacing. In addition, it was found that we can control the preferential growth direction of a dendrite by controlling the undercooling [7].

3. Morphological transformation of crystal/melt interfaces

To control the morphology of the crystal-melt interface during unidirectional growth processes is crucial to obtaining high-quality crystals because it affects the macro- and micro-structures and eventually the mechanical, optical, and electrical properties of materials.

Figure 3 shows the Si (100) crystal-melt interface whose growth velocity was 162 μm/s [8]. The morphology of the interface transformed from planar to zigzag facets during the growth. It was shown that a wavelike perturbation is introduced into a planar interface, the perturbation is amplified, and the zigzag facets are formed finally.

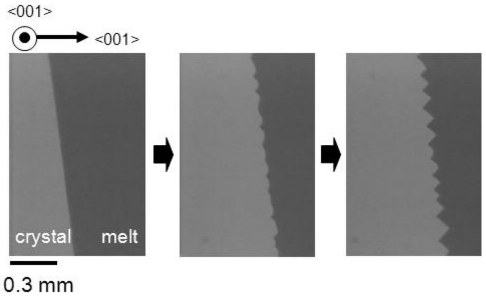

Fig. 3 Morphological transformation of Si (100) crystal-melt interface moving at 162 μm/s [8].

A similar morphological transformation from planar to zigzag facets of the moving interface was observed at the Si (112) and (110) crystal-melt interfaces at higher growth velocities [9]. On the other hand, when the growth velocity was lower, the planar interfaces were maintained throughout the crystallization. Generally, a crystal-melt interface becomes unstable, leading to the amplification of the perturbation, when the temperature gradient at the interface is negative along the growth direction [10]. We considered that the latent heat of crystallization increases the temperature at the crystal-melt interface, and that the temperature gradient in the Si melt at the interface becomes negative when growth velocity is high, because the amount of generated latent heat per unit time increases with growth velocity. The thermal fields of the Si crystal and melt during crystal growth for various growth velocities were calculated, as shown in Fig. 4 [8]. The temperature gradient in the Si melt at the interface changes from positive to negative as growth velocity increases. When the growth velocity is low, the temperature gradient is positive, this means that the interface is stable and the planar interface is maintained. On the other hand, when the growth velocity exceeds its critical value, the temperature gradient in the Si melt at the interface changes from positive to negative, and thus the perturbation introduced into the planar interface is amplified, forming zigzag facets at high growth velocities.

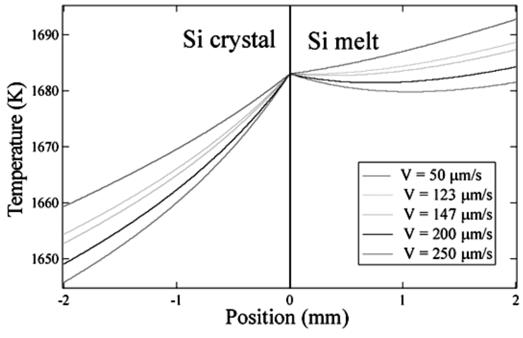

Fig. 4 Calculated thermal fields of Si crystal and melt during crystallization for V = 50, 123, 147, 200 and 250 μm/s. [8].

We found that twin boundary is generated on the {111} facet plane, when the interface become zigzag facets [11]. We also found that impurities segregated at the valleys of zigzag facets during unidirectional growth [12]. Therefore, we have to control the interfacial morphology during unidirectional growth by controlling temperature gradient and/or growth velocity to obtain a structure-controlled mc-Si ingot.

4. Summary

The Si-faceted dendrite growth and the morphological transformation of the crystal-melt interface were investigated by in situ observations. For the complete understanding of crystal growth mechanisms and the control of the macro- and micro-structures of mc-Si ingots, further data accumulation is required. It is expected that such ongoing studies will lead to the establishment of a technology for producing high-quality mc-Si ingots in the near future.

Acknowledgements

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