G02:Bulk crystal growth

Growth of Si large single bulk crystals for solar cells using small crucibles with a given diameter by the Noncontact Crucible method

Kazuo Nakajima[1]*, Kohei Morishita[2], and Ryota Murai[3]

1-3. Graduate School of Energy Science, Kyoto University, Yoshida-honmachi, Sakyo-ku, Kyoto 606-8501, Japan, 1. +81-75-753-9107/+81-075-753-9177/nakasisc@energy.kyoto-u.ac.jp,

2. k-morishita@energy.kyoto-u.ac.jp,

3. murai.ryota.82r@st.kyoto-u.ac.jp,

Abstract

We proposed the noncontact crucible method for reducing stress in Si bulk crystals [1, 2]. In this method, a Si melt has a large low-temperature region in its upper central part and a crystal grows in the region without contacting with the crucible wall. In order to confirm whether high-quality Si single bulk crystals with a large diameter can be obtained by controlling the low-temperature region, we prepared p- and n-type Si single bulk crystals using small crucibles with a given diameter. We obtained single crystals with a diameter as large as 60 % of the crucible diameter. The etch pit density of the Si single bulk crystals was 1.9×10^3 -1.2×10^4/cm^2, and the O concentration of the present ingots was relatively lower than that of the ingots grown by the CZ method.

1. Introduction

Generally, conventional crystal growth methods using silica crucibles cannot effectively control Si expansion due to Si melt solidification because silica crucibles have insufficient flexibility to reduce the stress. We proposed a noncontact crucible method for reducing stress in Si bulk crystals [1, 2]. In this method, a Si melt has a low-temperature region in its upper central part for natural Si crystal growth inside it. Nucleation occurs on the surface of the Si melt using a seed crystal, and a crystal grows inside the melt without touching the crucible wall. Then, the crystal continues to grow while being slowly pulled upward to ensure that the crystal growth remains in the low-temperature region. Therefore, the bottom of ingots was convex in the growth direction as shown in Fig. 1, which compares the present method with the CZ method. For the CZ growth, an ingot is grown above the surface of a Si melt and the solid-liquid interface of the ingot is generally concave in the growth direction.

Si single bulk crystals used for solar cells are mainly grown by the Czochralski (CZ) method. Generally, the diameter of the ingots grown by the CZ method was controlled within 30 % of the crucible diameter. In the present method, the crystal diameter was determined by the size of the low temperature region, and the method has a possibility of growing large ingots even using a small crucible with a given diameter. Therefore, it is very important to confirm whether a Si single crystal can be grown inside a Si melt without touching the crucible wall.

In this work, we used the noncontact crucible method to prepare p- and n-type Si single bulk crystals. The quality of the ingots was determined in terms of minority carrier lifetime, dislocation distribution and crystal structure. The diameter of ingots was investigated for many crucibles with and without coating Si_3N_4 particles as a function of temperature reduction.

2. Experiments

A large low-temperature region was formed in a Si melt by controlling the temperature distribution in the furnace. A Si crystal grew naturally from the seed crystal along the surface of and inside the Si melt, then continued to grow while being slowly pulled upward. The temperature reduction used for the growth was defined as the difference between the starting temperature and the final temperature. P- and n-type Si ingots were grown using silica crucibles of 30 and 33 cm diameters. The inner wall of a crucible for the growth of p-type Si multicrystals was coated with Si_3N_4 particles to prevent the oxidation of the ingots during crystal growth. The inner wall of crucibles for the growth of p- and n-type Si single bulk crystals was not coated with Si_3N_4 particles to prevent floating Si_3N_4 particles on the surface of Si melts during crystal growth.

To evaluate the quality of the ingots, minority carrier lifetime was measured by microwave photoconductive decay (PCD). The oxygen (O) and carbon (C) concentrations of the ingots were measured by Fourier transform infrared spectrometry (FTIR).

3. Results and Discussion

We can prepare an n-type Si single bulk crystal with a diameter of 20 cm even using a small crucible with a diameter of 33 cm as shown in Fig. 2. The temperature reduction used for the growth of this ingot was 45 °C. The surface structure exhibits a four-cornered pattern on its top surface. The surface orientation of the cross section was (100). The crystal diameter was as large as 60 % of the crucible diameter. For the growth of p-type Si multicrystals using crucibles with coating Si_3N_4 particles, we obtained the diameter of 26.8 cm and the crystal diameter as large as 81 % of the crucible diameter. The larger temperature reduction was required for the growth using crucibles without coating Si_3N_4 particles than for the growth using crucibles with coating Si_3N_4 particles to obtain ingots with the same diameter. The etch pit density of the n-type Si single bulk crystals was 2.0-3.2 x 10^4/cm^2 and that of the p-type Si single bulk crystals was 1.9 x 10^3 -1.2 x 10^4/cm^2 even using necking grown seeds of only 4.2-6.5 cm long. The O concentration of the present ingots was relatively lower than that of the ingots grown by the CZ method because the convection in Si melts may be suppressed by making a large low-temperature region [3]. The average minority carrier lifetime of an entire wafer cut from an n-type ingot was 83 μs (maxmum:165μs), which was much higher than that of p-type ingots [3].

4. Conclusions

It was confirmed that n- and p-type Si single bulk crystals can be grown using crucibles without coating Si_3N_4 particles by the noncontact crucible method. The surface orientation of the cross section was (100) for the ingots. Using crucibles without coating $Si_3N_{4,}$ we have obtained the crystal diameter as large as 72 and 81 % of the crucible diameter for a single bulk crystal and a multicrystal, respectively. The growth of Si ingots with a larger diameter using a crucible with a given diameter is a large merit for this method. It is effective to prepare Si single bulk crystals with a large diameter by the low cost method. The distribution of minority carrier lifetime was uniform in almost the entire cross section of the single bulk crystals except in the ring-shaped region. The dislocation density was 1.9 x 10^3/cm^2 in the cross section of a single bulk crystal except at the periphery of the cross section. The average minority carrier lifetime of a wafer cut from an n-type ingot was 82.8μs for the passivated surface, which was higher than those (7.3-16.0 μs) of p-type wafers. The O and C concentrations of the present ingots which were grown using crucibles without coating Si_3N_4 particles were 2.5-8.3 x 10^{17}/cm^3 and about 1 x 10^{16}/cm^3, respectively. The O concentration is relatively lower than that of the ingots grown by the CZ method.

References

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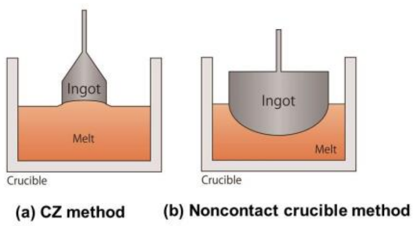

Fig. 1 Comparison between (a) the CZ method and (b) the present method

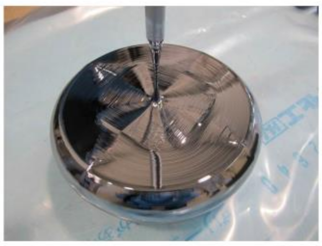

Fig. 2 N-type single crystal grown by the present method