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# Electrocoagulation of chemical mechanical polishing (CMP) wastewater from semiconductor fabrication

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## Abstract

Treatment of chemical mechanical polishing (CMP) wastewater is investigated. The CMP wastewater, as obtained from a large semiconductor manufacturer, was characterized by high suspended solids (SS) content, high Nephelometric turbidity unit (NTU), chemical oxygen demand (COD) concentration up to 500 mg/l, copper concentration over 100 mg/l and a milky color. This study was to explore the feasibility of treating the CMP wastewater by electrocoagulation with an aim of simultaneously lowering the wastewater turbidity, and copper and COD concentrations. Experiments were conducted to analyze the characteristics of the CMP wastewater and to evaluate the effects of electrocoagulation time, type of electrode pair and electrolyte dosage on the system performances. The fine suspended oxide particles were found to have an average size of 100 nm with narrow particle size distribution between 68 and 120 nm. The test results revealed that Al/Fe (anode/cathode) was a good electrode pair in terms of overall performances. Electrocoagulation with Al/Fe electrode pair was able to achieve 99% copper removal and 96.5% turbidity reduction in less than 100 min. The COD removal obtained in the treatment was better than 75%, with an effluent COD below 100 mg/l. The wastewater quality exceeded the direct discharge standard and the effluent can be considered for reuse.

*Keywords:* Semiconductor CMP wastewater; Electrocoagulation; Copper and COD removal; Turbidity reduction

## 1. Introduction

The semiconductor manufacturing involves a large number of complex and highly delicate processes, including silicon growth, oxidation, doping, dicing, ion implantation, photolithography, etching, stripping, metallization, planarization, washing and cleaning, etc. [1–4]. Over 200 of high purity organic and inorganic compounds are used in the manufacturing processes. Moreover, a large quantity of ultrapure water is consumed in various washing and cleaning steps of the semiconductor manufacturing processes [2–4]. Consequently, different types of wastewater are generated. These semiconductor wastewaters commonly contain organic solvents, acids, bases, salts, heavy metals, fine suspended oxide particles and other organic compounds [1–4]. Therefore direct discharge of these wastewaters into a water body is strictly regulated by water quality control authorities.

Among the various semiconductor manufacturing processes noted above, chemical mechanical polishing (CMP) represents an important operation [5,6]. The CMP technology has been commonly adopted for integrated circuit (IC) devices at or below 0.25 μm. However, the CMP technology has a major disadvantage of high operating cost in terms of ultrapure water consumption and wastewater treatment [7–10]. According to recent estimates, the CMP operation could account for as much as 40% of ultrapure water consumption in semiconductor manufacturing by 2005 [11]. The ultrapure water is used in the washing and cleaning step to remove the waste CMP slurry, metallic and organic substances from the wafer surface after a CMP operation is completed. The CMP slurries, the majority of them being proprietary, consist of suspension of very fine $SiO_2$ (silica), $Al_2O_3$ (alumina), $MnO_2$ (magnesia) or $CeO_2$ (ceria) abrasive particles in the pH-controlled solution which also contains oxidizing agents, such as $Fe(NO_3)_3$, $CuSO_4$, $H_2O_2$, $KMnO_4$, chlorates, hydroxylamine, etc. and various additives, such as $NH_4OH$, dispersants/surfactants, buffering inorganic and organic acids, metal complexing agents, corrosion inhibitors, etc. [5,6]. These organic and inorganic components of CMP slurries eventually end up in the wastewater when wafer washing is completed. Furthermore, in the more advanced IC devices below 0.15 μm using copper interconnects, the CMP wastewater also contains a copper concentration up to 100 mg/l or even higher [12–14].

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Due to its high solid content, the CMP wastewater is distinctly different from most of the industrial wastewaters. Removal of the total solids (TS) thus has received the most attention. Browne et al. [7] utilized ultrafiltration (UF) for removal of fine suspended oxide particles from the CMP wastewater. Using simulated alumina and silica suspensions, Belongia et al. [8] studied the various facets of particle removal phenomena by electrodecantation and electrocoagulation. Golden et al. [9] analyzed the effectiveness of chemical coagulation using a proprietary polymer coagulant followed by microfiltration. These researchers found that the combined process was effective in removing the fine suspended oxide particles. During the wafer surface cleaning process, the suspended oxide particle and chemical oxygen demand (COD) contents of the CMP wastewater effluent vary considerably with time. This prompted Browne et al. [10] and Corlett [11] to employ an online monitoring device in separating the cleaner CMP wastewater from the dirtier one so that the former can be recycled for non-critical applications. By combining chemical pretreatment, microfiltration and ion exchange, James et al. [12] were able to remove fine oxide particles and copper from the copper CMP wastewater. In a recent work, Kruilik et al. [13] found that the treatment process of Golden et al. [9] using proprietary polymer coagulant and microfiltration was also effective in removing copper ion from the copper CMP wastewater. All the previous investigations were successful in eliminating the fine oxide particles and copper ion from the CMP wastewater. However, the majority of the CMP wastewater from semiconductor fabrication contains more than just these two pollutants. As noted earlier, CMP wastewater effluents invariably contain significant amounts of organic compounds as well, as reflected by their chemical oxygen demand concentration as high as 500 mg/l. Without simultaneously removing the dissolved organic compounds, treatment of the CMP wastewater effluents would be inadequate and the recovered CMP wastewater by the previous processes could only have limited applications.

The purpose of this work is to utilize electrocoagulation in treating the CMP wastewater. Electrocoagulation (i.e. electrochemical treatment) has been a low-cost and efficient method for the treatment of water and wastewater. It was tested successfully to treat potable water [14,15], aquacultural wastewater [16], textile wastewater [17,18], industrial wastewater [19,20], landfill leachate [21], restaurant wastewater [22], municipal sewage [23] and urban wastewater [24]. It was also used to remove phenol [25] and surfactants [26] from industrial wastewaters. In the present work, electrocoagulation was employed to treat the CMP wastewater with an attempt to remove the fine oxide particles, improve the wastewater turbidity and lower the copper concentration in a single treatment. Operating conditions of treatment and CMP wastewater quality before and after treatment were monitored. The experimental results were evaluated to elucidate the coagulation mechanism and the performance characteristics of the electrocoagulation process.

## 2. Experimental studies

The CMP wastewater samples were obtained from a large semiconductor manufacturer in northern Taiwan. Information on the contents of the CMP wastewater samples was very difficult to ascertain due to different types and amounts of CMP slurries (all of them being proprietary) employed in the manufacturing process. The water quality of the CMP wastewater, such as the COD, turbidity (in terms of Nephelometric turbidity unit, NTU), pH, conductivity was measured by the standard methods [27] in conjunction with a GBC 916 UV-Vis spectrophotometer (GBC Scientific Equipment Ltd., Vic., Australia). The same methods were adopted for measuring the water quality after electrocoagulation. A Malvern zeta potential analyzer (Model Zetasizer 3000HS, Malvern Instrument Ltd., Worcester, UK) was used to determine the size distribution of the fine oxide particles of the CMP wastewater and the wastewater zeta potential during the electrocoagulation process. An atomic absorption spectrophotometer (Model 932, GBC Scientific Equipment Ltd., Vic., Australia) was used to measure the copper ion concentration.

The electrocoagulation experimental apparatus is shown in Fig. 1. The electrolytic cell was a 1 l pyrex glass reactor which was equipped with an external water jacket for temperature control. Cast iron (Fe), aluminum (Al) and titanium (Ti) plates (20 cm $L \times 3$ cm $W$) were used in five different combinations as the anode/cathode pair. The electrode pair was dipped in the CMP wastewater to a depth of 12 cm and was situated approximately 8 cm apart. The effective area of the electrode pair was 72 cm$^2$. The dc power supply was controlled by a voltameter and was maintained constant at 30 V for most test runs unless noted otherwise. A magnetic stirrer was employed in the reactor to maintain well mixing of the CMP wastewater during electrocoagulation process.

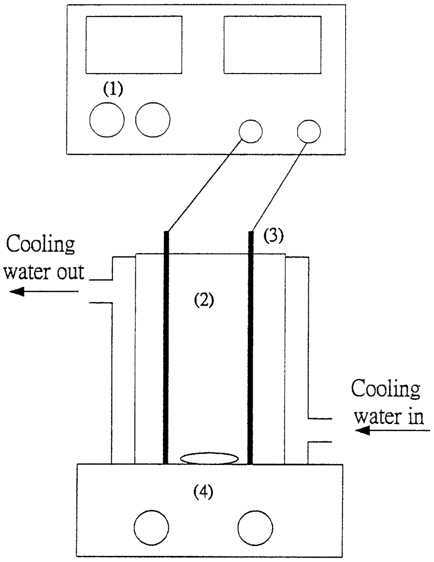

Fig. 1. Experimental schematic: (1) dc power supply; (2) electrolytic cell; (3) electrode pair; (4) magnetic stirrer.

For each test run, one liter of CMP wastewater was put in the reactor. The magnetic stirrer was turned on and set at 100 rpm. The stirrer speed was found sufficient to provide good mixing in the electrolytic cell and yet not so strong as to break up the flocs formed during the treatment process. A fixed amount of 25 or 50 mg/l electrolyte (NaCl) was added to the CMP wastewater to increase the wastewater conductivity and thus facilitated electrocoagulation treatment. The dc power supply was turned on and provided a constant 30 V. Wastewater samples were taken every 10 min for water quality measurements and the electrocoagulation process lasted about 2 h. The treated CMP wastewater was finally allowed to settle for 30 min. Samples were taken from the supernatant for water quality measurements.

## 3. Results and discussion

### 3.1. Characteristics of CMP wastewater

Table 1 lists the measured water quality of CMP wastewater samples obtained from a large semiconductor manufacturing facility during the experimental period. The CMP wastewater had a faint blue and milky color, indicative of the presence of copper ion and fine suspended oxide particles. The CMP wastewater had a high total solids content of 4000–5000 mg/l, but the suspended solids (SS) concentration was negligible, being less than 0.1–0.4 mg/l. The large difference between TS and SS concentrations was due to that the average size of the fine oxide particles (100 nm) was much smaller than the pore size (1.5 μm) of standard filter paper used for SS measurement [27]. Hence, most of these fine oxide particles passed through the filter paper during the SS measurement. In contrast, the TS concentration was measured by evaporating the wastewater sample in a crucible in an electric oven that retained all the fine oxide particles [27]. Comparing to over 120,000 mg/l TS in the original CMP slurry, the lower TS content of the CMP wastewater was apparently due to dilution of the waste CMP slurry during wafer washing and cleaning. It is further noted that the COD concentration of the CMP wastewater was much higher than the original slurry. This was attributable to mixing of the CMP wastewater with other process wastewater that contained organic pollutants, but contributed little total solids. Hence, the COD removal from the CMP wastewater is as important as the removal of fine suspended oxide particles and copper if the treated CMP wastewater is to be upgraded to a level for possible reuse.

Table 1
Water quality and properties of the CMP slurry and wastewater

| Parameter | CMP slurry | CMP wastewater |
|---|---|---|
| pH | 10.2 | 6–8.7 |
| TS (mg/l) | 124,100 | 4000–5000 |
| SS (mg/l) | 0.2 | 0.1–0.4 |
| Copper concentration (mg/l) | – | 45–120 |
| Oxide particle size (nm) | 100 | 100 |
| Conductivity (μs/cm) | – | 450–470 |
| COD (mg/l) | 98 | 210–480 |
| BOD/COD | NM | 0.11–0.15 |
| Color | Milky | Milky |

*Note*: NM: not meaningful because the CMP slurry is non-biodegradable. The brand name of CMP slurry not known.

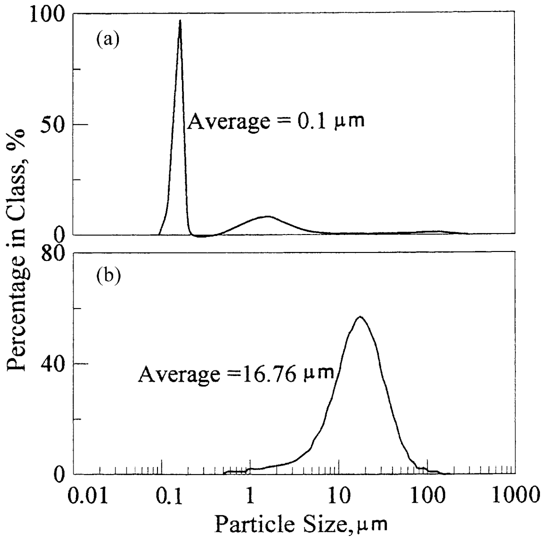

Fig. 2. Particle size distributions of the CMP wastewater with Al/Fe electrode pair, 288 mg/l initial COD, 158 NTU, 50 mg/l NaCl and 79 mg/l copper concentration: (a) in the original wastewater; (b) in the treated wastewater.

### 3.2. Performances of electrocoagulation process

Fig. 2a and b show the size distributions, as measured by the Malvern zeta potential analyzer, of fine oxide particles in the CMP wastewater before and after electrocoagulation. Fig. 2a reveals that the size of fine oxide particles in the original CMP wastewater falls within the narrow range between 68 and 120 nm with an average particle size around 100 nm, which was much smaller than the pore size of the filter paper for SS measurement, as noted earlier. The small peak at 159 nm was attributed to the aggregated oxide particles that are either removed by filtration or redispersed by sonication before the slurry is used. After electrocoagulation, the fine oxide particles originally in stable suspension in the CMP wastewater were destabilized. Aggregates of various sizes were formed, leading to a wide spread of particle size between 49 and 141 μm, as shown in Fig. 2b. The average aggregated particle size was estimated to be 16.76 μm and the peak occurred at 11.3 μm. Due to their good settability, these aggregated particles could be completely removed in less than 30 min, as further elaborated in a later section.

Coagulation of suspended fine oxide particles is governed by different mechanisms: double layer compression, surface charge neutralization, adsorption and interparticle bridging [28]. To ascertain the mechanism governing the present

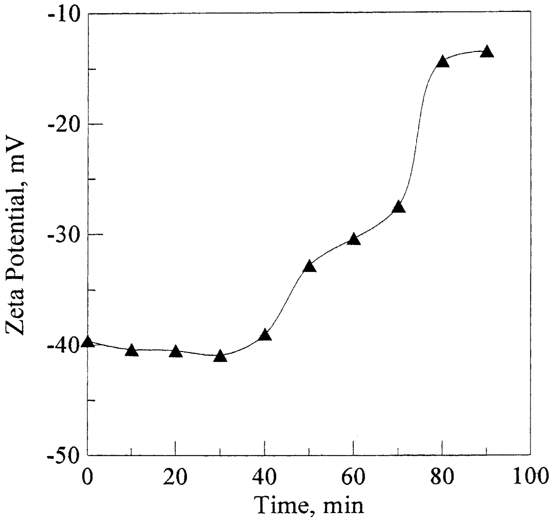

Fig. 3. Change of wastewater zeta potential during electrocoagulation with Al/Fe electrode pair, 288 mg/l initial COD, 158 NTU, 50 mg/l NaCl and 79 mg/l copper concentration.

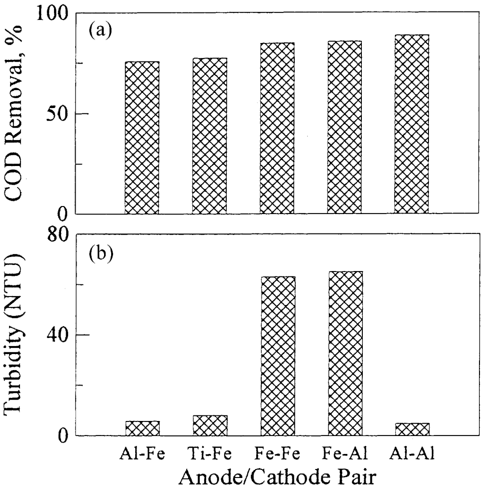

Fig. 4. The COD and turbidity removal for different electrode pairs with 288 mg/l initial COD, 158 NTU, 79 mg/l copper concentration, 50 mg/l NaCl, 60 min electrocoagulation and 30 min sedimentation: (a) COD removal; (b) turbidity (NTU) reduction.

electrocoagulation, the zeta potential of the oxide particles was measured during the treatment period and its change during the course of electrocoagulation is demonstrated in Fig. 3. The figure shows that the zeta potential remained relatively steady at about −40 mV for the first 30 min. It started to rise after that and this was the time the oxide particle aggregates started to form. The particle aggregation continued until the zeta potential reached a plateau at about −15 mV after 80 min. The same change pattern of zeta potential was also observed for other test runs. The steady increase in the zeta potential after a short initial slack period clearly indicates that the electrocoagulation is a double layer compression [28].

Different materials could be used as the electrodes for the present electrocoagulation and the type of electrode pair has been known to be an important factor influencing the performances of the electrocoagulation process [29]. Cast iron, aluminium and titanium plates in five different combinations were tested in the present work to identify the optimum electrode pair. Fig. 4a and b demonstrate the COD removal and final wastewater NTU, respectively, of electrocoagulation using five different combinations of the anode/cathode pair. The wastewater COD removal was caused by direct and indirect oxidation decomposition of organic pollutants in the electrocoagulation process, as elaborated in a latter section. Fig. 4a shows that the COD removal ranges from 75.3% for the Al/Fe pair to 88.4% for the Al/Al pair while those of the Ti/Fe, Fe/Fe and Fe/Al pairs fall within this range. All electrode pairs were able to lower the wastewater COD from initial 288 mg/l to below the industrial wastewater discharge standard (100 mg/l). Fig. 4b displays the corresponding final wastewater NTU for the same electrode pairs. The two cases with cast iron anode yielded a final wastewater NTU over 60 that was much higher than these of the others (all under 8). This could be due to that in the former cases, $Fe(OH)_2$ was formed during the electrocoagulation process and the aggregated particles in these case were observably finer and much more difficult to settle than those formed in the other cases.

The copper removal by electrocoagulation of the present CMP wastewater was demonstrated in Fig. 5a. All five electrode pairs had excellent copper removal over 98% with the two pairs with aluminum anode being almost 100% removal. With 79 mg/l initial copper concentration of the CMP wastewater, the final wastewater copper concentration was all below 1 mg/l, the discharge standard for industrial wastewater in Taiwan. It is noted that the copper removal was

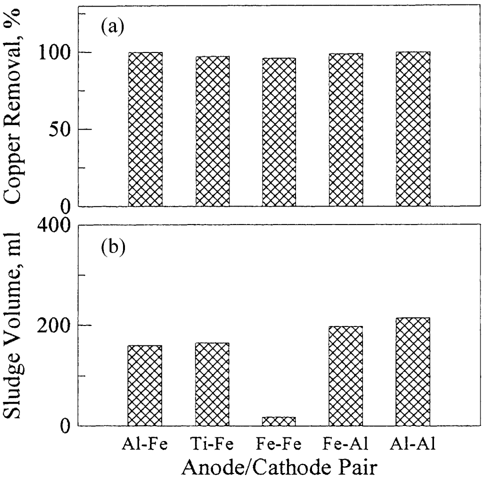

Fig. 5. The copper removal and sludge production for different electrode pairs with 288 mg/l initial COD, 158 NTU, 79 mg/l copper concentration, 50 mg/l NaCl, 60 min electrocoagulation and 30 min sedimentation: (a) copper removal; (b) sludge production.

primarily due to adsorption of copper ion onto the flocs that were removed in the final settling after electrocoagulation.

In all electrocoagulation test runs, 30 min were permitted for floc settling. The sludge volume produced after sedimentation hence could be an important factor in assessing the electrocoagulation performances. Fig. 5b shows that the Fe/Fe electrode pair generated only 18 ml of sludge from the original 1 l of CMP wastewater while the four other electrode pairs yielded a sludge ranging from 160 to 216 ml. The size distribution of aggregated particles generated by the Fe/Fe electrode pair after electrocoagulation was found to be similar to that shown in Fig. 2b except that the average particle size was 18.2 μm. This particle size was significantly smaller that those generated by other electrode pairs. Moreover, the settled sludge of the Fe/Fe electrode pair was considerably denser and more compact than the other cases due primarily to smaller particle size. It is noted that some very fine particles of the Fe/Fe electrode pair remained suspended in the wastewater after 30 min of settling, resulting in a much higher final wastewater NTU, as demonstrated in Fig. 3a. Considering all aspects of performance, the Al/Fe electrode pair is deemed to be a better choice out of the five electrode pair combinations tested in the present work. This Al/Fe electrode pair offers good overall COD and copper removal, low final wastewater NTU and reasonably low sludge production.

The electrocoagulation or electrochemical process is known to have two major oxidations on the wastewater: direct and indirect oxidation [21,29]. Direct oxidation occurs on the anodic surface and is due to surface adsorption and decomposition of pollutants. Indirect oxidation occurs in the bulk liquid phase and is caused by the strong oxidants. In general, the indirect oxidation, which is highly dependent of the wastewater conductivity, plays a much more important role in the electrochemical oxidation process [21,29]. In the present study, the original CMP wastewater obtained from a semiconductor plant had a conductivity less than 500 μs/cm as shown in Table 1 and this conductivity is not conducive to electrocoagulation treatment. To remedy this drawback, a small dosage of electrolyte can be very helpful. In fact, Belongia et al. [8] reported that enhanced electrocoagulation efficiency was obtained by a small $KNO_3$ dosage. For the present study, sodium chloride (NaCl) was chosen for this purpose. Fig. 6a and b, respectively, show the COD removal and final wastewater NTU as a function of the NaCl dosage. Fig. 6b reveals that excellent final wastewater NTU was achieved in less than an hour for a NaCl dosage of 50 mg/l. For a lower NaCl dosage of 25 mg/l and that without NaCl, the same excellent final wastewater NTU could also be attained in 80–90 min. However, in terms of COD removal, shown in Fig. 6a, 25 mg/l NaCl concentration attained the best result at 73.5% in 40 min, followed by 65% with 50 mg/l NaCl in 30 min and 63% without NaCl in 120 min.

Fig. 7a indicates that over 98% copper removal was obtained in 60 min with 25 mg/l NaCl. For a lower NaCl dosage of 10 mg/l, the same excellent copper removal could be attained in 90 min. Increase in the NaCl dosage, however, led to a steady increase in sludge production, as shown in Fig. 7b. Hence, considering the overall treatment performances shown in Figs. 6 and 7, 25 mg/l was considered as a good NaCl dosage, although 50 mg/l might offer an attractive alternative too.

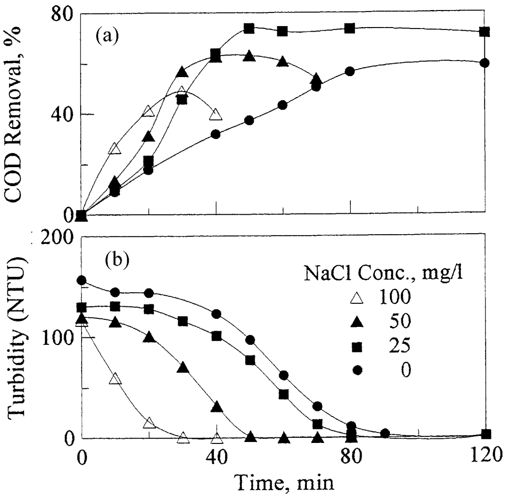

Fig. 6. Effect of NaCl dosage on the COD removal and final wastewater NTU of electrocoagulation with Al/Fe electrode pair, 95 mg/l initial copper concentration, 390 mg/l COD and 54 NTU: (a) COD removal; (b) final wastewater NTU.

In all test runs illustrated above, electrocoagulation was operated at a constant 30 V power supply. It would be of practical interest to see how the voltage affects the performances of electrocoagulation. Fig. 8a and b demonstrated the voltage effect on the copper removal and final wastewater NTU, respectively. Fig. 8a shows that 99% copper removal was achieved in 40, 50 and 90 min for 30, 20 and 10 V power supplies, respectively. For the same power supplies

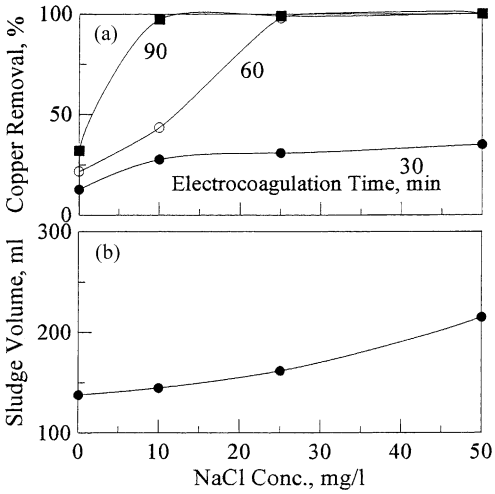

Fig. 7. Effect of NaCl dosage on the copper removal and sludge production of electrocoagulation with Al/Fe electrode pair, 95 mg/l initial copper concentration and 390 mg/l COD and 54 NTU: (a) copper removal; (b) sludge production.

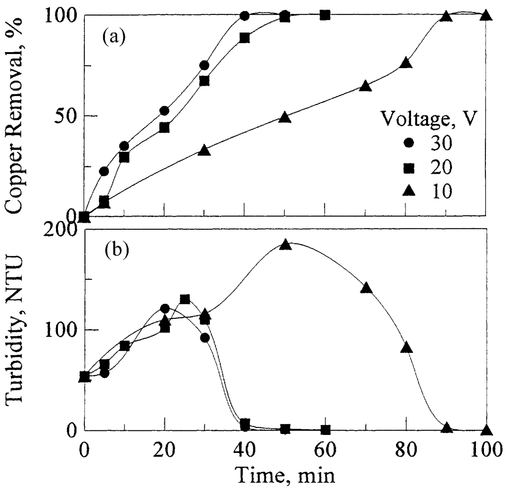

Fig. 8. Effect of operating power supply voltage on the copper removal and the final wastewater NTU with Al/Fe electrode pair, 95 mg/l initial copper concentration, 390 mg/l COD, 54 NTU and 25 mg/l NaCl: (a) copper removal; (b) final wastewater NTU.

and electrocoagulation times, the corresponding final wastewater NTUs realized were 3.3, 6.8 and 3.5, respectively, as shown in Fig. 8b, all these final wastewater NTUs being excellent indeed. Hence, both 20 and 30 V were good choices of power supply for the present electrocoagulation. However, under constant 10, 20 and 30 V power supplies, the corresponding current inputs registered were 0.18, 0.11 and 0.06 A, respectively. Based on 99% copper removal, the power input, which was the product of the power supply (V), current input (A) and operating time (h), was calculated to be 0.9, 1.83 and 3.6 W. Fig. 9 displays the electrocoagulation time to achieve 99% copper removal and the power input as a function of power supply. It is clear from this figure that 30 V power supply achieves 99% copper removal in a short time of 40 min, but requires the largest power input of 3.6 W. A power supply of 10 V consumes very little power input of just 0.9 W, but it requires longer time (90 min) to achieve 99% copper removal. Twenty volts power supply appears to offer a good overall balance with a reasonable treatment time for 99% copper removal and relatively low power input.

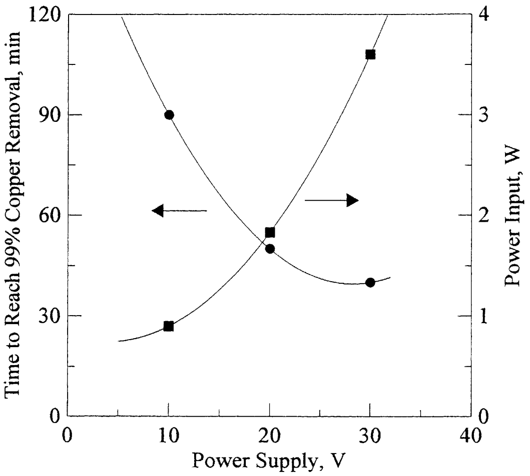

Fig. 9. The electrocoagulation time to achieve 99% copper removal and power input as a function of the power supply with Al/Fe electrode pair, 95 mg/l initial copper concentration, 390 mg/l COD, 54 and 25 mg/l NaCl.

## 4. Conclusions

Electrocoagulation treatment of chemical mechanical polishing wastewater from a semiconductor manufacturer was investigated. This method was employed for removal of fine oxide particles, chemical oxygen demand and copper ion. Based on the results of experimental tests, the following conclusions can be drawn:

1. The CMP wastewater was found to have a high total solids content over 4000 mg/l and a COD concentration up to 500 mg/l. The fine oxide particles in the CMP wastewater had an average particle size of 100 nm and a narrow particle size distribution between 68 and 120 nm.
2. For electrocoagulation of the CMP wastewater, cast iron cathode and aluminum anode were found to form a better electrode pair due to good COD, copper ion and fine suspended particle removal and relatively low sludge production. Adopting this electrode pair, electrocoagulation of the CMP wastewater was able to realize a 96% NTU reduction and 99% copper removal. The COD removal achieved was better than 75% with a final wastewater COD concentration below the discharge standard (100 mg/l). The final wastewater effluent was clear and can be considered for reuse.
3. Addition of electrolyte (i.e. NaCl) was found to be very effective in enhancing the fine oxide particle and copper removal and in improving the wastewater turbidity. A small NaCl dosage of 25 mg/l yielded the best overall performances.
4. In terms of low electrocoagulation time to reach 99% copper removal, low final wastewater NTU below 10 and low power expenditure, a 20 V power supply was found to offer the best overall results within the test range of power supply between 10 and 30 V.

## Acknowledgements

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