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Silicon and silicon carbide powders recycling technology from wire-saw cutting waste in slicing process of silicon ingots

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ABSTRACT

A study on recycling silicon and silicon carbide powders from wire-saw slurry containing silicon carbide, silicon kerfs, polyethylene glycol (PEG), and iron fragments in the wire-saw slicing silicon ingots process was conducted. Recycling technology was suggested. PEG from the slurry was removed using filter press, and iron particles from the slurry were removed using high gradient magnetic separator. To separate silicon carbide and silicon particles hydrocyclone assembly was used. Recovered silicon carbide powder (95 wt% SiC, 4 wt% Si and 1 wt% Fe) can be used again in the process of cutting silicon ingots. Obtained recovered powder with high Si content after additional purification further can be used as a feedstock together with the amorphous silicon for crystal growth by directional solidification in an induction furnace.

1. Introduction

The majority of PV (photovoltaic) cells are made of silicon, which is mainly produced from the energy-intensive Siemens process [1]. The wafer shares more than 65% of the cost for solar cells, but on the other hand, more than 40% of the high-purity silicon is wasted during wafer slicing. The silicon kerf loss is in the form of slurry mixed with the cutting fluid and abrasive silicon carbide particles, as well as the metals from the saw wire. Although the kerf loss silicon particles remain in high purity, recycling them from the slurry waste for their reuse has not yet been successful [2–4].

Recovery of SiC abrasive can be performed by several methods, including centrifugation [5], froth flotation technologies [6], through filtration [7]. Due to the larger particle size and lower acceptable purity of SiC, it is easier to recover SiC for recycling, the Si particles are much finer and more difficult to separate. Only a few articles have reported the recovery of silicon kerfs from the wire-saw slurry. Thus, the silicon kerfs in the former type slurry are more difficult to recover. It is known that the major challenge in recycling is the complete removal of small (<1 μm) SiC particles [4]. Kapur and Khanna [8] reported that the silicon particles might be recovered by combining flocculation and filtration techniques. Billiet and Nguyen [9] was granted a patent for a froth flotation method for the recovery of Si kerfs using particular surfactants as collector.

The aim of this study was to develop the process of recovering polyethylene glycol (PEG), silicon carbide and silicon kerfs from the sawing waste.

2. Experimental

In the present work, the sawing waste was obtained from the ukrainian manufacturer PJSC "Kvazar" (www.kvazar.com). For the preparation of the initial cutting suspension used original green silicon carbide (Table 1, powder 1) micropowder FEPA F 600 or F 500 (Shinano Electric Refining Co., Ltd., or other manufacturer) (Fig. 1). The initial sawing waste obtained after the sequential treatment of cutting slurry in two decanters was used as a feedstock. The sawing waste was washed in the water from the PEG and then the content of Si, SiC and metals in the dry powder was measured gravimetrically (powder samples weight was 1 g, laboratory balance AXIS AD100) after the sequential suspension treatment in hydrochloric acid and in an acid mixture containing HF and HNO_3. Other powders were analyzed similarly. The level of impurities in the powder 12 was measured by a glow discharge mass spectrometry (Model VG9000). The circularity and size measurements of particles before and after recycling of the slurry were carried out using the Sysmex FPIA-3000 Flow Particle Image

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Table 1
The composition and characterization of the powders.

No.	Powders	SiC (wt%)	Si (wt%)	Metals (mainly Fe) (wt%)	PEG (wt%)	Weight ratio	Circularity	Diameter (μm)
1	Original SiC powder	99	–	0.5	–	–	0.879	16.462
2	After the first decanter centrifuge	54	14	9	30	–		
3	After the second decanter centrifuge	34	33	10	23	–	0.930	13.535
							0.910[a]	14.316[a]
4	After PEG separation	43	42	13	2	–		
5	After magnetic separator	49	47	4	<1.5	–		
6	After the first stage treatment of powder 5 on hydrocyclones (underflow)	85	13	2	–	1:1		
7	After the first stage treatment of powder 5 on hydrocyclones (overflow)	13	81	6	–			
8	After the second stage treatment of powder 6 on hydrocyclones (underflow)	95	4	1	–	7:3	0.907	15.156
							0.887[a]	16.050[a]
9	After the second stage treatment powder 6 on hydrocyclones (overflow)	62	34	4	–		0.930	3.056
							0.915[a]	4.121[a]
10	After hydrochloric acid treatment of powder 7	14	86	–	–	–		
11	After the second stage treatment of powder 7 on hydrocyclones (underflow)	46	54	–	–	1:4		
12	After the second stage treatment of powder 7 on hydrocyclones (overflow)	6	94	–	–			
13	After silicon carbide sedimentation process using alcohol medium	3	97	–	–	–		

[a] Characterization of the powder treated with hydrochloric acid (10 wt%) and alkaline solution (3 wt%) to remove silicon and metal particles.

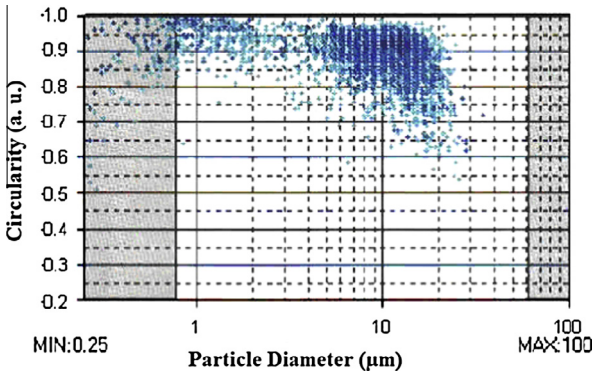

Fig. 1. Characterization of the original SiC (powder 1, Table 1).

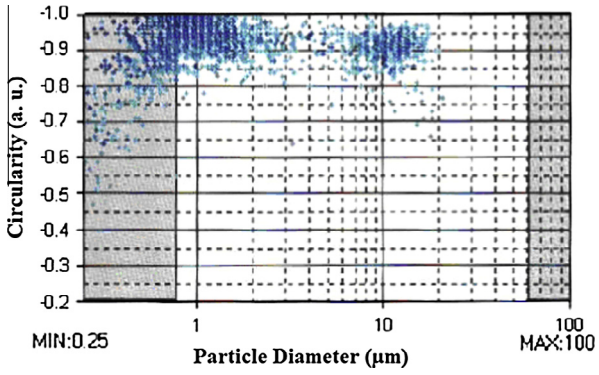

Fig. 2. Characterization of the initial sawing waste obtained after the second decanter centrifuge (powder 3, Table 1).

Analyzer (Malvern Instruments). Water content determination in PEG solution was conducted by Karl Fischer titration.

3. Results and discussions

The slurry was produced after cutting of single- or polycrystalline silicon ingots was pre-treated using two sequential decanter centrifuges. The initial sawing waste obtained after the first decanter centrifuge (powder 2) (Table 1) approximately contained 54 wt% SiC, 14 wt% Si, 30 wt%. PEG 9 wt% metals from cutting wire, mainly Fe and some amount (<1 wt%) of Cu and Zn (metals included in the alloy composition of wire saw), and after the second decanter centrifuge contained 34 wt% SiC, 33 wt% Si, 23 wt% PEG and 10 wt% metals (powder 3) (Table 1). It is efficiently to use decanter centrifuges for separation PEG from powders but it is inefficiently to use decanter centrifuges for separation Si and SiC particles with each other. We used the initial sawing waste after the second decanter centrifuge as a feedstock. Fig. 2 shows a typical particle size distribution in the slurry waste. The major area around 16 μm particle size was contributed by SiC particles, while the smaller area at 1 μm was mainly due to Si particle, small SiC and metal particles also share this peak.

3.1. Recovery process of polyethylene glycol

At first the sawing waste obtained after the second decanter centrifuge placed in the concrete mixer (FS-1000/750E «Ferrovial»), then warm water (about 50 °C) required to obtain 30–50 wt% solution of PEG in water were added. After homogenization, the suspension was fed to the filter press (ФОМ 52-1У-001, Berdichev Machine-Building Plant "Progress") and squeezed at pressure up to 30 bar. The resulting solution of polyethylene glycol was subjected to further purification, because it contained metal ions (mainly Fe^{3+}) and had brown color. To clean PEG solution we tried to use different methods: addition oxidizing agents (hydrogen peroxide), purification using adsorbents, the highest level of purification was achieved using as adsorbent activated carbon (БАУ-А ГОСТ 6217-74), as known it is a good adsorbent for heavy metal ions [10]. Activated carbon (2.5 wt%) was added to PEG solution then the suspension was stirred during 24 h at 25 °C and filtered. As a result, PEG solution had no color, or had light yellow color (because of the presence of residual metal ions) and was transparent, that allowed to use it again (after water removal by vacuum treatment). This method of purifying is also suitable for PEG with water content less than 0.5 wt%, because this method does not