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Enhancing the separation of silica nanoparticles from backside grinding (BG) wastewater with synthesized magnetite

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ABSTRACT

Fine nano-sized magnetite particles were synthesized and then used to treat backside grinding (BG) wastewater for the removal of silica nanoparticles (SiO_2). Batch experiments were conducted to examine the effects of the magnetite dosage, pH, stirring intensity (G) and duration (t_d), magnetic field strength and sedimentation time on the removal of SiO_2. The optimum dosage of magnetite was determined as 16 mg/L, which had the best SiO_2 removal of about 97%, under the conditions of initial pH 5.9, stirring intensity 900 S^{-1}, stirring duration 30 min, sedimentation time 30 min and magnetic field strength 317 mT. The synthesis of nano-sized magnetite particles 10 nm in diameter led to a reduction in the magnetite dosage needed for the removal of SiO_2. The pH was maintained at less than 10 to obtain the high SiO_2 removal mentioned above. When the pH was over 10, the Fe detached from magnetite particles which subsequently led to a decrease in SiO_2 removal. Magnetite aggregation was affected by both G and t_d. Increasing the values of G and t_d enhanced the removal of SiO_2. When the multiple values of G and t_d, i.e., Gt_d reached 12,000, a SiO_2 removal of 95% was achieved. The relationship between the magnetic field strength and sedimentation time was also evaluated. An increase in the magnetic field strength improved the removal of SiO_2. Specifically, the SiO_2 removal was greatly dependent on the sedimentation time when the magnetic field strength was weak. The results of this study can be used in the design and operation of a magnetite aggregation process to treat BG wastewater for the removal of SiO_2.

1. Introduction

Semiconductor manufacturing produces a large quantity of backside grinding (BG) wastewater containing substantial amounts of fine abrasive particles from the wafer backgrinding step which requires a high volume of ultra pure water [1]. BG wastewater has a high turbidity (varying from 110 to 2500 NTU) characterized by nano-size particles ranging from 0.06 to 7.72 μm [1,2]. Specifically, high levels of extremely stable silica (SiO_2) nanoparticles are found in BG wastewater [1–3]. Thus, the effective removal of SiO_2 from BG wastewater has received the most attention from an engineering aspect.

Conventional methods for the removal of SiO_2 from semiconductor wastewater use chemical coagulation, floatation, electrocoagulation, membrane filtration and adsorption. However, those applications not only produce tremendous amounts of sludge but also expensive costs for purification as shown in many previous studies [4–6].

Application of magnetite (Fe_3O_4) has emerged as a promising alternative approach for the removal of nanoparticles surpassing the limitations of conventional methods. Magnetite is a mineral, one of three common naturally occurring iron oxides. Magnetite is the most magnetic of all the naturally occurring minerals on Earth [7]. According to previous works, the magnetic force of magnetite enhances the removal of nanoparticles in a very short period of time reducing chemical waste sludge [2,3,8]. In addition, it was reported that magnetic nanoparticles were very effective in removing heavy metals [9–12].

Although it is believed that the application of magnetite is a viable technique in treating wastewater, very few related studies for the removal of nanoparticles have been found in previous literature. Only two papers have reported on the aggregation and precipitation of nanoparticles from BG wastewater using magnetic seeds [2,3]. The study of Wan et al. [2] focused on the combination effects of magnetite and polyaluminum chloride (PAC) on the removal of turbidity under different physical and chemical conditions, such as magnetic seed dosage, pH and applied magnetic field

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strength. Shen et al. [3] assessed the potential for the regeneration of used magnetic seeds employed in turbidity removal using ultrasound.

Although the earlier works try to suggest factors that influence the application of magnetite in treating BG wastewater, they were limited to only evaluating turbidity removal. Due to the high concentration of SiO_2 nanoparticles in BG wastewater [2,3], assessing the efficiency of magnetite in the removal of SiO_2 is therefore definitely necessary for the rational application of magnetite. Furthermore, to the best of our knowledge, the previous studies did not examine the effects of the magnetite particle size on the removal of SiO_2 even though some studies reported that the adsorption capacity of magnetite particles increased with decreasing the particle size or increasing the surface area in treating the wastewater contaminated with the metal ions, such as Ni(II), Cu (II), Cd(II) and Cr(II) [11,12]. In addition, unfortunately, the effects of the stirring intensity and duration on SiO_2 removal have not yet been studied although these parameters have an important role in flocculation chemistry [13–15].

In this study, batch experiments were conducted to determine the effectiveness of the synthesized magnetite in the removal of SiO_2 from BG wastewater. The evaluations mainly focused on identifying parameters including magnetite dosage, pH, stirring intensity, stirring duration, sedimentation time and magnetic field, which affect the removal of SiO_2. Moreover, the effect of the magnetite nanoparticle size was investigated in the separation of SiO_2 from BG wastewater.

2. Materials and methods

2.1. Characteristics of the BG wastewater

The BG wastewater samples were obtained from a large semiconductor manufacturer in Cheongju, Korea. The composition of the BG wastewater is shown in Table 1. A significantly high concentration of SiO_2 was observed. The particle size distribution in the BG wastewater was ranged from 51 to 5560 nm with a Z-averaged value of 171 nm as shown in Fig. 1.

2.2. Synthesis of magnetite nanoparticles

To synthesize the magnetite nanoparticles, a mixture of ferric and ferrous solution was prepared at a molar ratio of 2:1 by dissolving 16.2 g of $FeCl_3{\cdot}6H_2O$ and 5.97 g of $FeCl_2{\cdot}4H_2O$ in 135 mL of deionized water. Then, the mixture was vigorously stirred for 1 h at 40 °C. After that, 120 mL of 26% NH_4OH solution as a precipitating agent was added to the solution containing the mixture of Fe^{2+}/Fe^{3+} by stirring for 20 min at 400 rpm, at 70 °C. According to a previous study by Peternele et al. [16], the magnetite obtained with the precipitating agent NH_4OH was more uniform than that obtained with NaOH. Magnetite particles are formed by solid reaction according to the following chemical reactions:

$$FeCl_2 + 2FeCl_3 + 8NH_4OH \rightarrow Fe(OH)_2 + 2Fe(OH)_3 + 8NH_4Cl \rightarrow Fe_3O_{4(S)} \downarrow + 8NH_4Cl + 4H_2O \quad (1)$$

Table 1
Characteristics of the BG wastewater.

Parameter	Concentration range
Turbidity (NTU)	6684–6696
SiO_2 (mg/L)	234–250
Suspended solid (SS) (mg/L)	1130–2230
Conductivity (μS/cm)	1.1–2.6
pH	5.8–6.1

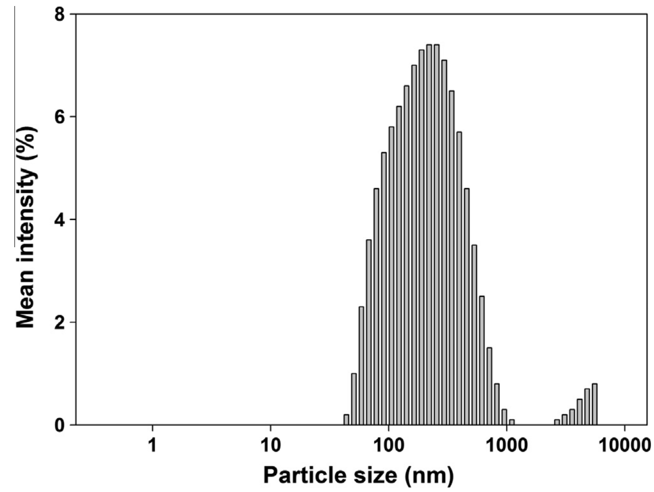

Fig. 1. Measured particle size distribution in BG wastewater.

This method has been shown to be an adequate procedure for obtaining nano-scale particles in previous studies [17,18]. The prepared magnetite particles were washed five times with deionized water. After the washing procedure, they were ultrasonicated at 200 W for 5 h at room temperature in order to disperse them into colloidal nanoparticles. Ultrasound was used in these experiments because the particle size of the magnetite decreased as the frequency of the mechanical vibration increased, as previously described by Rusianto et al. [19]. All chemicals used were of analytical grade.

2.3. Experiments on magnetite aggregation and sedimentation

To evaluate the synthesized magnetite in removing SiO_2 nanoparticles from BG wastewater, batch tests were done with a jar test apparatus. The paddle at the end of each stirrer shaft had a diameter of 7.6 cm and a height of 2.5 cm. The jars were made of acrylic plastic with the dimensions of 11.5 × 11.5 × 21 cm and held 2.0 L of liquid. In order to determine the optimum dosage of magnetite for SiO_2 removal, 2.0 L of BG wastewater was mixed with 0.07, 0.33, 0.67, 3.33, 6.67, 13.33 and 20 mL of magnetite slurry, which contained 0.64, 3.2, 6.4, 32, 64, 128 and 192 mg of magnetite nanoparticles, respectively. When studying the effects of pH on the removal of SiO_2, the pH was adjusted from 2 to 12 with 5 N H_2SO_4 and 4 N NaOH. The stirring intensity (G) and duration (t_d) were 900 S^{-1} and 30 min, respectively, followed by a settling of 30 min in the above experiments. The G value tested in this study varied from 327 to 2467 S^{-1}. The applied t_d ranged from 5 to 30 min. The applied G value was calculated based on a previous study by Cornwell and Bishop [20], in which the velocity gradient had a linear correlation to the impeller speed. During the sedimentation of aggregates, the appropriate magnetic field strength was tested when investigating the effects of magnetic field strength on the removal of SiO_2. Aggregates of SiO_2 nanoparticles were separated from aqueous solution with magnetic field strengths of 215, 247 and 317 milli Tesla (mT). A syringe was used to withdraw a sample from each jar at the end of the sedimentation period. The samples were filtered through membrane filters (Gelman GN-6 with an effective pore size of 0.45 μm) prior to the analysis. Experiments were carried out at ambient laboratory temperature (20 °C).

2.4. Analytical procedures

The particle size distribution in BG wastewater was determined with a particle size analyzer (Mastersizer 3000, MALVERN). SiO_2