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Separation of silicon and silicon carbide using an electrical field

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ABSTRACT

Recycling silicon from wiresaw slurry may be a good solution to reducing the high cost of silicon feedstock for solar cells. In this study, a cell was designed for separating Si and SiC particles in a buffer solution by gravity combined with electrical fields. According to the particle size distribution and zeta potential analysis, the average size of SiC particles was greater than that of Si particles in wiresaw slurries. The negative charges on the Si surfaces were more than that on SiC surfaces in buffer solution with a pH > 2.5, increasing the average settling velocity for SiC particles and attractive force toward the anode for Si particles. Therefore, the horizontal and vertical movement of Si and SiC particles occurred simultaneously when a horizontal electrical field was applied to the cell. Due to the small size, low density and increased charges, the electrical field enhanced greater displacement for Si particles, leading to a Si distribution on the bottom of separation cell. Analysis of carbon content at various positions on the cell bottom indicates that the experimental results and predicted result were consistent. The highest efficiency of separation, with only 7.15 wt% SiC remaining in the mixture, was obtained near the cell outlet; the original mixture had roughly 75.3 wt% SiC. The recovered material with high Si content can be transferred to an induction furnace to generate solar-grade Si.

1. Introduction

Photovoltaic (PV) technology is an important source of clean electrical energy. Although the PV industry has grown markedly, the current PV industry requires substantial amounts of silicon, which has caused a shortage of feedstock and increased feedstock price. Reusing waste silicon may increase the silicon supply. Thus, obtaining silicon from wiresaw slurries is a potential recovery method [1].

When manufacturing silicon wafers, a silicon ingot is sliced by a multi-wiresaw with an ethylene glycol-based solution and silicon carbide (SiC) abrasives. Although the cutting wires are thin, roughly 30% of the silicon ingot becomes waste in slurries during sawing. This waste slurry typically contains silicon kerf-loss, sawing solution, and many tiny iron fragments from the worn saw wire. To recovery Si feedstock from such wastes, removing metal impurities is necessary. This related study on the separation of metal fragments via electrokinetic techniques was investigated [2]. Several studies with regard to separation of silicon or silicon carbide have been done. Mühlbauer et al. utilized a directional solidification process to remove C and SiC from silicon materials produced via silica reduction in an arc furnace [3]. Nishijima et al. applied superconducting magnetic methods to acquire SiC from sawing waste [4]. Shibata et al. used flotation to obtain SiC from waste [5]. Zhang and Ciftja investigated the removal of SiC and Si_3N_4 inclusions from top-cut solar cell silicon scraps by filtration with foam filters [6]. Wang et al. developed a process for purifying the silicon obtained from slurry; this process comprised chemical treatment, centrifugation, high-temperature treatment and directional solidification [7]. However, these approaches did not use an electrical driving force to separate Si particles from wiresaw slurries. This study recovers Si from sawing waste by applying an electrical field.

In previous research, metal removal performance using an electrokinetic process was better than that of acid treatment. This study presents an electrical technique, instead of centrifugation, to subsequently separate Si from wiresaw slurries because heavy fluids used in centrifugation may be toxic to the environment. Based on the fact that Si and SiC particles in wiresaw slurry have different densities, surface charges and particle sizes, a new cell was designed for separating Si and SiC by applying an electrical field. Via this proposed method, the motion of particles was investigated, and the recovered Si was analyzed.

2. Experimental

2.1. Pretreatment for metal removal

The silicon wiresaw slurries (Fig. 1) contain Si, SiC, metal and liquid. These slurries were obtained from Sino-American Silicon

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Fig. 1. Slurries from wire sawing of a Si wafer.

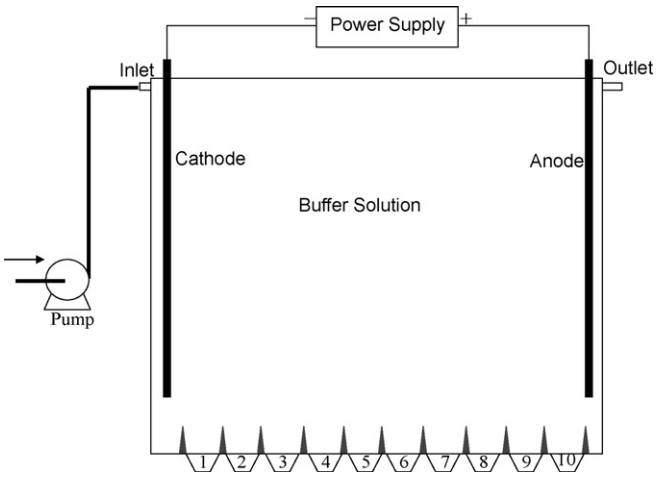

Fig. 2. Experimental setup for electrical and gravitational settling.

Products, Inc. (Chu-Nan, Taiwan). To analyze the separation of Si and SiC, the slurries were pretreated to remove metal impurities. This pretreatment included the approaches of electrokinetics and acid dissolution. After metals were removed by either method, the liquid components of the slurry were washed away by acetone and then centrifuged at 12,000 rpm for 40 min to obtain a wet cake, which was then dried in an oven at 150 °C to form solid powders. The solid components were 50–55 wt% of the original slurries.

2.2. Analyses of silicon and silicon carbide

Several properties of the dry powders were analyzed. First, microscopic images of Si and SiC particles were observed by SEM and qualified by energy dispersive X-ray spectroscopy (EDS). The particle size distribution (PSD) was measured using static light scattering (Beckman, Coulter LS230). The carbon content in the solid powders was measured using a Horiba carbon/sulfur analyzer (model CS-244; Leco). The SiC content was calculated using analytical results. Once the weight percentage of the residual metal was determined, the Si content in the powders was estimated. Furthermore, a zeta potential analyzer (Zetasizer 3000; Malvern) was used to determine the surface charges of these particles at various pHs. The solutions at different pHs were prepared using H_3PO_4 and NH_3.

2.3. Separation of silicon and silicon carbide

Samples comprised of 15 g of the solid powders were mixed with 5000 ml of buffer solution and transferred to the separation setup. Buffer solutions with pHs of 2, 7 and 8 were prepared using H_3PO_4 and NH_3. The flow rate was low enough to just overcome the height between the reservoir of the test solution and the inlet of the separation cell. The cell was 15-cm deep and 15-cm long (Fig. 2). The cell was designed to separate different particles by gravity and electrical fields. At the cell inlet, a metering pump input the solution containing SiC and Si particles. Moreover, a power supply and pair of platinized titanium electrodes were used to apply a constant electrical field across the solution in the cell. In this study, the applied electrical field was 1 V/cm. At the cell bottom, 10 regions were equally divided to collect the settling particles. After operation for 24 h, the solutions collected at different positions were dried and analyzed using static light scattering (Beckman, Coulter LS230) and a Horiba carbon/sulfur analyzer (model CS-244; Leco). The PSD and SiC content at each position was then determined to investigate separation efficiency.

3. Results and discussion

Fig. 3 shows an SEM image of dried solid samples after pretreatment. The sizes of Si and SiC particles differed. The EDS analysis at points A and B indicates that the SiC particles were larger than Si particles. The PSDs were measured (Fig. 4). The distributions peaked at roughly 1.4 μm and 14 μm for Si and SiC particles, respectively. To investigate the separation of these particles, settling behavior and sedimentation velocities were determined first based on the fact that Si and SiC particles have different densities (ρ_{Si} = 2.33 g/cm^3, ρ_{SiC} = 3.16 g/cm^3) and diameters.

According to sedimentation theory [8,9], settling behavior can be classified by the Reynolds number of a particle (Re_p), expressed as

$$Re_p = \frac{\rho_f v d_p}{\mu} \quad (1)$$

where ρ_f is fluid density, v is particle velocity, d_p is particle diameter, and μ is fluid viscosity. If we assume these particles are spherical, the settling behaviors of Si and SiC obey Stokes' law when $Re_p < 0.1$. Therefore, their terminal sedimentation velocities in vertical direction (v_t^0) can be calculated by

$$v_t^0 = d_p^2 \frac{(\rho_p - \rho_f)g}{18\mu} \quad (2)$$

where ρ_p is particle density and g is gravitational acceleration. Thus, the obtained terminal velocities of Si and SiC particles according to their primary particle sizes were

$$v_t^0(Si) = 1.42 \times 10^{-4} \text{ cm/s} \quad (3)$$

$$v_t^0(SiC) = 2.30 \times 10^{-2} \text{ cm/s} \quad (4)$$

Through the prediction of terminal velocity, the Reynolds numbers of Si and SiC should be examined to determine whether settling behavior obeys Stokes' law. Consequently, the calculated Re_p values were

$$Re_p(Si) = 1.99 \times 10^{-6} \quad (5)$$

$$Re_p(SiC) = 3.22 \times 10^{-3} \quad (6)$$

Clearly, the Re_p values were far less than 0.1, *i.e.*, sedimentation obeys Stokes' law.

In addition to the effects of size and density of Si and SiC particles, their zeta potentials also affect separation. Fig. 5 shows measured results for the zeta potentials. The isoelectric points of Si and SiC were both at a pH of 2.5, indicating that the surfaces of Si and SiC were positively charged at pH < 2.5, and negatively charged