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Effect of alternating magnetic field on the removal of metal impurities in silicon ingot by directional solidification

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ABSTRACT

Multicrystalline silicon ingots without and with alternating magnetic field during directional solidification process under industrial system were obtained from metallurgical grade silicon (MG-Si). The concentrations and normalized concentrations of metal impurities in the two silicon ingots were studied. The result shows that the concentrations and normalized concentrations in high-purity area of the silicon with alternating magnetic field are lower than those of the ingot without alternating magnetic field. The transport mechanism for metal atoms in the diffusion layer area has been changed due to the alternating magnetic field. Alternating magnetic field introduces a convection to reduce the thickness of diffusion layer in the molten silicon, which results in a decreased effective segregation coefficients. Enhancing transport driving force of metal atoms in molten silicon is the effective way to improve the removal rate of metal impurities.

1. Introduction

So far, cast multicrystalline silicon has been the main photovoltaic materials for solar cell [1]. Concentrations of metal impurities in the solar grade silicon (SoG-Si) play a crucial rule on the performance of solar cell [2]. SoG-Si prepared directly from MG-Si by metallurgical route, attracts more and more attentions due to its characteristics of low cost, low energy consumption and environment friendly [3]. Metal impurities are removed by directional solidification because of their low segregation coefficients [4,5], which is an extensive method in the metallurgical route [6]. The effective segregation coefficients of metal impurities can be influenced by many factors during the directional solidification, such as alternating magnetic field, the crystal growth rate and vacuum condition [7–9].

Magnetic field has been introduced to control the flow of molten silicon during the direction solidification process [10–14]. For example, Sun et al. [15] have obtained a silicon ingot with low metal impurities by directional solidification under the alternating magnitude field condition. Dadzis et al. [16] have revealed that the unsteady segregation process of carbon and oxygen impurities exhibits a non-uniform concentration distribution under magnetic field condition. Liu et al. [17] have revealed the effects of different types of traveling magnetic field on the molten silicon mixing and the solid–liquid interface shape at different directional solidification stages. The above mentioned results indicate that magnetic fields have an important role on the shape of the solid–liquid interface and the vortex flow of molten silicon during the directional solidification process under laboratory system. Furthermore, Kudla et al. [18] have studied the effect of magnetic field on the distribution of SiC and Si_3N_4 in silicon ingot under industrial system. However, the research about the effect of magnetic field on the removal of metal impurities under industrial system has not been reported. Moreover, the influence of magnetic field on the transport and distribution mechanism of metal impurities during directional solidification process under industrial system has not been revealed.

In this paper, silicon ingots with and without magnetic field during directional solidification process under industrial system were obtained. The effects of alternating magnetic field on metal impurities concentration during directional solidification process were also revealed.

2. Experiment

Silicon ingots were prepared by the industrial directional solidification furnaces, which were schematically shown in Fig. 1. The main difference of apparatus is the heating mode. In Fig. 1(a), the

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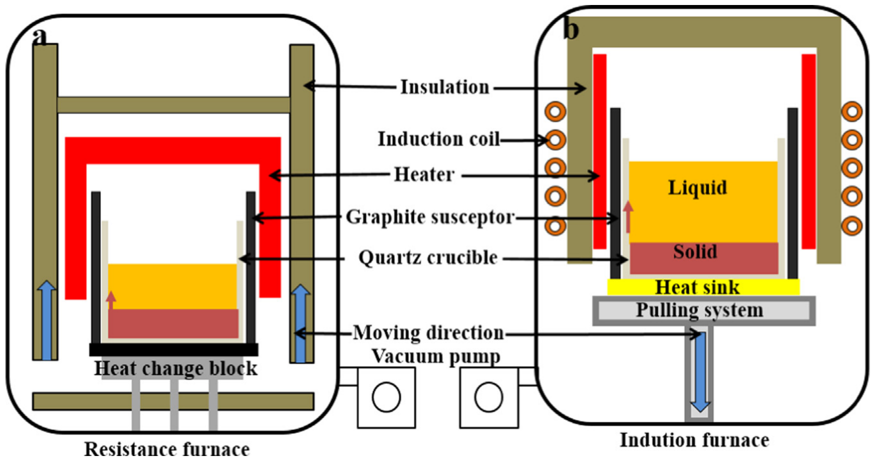

Fig. 1. Schematic diagram of the experimental apparatus (a) without and (b) with alternating magnetic field.

heating mode of experimental apparatus without magnetic field is resistance heating. It consists of vacuum system, melting system, insulation system, heating exchange system. During the directional solidification process, the temperature slowly decreases while the insulation system moves upwards and the quartz crucible remains stationary on the heat exchange block. Then the heat in molten silicon flows only along axial direction and is taken away by the heat exchange block, so the silicon crystal grows from the bottom to the top.

In Fig. 1(b), the apparatus also consists of vacuum system, melting system, insulation system, pulling system and cooling system. However, the heating mode of experimental apparatus with magnetic field is the graphite induction heating. The induction coil in the experimental apparatus can create alternating magnetic field under an applied alternating current condition. The heater which is graphite can provide huge power to melt silicon under the alternating magnetic field condition. The graphite susceptor is cut into several pieces to avoid restraining the magnetic field. Magnetic field strength of the melting system is up about 4×10^3 A m from the numerical simulation result. The insulation system is used to prevent the heat flowing across the crucible wall, eliminate the radial temperature gradient. During the directional solidification process, the quartz crucible moves downwards with the heat sink while the heating power remains stationary. Then the heat in molten silicon flows only along axial direction and is taken by the cooling system so that the silicon crystal grows from the bottom to the top.

In this industrial production, two silicon ingots were obtained by the two apparatuses under the same average crystal growth rate condition. MG-Si with an initial purity of 99.8% were used in this industrial production. Firstly, they were placed in two Si_3N_4 coated quartz crucibles and then the chamber was full of flowing argon gas with a certain pressure of 6×10^4 Pa. Si_3N_4 powder were sprayed on the both quartz crucible under the same process condition. The size of quartz crucible for silicon ingot with alternating magnetic field is $\phi 810 \times 655$ mm, which loads about 300 kg feedstock. The size of quartz crucible of silicon ingot without alternating magnetic field is $840 \times 840 \times 480$ mm^3, which loads about 350 kg feedstock. The morphology of silicon raw material is irregular block, and they were simply listed inside the crucible. Secondly, the two silicon ingots with and without alternating magnetic were melted by heat power and maintain melt state for about an hour. Lastly, the whole crucible with alternating magnetic field is in the coil inner during silicon melting process. The crucible on the pulling system moved down during directional solidification process. The rate of the pulling system was slow and adjustable. The rate of the crystal growth was measured by a quartz rod and controlled by setting temperature. The rate of pulling system was slower than the rate of the crystal growth during directional solidification process. However, the silicon without alternating magnetic field realize the directional solidification by moving the insulation system. The silicon crystal with and without alternating magnetic grew with an average rate of 2×10^{-6} m/s until the molten silicon solidified completely.

The solid–liquid interface shape during growth of the two silicon ingots is slightly convex. However, it can be seen as a flat in the center of the two silicon ingots. So the shape of solid–liquid interface in the center of the two silicon ingots is the same. The silicon chips were cut in the center of the two silicon ingots, which parallel along the growth direction. Some small silicon blocks were cut in different heights of the two silicon chips as sample. The metal impurities concentrations of each sample were determined by Inductively Coupled Plasma Mass Spectrometer (ICP-MS).

3. Results

3.1. The concentration of Fe, Ni, Ti and Cu

The concentration of metal impurities is less than 5 ppmw after the directional solidification process. The metal impurities concentrations of the feedstock for the silicon ingots without and with alternating magnetic field are different, as shown in Fig. 2. The metal impurities concentrations of the samples in different positions of the silicon ingots without and with alternating magnetic field are shown in Fig. 2. Compared with those in the raw material, most of the impurities are removed into the top of the two silicon ingots. There is a high-purity area in the bottom and middle of the two silicon ingots. The metal impurities concentration of the high-impurity area is less than 5 ppmw. The percentage of the high-purity area in the silicon ingot with alternating magnetic field is about 87.9%, which is higher than that in the silicon ingot without alternating magnetic field.

The average concentrations of Fe, Ni, Ti and Cu in the high-purity area in the silicon ingot without alternating magnetic field are shown in the Table 1. The concentrations of Fe, Ni, Ti and Cu in the silicon ingot with alternating magnetic field are significantly lower than those in the silicon ingot without alternating magnetic field in the high-purity area. The average concentrations of Fe, Ni, Ti and Cu in the silicon ingot without alternating magnetic field are about 1.66, 0.22, 0.14 and 0.18 ppmw in the high-purity area, respectively. However, the average concentrations of Fe, Ni, Ti and Cu in the silicon ingot with alternating magnetic field are decreased to 0.42, 0.02, 0.08 and 0.02 ppmw, respectively.