The effluent discharge regulations for paper mills have become increasingly stringent to reduce water pollution in the environment and in receiving waters. Together with a higher proportion of water reuse, traditional wastewater treatment units are often unable to remove the accumulated electrical conductivity (ionic species), dissolved solids and colloidal substances in the process water, thus leading to disturbances in papermaking wet-end systems (Sundholm 2000). The treatment units often installed in typical paper mills are mostly sedimentation or dissolved air flotation processes for primary treatment and activated sludge processes for secondary treatment (Thompson et al. 2001; Pokhrel and Viraraghavan 2004).

To date, there is no known case of a commercialized full-scale application of electrocoagulation technology in the pulp and paper industry. Oloman (1992) reviewed the potential applications of electrocoagulation in the pulp and paper industry. For the purpose of lowering the electrical conductivity in kraft mill effluents, Gupta (1979) studied the direct electro-oxidation of effluents, using a dip-bed electrode (HAS graphite fibre electrode). Nassar et al. (1983) used an electro-oxidative method to decolorize pulp bleaching effluents. Tabakov (1983) studied the use of electro-flotation and electrocoagulation to treat kraft pulp mill effluents. With the use of electrolysis, Brinecell Inc. added a small amount of sodium chloride to wastewater to generate oxidants, hypochlorite and ozone, which were then used to treat the wastewater (Oloman 1992). Springer et al. (1995) employed an electrochemical method to treat pulp bleaching effluents, by which 85 to 90% of the color and 90% of the toxicity were removed with treatment costs of between US$0.13 and $0.61/m³. Bejankiwar (2002) studied the use of an electrochemical method to treat cigarette paper mill effluent, and COD removal of 58% and BOD removal of 84% were achieved. Melcer and Maltby (1998) evaluated 26 treatment strategies for removing metal ions from pulp and paper industry effluents, and found that electrocoagulation is one of the technically and economically feasible methods. Donini et al. (1994) showed that treatment operating costs were US$0.65/1,000 gal (ca. US$0.17/m³) for bleaching effluent and US$0.56/1,000 gal (ca. US$0.15/m³) for the entire mill effluent.

The electrocoagulation method uses an iron or aluminum anode as a sacrificing electrode. In addition to generating oxygen, the method also dissolves metals into ions in solution to serve as the coagulant, replacing the traditional alum, ferric chloride or polyelectrolytes. At the same time, hydrogen gas and hydroxyl free radicals are formed at the cathode. Vik et al. (1984) reported that an electrocoagulation method was first initiated in 1889 when London used seawater and an electrolysis method to treat its sewage water. Holt et al. (1999) reviewed the applications of electrocoagulation technology to wastewater treatment and noted that: (1) current density is generally operated at 10 to 2,000 A/m², more typically between 10 and 150 A/m²; and (2) cation production was 10 to 30 μmol for Fe³⁺ and 0.05 to 1 μmol for Al³⁺. The main operating parameter for the setup of an electrocoagulation method is the surface area to volume ratio.
(S/V), which is typically between 18.8 and 42.5 m³/m³ (Zolotukhin 1989; Mameri and Yeddou 1998; Holt et al. 1999). Jüttner et al. (2000) reviewed the types of reactors used in the electrochemical treatments of wastewaters and their potential for applications to various industries. Robinson (1999, 2000) utilized two types of electrocoagulation reactors to treat agricultural wastewater, poultry farm effluent and municipal wastewater. Vlyssides et al. (2000) and Shin et al. (2004) employed electrocoagulation methods to treat the dyeing wastewaters of textile mills. Phutdhawong et al. (2000) and Rajkumar and Palanivelu (2004) investigated the use of electrocoagulation technology to treat wastewaters containing phenol and phenolic compounds.

Pulsed electrocoagulation technology is a novel method for wastewater treatment. It uses the interactions of electrochemical and polarity in an electrical field to induce dipole formation in nonpolar particles in the wastewater, thus enabling the formation of micro-aggregates of insoluble substances. The aggregates formed are further assisted in forming macro-aggregates; Fe(OH)₃ or Al(OH)₃ are produced and act as coagulants. Charge neutralization of ions or charged materials also takes place in the electrochemical reactor, turning them into insoluble, suspended substances in the wastewater. The neutralization process enhances the efficiency of removing electrical conductivity.

The purpose of this study was to assess the possibility of using pulsed electrocoagulation technology to treat the wastewater of an old corrugated containerboard (OCC)-based paper mill for water reuse. Untreated wastewater was pumped to the homogenization tank for equalization so as to avoid the confounding effect of inflow wastewater quality variations on treatment results. Each set of experiments was carried out with 1,000 L of equalized wastewater that was fed to the reactor for treatment. Further, the treated wastewater passed through the aerator and the flocculation tank, and then flowed into the sedimentation unit for sludge settling. Untreated wastewater was further supplemented with coagulant to study the effect of coagulant addition.

**Materials and Methods**

**Wastewater Characteristics**

The wastewater used in the experiments was a combined effluent of the mill’s untreated wastewater. As there were substantial variations in water quality measurements, even from moment to moment, a combined effluent of 1,000 L was equalized in a homogenization tank before use in each set of experiments so as to avoid variations that might skew the treatment results.

**Apparatus**

**Treatment system.** Figure 1 presents the schematic diagram of the treatment system used in the study. The system was composed of: a perfectly mixed homogenization tank of 1,000 L; a 65-L pilot-scale alternating electrocoagulation reactor; a 78-L aerator; a 100-L flocculation tank, to allow flocs sufficient time to grow; and a 120-L sedimentation unit.

**Pulsed electrocoagulation reactor.** A pulsed electrochemical reactor (Model JC-10), developed by Jye-shi Environmental Technologies & Engineering Inc. of Taiwan, was used in the study (Fig. 2). The reactor is capable of treating 0.1 to 0.5 m³/h of wastewater with an HRT of 0.13 to 0.65 h.

There were 16 electrolytic cells inside the reactor, separated by cast iron plates as electrodes. Figure 3 shows the setup of the electrodes. For the reactor, the relationship between current density and voltage is indicated in Table 1. The operational current density for the reactor was between 160 and 267 A/m², and the electrical potential per cell was 7.2 to 10.9 V.

**Operating Procedures**

Untreated wastewater was pumped to the homogenization tank for equalization so as to avoid the confounding effect of inflow wastewater quality variations on treatment results. Each set of experiments was carried out with 1,000 L of equalized wastewater that was fed to the reactor for treatment. Further, the treated wastewater passed through the aerator and the flocculation tank, and then flowed into the sedimentation unit for sludge settling. With the use of a 3-hp compressor and air diffuser, air was supplied to the
aerator. Based on a particular set of operating conditions chosen to carry out an experiment, polymer coagulant was added to the flocculation tank. The sludge settling in the sedimentation unit was collected and subsequently dewatered.

During operation, the polarity of the electrodes was switched every 30 to 60 s to avoid excessive iron plate erosion. The switching of polarity also produced a pulsed effect that caused dipole inductions in non-polar insoluble substances in the wastewater.

Three sets of experiments were conducted, and each set was performed with one specific HRT, five current densities and four coagulant dosages (excluding the conditions of zero current density and zero coagulant dosage), such that each set contained nineteen experiments.

Operating Conditions

The values of three operating variables studied are listed below:

- Current density (A/m²): 0, 40, 107, 187 and 240;
- HRT in the reactor (min): 8, 10 and 16; and
- Coagulant (an anionic polyacrylamide) dosage (mg/L): 0, 10, 20 and 30.

pH and temperature were not controlled in any of the experiments, because these two parameters varied with the untreated wastewater quality, the HRT and the current density that was applied. Consequently, the operative pH ranged between 6.1 and 7.7, and temperature varied between 31 and 34°C.

Instruments and Methods for Chemical Analyses

The following instruments and methods were used to perform the chemical analyses:

- Conductivity meter: Suntex, SC-170; NIEA W203.51B
- SS: NIEA W210.56A
- COD: NIEA W517.50B
- Turbidity meter: Hach, Model 2100P; NIEA W219.51C
- pH meter: Mettler-Toledo, MP220; NIEA W217.51A
- Ion chromatography: Hitachi L-7470 using a #2740 column

The electrical conductivity, SS content, COD, turbidity, pH and temperature of the equalized wastewater were measured before and after treatment. Measurements of the iron content of untreated and treated wastewaters were made for a set of experiments run at different current densities for a fixed HRT and without the addition of coagulant.

Results and Discussion

Wastewater Characteristics

Table 2 shows combined wastewater properties observed during a period of two months. As shown, the variations in electrical conductivity, SS, filtered COD, turbidity, pH and temperature properties were substantial.
Iron Content of the Treated Wastewater

A major concern of using electrocoagulation for wastewater treatment is that iron will be released from the anodes and enter the wastewater as Fe\(^{2+}\) ions. If aeration is unable to oxidize and convert most of the Fe\(^{2+}\) ions coming from the reactor to Fe(OH)\(_3\), which combines with insoluble substances and leaves the system, then the residual iron may hinder subsequent water reuse due to the potential staining and scaling characteristics of the Fe ions. Table 3 indicates that in all current densities tested, the iron content of the treated wastewater was considerably lower compared with that of the untreated wastewater. Thus, this possible hindrance to the reuse of the treated wastewater did not pose a problem.

Effects of Operating Variables on the Treatment Performance

(A) Effects of current density and HRT without coagulant addition.

Electrical conductivity removal. Figure 4 shows the effects of current density and HRT on the removal of electrical conductivity. The results indicate that the removal increases with increasing current density and HRT. The highest removal was 47.7\% (from the untreated value of 2,070 \(\mu\)S/cm to the treated value of 1,082 \(\mu\)S/cm), obtained at 240 A/m\(^2\) with an HRT of 16 min. Electrical conductivity is a reflection of the concentration of ions in a solution. The decrease in conductivity indicates a reduction in the amount of ions. Thus, during treatment in the reactor, the reduction in the conductivity of wastewater was due to the charge neutralization of ionic species contained in it. The charge neutralization de-charges ionic species, causing them to become insoluble substances. These are removed through induced micro-aggregation and coagulated macro-aggregation (floc formation).

SS removal. Effects of current density and HRT on the removal of effluent SS are shown in Fig. 5. As illustrated, the removal increased with an increase in current density and in HRT; more than 95\% of the SS were removed from the wastewater after the electrocoagulation treatment followed by aeration. The highest removal obtained with a current density of 240 A/m\(^2\) and an HRT of 16 min was 99.3\% (from the untreated value of 2,690 mg/L to treated value of 20 mg/L), and the obtained value of 20 mg/L SS meets most national and regional effluent discharging standards.

The effectiveness of the SS removal is deemed to result from the following mechanisms. In the reactor, SS and colloidal impurities become polarized due to the electrical field, and form micro-aggregates through a bridging effect. The micro-aggregates then coagulate to form flocs due to the flocculation action of the Fe(OH)\(_3\), generated by the aeration of dissolved ferrous ions (Holt et al. 1999; Robinson 1999).

COD removal. Figure 6 shows the effects of current density and HRT on the removal of COD. Similar to cases of conductivity and SS removal, COD removal also increased as current density or HRT increased. The highest removal was 75\% (from the untreated value of 2,280 mg/L to the treated value of 570 mg/L) obtained at 240 A/m\(^2\) with an HRT of 16 min. The removal of COD might have been due to the

<table>
<thead>
<tr>
<th>TABLE 1. Current density vs. voltage for the 16 electrolytic cells</th>
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<tbody>
<tr>
<td>Current density</td>
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<tr>
<td>-----------------</td>
</tr>
<tr>
<td>12 A/m(^2)</td>
</tr>
<tr>
<td>14 A/m(^2)</td>
</tr>
<tr>
<td>16 A/m(^2)</td>
</tr>
<tr>
<td>18 A/m(^2)</td>
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<tr>
<td>20 A/m(^2)</td>
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<table>
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<th>TABLE 2. Basic properties of the untreated wastewater</th>
</tr>
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<td>Conductivity ((\mu)S/cm)</td>
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<tr>
<td>-----------------------------</td>
</tr>
<tr>
<td>Range</td>
</tr>
<tr>
<td>Average</td>
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<th>TABLE 3. Iron content of the treated wastewatera</th>
</tr>
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<tbody>
<tr>
<td>Current density (A/m(^2))</td>
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<td>11.1</td>
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aHRT: 6.5 min, without the use of coagulant
attachment of dissolved substances to polarized suspended solids and polarized colloidal particles, which form flocs with the flocculation action of the Fe(OH)$_2$ in the reactor.

(B) Effects of adding coagulant to the flocculation tank. The coagulant added to the flocculation tank was an anionic polyacrylamide polymer (Rong-gi Co., Taiwan). The purpose of adding coagulant was to apply a supplementary treatment step after electrocoagulation and aeration, to further reduce the amounts of pollutants remaining in the wastewater. When ferrous ions generated by electrochemical reaction served as the coagulant, the maximum theoretical yield at a current density of 240 A/m$^2$ (18 A) and an HRT of 16 min would be 76.9 mg/L (ferrous ion concentration = 18 A x 16 min x 60 s x 55,850 mg/mole ÷ (2 x 96,500) ÷ 65 L = 76.9 mg/L). The theoretical yield is a linear function of the current density and HRT. Thus, the electrochemical generated coagulant and the traditional anionic polyacrylamide polymer dosage of 0 to 30 mg/L were in the same order of magnitude.

**Fig. 4.** Effects of current density and HRT on the removal of effluent electrical conductivity.

**Fig. 5.** Effects of current density and HRT on the removal of effluent SS.

**Fig. 6.** Effects of current density and HRT on the removal of effluent COD.

**Electrical conductivity removal.** Since the results indicated that high removal was obtained at an HRT of 16 min, it was decided to study the effects of current density and coagulant dosage on the removal of conductivity at an HRT of 16 min (Fig. 7). The removal increased with increases in both current density and coagulant dosage. Under the non-electrocoagulation condition, removal increased from 3% to 24% with an increase in coagulant dosage of from 0 to 30 mg/L; with electrocoagulation, the addition of the coagulant promoted further removal of conductivity. Using 30 mg/L of coagulant dose, removal was enhanced by 19% and 17.7% at current densities of 107 A/m$^2$ and 187 A/m$^2$, respectively, and the reduction in conductivity was due to charge neutralization of part of the positive ions remaining in the wastewater after electrocoagulation. No increase in conductivity removal was observed at a coagulant dosage of 30 mg/L and current density greater than 40 A/m$^2$. The results indicated that with the use of 30 mg/L of coagulant, a much lower current density (107 or 187 A/m$^2$) can be used to achieve a removal (50.5% or 52.1%) which is similar to that obtained with the highest current density of 240 A/m$^2$. The diminishing effectiveness of the increasing current density was probably due to intrinsic limitations in the capability of charge neutralization and mineralization to remove ionic species. Hence, 30 mg/L of coagulant should be considered an optimal dosage.

**SS removal.** Figure 8 shows the effects of current density and coagulant dosage on the removal of SS. As indicated, removal increased with an increase in current density and coagulant dosage. Without electrocoagulation, removal increased from 66% to 77% (from the untreated value of 2,040 mg/L to the treated value of 471 mg/L) with increasing dosage from 0 to 30 mg/L. The removals obtained at a current density of 40 A/m$^2$ were more than 95% for all dosages studied. For all dosages, the increases in removal obtained with current densities exceeding 40 A/m$^2$ were insignificant. As shown in Fig. 5, the removal of SS obtained without adding coagulant to the flocculation tank were also higher than 95%. Hence, the addition of coagulant has little or no effect on SS removal. The comparison suggested that the SS in the wastewater can be removed completely through electrocoagulation without
the use of a traditional coagulant which acts through charge neutralization, patch formation and bridging action on impurities. The highest removal rate was 99.6% (from the untreated value of 2,040 mg/L to the treated value of 8 mg/L) obtained with a current density of 240 A/m², an HRT of 16 min and a dosage of 30 mg/L.

**COD removal.** Figure 9 illustrates the effects of current density and coagulant dosage on the removal of effluent COD. As illustrated, removal increased with increases in both current density and coagulant dosage. Under the condition without electrocoagulation, the removal increased from 5% to 30.7% with the increase in coagulant dosage of 0 to 30 mg/L. With electrocoagulation, the addition of coagulant exhibited a remarkable effect on the increase in COD removal, particularly at current densities of 107 and 187 A/m²; the removals were enhanced by 30% (from 52% to 82%) and 32% (from 56.7% to 88.7%), respectively, with the addition of 30 mg/L of coagulant. As shown in Fig. 7, 30 mg/L of coagulant was the dosage needed to obtain an optimal COD removal of 88.7% with a lower current density of 187 A/m², compared with the removal of 92.7% of COD achieved with a current density of 240 A/m². The excess removal of COD might have been due to the formation of complexes of dissolved COD-containing substances with the coagulant polymer molecules. The highest removal was 92.7% (from the untreated value of 3,000 mg/L to the treated value of 220 mg/L), obtained with a current density of 240 A/m², an HRT of 16 min and a dosage of 30 mg/L.

Possible Chemical Reactions in the Treatment System

In accordance with the knowledge of electrochemistry that takes place inside electrolytic cells, the possible chemical reactions in the main components of the treatment system can be described as follows (Potter 1970; Holt et al. 1999):

(A) **Reactions at the anode.**

- Dissociation of iron from the electrode:
  \[ \text{Fe} \rightarrow \text{Fe}^{2+} + 2e^- \]  

(B) **Reactions at the cathode.**

- Generation of oxygen gas:
  \[ 4\text{OH}^- \rightarrow 2\text{H}_2\text{O} + \text{O}_2 + 4e^- \]  

(C) **Reactions in the electrocoagulation reactor.**

- Conversion of Fe²⁺ ions to Fe(OH)₃:
  \[ \text{Fe}^{2+} + 2\text{HO}^- \rightarrow \text{Fe(OH)}_3 \]
• Conversion of most of the Fe(OH)_2 to Fe(OH)_3 by oxidation:

\[ 4 \text{Fe(OH)}_2 + \text{O}_2 + 2\text{H}_2\text{O} \rightarrow 4 \text{Fe(OH)}_3 \quad (6) \]

• Formation of ferric hydroxide into a bulky, gelatinous floc due to its insolubility in water.

• Neutralization of the charges of the ionic species of dissolved substances, rendering them insoluble, suspended substances.

• Self-aggregation of suspended substances and colloidal particles due to the formation of dipoles and charge realignment.

• Formation of a substantial amount of the aggregates into coagulates as flocs, as the result of the Fe(OH)_3 produced.

(D) Reactions in the aerator.

• Conversion of the remaining Fe(OH)_2 to Fe(OH)_3, as a result of contact with air bubbles:

\[ 4 \text{Fe(OH)}_2 + \text{O}_2 + 2\text{H}_2\text{O} \rightarrow 4 \text{Fe(OH)}_3 \quad (7) \]

• Coagulation of the remaining micro-aggregates to form flocs, due to the extra Fe(OH)_3 produced.

• Flotation of the coagulated aggregates to the aerator surface.

Statistical Analysis of the Effects of Operating Conditions on Treatment Performance

The effects of the three operating variables on the treatment performance were also analyzed statistically, using a factorial experimental design of 2^3 (three variables at two levels). The two levels of current density (X1), HRT (X2) and coagulant dosage (X3) were 40 and 240 A/m^2, 8 and 16 min and 0 and 30 mg/L, respectively. The three removal results according to the design are presented in Table 4. The main effects and the interactions on the removals as well as their statistical significances as represented by the F-values are shown in Table 5. The significance was analyzed with the F-test.

For conductivity and COD removal, the effects of current density and HRT were both highly significant at \( p < 0.01 \). The effect of current density on SS removal was highly significant at \( p = 0.01 \), while that of HRT was insignificant. All interactions were insignificant. These results indicate that the higher the current density, the greater the removals of conductivity, SS and COD became. The conductivity and COD content of wastewater are related to the amount of soluble substances present in it. The removal of these two pollutants suggests a reduction in the amount of soluble substances as well.

The increases in removal efficacies were due to the higher power input (current density x voltage x HRT), which probably induced the following mechanisms: (1) charge neutralization took place, (2) a significant quantity of iron ions was released from the anodes, (3) a high amount of Fe(OH)_3 was generated, (4) more polarization of both dissolved and colloidal particles occurred, and (5) a high amount of flocs (macro-aggregates) formed.

Practical Implication on the Treatability of OCC-based Paper Mill Process Streams for Reuse

Ackermann et al. (2000) noted that with brown paper grades, including those from OCC-based paper mills, effluent discharges of 0 to 2.5 m^3/t can be reached and COD values above 10,000 mg/L are common in closed OCC lines. Diedrich et al. (1997) reported that in a totally closed board mill, COD concentration in the process increased to 35,000 mg/L and a combination of anaerobic-aerobic treatment produced minor reduction in conductivity. Further, the system had a high salt content. These reports suggested that high concentrations of COD and salts in the process water of OCC mills were tolerable to certain extent. Although the electrocoagulation treatment could not achieve optimal effluent water quality for many parameters, if the unit was used to treat the entire stream of the process water internally, the removal rates for COD and conductivity obtained could allow perpetual recirculation of the process white water. Thus, the pulsed electrocoagulation process can be an economic and viable technology to minimize OCC mill water use.

<table>
<thead>
<tr>
<th>X1 (Current density (A/m^2))</th>
<th>X2 (HRT (min))</th>
<th>X3 (Coagulant dosage (mg/L))</th>
<th>Conductivity removal (%)</th>
<th>SS removal (%)</th>
<th>COD removal (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
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<td>0</td>
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<td>47.7</td>
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<td>30</td>
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Conclusions

The results obtained during the study were favourable and showed that electrocoagulation technology can be used to treat the wastewater of an OCC-based paper mill for water reuse. The technology was effective for SS removal and more than 95% of the SS could be removed using a current density of only 40 A/m² and an HRT of 8 min, and without addition of a coagulant. Under the operating conditions studied without adding coagulant, the removals of conductivity and COD always increased with an increase either in current density or HRT. The highest removals obtained for conductivity, SS and COD were 47.7%, 99.3% and 75%, respectively.

The conductivity and COD removal could be further improved using a coagulant. Enhancements of up to 6.8% and 17.7% for conductivity and COD removal were obtained. The highest removals obtained with the optimal coagulant dosage of 30 mg/L for conductivity, SS and COD were 34.5%, 99.6% and 92.7%, respectively. Improvement in water quality rates obtained by the treatment indicated that at least a good part of the treated wastewater can be effectively reused as makeup process water in papermaking.

Acknowledgments

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### Table 5. Main effects, interactions and F-values from the analysis of variance (ANOVA)

<table>
<thead>
<tr>
<th>ANOVA</th>
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<th>Interaction</th>
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<tr>
<td></td>
<td>X1</td>
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<td>Conductivity</td>
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Electrocoagulation of OCC Paper Mill Wastewater


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