Short Communication

Removal of silicon carbide from kerf loss slurry by Al–Si alloying process

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A B S T R A C T

A novel method to separate silicon and silicon carbide from kerf loss slurry by Al–Si alloying process has been reported in this paper. The kerf loss slurry was washed and dried, and then aluminum was added on the top of these dry powders with silicon and silicon carbide. The Al–Si alloying process was performed in argon atmosphere using a vacuum carbon tube furnace at 1773 K. In this way, an Al–Si ingot was obtained, on the surface of which a lot of hexagonal crystals were observed. The Al–Si ingot was characterized by X-ray diffraction, scanning electron microscopy, X-ray fluorescence and electron probe micro-analyzer. The X-ray results indicated that the Al4C3 phase was obtained on the top of the cast. The scanning electron microscopy, X-ray diffraction and electron probe micro-analyzer results revealed that the Al–Si alloy without silicon carbide phase formed in the cast, which indicated that silicon and silicon carbide can be separated from slurry by this alloying process.

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1. Introduction

The crystalline silicon material, which is mainly produced from the energy-intensive Siemens process [1], has been found an increasing number of applications in the photovoltaic (PV) industry. During silicon wafer manufacturing, the silicon crystal is sliced by a multi-wire saw with an ethylene glycol or glycerol solution containing silicon carbide (SiC) abrasives. Therefore, more than 30 wt.% of silicon goes into slurry waste due to kerf loss during this processing [2–4]. With the reduction of wafer thickness, the loss of silicon material would be increased further [5]. Consisted of fine silicon particles, silicon carbide (SiC) particles, metal impurities from cutting wire, lubricating oil (poly-ethylene glycol), and the additives for better particle suspension [6], the kerf loss slurry waste has recently attracted attention on the separation of the components due to the high purity silicon material [7]. Several purification methods have been developed for the removal of SiC including centrifugation [8,9], magnetization [10], filtration [11–13], directional solidification [4,14], phase-transfer separation [15], acid treatment and electrokinetic separation [16], and froth flotation technologies [17,18]. However, the high-gravity centrifugation is not an effective separating method because of the small particle sizes of the Si and SiC. On the other hand, SiC is too stable to be removed by acid leaching.

In this paper, we report a novel approach to remove SiC from the slurry by solidification refining with Al addition; furthermore, high purity silicon can be recovered from Al–Si alloy.

2. Experimental

The kerf loss slurry waste, which was comprised of Si, SiC, metal fragments and glycol solution, was provided by a semiconductor company. After the glycol solution and metal fragments were removed by acetone and nitric acid leaching, respectively, the dried powders containing 74 wt.% Si and 26 wt.% SiC were applied as the starting material.

During the Al–Si alloying process, aluminum powders were placed on the dried powder in a quartz crucible, which was heated at 1773 K for 1 h in argon atmosphere with a vacuum carbon tube furnace. And an Al–Si ingot was prepared. As shown in Fig. 1b, a small sample with 1 × 0.8 × 0.6 mm was taken out from the central part of the ingot, followed by polishing the bottom of sample with 1 μm diamond paste. And then, the surface and bottom of the sample were characterized by XRD(D8 Discover, German), SEM(S-4800, Japan), XRF(XRF-1800, Japan), EPMA(EPMA-1600, Japan), respectively.

3. Results and discussion

As shown in Fig. 1, Al–Si ingot (Fig. 1a) can be obtained after the Al–Si alloying process and a lot of hexagonal crystals with yellow-gold color were observed on the top surface of it (Fig. 1b). The SEM images of the top sample surface indicate that the diameter of hexagonal crystallizes was in the range of 0.1–2 mm (Fig. 1c). As shown in Fig. 1d, the bottom of sample after the polishing and

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1 For interpretation of color in Fig. 1, the reader is referred to the web version of this article.
acid etching was investigated. The energy dispersive spectroscopy (EDS) proves that the ingot was consisted of silicon and aluminum and the carbon element can not be observed in the bottom of sample.

The XRD patterns of raw powders and Al–Si ingot are shown in Fig. 2 and the phase constitutions can be clearly confirmed by the results of XRD analysis. The major peaks of the raw powders are in agreement with those of the JCPDS cards Si (No. 27-1402) and SiC (No. 29-1131), respectively. But no diffraction peaks of silicon carbide were observed in the XRD pattern of the sample (neither on the top nor on the bottom), which illustrates that the silicon carbide had been completely removed after the Al–Si alloying process.

It is noted that the intermediate phase Al₄C₃ is detected on the top surface of Al–Si ingot, which is attributed to the formation of Al₄C₃ with the hexagonal crystalline structure via the following reaction [19,20]:

\[
4\text{Al} + 3\text{SiC} \rightarrow \text{Al₄C₃} + 3\text{Si}
\]

The above is consistent with the result observed by EPMA. However, Al₄C₃ diffraction peaks were not detected on the bottom of sample, which indicates that the Al₄C₃ phase was synthesized on the surface of Al–Si alloy ingot.

EPMA was used to describe the cross section composition of the interface between Al–Si alloy ingot and raw powders and the results are shown in Fig. 3. A mixed layer with thickness of about 200 µm was observed and the EDS analysis reports that the composition of mixed layer were Al–Si alloy and Al₂O₃. The corresponding element maps of Al, Si and C implied that the solidified structure consisted of piled Si needle-like crystals and Al–Si eutectic intergrains. Meanwhile, the C element was not detected in the Al–Si ingot, and it proves that the SiC had been removed in the Al–Si alloy. This result was also determined by the XRF analysis.

Table 1 shows a typical XRF result about the elemental concentration of the raw material and sample. The results clearly demonstrated that the C element was completely removed after the Al–Si alloying process. But due to the formation of Al₄C₃ phase, the C element can be detected on the top surface of the ingot and it disappeared after polishing treatment. On the other hand, we also find that the microstructure of the sample top is Al–Si hypoeutectic, and that of the sample bottom is Al–Si hypereutectic. It indicates that the liquid aluminum may “absorb” silicon from the kerf loss slurry step by step and form the Al–Si alloys. And Si grains can be found on the bottom of sample.

4. Conclusions

Conclusively speaking, an Al–Si ingot was successfully prepared by the kerf loss slurry and Al. SiC was completely removed from the Al–Si alloy ingot by Al–Si alloying process at 1773 K in argon atmosphere. Meanwhile, Al₄C₃ phase with hexagonal structure and yellow-gold color was also formed on the top surface of the ingot. So Al–Si alloying process is a novel and simple method to recycle the kerf loss slurry and the solar grade Si can be obtained from this Al–Si alloy ingot by electrolysis or acid leaching.
Acknowledgments

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References


Table 1
Elemental concentration for the raw material and sample characterized by XRF.

<table>
<thead>
<tr>
<th>Elements</th>
<th>Raw material (%)</th>
<th>Top surface of sample (%)</th>
<th>Top surface without hexagonal crystal (%)</th>
<th>Bottom of the Sample (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>85.2332</td>
<td>11.6035</td>
<td>7.8971</td>
<td>14.2696</td>
</tr>
<tr>
<td>Al</td>
<td>-</td>
<td>79.0055</td>
<td>88.1863</td>
<td>84.9997</td>
</tr>
<tr>
<td>C</td>
<td>3.9703</td>
<td>9.1280</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Other</td>
<td>8.2402</td>
<td>0.2629</td>
<td>0.5166</td>
<td>0.7307</td>
</tr>
</tbody>
</table>

Fig. 3. Surface scanning of elements Al, Si and C on the interface between the ingot and rest powders by EPMA.