# Treatment of papermaking tobacco sheet wastewater by electrocoagulation combined with electrochemical oxidation

Xiangjuan Ma, Yang Gao and Hanping Huang

## ABSTRACT

Attempts were made in this study to examine the efficiency of electrocoagulation (EC) using aluminum (Al) anode and stainless steel net cathode combined with electrochemical oxidation with a β-$PbO_2$ anode or a mixed metal oxide (MMO) anode for treatment of papermaking tobacco sheet wastewater, which has the characteristics of high content of suspended solids (SS), intensive color, and low biodegradability. The wastewater was first subjected to the EC process under 40 mA/$cm^2$ of current density, 2.5 g/L of NaCl, and maintaining the original pH of wastewater. After 6 minutes of EC process, the effluent was further treated by electrochemical oxidation. The results revealed that the removal of SS during the EC process was very beneficial to mass transfer of organics during electrochemical oxidation. After the combined process, 83.9% and 82.8% of chemical oxygen demand (COD) removal could be achieved on the β-$PbO_2$ and MMO anodes, respectively. The main components of the final effluent were biodegradable organic acids, such as acetic acid, propionic acid, butyric acid, valeric acid, and hexahyl carbonic acid; the 5-day biochemical oxygen demand/chemical oxygen demand ($BOD_5$/COD) ratio increased from 0.06 to 0.85 (Al + β-$PbO_2$) or 0.80 (Al + MMO). Therefore, this integrated process is a promising alternative for pretreatment of papermaking tobacco sheet wastewater prior to biological treatment.

**Key words** | biodegradability, electrochemical oxidation, electrocoagulation, papermaking tobacco sheet, wastewater

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## INTRODUCTION

Papermaking tobacco sheet is a kind of reconstituted tobacco sheet (RTS) prepared via recomposing and processing, utilizing tobacco waste such as tobacco stems, leaf scraps, tobacco dust, and some parts of low-grade tobacco leaf (Potts *et al.* 2010). RTS has been widely used by the tobacco industry due to its advantageous economic impact on the manufacturing cost of cigars and cigarettes (Zhou *et al.* 2013). Presently, the papermaking process is a widely used method to manufacture RTS. The main procedures include extraction of tobacco waste by water, the concentration of extracts, pulping, papermaking, dip-coating, drying, and so on (Raquel *et al.* 2008; Wang *et al.* 2014). During the papermaking process, some organic components in tobacco waste transfer from the solid phase to an aqueous solution and then exist as organic pollutants, thus resulting in the complicated components of wastewater. According to the statistics of a tobacco sheet manufacturing company, approximately 60–80 $m^3$ of wastewater will be generated in the papermaking step for yielding 1 ton of tobacco sheet. Currently, the biological method is the preference choice of manufacturing companies due to the fact it is efficient and cost-effective. However, nicotine, solanone, and suspended solids (SS) that exist in tobacco sheet wastewater are biorefractory and even toxic to most microorganisms (Civilini *et al.* 1997; Wang *et al.* 2011b). In addition, large amounts of fine lignin, cellulose, and hemi-cellulose suspended in wastewater are recalcitrant. For these reasons, biodegradation is always unsatisfactory for effective abatement of tobacco sheet wastewater and causes a significant residual color in effluent. Recently, other approaches concerning the treatment of papermaking tobacco sheet wastewater have included coagulation–flocculation (Wang *et al.* 2014), the Fenton process (Ma 2009; Wang *et al.* 2011a), and the electrocoagulation (EC) process (Gao 2012). Among them, the EC process only utilizes electrons to facilitate wastewater treatment rather than

doi: 10.2166/wst.2015.057

using chemicals and microorganisms (Mollah *et al.* 2001). EC involves the generation of coagulants such as metal ions and different species of metal hydroxides *in situ* by dissolving sacrificial anodes such as aluminum (Al) and iron (Fe) upon application of a direct current, which causes the destabilization and aggregation of SS or precipitation and adsorption of dissolved pollutants in wastewater. In comparison, Al has been proven to be a more efficient performer than Fe due to the fact that polynuclear hydrolytic complexes (Equations (1) and (2)) ensure better adsorption of soluble and colloidal species from wastewater (Zidane *et al.* 2008). Simultaneously, hydrogen ($H_2$) released from the cathode (Equation (3)) during the EC process will be very beneficial to the floatation of SS out of wastewater (Bockris & Minevski 1994; Chen 2004).

Anode:

$$Al(s) \rightarrow Al^{3+} + 3e^- \quad (1)$$

$$Al^{3+} + nH_2O \rightarrow Al(OH)_n^{3-n} + nH^+ \quad (2)$$

Cathode:

$$2H_2O + 2e^- \rightarrow H_2(g) + 2OH^- \quad (3)$$

Accordingly, electrochemical oxidation with high oxygen evolution over-potential such as β-$PbO_2$ and boron-doped diamond (BDD) can generate highly reactive hydroxyl radicals (·OH) by water discharge (Equations (4)–(7)) under the mild conditions (Panizza & Cerisola 2009). It has been proven that toxic or biorefractory organics could be effectively destroyed under the successive attack of ·OH and converted to nontoxic or biodegradable organic acids, and even complete mineralization to $CO_2$ and $H_2O$ (Wu & Zhou 2001; Ma *et al.* 2012). Even so, when electrochemical oxidation is employed as the only treatment process, the high energy consumption will restrict its practical application. Furthermore, SS always existing in wastewater will influence the mass transfer during oxidation (Panizza & Cerisola 2010a, b). For that reason, using electrochemical oxidation as a refining technology in an integrated process consisting of EC followed by electrochemical oxidation will be more promising.

$$PbO_2 + H_2O \leftrightarrow Pb{*}O(OH)_2 \leftrightarrow H_2PbO_3 \quad (4)$$

Crystal layer hydrated (gel) layer

$$Pb{*}O(OH)_2 \leftrightarrow Pb{*}O(OH)^+(OH)^{\bullet} + e^- \quad (5)$$

$$Pb{*}O(OH)^+(OH)^{\bullet} + H_2O \rightarrow Pb{*}O(OH)_2 \cdots (OH)^{\bullet} + H^+ \quad (6)$$

$$BDD + H_2O \rightarrow BDD(\cdot OH) + H^+ + e^- \quad (7)$$

In this study, the treatment of papermaking tobacco sheet wastewater using a two-step process consisting of EC with Al anode and stainless steel net cathode followed by electrochemical oxidation with a β-$PbO_2$ anode modified with fluorine resin (Zhou *et al.* 2005) was investigated. In addition, a mixed metal oxide (MMO) anode, Ti/$TiO_2$-$RuO_2$-$IrO_2$, was chosen as the active anode not only due to its dimensional stability, low cost, and the ability to generate active chlorine *in situ*, but also because it has been commercially applied by the chlorine alkali industry and in other electrochemical processes (Rajkumar *et al.* 2005; Panizza & Cerisola 2009). The total treatment performance, biodegradability improvement, and the main components of effluent were investigated and compared. The experimental results can shed light on understanding the potential application of this integrated process for papermaking tobacco sheet wastewater treatment.

## METHODS

### Papermaking tobacco sheet wastewater

Wastewater used in this study was collected from the papermaking process of a tobacco sheet manufacturing company situated in Hangzhou (China), which produced approximately 1,600 $m^3$/day of wastewater; the characteristics of wastewater were relatively stable with the change of seasons. The wastewater was stored in a freezer at 4 °C in order to avoid deterioration, and the experiments were carried out within 7 days for each batch. The ranges of pH value, conductivity, and initial transmittance value of this wastewater were 6.7–7.0, 2.0–2.5 mS/cm, and 0.8%–1.0%, respectively. Cellulose, hemi-cellulose, and lignin existed extensively as the SS. Some non-volatile organic acids, alkaloids, tar, and other pollutants darkened the color of the wastewater. The other characteristics of the wastewater are presented in Table 1.

### Electrocoagulation and electrochemical oxidation processes

EC and electrochemical oxidation experiments were conducted under galvanostatic conditions in an 800 mL monopolar batch reactor (10 × 8.0 × 10 cm) made of

**Table 1** | Characteristics of papermaking tobacco sheet wastewater

| Parameters | Value |
|---|---|
| Total chemical oxygen demand ($COD_t$, mg/L) | 6,032–6,850 |
| Soluble chemical oxygen demand ($COD_s$, mg/L) | 2,178–2,959 |
| Five-day biochemical oxygen demand ($BOD_5$, mg/L) | 320–418 |
| SS (mg/L) | 1,588–2,086 |
| Turbidity (NTU) | 1,215–1,890 |

Plexiglas. In both processes, the anode and cathode were positioned vertically and parallel to each other with an inter-electrode gap of 15 mm. During experiments, the wastewater was stirred by the $H_2$ bubbles generated on the cathode.

In the EC process, the parameters, including current density, NaCl dosage, reaction time, and pH were controlled at the optimized conditions as in our previous work (Gao 2012). Six-hundred and fifty millilitres of wastewater maintaining the original pH value was primarily added into the reactor and 2.5 g/L of NaCl was used as supporting electrolyte. Then, an aluminum plate anode ($7.5 \times 5.8 \times 0.2$ cm) and a stainless steel net cathode ($7.5 \times 5.8 \times 0.1$ cm) were connected to a digital DC power supply (WYL3015, Hangzhou, China) and the current density was maintained at 40 mA/cm$^2$ with only slight adjustment of the applied voltage. After 6 minutes of reaction and 15 minutes of settlement, the effluent was withdrawn and analyzed.

Prior to the electrochemical oxidation process, the pH of the effluent pretreated by EC was adjusted to the desired value using NaOH or HCl solutions, then 650 mL of the effluent was added into the above-mentioned reactor. A β-$PbO_2$ (28.78 cm$^2$) with one side only exposed to the solution or an MMO plate ($7.5 \times 5.5 \times 0.1$ cm) operating on both sides and a corresponding size of stainless steel net were used as anode and cathode, respectively. The MMO electrode was prepared by a titanium equipment and manufacturing company located in Suzhou, China. During oxidation, samples were periodically withdrawn for chemical oxygen demand (COD) analysis. Three repetitions were performed for each treatment.

At the end of the integrated process, the main components and biodegradability enhancement of the final effluent were also investigated.

All chemicals used in this study were of analytical grade and purchased from Huadong Medicine Group Co. Ltd (Hangzhou, China).

### Analytical methods

During experiments, samples were periodically taken from the sampling port located about 4 cm below the liquid level and then maintained at a standstill for 15 minutes. The supernatant solution was taken for COD, SS, turbidity, and transmittance value analyses. Samples for determination of $COD_s$ were first filtered with 0.45 μm membrane. $COD_t$ and $COD_s$ were determined in accordance with the method 508 C (closed reflux, colorimetric method) in *Standard Methods* (APHA 1998). The pH was measured with a pH meter (pH3110 SET2, Germany). The transmittance value (in per cent) of wastewater or effluent was measured using a visible spectrophotometer (S23A, Shanghai, China) at the wavelength of 630 nm, which was standardized at 100% transmittance with distilled water. The turbidity and conductivity of wastewater were determined using the SGZ-2 digital turbidity meter (Shanghai, China) and YSI Model 30 conductivity meter (USA), respectively. The SS of wastewater was measured by gravimetric method. Five-day biochemical oxygen demand ($BOD_5$) was measured in an OxiTOP system (WTW, Germany) for investigating the biodegradability improvement of wastewater.

Dichloromethane (DCM) was used to extract the main organic components from wastewater after treatment; the ratio of water sample and DCM was 1:1 (v/v). After extraction, the DCM extract was concentrated to 10 mL with a rotary evaporator (R201BL, Shanghai, China) and analyzed by gas chromatography–mass spectrometry (GC–MS) using an Agilent 6890 N chromatograph coupled to a quadrupole Agilent 5975B inert XL mass selective spectrometer, which is equipped with an HP-Innowax column (Agilent 19091N-233, capillary 30.0 m × 250 μm × 0.5 μm). Helium (99.999%) was used as carrier gas at a constant flow of 0.7 mL/minute. The temperature of the injector was 250 °C and the injection volume was 2.0 μL in splitless mode. The column was held at 80 °C for 1 minute and then heated at 5 °C/minute to 240 °C and maintained at this temperature for 27 minutes. The characterization of the obtained spectra was conducted by comparing the mass spectra with those reported in the GC–MS library (NIST).

## RESULTS AND DISCUSSION

### Treatment of papermaking tobacco sheet wastewater by electrocoagulation

The papermaking tobacco sheet wastewater with 2.5 g/L of NaCl and maintaining the original pH was first treated by EC using Al anode and stainless steel net cathode at 40 mA/cm$^2$ of current density. After 6 minutes of EC, the

pH and conductivity of the effluent were 6.7 and 8.0 mS/cm, respectively. The characteristics of wastewater before and after EC are presented in Table 2.

As described in Table 2, almost all SS was removed from wastewater under the dual function of coagulation of polynuclear hydrolytic complexes and electrofloatation of $H_2$ generated in the cathode (Chen *et al.* 2000a), resulting in equal values of $COD_t$ and $COD_s$. Thus, it is unnecessary to distinguish the $COD_t$ and $COD_s$ in the following oxidation experiments. With the removal of SS, nearly 100% of turbidity removal was achieved and the transmittance value of wastewater increased from 0.9 to 86.4%. However, only 17.5% of $COD_s$ was removed, indicating that EC is more effective for the removal of SS, but not for the removal of dissolved organic compounds. This may be due to the fact that they are not entrapped and bridged by the flocs formed during EC (Chen *et al.* 2000b), consequently leading to the residual color of the effluent. The $BOD_5$/COD ratio increased from 0.06 to 0.42 after 6 minutes of the EC process. On the basis of this phenomenon, an electrochemical oxidation process was needed to further reduce the organic pollutants and enhance the biodegradability of effluent.

## Electrochemical oxidation of the effluent treated after EC

The effluent pretreated by EC with a residual COD of 2,350 mg/L was further treated by electrochemical oxidation using β-$PbO_2$ or MMO as anode. Figure 1 presents the effect of current density on the COD removal at the surface of the β-$PbO_2$ electrode. Considering that the dosage of NaCl added in the EC process was sufficient, only the pH needed to be adjusted to 5.0 to ensure good performance of β-$PbO_2$ before electrochemical oxidation (Gao 2012).

**Table 2** | Characteristics of the wastewater before and after treatment of electrocoagulation

| Parameters | Before treatment | After EC process | Removal (%) |
|---|---|---|---|
| $COD_t$ (mg/L) | 6,715 | 2,350 | 65.0 |
| $COD_s$ (mg/L) | 2,850 | 2,350 | 17.5 |
| Turbidity (NTU) | 1,750 | 0.9 | 99.9 |
| SS (mg/L) | 1,860 | 66 | 96.5 |
| $BOD_5$ (mg/L) | 402 | 980 | – |
| Transmittance value (%) | 0.9 | 86.4 | – |

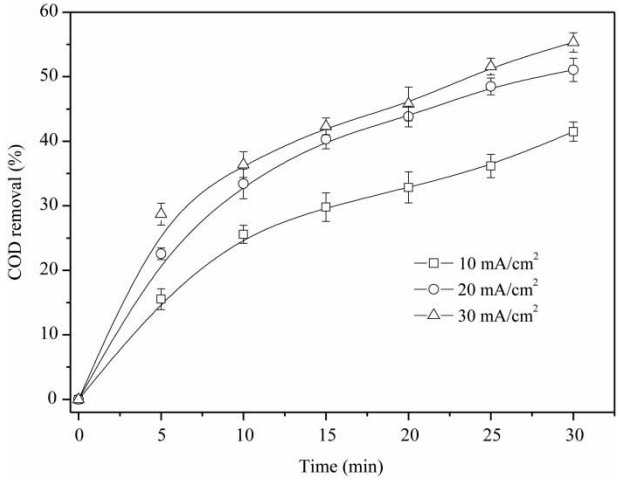



**Figure 1** | Effect of current density on COD removal of effluent at the surface of β-$PbO_2$ anode. Conditions: pH 5.0 and conductivity 8.0 mS/cm.

As shown in Figure 1, after 30 minutes of reaction, an increase in current density from 10 to 20 mA/$cm^2$ yields an increase of COD removal from 41.5 to 51.1% ascribing to much more ·OH generation at higher current density. A further increase of current density up to 30 mA/$cm^2$ does not produce significant improvements in the COD removal, but only an excessive energy consumption. For example, when the current density increased from 20 to 30 mA/$cm^2$ and 30 minutes of reaction, the COD removal increased from 51.1 to 55.3%. This may be due to the fact that the oxidation is controlled by the rate at which organic molecules are transferred from the bulk solution to the electrode surface rather than the rate at which hydroxyl radicals (•OH) are produced according to Equations (4)–(6). As a result, further increase of current density only results in the enhancement of the side reaction of oxygen evolution (Panizza & Cerisola 2010a). Taking into account both the COD removal and the energy consumption, 20 mA/$cm^2$ of current density was appropriate in the following experiments in the case of β-$PbO_2$ anode.

Figure 2 demonstrates the effect of current density on COD removal at the surface of the MMO anode and maintaining the pH and conductivity of the effluent pretreated by the EC process.

As shown in Figure 2, when MMO was the anode, the COD removal was not dependent on current density beyond 30 mA/$cm^2$, showing that the electrochemical oxidation of organics on the MMO anode was also limited by the rate of mass transfer. Thus, the appropriate current density was 30 mA/$cm^2$ in the case of the MMO anode.

Comparing Figures 1 and 2, it can also be found that the β-$PbO_2$ anode enabled significantly faster COD removal

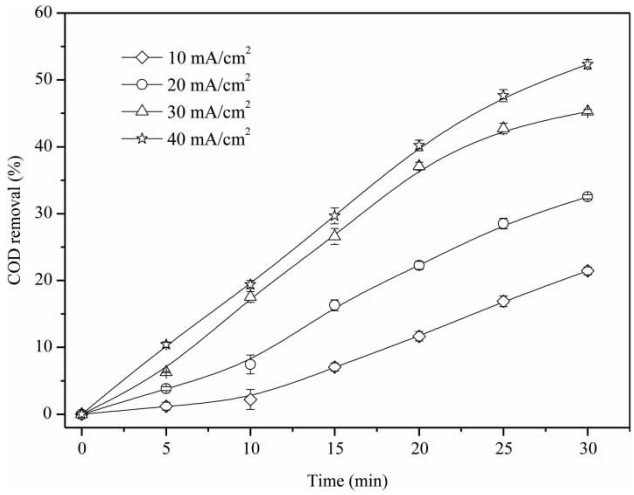



**Figure 2** | Effect of current density on COD removal of effluent at the surface of MMO anode. Conditions: pH 6.7 and conductivity 8.0 mS/cm.

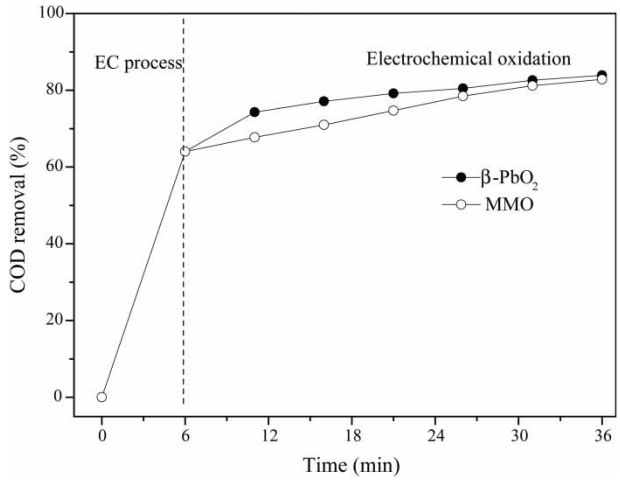



**Figure 3** | Evolution of COD removal during the combined process. Conditions: EC process: current density 40 mA/cm$^2$, reaction time 6 minutes, and pH 6.8. Electrochemical oxidation: reaction time 30 minutes, current density 20 mA/cm$^2$ (β-$PbO_2$) and 30 mA/cm$^2$ (MMO), and pH 5.0 (β-$PbO_2$) and 6.7 (MMO).

than the MMO anode. For example, at 30 mA/cm$^2$ of current density and after 30 minutes of reaction, 55.3% and 45.1% of COD removal were obtained on the β-$PbO_2$ and MMO anodes, respectively. The greater oxidation ability of β-$PbO_2$ can be explained by the higher reactivity of ·OH electrogenerated on this electrode. However, the MMO anode could perform well in a wide range of pH, namely, no pH adjustment was needed, implying that this could save operating costs in practical applications.

## Treatment of papermaking tobacco sheet wastewater by EC combined with electrochemical oxidation

On the basis of the above results, the combined process used for treatment of papermaking tobacco sheet wastewater consists of 6 minutes of EC at 40 mA/cm$^2$ of current density followed by 30 minutes of electrochemical oxidation at 20 mA/cm$^2$ (β-$PbO_2$) or 30 mA/cm$^2$ (MMO) of current density. The variation of COD removal during this combined process is presented in Figure 3. It can be observed that COD decreased rapidly during the EC process and 64% of COD could be removed; the residual organics in the pretreated effluent were further degraded by electrochemical oxidation.

After the integrated process, the COD of wastewater was decreased from 6,715 to 1,081 mg/L (β-$PbO_2$) and 1,155 mg/L (MMO); the $BOD_5$ of the effluent was 918 mg/L (β-$PbO_2$) and 924 mg/L (MMO). Therefore, the total COD removal was 83.9% (β-$PbO_2$) and 82.8% (MMO); the corresponding $BOD_5$/COD ratio increased from 0.06 to 0.85 (β-$PbO_2$) and 0.80 (MMO), respectively.

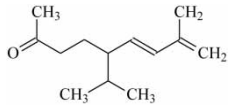
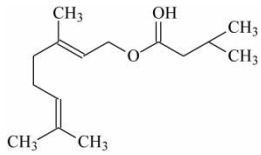
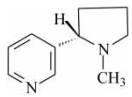
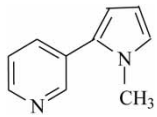
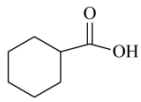
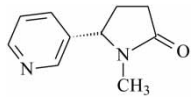
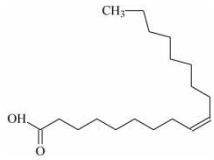
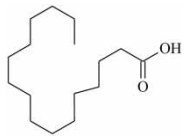
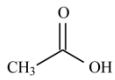
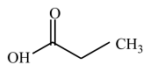
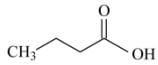
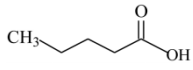
## Comparison of the main components in effluent after integrated process

To examine the main components in the effluent pretreated by EC and integrated process, the effluent was extracted by liquid–liquid extraction using DCM as an extractant and then characterized by GC–MS. The analysis results are listed in Table 3.

After EC, solanone, nicotine, β-nicotyrine, cotinine, geranyl isovalerate, hexahyl carbonic acid, oleic acid, and palmitic acid still existed in the effluent, confirming that the main dissolved organic pollutants and organic acids in papermaking tobacco sheet wastewater were difficult to eliminate through only the EC process. According to the comparison of peak area prior to and after EC, 40.5% of nicotine and 47.5% of solanone could be removed through the entrapping of coagulants generated. Whereas, after further electrochemical oxidation of the effluent on the β-$PbO_2$ anode, hexahyl carbonic acid, acetic acid, propionic acid, butyric acid, and valeric acid were detected as the main organic acids, indicating that solanone, nicotine, β-nicotyrine, cotinine, and geranyl isovalerate were transformed into small molecular organic acids under the successive attack of non-selective ·OH generated on the surface of β-$PbO_2$. With the elimination of these recalcitrant and toxic components, the inhibition and the toxicity of this wastewater to microorganisms were reduced, thus the biodegradability of wastewater was significantly enhanced and the $BOD_5$/COD ratio increased to 0.85.

Compared with the β-$PbO_2$ anode, the MMO anode seemed to be of low efficiency due to the lower oxidation

**Table 3** | Comparison of the main components of effluent treated after EC and integrated process

| Compounds | CAS no. | Structure | After EC process | After integrated process | |
|---|---|---|---|---|---|
| | | | | Al + β-$PbO_2$ | Al + MMO |
| Solanone | 54868-48-3 |  | √ | × | × |
| Geranyl isovalerate | 109-20-6 |  | √ | × | × |
| Nicotine | 54-11-5 |  | √ | × | × |
| β-Nicotyrine | 487-19-4 |  | √ | × | × |
| Hexahyl carbonic acid | 98-89-5 |  | √ | √ | √ |
| Cotinine | 486-56-6 |  | √ | × | × |
| Oleic acid | 112-80-1 |  | √ | × | √ |
| Palmitic acid | 57-10-3 |  | √ | × | × |
| Acetic acid | 64-19-7 |  | × | √ | √ |
| Propionic acid | 79-09-4 |  | × | √ | √ |
| Butyric acid | 107-92-6 |  | × | √ | √ |
| Valeric acid | 109-52-4 |  | × | √ | × |

*Note:* √ and × refer to detected and undetected, respectively.

power of oxidants generated on the MMO anode, such as active chlorines. Except for acetic acid, propionic acid, and butyric acid, part of hexahyl carbonic acid and oleic acid were detected in the effluent after treatment by the integrated process. Considering that the MMO anode could be applied in a wide range of pH and no pH

adjustment was needed, and the $BOD_5$/COD ratio of the effluent also increased to 0.80, the MMO anode may be more suitable for practical application.

According to the above analysis results of components remaining in the effluent after the integrated process, although the final COD was still above 1,000 mg/L and $BOD_5$ above 900 mg/L, the effluent could be directly treated by biological method due to the fact that the residual low molecular mass organics are biodegradable.

## CONCLUSIONS

The experimental results indicated that papermaking tobacco sheet wastewater can be effectively treated by EC combined with an electrochemical oxidation process. In such a combined process, EC primarily plays the role of destabilizing and aggregating the fine suspended particles, whereas electrochemical oxidation is responsible for eliminating the recalcitrant compounds existing in the effluent pretreated by EC. SS existing in wastewater could be almost completely removed through the EC process. This is beneficial for enhancing the performance of electrochemical oxidation and reducing its energy consumption. Moreover, both anodes have evidenced their great ability to remove the dissolved organic compounds and improve biodegradability. After the integrated process treatment, the effluent was transparent, the total COD removal was 83.9% (β-$PbO_2$) and 82.8% (MMO), the $BOD_5$/COD ratio increased from 0.06 to 0.85 (β-$PbO_2$) and 0.80 (MMO). This study also showed that EC for treatment of papermaking tobacco sheet wastewater has the advantages of short reaction time, no addition of chemicals, and no adjustment of pH. Electrochemical oxidation could effectively convert the recalcitrant compounds into organic acids through direct and indirect oxidation, and thus the combination of EC and electrochemical oxidation could be used as a promising technology for pretreatment of papermaking tobacco sheet wastewater ahead of biological methods.

## ACKNOWLEDGEMENT

This work was supported by Zhejiang Provincial Natural Science Foundation of China (Grant Nos Y4080335 and LY12E08014).

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First received 24 July 2014; accepted in revised form 26 January 2015. Available online 21 February 2015