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Recovery of cutting fluids and silicon carbide from slurry waste

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ABSTRACT

The wafer slicing process generates large amounts of slurry waste. The recovery of cutting oil and abrasives from slurry waste can reduce both the cost and environmental damage. A process combining magnetic precipitation and flocculation was developed for the recovery of cutting oil. A magnetic precipitation tank was employed for storage of the slurry and acceleration of the settlement of suspended particles. The larger particles further aggregate upon adding a non-aqueous flocculant comprising polyacrylamide (PAM) and ethylene glycol (EG). The recycled oil product is obtained by centrifugation and bag filtration. The physical properties and wafer dicing tests indicate that the recycled oil is qualified. Wafer manufacturers can thus reduce costs by using this process to produce recycled oil. A magnetic reactor with alkaline aqueous successfully recycled the SiC powder from the slurry waste by converting all the silicon species into sodium silica for further use. The results demonstrate that the magnetic reactor is able to remove most metal species and that the alkaline aqueous medium can recover all the Si substances in a sodium silicate solution, also called water glass.

1. Introduction

Solar cells based on silicon wafers are the most widely used in the photovoltaic industry. Silicon wafer fabrication involves the growth of the crystal ingot, wafer slicing, surface treatment, edge rounding, polishing, cleaning, etc. [1–3]. Fig. 1(a) shows the silicon wafer production process. Single-crystal ingots are sliced into wafers by using a brass-coated steel wire inside a cutting slurry pool. The cutting slurry contains diethylene glycol (DEG, as the cutting oil), silicon carbide (SiC, as an abrasive), and dispersion agents. During the wafer slicing process, fine Si particles and metal species are stripped from the silicon ingots and steel wire. In addition, some SiC particles are broken into smaller SiC debris. Such spent cutting slurry contains many impurities and cannot be reused, thus becoming waste.

In Taiwan, more than 30,000 tons of silicon slurry waste per year are generated from the wafer and semiconductor industry. The cutting slurry accounts for most of the wafer production cost. In addition, such a cutting slurry waste is hazardous to our environment and health. The toxicity of DEG has been reported to have a detrimental impact on human beings and animals [4–8]. Clinical reports have revealed that DEG poisoning can cause severe complications, such as kidney failure, liver disease, and central nervous system depression. Due to environmental and economic concerns, wafer-slicing companies are required to reduce the amount of cutting slurry waste. As shown in Fig. 1(b), the slurry waste is treated with a screw decanter centrifuge machine for separation into two fractions: light slurry and heavy slurry. The light slurry consists of about 65% DEG and 35% solids of small size, while the heavy slurry contains abundant SiC particles of suitable (larger) size for its reuse in the preparation of cutting slurry. However, the light slurry needs to be treated by a recycler to recover the cutting oil as it contains too many impurities. The recycling process is expensive, and wafer-slicing companies have to pay for such recycled cutting oil. The use of recycled cutting oil and heavy slurry to replace a portion of new cutting slurry is the main approach to reduce production costs. In order to further reduce the cost of recycled cutting oil, wafer-slicing companies would rather build a recovery process to recycle the cutting oil than purchase the oil from the recycler. However, the separation of DEG and SiC from slurry waste is challenging because the particles (Si, SiC, and metal fragments) are well-dispersed in the DEG liquid. Numerous methods for the recovery of cutting oil have been developed [9–12]. The most common methods applied in industry to recycle cutting oil are by distillation and filtration. The energy consumption during distillation and the large amount of consumables used for filtration result in higher costs for cutting oil recycling and create even more waste.

Due to the strong surface charge, it is difficult for these fine particles to come closer. The electrostatic repulsion causes the well-dispersion of fine particles in the cutting oil. In order to make the fine particles contact with each other, the repulsive potential energy barrier must be overcome. This may be accomplished by electrostatic destabilization (coagulation) or steric destabilization (flocculation). Flocculation is a

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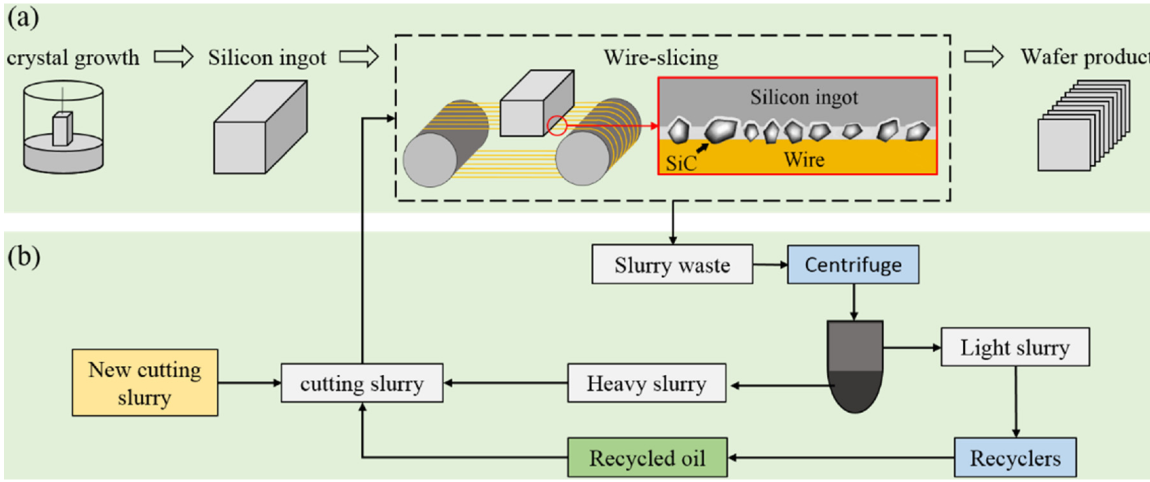

Fig. 1. Processing flow diagram for (a) solar wafer production and (b) treatment of slurry waste in the plant.

cheaper method to remove solids from liquids [13–18]. Fine suspended particles are bridged together by polymer chains bringing the particles closer together forming a heavier bulk for settlement and filtration. Polyacrylamide (PAM) is widely used as a flocculant. PAM is a water-soluble polymer widely used in wastewater treatment and solid–liquid separation processes as an aqueous flocculant [19–24]. However, the use of aqueous flocculants increases the water content (i.e., decreases the viscosity) of the cutting oil. It is well known that the water content has a great influence on the production of wafers. Finding an alternative solvent to replace water is necessary if we want to use PAM as a flocculant in the slicing process. In this study, following the van Oss, Chaudhury, and Good (VCG) theory [25–27], a series of commercially available alcohols and ethylene glycol derivatives were evaluated to replace water as the solvent to dissolve PAM. The best solvent was chosen based on the experimental results.

The recovery of solids from spent slurry is also an important issue from the environmental point of view due to their potential application [28–30]. A variety of processes for Si and SiC recycling have been proposed in the literature, including alloying methods [31], centrifugation [32,33], electrical field [34–36], flotation [37], hydrobromination [38], hydrocyclone [39], liquid–liquid extraction [40], phase-transfer separation [41–43], rapid thermal processes [44], sonication [45], and supercritical water methods [46]. In the recycling industry, the spent slurry is treated by chemical methods. A strong basic reactant dissolves silicon in the form of silicates [47] or a strong acid solution containing hydrofluoric acid (HF) and hydrogen peroxide (H_2O_2) transforms silicon into hexafluoro silicic acid. In the industry, the metal species in the slurry are usually treated by an acid wash process. Using too much acid may lead to pipe corrosion and increase the higher cost. So the researchers have developed the magnetic method for the removal of the metal species from the slurry waste to replace the acid washing. However, it is difficult to apply the superconducting magnet for the removal of metal species in SiC recovery process. Using the permanent magnets is practicable. For example, the magnetic filters have widely been used to remove magnetic substances from industrial process streams [48–50].

In this study, we report a low-cost and simple process to recycle DEG and SiC powders from slurry waste. A magnetic precipitation process and a flocculation system were combined to produce qualified recycled oil. In order to reduce the dosage of flocculant, the magnetic precipitation tank was installed in line with the flocculation system. The magnetic precipitation tank was designed based on our previous invention [51] for slurry storage and acceleration of the settlement of suspended particles. PAM is a widespread flocculant in wastewater treatment but there is no previous literature on the use of PAM in oil recovery. A novel flocculation solution comprising PAM and EG was prepared and used in this study to avoid increasing the water content in the recycled DEG. The cost of the oil recycled by our process is much lower than that of commercial recycled oil. Moreover, the recovery process of SiC powder was tested by combining a magnetic method and an alkali solution. Instead of acid leaching, we integrated the alkali solution washing to remove the Si powders and the magnetic method for the metal species removal in one step. To study the feasibility of the physico-chemical separation technique for slurry waste, we used a separating funnel equipped with magnetic bars as a magnetic reactor. The results show that magnetic bars are able to remove almost all the metallic species, the alkali solution collects all the Si compounds in the form of a sodium silicate solution, i.e., water glass, which is a useful raw material [52] and the SiC powders with a purity of 95 wt% were recycled.

2. Experimental section

2.1. Materials and reagents

The slurry was supplied by Utech Solar Co., Ltd. PAM (Mw 5,000,000–6,000,000) was purchased from He-Cheng Chemical Corporation, while EG (99.9%), propylene glycol (PG), DEG, glycerol, and sodium hydroxide (NaOH, 99.38%) were obtained from Choneye Pure Chemicals. The centrifuge separator (Model No: HTC-25) was a product of Zhen-Mei Machinery Industrial Co., Ltd. The magnet bars (6000 G) were purchased from Amoeba Magnetic Co., Ltd and the magnetic precipitation device was designed and built by ourselves. The bowl-tilting type mixing machine (Model No:LB-2.4-1) was manufactured by Win-shin-gi Machinery Co., Ltd. The stainless steel bag-filter housing and polypropylene filter bags were purchased from Taiwan Grace International Corp. The wafer slicing tests were performed by Utech Solar Co., Ltd with a saw machine (Model No: MB-DS271) manufactured by Meyer Burger.

2.2. Recovery process of cutting oil and SiC

A flowsheet of the recovery process for cutting oil and SiC is schematically presented in Fig. 2. The main blocks are the precipitation system, flocculation system, and SiC recovery system. The recycling process is conducted in the magnetic precipitation device as the first step to obtain two kinds of slurry: the upper half of the tank (UH slurry) with a larger DEG volume and the sludge with a larger amount of solid. DEG is recycled by treatment of the UH slurry in the flocculation system and SiC powder was obtained by treatment of the sludge in a magnetic reactor. The detailed procedure is as follows.