ELECTROCHEMICAL OXIDATION OF LEACHATE PRETREATED IN AN UPFLOW ANAEROBIC SLUDGE BLANKET REACTOR

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(Received 3 April 2000; Accepted 8 October 2000)

ABSTRACT

This study was conducted to develop a two-stage process for the treatment of Hong Kong leachate. The leachate, containing 4750 mg l¹¹ of chemical oxygen demand (COD) and 1310 mg l¹¹ of ammoniacal nitrogen (NH₃-N), was first treated in a UASB (upflow anaerobic sludge blanket) reactor at 37°C. The process removed on average 66.1% of COD with 6.1 days of hydraulic retention time (HRT), corresponding to an organic loading rate of 0.78 g-COD (l·day)¹. The UASB effluent, which had an average pH of 8.9, COD of 1610 mg l¹ and NH₃-N of 1480 mg l¹, was then further treated by electrochemical oxidation in a batch reactor with recirculation. With the addition of 2000 mg l¹ of chloride (in the form of sodium chloride) and at a current density of 32.3 mA cm², 87% of COD and 100% of NH₃-N were removed from the UASB effluent in six hours. The energy consumption was <55 kWh kg-COD¹. The final effluent contained no residual NH₃-N and only 209 mg l¹ of COD.

Keywords: Ammoniacal-nitrogen, chloride, electrochemical-oxidation, leachate, UASB

INTRODUCTION

Solid wastes are degraded anaerobically in landfills in two distinct phases. Young landfills are normally operated in the acidogenic phase, the leachate of which contains high levels of readily biodegradable organics. Mature landfills are, however, operated at the methanogenic phase. Leachate from mature landfills has relatively lower organic strength, as anaerobes have converted easily biodegradable matters into methane inside the landfill. The organic residues in such a leachate are refractory to further biodegradation. Furthermore, such a leachate often contains high a concentration of ammoniacal nitrogen (NH3-N) which is toxic to many aquatic species and costly to remove. Hong Kong has 6.8 million inhibitants, producing 9500 tonnes of municipal solids waste daily. All of these wastes are collected and disposed of at three landfill sites. The landfill leachate, rich in organic residues and NH3-N, is treated in oxidation ponds on site continuously for 30 days [1]. The process is energy intensive and costly. It is, thus, desirable to develop a more cost-effective alternative.

This study was conducted to develop a two-stage

process for the effective treatment of Hong Kong landfill leachate. At first, the organics in the leachate are degraded using the upflow anaerobic sludge blanket (UASB) process. The residual organics and NH3-N in the UASB effluent were then further treated by electochemical oxidation. The UASB process was chosen as the first stage of leachate treatment because it has been successfully commercialized in the past decade for the treatment of wastewater from various industries. The process was effective in degrading organic chemicals of different nature, including not just fatty acids [2] and starch [3], but also refractory proteins [4] and even aromatics [5]. However, since the anaerobic process alone cannot effectively remove NH3-N and the refractory organics, a further treatment is often needed. Advanced oxidation processes, such as Fenton coagulation and ozonation, have been demonstrated to be effective for the post-treatment of the non-biodegradable organic pollutants [6, 7], but they are ineffective for NH₃-N removal. Traditionally, NH3-N in wastewater is removed by biological nitrification/denitrification, air-stripping, ion exchange, membrane processes or chemical precipitation [8]. All of these processes are, however, costly.

In recent years, electrochemical oxidation has been demonstrated to be a promising means for the destruction of refractory organics [9] and NH₃-N [10]. Pollutants can be oxidized electrochemically either directly or indirectly. In the former process, pollutants are oxidized by electron transfer at the anode surface [11, 12]. For example, ammonia can be directly oxidized to nitrogen gas at the anode as follows:

$$2NH_3 + 6OH^- \rightarrow N_2 + 6H_2O + 6e^-$$
 (i)

In the latter process, pollutants are oxidized by hypochlorite [12], a strong oxidant produced by the electrochemical reaction in the presence of chloride. Since landfill leachate often has high concentrations of chloride [13, 14], the electrochemical oxidation process was thus chosen for the post treatment of the UASB effluent. The effects of operating factors, such as initial pH, current density, flow velocity and chemical dosage on the organic and NH₃-N removal efficiency, as well as the current efficiency, energy consumption and the power requirement, were evaluated.

MATERIALS AND METHODS

UASB process

Leachate from a Hong Kong landfill was first treated in a 2.8-liter UASB reactor [15] with an internal diameter of 84 mm and a height of 500 mm. On top of the reactor was a gas-liquid-solid separator with an internal diameter of 114 mm and a height of 250 mm, making a filled volume of 2.0 l. Volumetric loadings were calculated based on the reactor volume alone, excluding the volume of the separator. The reactor was water-jacketed and operated at a constant temperature of 37°C for 354 days. The reactor was seeded with one liter each of UASB sludge (26.7 g l-1) and settled sludge (10.8 g l-1) from the oxidation pond at the landfill site. The UASB seed sludge was obtained from a previous study treating wastewater containing proteinaceous carbohydrate pollutants. A variable-speed peristaltic pump (Cole-Parmer, Masterflex) was used to feed leachate into the reactor, forcing an equal volume of supernatant out of the effluent line.

Electrochemcial oxidation

The experiments were conducted batchwise in a 265-ml batch electrolytic cell (Figure 1) under a constant direct current density. The cylindrical electrolytic cell was 235 mm in height with an internal diameter of 40 mm. The outer wall served as the cathode, whereas the 12-mm anode was located at the center. Both electrodes were made of titanium, coated with SnO₂. The surface areas of anode and cathode were about 87 cm² and 293 cm², respectively. Current density in this study was calculated based on the anode surface area. In each batch, 500 ml of UASB effluent in a 2-l holding tank was fed to the bottom of the electrolytic cell using a variable-speed peristaltic pump (Cole-Parmer, Masterflex). An equal volume of solution was forced out and recirculated back to the holding tank.

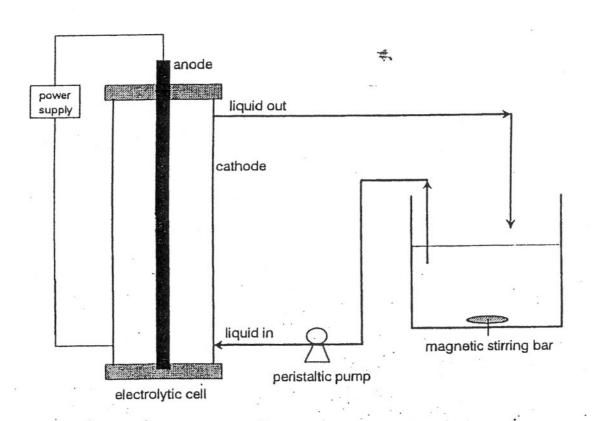


Figure 1. Schematic diagram of electrochemical oxidation experimental setup.

Wastewater parameters, such as COD (chemical oxygen demand), BOD₅ (5-day biochemical oxygen demand), chloroform and NH₃-N, were measured following the procedures of the *Standard Methods* [16]. Contents of VFA (volatile fatty acids) were measured following the procedures reported previously [15]. Nitrate and chloride contents were analyzed using an ion chromatograph (LC-10, Shimadzu) equipped with the AllsepTM anion column.

RESULTS AND DISCUSSION

UASB performance

The UASB reactor was operated at 37°C continuously for 354 days treating leachate with COD ranging 4750-15700 mg l⁻¹. Thoughout the experiment, the pH of mixed liquor in the reactor was pH 7.1-8.5, which was mainly due to the high alkalinity buffer in the leachate. When the COD loading was kept at less than 1 g-COD (l·day)⁻¹, the COD removal efficiency was maintained at 60%. It increased to 89-91% at the organic loading rate of 1.9-2.4 g-COD (l·day)⁻¹. Beyond this loading rate, the performance of the reactor deteriorated sharply; the COD removal efficiency declined linearly with the increase in organic loading rate and levelled off at 45%.

Table 1 summaries the characteristics of leachate and the UASB effluent under steady-state condition during days 269-354. During this period, the average loading rate was 0.78 g-COD (l·day)⁻¹, and the leachate contained on average 4750 mg-COD l⁻¹ and 1310 mg-NH₃-N l⁻¹. Although a high concentration of ammonia could inhibit the methanogenic activity [17], the UASB process was still capable of removing 66.1% of COD and 96.0% of BOD₅ at a hydraulic retention time (HRT) of 6.1 days The pH and the NH₃-N concentration in the mixed liquor were both increased after the UASB treatment, due to the conversion of organic nitrogen into NH₃-N. The UASB effluent containing 1610 mg-COD l⁻¹, 91 mg-BOD₅ l⁻¹ and 1480 mg-NH₃-N l⁻¹ was collected for the subsequent treatment of electrochemical oxidation.

Optimization of electrochemical oxidation

The electrochemical reactions, which take place during

Table 1. Characteristics of leachate and UASB effluent.

		Leachate	UASB effluent
pН		7.9 ± 0.4	8.9 ± 0.3
COD	(mg l-1)	4750 ± 510	1610 ± 170
NH ₃ -N	(mg l-1)	1310 ± 120	1480 ± 150
Chloride	(mg l-1)	1890 ± 30	2010 ± 40
BOD ₅	(mg l ⁻¹)	2270 ± 190	91 ± 25

the electrolysis of a brine solution, are complicated. The main electrolytic reactions for a chloride-rich aqueous solution can be described below:

Solution:
$$Cl_2 + H_2O \rightarrow HClO + Cl^- + H^+$$
 (ii)

$$HClO \rightarrow H^{+} + OCl^{-}$$
 (iii)

$$H_2O \rightarrow H^+ + OH^-$$
 (iv)

Anode:
$$2 \text{ Cl}^- \rightarrow \text{ Cl}_2 + 2e^-$$
 (v)

$$6OCl^{-} + 3H_{2}O \rightarrow 2ClO_{3}^{-} + 4Cl^{-} + 6H^{+} + 1.5O_{2} + 6e^{-}$$
(vi)

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$$
 (vii)

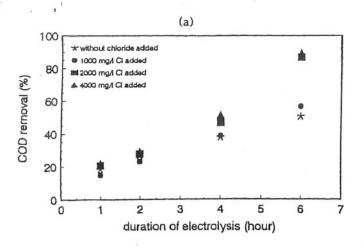
Cathode:
$$2H_2O + 2e^- \rightarrow 2OH^- + H_2$$
 (viii)

$$OCl^{-} + H_2O + 2e^{-} \rightarrow Cl^{-} + 2OH^{-}$$
 (ix)

In this study, a current density of 32.3 mA cm⁻², the maximum provided by the power supply, was used in most of the experiments, unless specified otherwise. A series of experiments were conducted to determine the optimum conditions for electrochemical oxidation of the UASB effluent.

Effect of chloride concentration:

During the electrochemical oxidation process, chloride in wastewater is oxidized to form chlorine (Reaction v), which is further hydrolyzed forming hydrochlorous acid (Reaction ii) and hypochlorite (Reaction iii). All of these species are very strong oxidants. Thus, in order to enhance the oxidizing effect, the addition of chloride has often been suggested for the electrochemical oxidation process [18, 14]. Even though the UASB effluent contained 2010 mg l-1 of chloride, experiments were conducted to determine if the addition of extra chloride (in the form of sodium chloride) would improve the COD removal efficiency for the electrochemical oxidation of UASB effluent. All experiments were conducted at the recirculation velocity of 4.0 1 h-1. Figures 2a and 2b illustrate that the COD removal efficiencies increased with the dosage of chloride. Without extra dosage of chloride, 45.5% of COD and 50.4% of NH3-N in the UASB effluent was removed after six hours of electrolysis. However, by adding 2000 mg l-1 of extra chloride, the corresponding removal effiencies were nearly doubled to 87% and 100%. Further addition of chloride to 4000 mg l-1 of chloride did not show any noticeable improvement. Thus, the addition of 2000 mg l-1 of extra chloride was adopted in the subsequent experiments. This dosage level was considerably lower than the 7500 mg-Cl l⁻¹ of extra dosage reported by Chiang et al. [19] in their study of electrochemical oxidation of leachate. This was most likely due to the lower organic content in the · UASB effluent of this study, compared to the wastewater treated in Chiang's study.



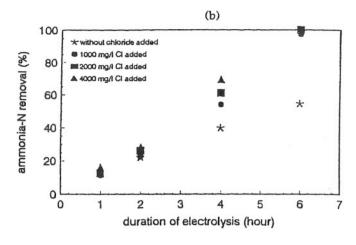


Figure 2. Effects of chloride concentration at initial pH 8.9 on (a) COD removal, and (b) NH₃ removal.

Effect of adding oxidation enhancers:

Experiments were conducted to compare the effects of adding two other oxdiation enhancers - sulfate and H_2O_2 - on the removal of COD and NH_3 -N. Sulfate was chosen because it is an electrolyte with strong ionic strength, and would produce oxygen at the anode [19], and thus has the potential to enhance oxidation. On the other hand, H_2O_2 was believed to be capable of accelerating electrochemical oxidation by generating hydroxyl radicals during electrolysis [20, 21]. Figures 3a and 3b illustrate the removal of COD and NH_3 -N, respectively, by the addition of sulfate (2700 mg I^{-1}) and H_2O_2 (200 mg I^{-1}) compared to the addition of chloride (2000 mg I^{-1}) and the control. All of these experiments were conducted at the initial pH of 6.0 and with a recirculation velocity of 4.0 l h⁻¹.

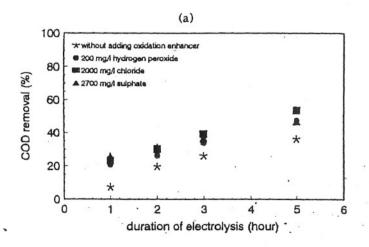
Figure 3 illustrates that without addition of any oxidation enhancers, the control electrochemical oxidation process removed 36% of COD and 47% of NH₃-N after five hours. Adding sulfate increased the COD removal to 47%, but did not have any noticeable effect on NH₃-N removal. On the other hand, adding H₂O₂ increased the COD removal to 47% and NH₃-N removal to 53%. In both cases, the increases of COD and NH₃-N removal were not as good as the

respective removals of 54% and 89% achieved by the addition of chloride. The superior performance of chloride addition, especially in NH₃-N removal, was due to indirect oxidation, caused by the formation of chlorine and its hydrolytic products, hypochlorous acid and hypochlorite, as shown in Reactions (ii) and (iii). NH₃-N was oxidized by hypochlorous acid forming chloramines and to nitrogen, when hydrochlorous acid is in excess [8].

Effect of initial pH:

Under acidic conditions, electrolysis produces hydrogen [22], which has a very strong reducing potential. Thus, in general, electrochemical oxidation prefers basic conditions. Three experiments were conducted at pH 7.5, pH 8.9 (the asis pH of the UASB effluent), and pH 10.0 to evaluate the effect of pH on COD and NH₃-N removals. All experiments were conducted by dosing 2000 mg l⁻¹ of extra chloride and with a recirculation velocity of 4.0 l h⁻¹. Figures 4a and 4b illustrate that at the as-is pH of 8.9 and pH 10.0, all the NH₃-N and 86-90% of COD were removed within six hours. At pH 7.5, the corresponding removal efficiencies were only 86% and 76%.

High pH favors the formation of hypochlorous acid, as



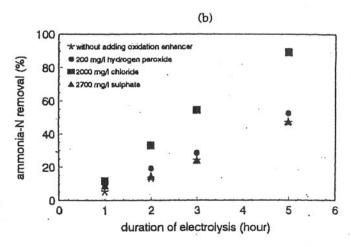
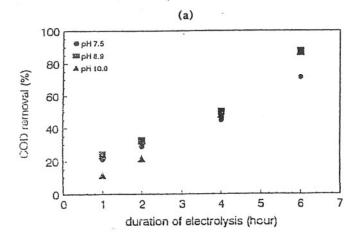


Figure 3. Effects of adding oxidation enhancer at initial pH 6 on (a) COD removal, and (b) NH₃ removal.



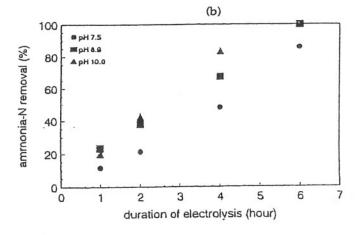


Figure 4. Effects of initial pH on (a) COD removal, and (b) NH₃ removal.

shown in Reaction (ii), which has a very high oxidation potential. Furthermore, at high pH most of the NH₃-N exists in the undissociated form, which is much easier to oxidize than in the dissociated form [23]. On the other hand, high pH also favors the formation of the stable ClO₃-, which does not have much oxidation potential. The results of this series of experiments suggest that the UASB effluent could be effectivley treated as-is without any pH adjustment.

Effect of recirculation flow velocity:

Three experiments were conducted at 0.4, 2.0 and 4.0 l h⁻¹ of recirculation velocity at pH 8.9 and with the addition of 2000 mg l⁻¹ of extra chloride. Increase of flow velocity would produce smaller chlorine bubbles, enhance mass transfer and reduce the diffusion layer. However, results illutrated in Figures 5a and 5b show that increase of recirculation velocity did not significantly improve the COD and NH₃-N removals.

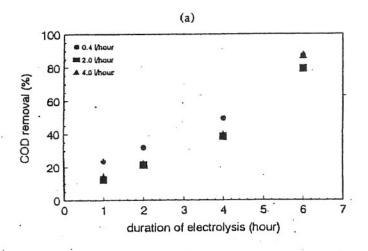
Effect of current density:

In this study, all but one of the experiments were

conducted at the current density of 32.2 mA cm⁻², the maximum density provided by the power supply. The exceptional experiment was conducted at the current density of 23.1 mA cm⁻², which is the minimum current which could be generated steadily. Figures 6a and 6b illustrate that the degradation of pollutants was independent of these two density currents.

Electrochemical oxidation of leachate and UASB effluent

Electrochemical oxidation was also conducted for the raw leachate. The experiment was conducted, with the addition of 2000 mg l⁻¹ of chloride but without pH adjustment, at 4.0 l h⁻¹ of recirculation and a current density of 32.3 mA cm⁻². About 69% COD and 67% NH₃-N were removed from the raw leachate after ten hours of electrochemical oxidation, compared to 87% COD and 100% NH₃-N removals from the UASB effluent after six hours, as illustrated in Figures 7a and 7b. Table 2 summarizes the compositions of leachate and UASB effluent after the electrochemical oxidation process.



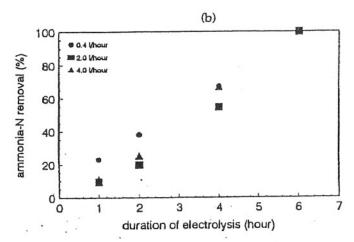


Figure 5. Effects of recirculation flow velocity on (a) COD removal, and (b) NH₃ removal.

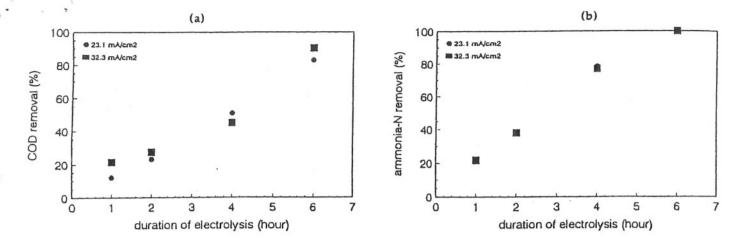


Figure 6. Effects of current density on (a) COD removal, and (b) NH₃ removal.

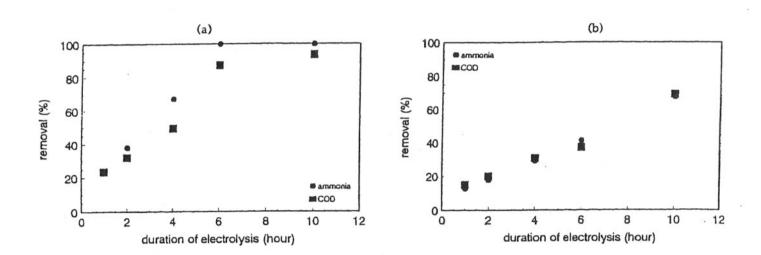


Figure 7. Electrochemical oxidation of (a) UASB effluent, and (b) raw leachate.

Table 2. Characteristics of leachate and UASB effluent after electrochemcial oxidation process.

	Leachate after	UASB effluent after 6-hour electrochemical
3 .	oxidation	oxidation .
COD (mg l ⁻¹)	1473	209
COD of total VFA (mg l-1)	96	207
NH ₃ -N-(mg l ⁻¹)	432	0
NO ₃ -N (mg l ⁻¹)	236	250
Chloride (mg l ⁻¹)	493	1288

The VFA in the UASB effluent after electrochemical oxidation, mainly consisting of acetate, butyrate, propionate, valerate, and caproate, accounted for 98.8% of the total COD remaing. It shows that most of the toxic, refractory pollutants in the leachate were converted into the harmless, easily biodegradable products. The remaining 1.2% of COD (about 2 mg-COD l⁻¹) was mainly due to the formation of

chlorinated organics, over 90% of which was chloroform. Similar observations of increased biodegradability were also reported in the other studies of electrochemical oxidation, including destruction of high molecular weight organics [9], detoxifications of benzoquinone [18] and tannery waste liquors [6].

Current efficiencies and energy consumptions

During carbonaceous oxidation, organic carbons are oxidized by oxygen forming carbon dioxide. Four electrons are transferred for each mole of oxygen consumed, as shown in Reaction (x).

$$O_2 + 4 e^- \rightarrow 2 O^{2-}$$
 (x)

Thus, removing every 32 g of COD (equivalent to consuming 1 g-mole of O_2) from wastewater requires four electrons. On the other hand, three electrons are transferred for each mole of NH₃-N oxidized to N₂ gas as shown in Reaction (i). Based on Reactions (i) and (x), removing each gram of COD from wastewater theoretically requires 12063 coulombs (96500x4/32), while oxidizing each gram of NH₃-N theoretically requires 20679 coulombs (96500x3/14). Thus, the

current efficiency of the electrochemical process can be calculated by comparing the current required for COD and NH₃-N removal with the actual current applied. Figure 8 illustrates that the current efficiency was 84% during the first hour of electrolysis of the UASB effluent, but it was gradually lowered to 58% after 4-6 hours. It also illustrates that the current efficiency was considerably lower for the electrochemical treatment of raw leachate. It is, thus, desirable to pretreat the leachate by the UASB process, instead of direct electrochemical oxidation.

Figure 9 illustrates that the energy consumption for the electrochemical oxidation of the UASB was <55 kWh (kg-COD)⁻¹ for six hours of treatment, but it was increased to 91 kWh (kg-COD)⁻¹ for ten hours, due to lowering of current efficiency. On the other hand, the energy consumption for raw leachate was at the level of 80 kWh (kg-COD)⁻¹ for 4-6 hours of treatment.

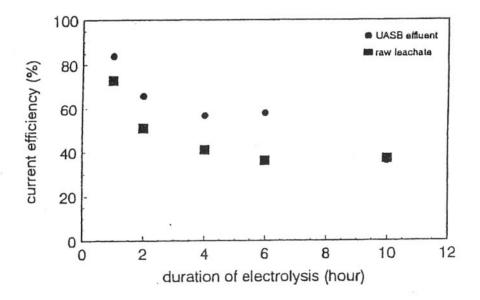


Figure 8. Current efficiency of electrochemical oxidation.

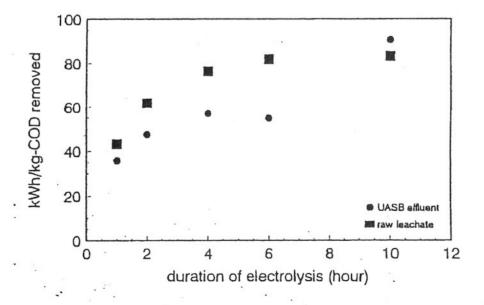


Figure 9. Energy consumption in electrochemical oxidation.

Energy requirement

Based on the energy consumptions for six hours of electrochemical oxidation, the energy requirements in treating the UASB effluent to meet various discharge standards are summarized in Table 3. It shows that about 210 kWh $\rm m^{-3}$ of energy was required to achieve the discharge standards of <200 mg-COD $\rm l^{-1}$ and the complete removal of NH₃-N.

CONCLUSIONS

The UASB pretreatment of leachate at 37°C removed 66.1% of COD with 6.1 days of HRT, corresponding to a loading rate of 0.78 g-COD (l·day)-1. The UASB effluent, which had a pH of 8.9, 1610 mg-COD l-1 and 1480 mg-NH₃-N l-1, was further treated by electrochemical

oxidation. With the addition of 2000 mg l⁻¹ of extra chloride (in the form of sodium chloride) and at the current density of 32.3 mA cm⁻², the process removed 87% of COD and 100% of NH₃-N from the UASB effluent within six hours. The energy consumption was <55 kWh (kg-COD)⁻¹. The final effluent contained no residual NH₃-N and only 209 mg l⁻¹ of COD.

ACKNOWLEDGEMENT

The authors are grateful to the Hong Kong Research Grants Council for the partial financial support of this study, to the Croucher Foundation for granting Herbert H. P. Fang the Croucher Senior Research Fellowship and Peng Wang the Croucher Chinese Visitorship, and to Dr. X. Y. Li for his technical assistance.

Table 3. Energy requirement of electrochemical oxidation of UASB effluent.

-	Energy requirements (kWh m ⁻³)			
Discharge standards	with 2000 mg l ⁻¹ of extra chloride		without extra chloride	
NH ₃ -N < 200 mg l ⁻¹	190	(COD < 450 mg l ⁻¹)*	390 (COD < 400 mg l ⁻¹)	
$NH_3-N \sim 0 \text{ mg } l^{-1}$	210	$(COD < 200 \text{ mg l}^{-1})$		
COD < 500 mg l ⁻¹	180	$(NH_3-N < 250 \text{ mg l}^{-1})$	350 (NH ₃ -N < 350 mg l ⁻¹)	
COD < 200 mg l ⁻¹	210	$(NH_3-N \sim 0 \text{ mg } 1^{-1})$	·	

^{*}The corresponding COD or NH₃-N residue is stated in brackets.

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