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Distributions of substitutional and interstitial impurities in silicon ingot with different grain morphologies

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ABSTRACT

A multicrystalline silicon ingot with columnar and irregular grains was obtained from metallurgical-grade silicon (MG-Si) by directional solidification. The segregation behaviors of substitutional and interstitial impurities in different grain morphologies have been studied. The concentration distribution of substitutional impurities (B and Al) in the silicon ingot was accord with the Scheil's equation, which depended on the grain morphology. However, the concentration distribution of interstitial impurities (Fe, Ti, Cu, and Ni) was only accord with the Scheil's equation under the columnar grains growth condition. The difference lattice sites of the impurities will result in the disparate segregation behavior of impurities for columnar and irregular grains growth, which leads to the diverse concentration distribution of substitutional and interstitial impurities in the silicon ingot. Furthermore, the transport mechanism of interstitial and substitutional impurities in front of the solid-liquid interface boundary has been revealed.

1. Introduction

Multicrystalline silicon has been widely used as a source of raw material for solar cells [1]. The impurities in silicon affect the electrical properties of solar cells such as resistivity, minority carrier lifetime and conversion efficiency [2]. According to the lattice site, impurities can be classified into interstitial impurity and substitutional impurity with different physical characteristics. For example, Al and B as substitutional impurities [3] are usually used to adjust the resistivity of silicon; Elements of 3d transition-metal (TM) such as Fe, Cu, Ni, and Ti [4–6] can reduce the minority carrier lifetime and conversion efficiency of crystal silicon solar cells as interstitial impurities [7].

Directional solidification technology is usually used to remove the impurities in silicon due to the low segregation coefficient they have [8,9]. The segregation behavior of impurities in silicon has been widely studied under the directional solidification process [10–12]. For example, Ma et al. have studied the segregation mechanism of metal impurities during vacuum directional solidification [12,13]. Luo et al. have revealed the redistribution of metal impurities and insoluble inclusions during directional solidification [14,15]. In our previous work, the effect of diffusion layer on the distribution of impurities has been discussed [16]. By now, the difference of the segregation behaviors between substitutional and interstitial impurities has not been revealed during the directional solidification process.

The silicon ingot usually includes irregular grains and columnar grains, in which the segregation behavior of impurities is different [17]. In this paper, a multicrystalline silicon ingot with columnar and irregular grains was obtained by directional solidification. The segregation behaviors of substitutional and interstitial impurities in different grain morphologies have been studied, respectively. The effect of grain morphology on the transport mechanism of impurities in solid-liquid interface is also discussed.

2. Experimental

Experiment was conducted in an industrial directional solidification furnace. Fig. 1 shows the schematic diagram of the experimental apparatus. It consisted of a vacuum system, a melting system, an insulation system and a heating exchange system. During the directional solidification process, the temperature slowly decreased, while the insulation system moved upwards and the quartz crucible remained stationary on the heat exchange block. The heat of molten silicon flowed only along the axial direction and is taken away by the heat exchange block so that the silicon crystal grows from the bottom to the top. In this experiment, the grain growth mode of the silicon ingot could be controlled by the temperature of heater.

The initial concentrations of impurities in the raw material were about 811.34 ppmw. Prior to processing by directional solidification

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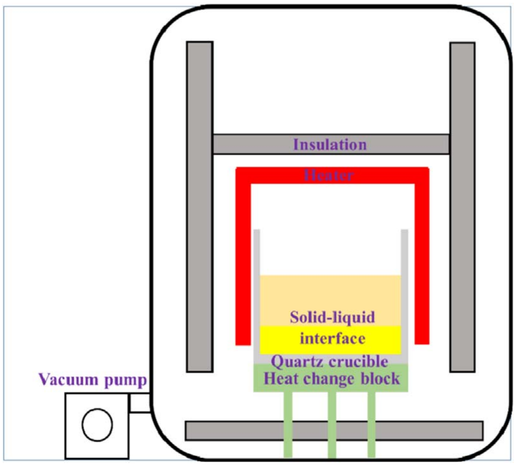

Fig. 1. Schematic diagram of the experimental apparatus.

under low vacuum condition, about 400 kg MG-Si was washed sufficiently with water to remove possible solid residues and extraneous impurities from the surface. Subsequently, it was placed in a quartz crucible with an inner dimensions of 800 × 800 × 480 mm, and then the chamber was full of flowing argon with a certain pressure of 6×10^4 Pa. Then the insulation was pulled upwards so that the speed of crystal growth is from 6×10^{-7} m/s to 4×10^{-6} m/s until the molten silicon solidified completely.

The silicon ingot which was obtained by a certain production process had a dimension of 800 × 800 × 240 mm, as shown in Fig. 2(a). The grain morphology was columnar in the middle of the silicon ingot and irregular in the bottom and top of the silicon ingot, as shown in Fig. 2(b). A block of silicon ingot was cut from the center region to observe the clear grain morphology and detect resistivity, which was outlined in black line, as shown in Fig. 2(a). These samples were cut from the silicon ingot by diamond saw to measure the concentration of impurities, which were outlined in purple line. The impurity concentration of each sample was determined by inductively coupled plasma mass spectrometer (ICP-MS).

3. Result and discussion

3.1. Grain morphology

The macro grain morphology of the sample along the direction of crystal growth is shown in Fig. 3(a). The silicon ingot consists of irregular grains and columnar grains, which are outlined in white line. The boundaries are based on limit position of the columnar grains. The boundary at 150 cm is the maximum of the columnar grains. The boundary at 60 cm is the minimum of the columnar grains. The columnar grains belong to the middle area of the silicon ingot, while the irregular grains belong to the bottom and top areas of the silicon ingot. The distribution range of the columnar grains is between 60 mm and 150 mm of the ingot height. Usually, the shape of the solid-liquid interface can be estimated from the distribution of the macro grain morphology [18]. During the columnar grains growth process, the shape of the solid-liquid interface is planar and steady, in which there are a few grooves which are introduced by the columnar grain boundaries. However, the solid-liquid interface with irregular grains growth is not a planar, in which there are two interface boundaries (the interface boundary of solid and mushy zone and the interface boundary of liquid and mushy zone) [9]. The shape of the solid-liquid interface is unsteady, because the mushy zone introduces a large number of fine grains in front of the solid-liquid interface boundary.

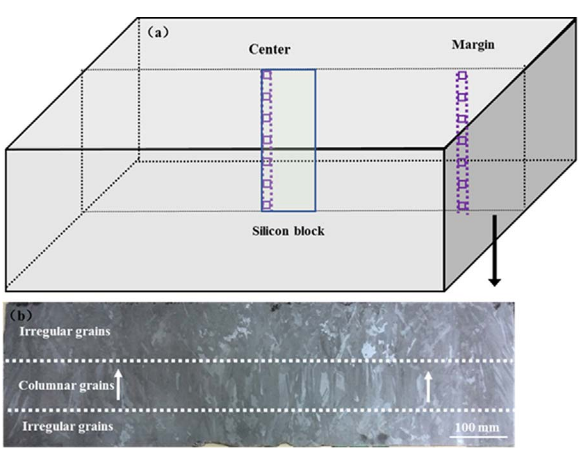

Fig. 2. (a) Schematic diagram of sampling and (b) longitudinal section of silicon ingot.

At the beginning of directional solidification process, a large number of crystal nuclei grow up on the bottom of crucible, which is shown as red dotted line in Fig. 3(a). The grains on the crucible should be regarded as columnar grains. The appearance of mushy zone hinders the growth of columnar grains and leads the growth of irregular grains. The mushy zone begins to disappear slowly, which leads to the growth of the columnar crystal upward. At the end of the directional solidification, the mushy zone begins to emerge once again so that the irregular grains can grow up in this region [17].

The solid-liquid interface for columnar grains growth is planar , which can be defined as a line perpendicular to the direction of crystal growth. However, the solid-liquid interface for irregular grain growth is rugged and unsteady due to the effect of mushy zone. The different structure of solid-liquid interface boundary will impact the segregation behavior of impurities during the directional solidification process.

The corresponding resistivity distribution of the silicon ingot is shown in Fig. 3(b). The average resistivity of the irregular grains area is about 0.052 Ω cm. The average resistivity of the columnar grains area is about 0.076 Ω cm. The resistivity is mainly affected by the impurities and grain boundary in silicon. Grain boundaries can increase the resistivity while impurities can reduce the resistivity. There are a large number of grain boundaries in the irregular grains area. So the resistivity of the irregular grains area should be higher than that of the columnar grains area. However, the resistivity of the irregular grains area is lower than that of the columnar grains area. There is a certain relationship between concentration and grains morphology. The concentration of impurities in the silicon is measured by ICP-MS for further study.

3.2. Concentration distribution of impurities

The concentrations of impurities in the edge and center areas of the silicon ingot are measured along the crystal growth direction. The result is shown in Table 1. The concentrations of B and Al are uniform in the bottom and middle of the silicon ingot. The average concentrations are about 0.17 ppmw and 0.23 ppmw, respectively. They are up to 0.30 ppmw and 3.39 ppmw in the height of 245 mm, respectively.

It can be concluded that the concentrations of B and Al increase with the increase of the silicon ingot height. As a result, the growth process of the silicon ingot can be divided into two areas, which consists of the segregation area of impurities in the ingot bottom and middle and the enrichment area of impurities in the ingot top. Although the segregation coefficient of B is large than other impurities, it can also be removed by directional solidification in a certain extent. Al can be clearly removed by directional solidification. The concentration distribution of B and Al is not depended on the grain morphology.

The concentration distribution discipline of the four interstitial metal impurities is similar with each other in the whole silicon ingot. At the beginning of directional solidification, the average concentrations of Fe, Ti, Cu and Ni near the bottom of crucible area are 3.08 ppmw, 0.16 ppmw, 0.69 ppmw and 1.57 ppmw, respectively. The concentra-

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