



Silica particles settling characteristics and removal performances of oxide chemical mechanical polishing wastewater treated by electrocoagulation technology

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ABSTRACT

The purpose of this study was to explore the feasibility of removing silica particles and reducing turbidity from oxide chemical mechanical polishing (oxide-CMP) wastewater. Based on the dynamic characteristics of batch electrocoagulation, three operating stages (lag, reactive, and stabilizing) are proposed to identify the relationships among the zeta potential of the silica particles, solution turbidity, and the corresponding mean particle size of the silica. Experimental results show that the silica particles were destabilized and settled at the critical mean particle size, which was estimated to be above 520 nm after 10 min, and the corresponding turbidity removal mostly occurred during the reactive stage. Furthermore, the corresponding mean particle size varied from 520 to 1900 nm as the treatment time progressed from 10 to 20 min, which also occurred during the reactive stage. Several parameters, including different electrode pairs, electrolyte concentration, applied voltage, and the optimum condition of power input were investigated. Experimental results indicate that a Fe/Al electrode pair is the most efficient choice of the four electrode pair combinations in terms of energy consumption. The optimum electrolyte concentration and applied voltage were found to be 200 ppm NaCl and 30 V, respectively.

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1. Introduction

Chemical mechanical polishing planarization (CMP), developed by IBM in 1985, is a rapidly growing process in semiconductor manufacturing. The process is mainly used for “polishing” the device side of a semiconductor wafer via the mechanical downward force of a slurry abrasive in association with the chemical oxidation of the wafer surface. In addition, CMP is an effective process for reducing topological wafer imperfections and for improving depth of the focus of lithography processes [1]. Owing to its capability of achieving a uniform surface topography with high throughput, the employment of CMP technology has extended from its traditional application in metal interconnections (i.e., metal CMP) to complicated applications in dielectric barrier layer planarization (i.e., oxide-CMP). Due to the ongoing pursuit of stronger functional devices with small feature sizes (sub 0.15 μm), device fabrication will depend on CMP processes in the foreseeable future [2]. Because of the polishing slurry used in the

CMP process, a large amount of ultrapure water is used to wash away abrasives that adhere to the surface of wafers during the polishing process [3,4]. According to a previous report [4], CMP processes may account for 40% of water consumption in the semiconductor industry. In proportion with the water used in the CMP process, generally, the same quantity of wastewater would be generated [5]. As stated in the International Technology Roadmap for Semiconductors specifies and with a greater than 80% water recycling capability for new plants, the objective of reducing total water consumption by roughly five-fold for fabrication processes must be conformed according to 2005. Consequently, the resulting wastewater stream needs proper treatment(s) to comply with local regulations before being discharged into surface water systems.

In general, inorganic and organic contaminants in the CMP effluent derive mostly from the CMP slurry [6]. The content of insoluble inorganic contaminants depends on the CMP slurry used. Inorganic contaminants in CMP wastewater may include suspended solids (in the range of nanometers to micrometers) of SiO₂, Al₂O₃, or CeO₂ that typically have concentrations in the range of 50–500 mg/L. On the other hand, organic contaminants may include metal complexing agents, surfactants, stabilizers, and

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rheology control agents [1]. Currently, chemical coagulation or membrane separation processes are used widely to treat the CMP wastewater in the semiconductor industry. Ultrafiltration (UF) is utilized to remove fine oxide particles [3]. Golden et al. [1] investigated the effectiveness of a combined chemical coagulation and microfiltration process in removing suspended oxide particles. In general, CMP wastewater possesses highly negative surface charges that repel adjacent particles when they are submerged in alkaline solutions. Due to these wastewater characteristics, removing such nano-scale particles by conventional chemical coagulation is not ideal.

To overcome these problems, electrochemical treatment techniques have attracted a great deal of attention because of their versatility and environmental compatibility. Using the electron, which is a “clean reagent” [7], as the main reagent, an electrochemical treatment (i.e., electrocoagulation) method has been proposed and investigated to treat wastewater containing dispersed fine particles without chemical additions [8–12]. Belongia et al. [13] first investigated the particle removal phenomena of synthetic alumina and silica solution using electrodecaantation and electrocoagulation. Electrocoagulation is an electrochemical method of treating polluted water whereby sacrificial anodes dissolve due to an applied voltage, producing active coagulant precursors (usually aluminum or iron ions). Furthermore, electrocoagulation offers the possibility of anodic oxidation and in situ generation of adsorbents (such as hydrous ferric oxides and hydroxides of aluminum). When cathodic reactions occur, in many cases, the evolution of hydrogen gas is involved. Sacrificial anodic electrodes of iron and aluminum are commonly used to continuously supply metallic ions as the source of coagulation. These electrochemically generated metallic ions can hydrolyze near the anode to form a series of metallic hydroxides capable of destabilizing dispersed particles. Electrocoagulation is a complicated process with a multitude of mechanisms operating synergistically to remove the pollutants (e.g., silica particles) in the vicinity of the anode from the suspended solution. The turbulence also created by oxygen (generated as a secondary reaction in the anode), and hydrogen (produced due to water reduction in the cathode) generates a mixture that helps to destabilize particles resulting in the coagulation of bigger particles. Pollutants are then removed by sedimentation and become the slurry. Fig. 1 schematically demonstrates the complex and interdependent nature of the electrocoagulation process. Electrocoagulation is an emerging water treatment technology that has been applied successfully to treat various wastewaters. This technique has been applied for the treatment of potable water [14,15], urban wastewater [16], heavy metal laden wastewater [17], restaurant wastewater [18], colored water [19], wastewater containing phosphate [20,21], and mechanical cutting oil [22]. Previous researchers have mostly studied the removal characteristics and CMP wastewater quality before and after electrocoagulation treatment [8,9]. However, in this study, electrocoagulation was employed to treat synthetic oxide-CMP wastewater with an attempt to remove the suspended silica particles and to reduce the wastewater turbidity. Additionally, the dynamic characteristics (e.g., zeta potential and mean particle size) of suspended silica particles in the oxide-CMP wastewater during electrocoagulation were monitored. Several parameters, including different electrode pairs, concentration of supporting electrolyte, applied voltage, and the optimum condition of power input, all of which govern the removal performance and throughput of the electrocoagulation procedure, were also investigated. These characteristics were experimentally investigated in this study to determine the optimum treatment conditions and to elucidate the dynamic nature of batch electrocoagulation by three stages, including the lag, reactive, and stabilizing stages.

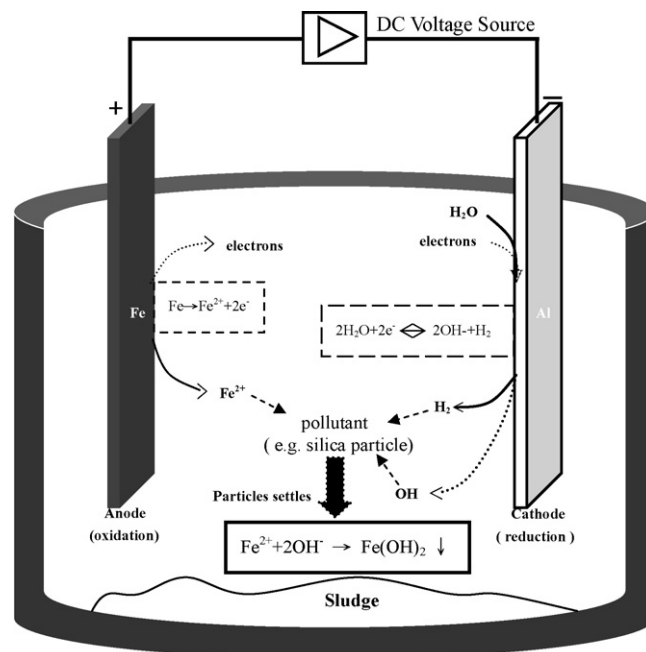


Fig. 1. Interactions occurring within an electrocoagulation reactor.

2. Experimental

2.1. Preparation of the synthetic oxide-CMP wastewater

Suspended silica solutions were prepared by diluting a commercially available, KOH-based CMP slurry containing 25 wt.% fume silica (Semi-Sperse[®] 25 slurry, Cabot Microelectronics Corp., USA). The original slurry dilution concentration was diluted to 0.2% and 2.4% with deionized water, yielding a suspension turbidity of approximately 30 and 300 NTU, respectively. Table 1 presents the chemical and physical properties of the synthetic oxide-CMP wastewater used in this study. The synthetic oxide-CMP wastewater was mainly composed of the diluted silica slurry with a turbidity ranging between 90 and 130 NTU, a conductivity of between 180 and 210 $\mu\text{S}/\text{cm}$, a pH of between 9 and 10, a zeta potential of between -35 and -40 mV, and a mean particle size between 150 and 160 nm.

2.2. Equipment

The electrocoagulation experimental apparatus is shown schematically in Fig. 2. The electrolytic cell was a 1-L Pyrex glass reactor which was equipped with water jacket and magnetic stirrer. The temperature of the electrolytic cell was controlled by circulating water continuously through the water jacket from a refrigerated circulating bath (Model BL-720, Taiwan). A magnetic stirrer (Corning, PC-420D, MALAYSIA) was employed in the reactor to maintain adequate mixing of the CMP wastewater during the

Table 1
Water properties of the synthetic oxide-CMP wastewater

Parameter	Synthetic oxide-CMP wastewater
Mean particle size (nm)	150–160
Zeta potential (mV)	-35 to -40
pH	9–10
Turbidity (NTU)	90–130
Conductivity ($\mu\text{S}/\text{cm}$)	180–210
TS (mg/L)	2000–2200

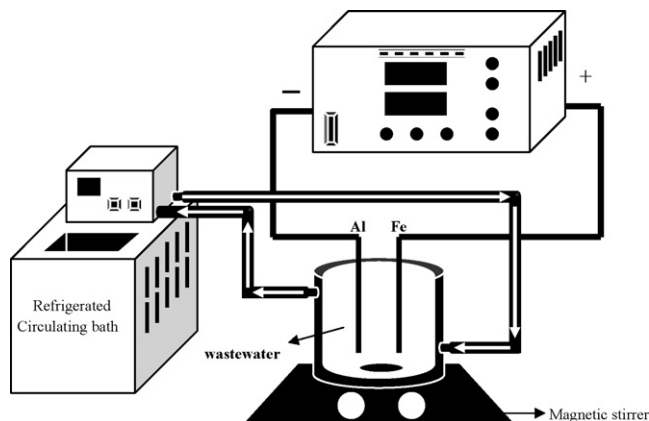


Fig. 2. Apparatus of the electrocoagulation process.

electrocoagulation process. Cast iron (Fe) and aluminum (Al) plates ($8\text{ cm} \times 4\text{ cm} \times 0.5\text{ cm}$) were used in four different combinations as the anode/cathode pair. The electrode pair was dipped in the CMP wastewater to a depth of 4.5 cm, with the electrodes approximately 2 cm apart. The effective area of the electrode pair was 18 cm^2 . Electrical voltage was provided by a manually controllable DC power supply (Model GC50-20D, Taichung, Taiwan) operating in the constant-voltage mode (range: 0–50 V). Characterization of the CMP wastewater, such as pH (Y.S.I, pH 10, USA), conductivity (EUTECH, cyberscan 510, Singapore), turbidity (in terms of Nephelometric turbidity unit, NTU), and total solids (TS), was determined using ROC EPA standard methods [23]. A Malvern zeta potential and size analyzer (Malvern Instruments, Zetasizer Nano Series, UK) was used to determine the zeta potential and particle size distribution of the suspended silica particles in the CMP wastewater.

2.3. Chemicals and reagents

The pH of the solution was adjusted by adding either dilute sulfuric acid (H_2SO_4) or sodium hydroxide (NaOH). The stock H_2SO_4 and NaOH solution of 0.1 M was freshly prepared by dissolving a reagent grade H_2SO_4 (Scharlau, Chemie, S.A., Spain) and NaOH (Sigma–Aldrich, Germany) in deionized water. The conductivity of solutions was raised and adjusted to different values by the addition of NaCl (Tedia Company, USA).

2.4. Method

Before each experiment, the electrodes were abraded with sand paper to remove scale, then dipped in 3 M H_2SO_4 to a depth of 6 cm for 30 min, and finally cleaned with successive rinses of water. During each test run, 0.5 L of CMP wastewater was placed in the reactor. The magnetic stirrer was turned on and set at 600 rpm. A steady temperature of $25\text{ }^\circ\text{C}$ was maintained for all test runs. The stirrer speed was found to be sufficient to provide good mixing in the electrolytic cell and yet not strong enough to break up the flocs formed during the treatment process. A fixed amount of between 100 and 300 mg/L (NaCl) was added to the CMP wastewater to increase the wastewater conductivity and thus facilitate the electrocoagulation treatment. The DC power supply was turned on with constant voltages at 20, 30, and 40 V, corresponding to current densities of 100, 180, and 260 A/m^2 , respectively. An electrocoagulation test run lasted no more than 35 min in all experiments. At the end of electrocoagulation, all samples were allowed to settle for 5 h in a Pyrex glass column of 25 mL. The turbidity of the CMP wastewater after electrocoa-

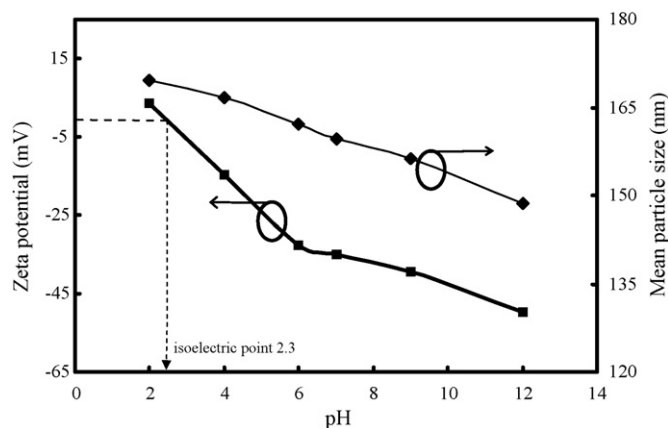


Fig. 3. Effect of solution pH on the zeta-potential and mean particle size of the synthetic CMP wastewater.

ulation treatment was determined using a turbidity meter (HACH, 2100P, USA).

3. Results and discussion

3.1. Characterization of synthetic CMP wastewater

The diluted silica solution was representative of synthetic oxide-CMP wastewater. It was prepared by diluting a commercially available oxide slurry with deionized water to 1%. The concept of surface potential (i.e., zeta potential) appears to be a valuable practical parameter for coagulation and the resultant separation from the aqueous solution, providing a direct indicator of solution stability. Fig. 3 shows the zeta potential of the diluted silica slurry and the corresponding particle size as a function of the solution pH. The solution pH was adjusted and maintained by the addition of the required amount of NaOH or H_2SO_4 . As can be seen from Fig. 3, an increase in pH resulted in a greater degree of negative zeta potential and a decrease in the corresponding particle size. For example, by increasing the pH of the solution from about 4 to 12, the zeta potential varied from -14.8 to -49.7 mV and the mean particle size decreased from 166.7 to 148.6 nm. This may be because the pH increase in the synthetic silica CMP suspension tended to adsorb more OH^- ions on the particle surface and created Si-O^- on the surface, thus causing greater negative zeta potential. In other words, electrostatic repulsion between the particles of the electric double layers with greater negative potential drives suspended particles apart, achieving particle stability and reducing a smaller particle size. Therefore, a relatively smaller mean particle size associated with the synthetic silica CMP suspension could be attributed to its relatively large negative zeta potential. Additionally, the isoelectric point at which charge reversal is observed for the silica CMP suspension was about 2.3, corresponding to a mean particle size of approximately 168 nm. The turbidity of the silica suspension indicated a linear relationship with a diluted slurry concentration of less than 2.4%, as demonstrated in Fig. 4. Additionally, an increase in the conductivity of the solution linearly increased the slurry concentration due to the presence of additives or oxidizers (e.g., NH_4OH , KOH) in the original slurry. The typical range of turbidity of the synthetic CMP wastewater was 250 to 35 NTU, which corresponds to a dilution level of between 50 and 500 times that of the original slurry. The range of these values agrees with those reported for actual oxide-CMP wastewater [8,9], and thus validates the appropriateness of using the synthetic CMP wastewater as the model wastewater. In this study, the synthetic oxide-CMP wastewaters were prepared by diluting original silica

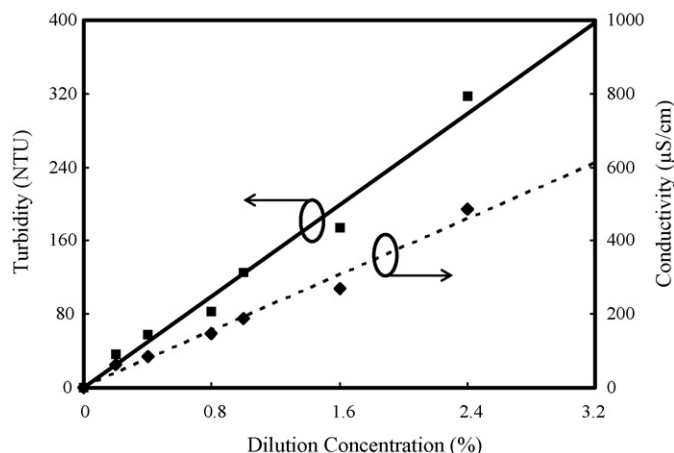


Fig. 4. Effect of the slurry diluted concentration on the solution turbidity and conductivity for preparation of the synthetic oxide-CMP wastewater.

slurry to around 1%, yielding a corresponding suspension turbidity of approximately 125 NTU, and then applying it in the subsequent experiments.

3.2. Comparison of electrode pairs

In any electrochemical process, different electrode materials and the type of electrode pair are regarded as significant factors affecting the performance of the electrocoagulation process [24]. Therefore, appropriate selection of the electrode pair is important. Iron and aluminum plates in four different combinations were investigated in this study to determine the optimum electrode pair. In order to investigate the effect of different electrode pairs on the electrocoagulation time to achieve 95% turbidity removal, an applied voltage of 30V was used and a fixed amount of 200 ppm electrolyte (NaCl) was added to the synthetic oxide-CMP wastewater. The pH values for the four different electrode pairs did not vary significantly. They were in the range of 9–10 during the electrocoagulation. Fig. 5 illustrates the effect of different electrode pairs on the electrocoagulation time to achieve 95% turbidity removal. As can be seen in this figure, the Fe/Al electrode pair accomplished 95% turbidity removal in the short time of 12 min, while 95% turbidity removal was achieved in approximately 17, 20, and 28 min

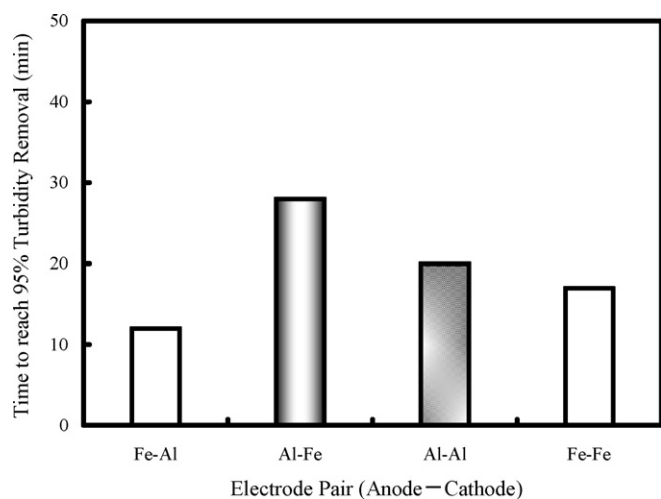


Fig. 5. Effect of different electrode pairs on the electrocoagulation time to achieve 95% turbidity removal with 125 NTU initial turbidity, 30V applied voltage, 200 mg/L NaCl, and 5 h sedimentation.

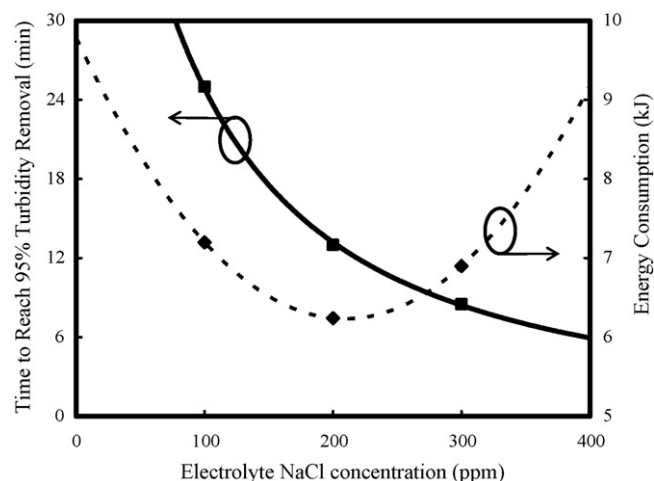
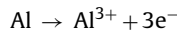


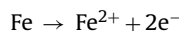
Fig. 6. The electrocoagulation time to reach 95% turbidity removal and energy consumption as a function of the electrolyte NaCl concentration with the Fe/Al electrode pair, 125 NTU initial turbidity, 30V applied voltage, and 5 h sedimentation.

for Fe/Fe, Al/Al, and Al-Fe electrode pairs, respectively. The energy consumption for the four different electrode pairs were calculated to be 5.8, 8.6, 10.1, and 12.1 kJ for Fe/Al, Fe/Fe, Al/Al, and Al-Fe electrode pairs, respectively. Considering the performance with respect to energy consumption, the Fe/Al electrode pair was regarded as the optimum choice of the four electrode pair combinations verified in this study, and was therefore applied in the subsequent experiments. The reason for the better performance when using Fe anodes can be explained by the chemical reactions taking place at the aluminum anode and the iron anode as follows.

For Al anode:



For Fe anode:



The nascent aluminum and iron ions are very efficient coagulants for particulates to flocculate. From the above two reactions, we can calculate the electrochemical equivalent mass for Al and Fe. Consequently, for aluminum, the electrochemical equivalent mass is 335.6 mg/Ah. For iron, the value is 1041 mg/Ah, about three times that for aluminum. That is, more coagulants are theoretically produced for iron anodes when passing the same electric charges. This was probably the reason for the removal efficiency being higher when using iron anodes in this study.

3.3. Effect of concentration of supporting electrolyte

The electrical conductivity of the solution is an important parameter for saving electric energy in an electrochemical cell. More energy is required for overcoming a high ohmic resistance between an anode and a cathode when electrical conductivity of the solution is low. In this study, the original CMP wastewater had a conductivity of less than 200 μS/cm, as displayed in Table 1. This conductivity is not conductive enough for electrocoagulation treatment. The most common method used to overcome this problem is to add a small amount of electrolyte, which increases the electric conductivity of the solution and thus reduces the energy consumption during electrocoagulation [25,26]. In this study, sodium chloride (NaCl) was used as the supporting electrolyte for increasing the conductivity of the CMP wastewater. Fig. 6 indicates the effect of the NaCl concentration on the time to reach 95% turbidity removal and the required power input. It can be seen evidently that

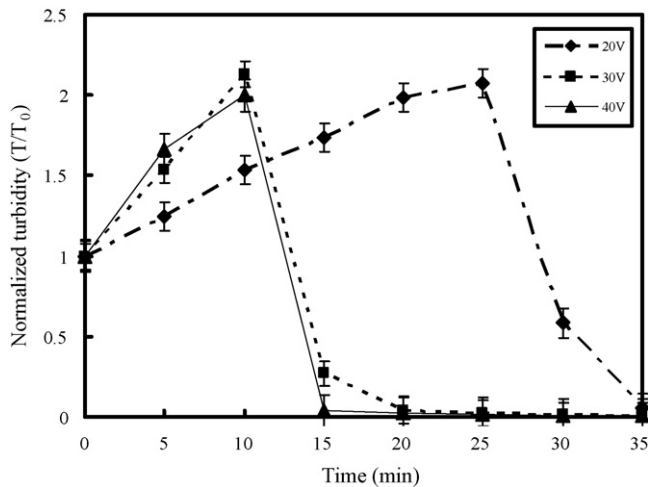


Fig. 7. Effect of various applied voltages on the normalized turbidity during electrocoagulation with the Fe/Al electrode pair, 125 NTU initial turbidity, 200 mg/L NaCl, and 5 h sedimentation.

increasing the electrolyte concentration from 100 to 300 ppm led to a decrease in the time to reach 95% turbidity removal from 25 to 8.5 min, respectively. This was probably because the Cl^- anions can destroy the passivation layer and increase the anodic dissolution rate of metal, either by the incorporation of Cl^- into the oxide film or by the participation of Cl^- in the metal dissolution reaction [27]. In addition, the increase of the chloride ion concentration in the solution can reduce the overpotential in the anodic oxidation of chloride ions, and thus results in a higher current efficiency [28,29]. The de-passivation effect was more significant when more Cl^- anions were added to the solution. Therefore, the problem of electrode passivation was partially solved when using NaCl as the electrolyte.

Once the required voltages and the corresponding currents were obtained from the experimental tests, it was possible to estimate the amount of Watts required. The amount of required Watts was multiplied by the process time to give the total amount of energy consumed for the electrocoagulation treatment. Fig. 6 also demonstrates that the energy consumption decreased from 7.2 to 6.24 kJ when the electrolyte (NaCl) concentration was increased from 100 to 200 ppm. There was an upward trend when the electrolyte (NaCl) concentration was above 200 ppm. For example, by increasing the electrolyte (NaCl) concentration from 200 to 300 ppm, the energy consumption increased from 6.24 to 6.9 kJ. Considering the energy consumption performance illustrated in Fig. 6, 200 ppm NaCl was regarded as the optimum balance with a reasonable treatment time for 95% turbidity removal at relatively low energy consumption.

3.4. Effect of applied voltage

Among the various operating variables, the applied voltage is an important factor which strongly influences the performance of electrocoagulation [24]. Fig. 7 illustrates the effect of the applied voltage on the normalized turbidity of wastewater for various lengths of time where the sludge was allowed to settle for over 5 h. As can be seen in Fig. 7, the final turbidity removal of wastewater was achieved to over 95% in less than 15 and 20 min of electrocoagulation for 40 and 30 V, respectively. As the applied voltage increased, the length of each electrocoagulation process decreased. Due to the sufficient voltage through the solution, the metal ions generated by the dissolution of the sacrificial electrode were hydrolyzed to form a series of metallic hydroxide species. These species neutralized the electrostatic charges on dispersed particles to reduce

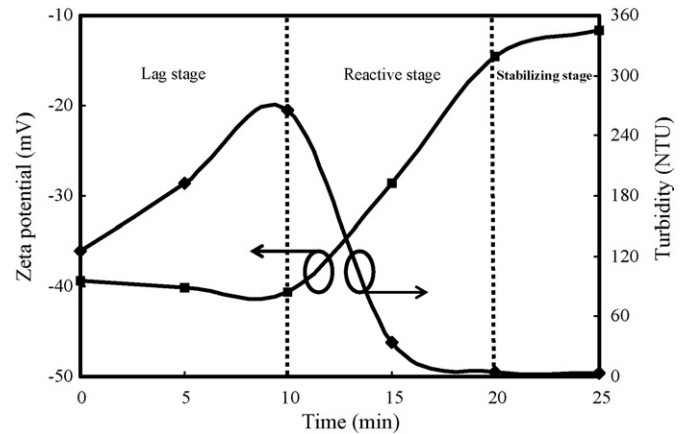


Fig. 8. Effect of wastewater zeta potential and turbidity during electrocoagulation with the Fe/Al electrode pair, 125 NTU initial turbidity, 30V applied voltage, 200 mg/L NaCl, and 5 h sedimentation.

the electrostatic interparticle repulsion enough so that the van der Waal's attraction predominates, thus facilitating agglomeration [30]. However, it took a longer time (above 35 min) to achieve over 95% turbidity removal for an applied voltage of 20 V. This is because the applied voltage of 20 V was unable to completely destabilize the suspended oxide particles in the solution. Therefore, both 30 and 40 V are suitable operating voltages for this electrocoagulation experiment, requiring treatment times of less than 20 min. Interestingly, and somewhat surprising, was the influence of the applied voltage, regardless of voltage value, in the first range of 10–25 min on the normalized turbidity. Fig. 7 indicates that the normalized turbidity was around 2 in the first 10, 10, and 25 min for 40, 30, and 20 V, respectively. This could be explained by the insufficient coagulant dissolving from the iron anode in the beginning period of electrocoagulation to effectively reduce the double layer of the suspended particles to destabilize the particles, resulting in enhanced turbidity. As time progressed and dissolved coagulants at the iron electrode increased, there was an abrupt decrease in turbidity, which eventually leveled out above zero. In order to study and explain this phenomenon, the dynamic characteristics of suspended silica particles during electrocoagulation are investigated in the subsequent section.

3.5. Dynamic characteristics of silica particles during the electrocoagulation process

A number of coagulation mechanisms of suspended particles, including surface charge neutralization, double layer compression, and adsorption and bridging, have been proposed that depend on the physical and chemical properties of the solution, pollutant, and coagulant [31,32]. As previously discussed, colloid particles were maintained in stable suspension by electrostatic repulsion between particles. The zeta potential is regarded as an effective measurement of the charge of the particle while it migrates to the solution, thus providing an indicator of solution stability. In general, the electrocoagulation process is inherently dynamic, with ferrous ions entering the system in this study being hydrolyzed with suspended particles, and then removed from the system. In order to investigate the dynamic characteristics of silica particles during electrocoagulation, the zeta potential of the oxide particles and the corresponding turbidity were measured during the treatment period. Its change during the course of electrocoagulation for the applied voltage of 30 V is demonstrated in Fig. 8. It can be obviously seen from this figure that there were three stages, defined as a lag, a reactive, and a stabilizing stage during the electrocoagulation

progress [15]. Fig. 8 shows that the zeta potential of silica particles remained relatively steady at about -40 mV for the first 10 min—a response that is considered as the lag stage. An increase in turbidity was observed in the lag, which might be ascribed to the possibility that the aggregated particle size in this period was inadequately large to settle. As time progressed and dissolved coagulants from

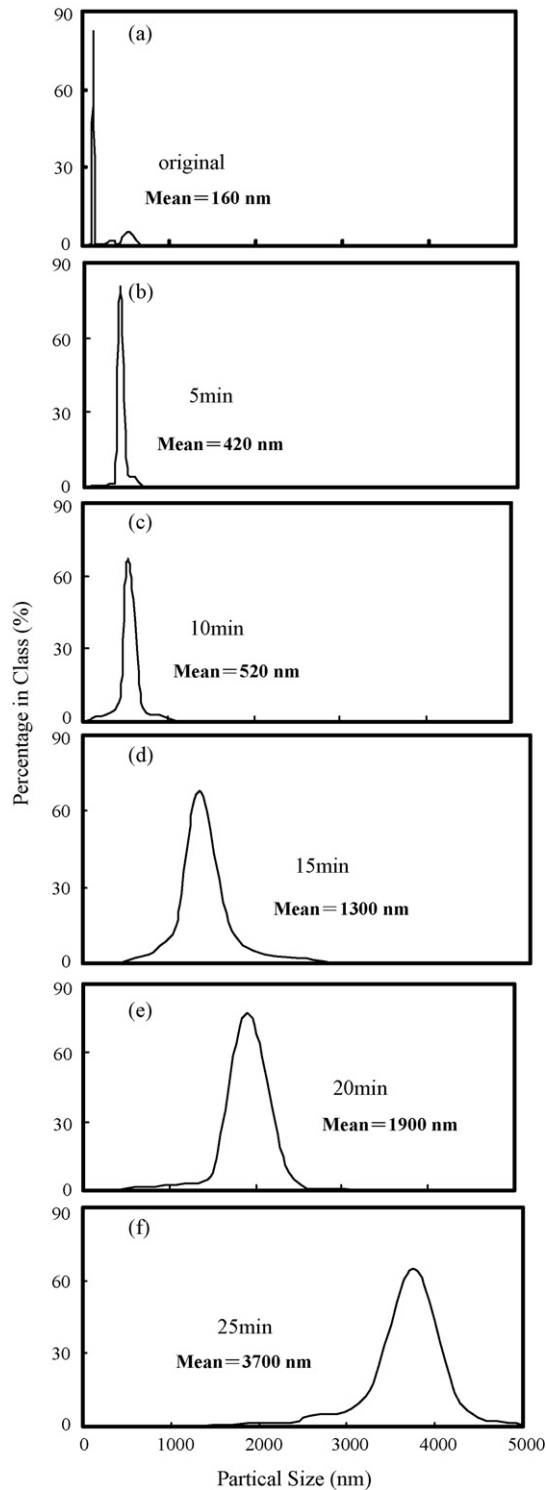


Fig. 9. Change of particle size distributions of the synthetic oxide-CMP wastewater during electrocoagulation with the Fe/Al electrode pair, 125 NTU initial turbidity, 200 mg/L NaCl, and 30 V applied voltage: (a) in the original wastewater; in the treated wastewater (b) 5 min, (c) 10 min, (d) 15 min, (e) 20 min, (f) 25 min.

the iron electrode increased, a period was reached where there was an abrupt increase in the zeta potential (i.e., a small degree of negative zeta potential) and the majority ($\sim 95\%$) of turbidity removal occurred during the reactive stage. Therefore, the beginning of the reactive stage might be defined as the period where a rapid decrease in turbidity started. As the system moved into the stabilizing stage, the rate of turbidity removal continued to decrease with the turbidity ultimately approaching zero, while the corresponding zeta potential approached a plateau in the range of -15 to -12 mV after 20 min. The stable increase in the zeta potential after a short steady period reveals that the electrocoagulation appeared as a double layer compression [33]. As time progressed and the amount of coagulants (i.e., iron cations) produced at the anode increased, these iron coagulants could suppress the electric double layer suspended silica particles and destabilize these particles, thus promoting the aggregation of the suspended silica particles. Fig. 9 demonstrates the dynamic variation of particle size distributions of the suspended silica in oxide-CMP wastewater during electrocoagulation. The mean particle size increased continually as the electrocoagulation treatment progressed, as shown in Fig. 9. As can be seen from Fig. 8, at the beginning of the reactive stage, it was observed that the turbidity started to decrease rapidly after 10 min. At this time, the zeta potential increased (i.e., a small degree of negative zeta potential), which resulted in the aggregation of the silica particles and the generation of bigger particles. The corresponding mean particle size varied from 520 to 1900 nm as the treatment time progressed from 10 to 20 min (i.e., in the reactive stage), as shown in Fig. 9. These observations imply that the suspended silica particles were destabilized to settle at the critical mean particle size that was estimated to be above 520 nm after 10 min, and that the corresponding turbidity removal mostly occurred during this period (i.e., the reactive stage).

3.6. Optimum condition of power input

Fig. 10 shows the electrocoagulation time required to reach 95% turbidity removal and the power input as a function of the applied voltage. It shows that 95% turbidity removal was achieved in 15, 20, and 35 min for 40, 30, and 20 V, respectively. Therefore, both 30 and 40 V are suitable choices of the applied voltage for the electro-

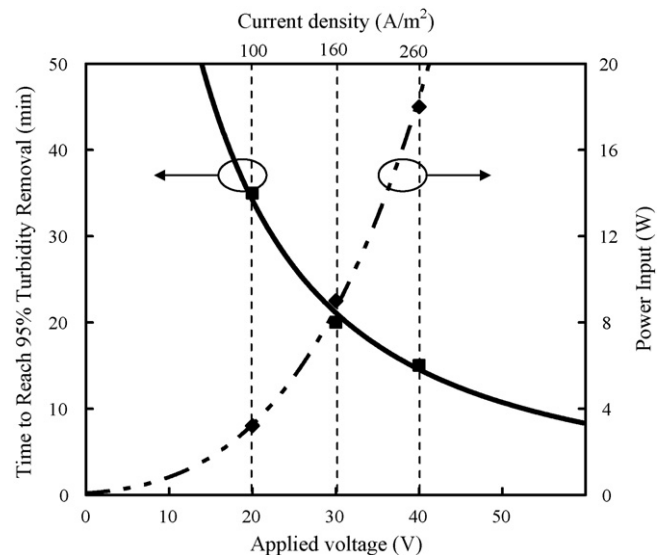


Fig. 10. The electrocoagulation time to reach 95% turbidity removal and power input as a function of the applied voltage with the Fe/Al electrode pair, 125 NTU initial turbidity, 200 mg/L NaCl, and 5 h sedimentation.

coagulation which requires a treatment time of less than 20 min. The power supply was turned on with constant voltages at 20, 30, and 40 V, corresponding to current densities of 100, 180, and 260 A/m², respectively. Under constant 20, 30 and 40 V, the corresponding currents were 0.16, 0.3 and 0.45 A, respectively. Based on a 95% turbidity removal, the power input, which was the product of the applied voltage (V) and current input (A), was found to be 3.2, 9, and 18 W. The applied voltage of 40 V achieves a 95% turbidity removal in a short time of 15 min, but requires the largest power input of 18 W. Conversely, the applied voltage of 20 V consumes little power, approximately 3.2 W, but requires a longer time (35 min) to reach 95% turbidity removal. The optimal applied voltage in this study is defined as the applied voltage with a low power input and acceptable treatment time for 95% turbidity removal. Consequently, when considering the above two factors simultaneously, the applied voltage of 30 V seems to offer the best overall performance for the present electrocoagulation with a reasonable treatment time for 95% turbidity removal and a relatively low power input.

4. Conclusions

The electrochemical removal performance of the turbidity of synthetic oxide-CMP wastewater was investigated in a batch electrocoagulation system. The diluted silica solution was representative of real oxide-CMP wastewater, rendering a discharge pH of between 9 and 10, a zeta potential of between –35 to –40 mV, a conductivity of approximately 180–210 $\mu\text{S}/\text{cm}$ at 1% dilution from original silica CMP slurry, and a corresponding initial turbidity and mean particle size of around 125 NTU and 160 nm, respectively. The time to reach 95% turbidity removal was achieved in approximately 12, 17, 20, and 28 min for Fe/Al, Fe/Fe, Al/Al, and Al–Fe electrode pairs, respectively. Considering the necessity of energy conservation, the Fe/Al electrode pair was regarded as the optimum choice of the four electrode pair combinations verified in this study. The addition of the NaCl electrolyte at 200 ppm was regarded as the optimum balance with a reasonable treatment time for achieving 95% turbidity removal and relatively low energy consumption. In terms of low electrocoagulation time to reach 95% turbidity removal and low power expenditure, the applied voltage of 30 V was found to offer the optimum overall considerations within the test range of between 20 and 40 V. In order to investigate the dynamic characteristics of silica particles during electrocoagulation, three operating stages (lag, reactive and stabilizing) were proposed to identify the relationships among the zeta potential of the silica particle, the turbidity of the suspended solution, and the corresponding mean particle size of the suspended silica. The experimental results revealed that the suspended silica particles were destabilized to settle at the critical mean particle size, which was estimated to be above 520 nm after 10 min, and the corresponding turbidity removal mostly (95%) occurred during this period (i.e., the reactive stage). Furthermore, the corresponding mean particle size varied from 520 to 1900 nm as the treatment time progressed from 10 to 20 min, which also occurred during the reactive stage. Based on the low time to reach 95% turbidity removal, the applied voltage of 30 V seems to offer the best overall performance for the electrocoagulation of 95% turbidity removal and relatively low power input.

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