RESEARCH ARTICLE



Industrial-scale application of the plunger flow electro-oxidation reactor in wastewater depth treatment

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Abstract Effluents after biochemical treatment contain pollutants that are mostly non-degradable. Based upon previous pilot-scale test results, an industrial-scale electro-oxidation device was built to decompose these refractory materials in the effluent from a park wastewater treatment plant. The electrooxidation device comprised a ditch-shaped plunger flow electrolysis cell, with mesh-plate Ti/PbO₂ electrodes as the anode and the same size mesh-plate Ti as the cathode. Wastewater flowed vertically through electrodes; the effective volume of the cell was 2.8 m³, and the surface-to-volume ratio was 17.14 m² m⁻³. The optimal current density was 100 A m⁻², and a suitable flow velocity was 14.0 m h^{-1} . The removal efficiencies for chemical oxygen demand and color in the effluent were over 60.0 and 84.0 %, respectively. In addition, the electro-oxidation system offered a good disinfection capability. The specific energy consumption for this industrialscale device was 43.5 kWh kg COD^{-1} , with a current efficiency of 32.8 %, which was superior to the pilot-scale one. To meet the requirements for emission or reuse, the operation cost was \$0.44 per ton of effluent at an average price for electricity of \$0.11 kWh⁻¹.

Keywords Electro-oxidation \cdot Plunger flow \cdot Industrial scale \cdot Wastewater depth treatment \cdot Disinfection \cdot Economic evaluation

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Introduction

Most pollutants in wastewater can be removed efficiently through physical, chemical, and biological processes. However, some non-degradable organic contaminants (Kumar et al. 2008; Maljaei et al. 2009; Zhang et al. 2015), which might have potential toxic, mutagenic, and endocrine effects on living things, remain in the effluent from wastewater treatment plants (Ventura et al. 2008; Fatima et al. 2007; Pomati et al. 2008). It is critical to remove these pollutants before discharging the effluent into the environment, and increasingly strict standards have been imposed to further reduce emission of these pollutants. Various advanced technologies have been developed to meet the requirements for wastewater depth treatment, such as Fenton's reagent (Barros et al. 2006; Pliego et al. 2015), ozonation (Chys et al. 2015; Legube and Leitner 1999; Zhang et al. 2014), adsorption (Ramesh et al. 2005), membrane separation (Shen et al. 2014; Suárez et al. 2015), electro-oxidation ((Ángela et al. 2009; Zhu et al. 2015), and biofilters (Greenman et al. 2009).

Electro-oxidation, an electrochemical technology, has received growing attention because it requires no chemicals during pollutant removal (Chatzisymeon et al. 2006; Meas et al. 2010; Su et al. 2011; Zhu et al. 2011). The main uses of electro-oxidation are as a pretreatment to enhance the biodegradable properties of raw wastewater and/or as a deep treatment to decompose residual (refractory) matter (Katsoni et al. 2014). Garcia-Segura indicated that electro-oxidation processes have a greater effect on persistent and non-degradable pollutants from secondary biological treatment than traditional physicochemical methods (Garcia-Segura et al. 2015). Some researchers have systematically assessed the operation or capital costs of electro-oxidation in comparison with ozonation, Fenton processes, and membrane processes (Canizares et al. 2009; Suárez et al. 2015). These analogous explorations have established a sound foundation for the application of electro-oxidation technologies. The current efficiency and operation cost are the two key factors that determine its industrial-scale application on wastewater treatment.

Combining a plunger flow pattern and mesh-plate electrodes, a novel plunger flow electro-oxidation reactor (PFER) has been proposed to increase the current efficiency during the overall electrolysis process (Djati et al. 2001). Plunger flow, which flows like a piston through the mesh electrode assemblies with no backmixing and ensures every fluid flow unit receives the same electrolysis time, achieves excellent removal of contaminants in a serial flow device. Hydrodynamic studies indicate that the mesh-plate electrodes enable uniform distribution of the liquid flow field with little backmixing in the reacting field (Santos et al. 2010).

In our previous bench-scale and pilot-scale studies, some essential factors such as the electrode materials, flow patterns, and suitable current densities on the wastewater depth treatment by the novel PFER were presented detailedly. The economic efficiency was also preliminarily estimated (Zhu et al. 2015). These experimental results provided the basis for the subsequent industrial-scale application of the electrooxidation device in wastewater depth treatment. Detailed information on its industrial-scale application for park wastewater depth treatment is presented, which also provides valuable information for other effluent treatments, especially by electro-oxidation. 18289

Experiment

Industrial-scale plunger flow electro-oxidation device

Based on the plug flow model for reaction kinetics of pollutants in wastewater with electro-oxidation, the structure sizes of the electrochemistry reactor were calculated; the sectional area of the wastewater flowing through the reactor was designed due to the mass transfer (Wang et al. 2015). The design capacity of this industrial-scale electro-oxidation device was 500 t wastewater per day, consisting of cell bodies, electrodes, and constant current DC power supplements similar to the pilot-scale test, shown in Fig. 1. A ditch-shaped cell body with a polypropylene copolymer was designed to form piston-like flow, with an effective volume of 2.8 m³. The electrodes, mesh-plate Ti/PbO₂ electrodes as the anode and the same size mesh-structure titanium (Ti) plates as the cathode, were positioned alternately at distances of 10 mm, perpendicular to the water flow direction in each electrolytic unit. The Ti/PbO₂ anodes had been applied on large scale for several years, with no leaching of lead from the anodes into the effluent water. The surface-to-volume ratio, which was defined as the ratio of the working electrode (anodes in this device) surface area relative to the effective cell volume, was 17.14 m² m⁻³. A commercial DC power supplement was supplied with a maximum current range of 1000 A and a maximum voltage range of 50 V.

The effluent from the secondary sedimentation tank of a park wastewater treatment plant flowed into the ditch-shaped cells and perpendicularly through the electrodes. Samples were taken at four different positions in the cell body along

Fig. 1 Industrial-scale plunger flow electro-oxidation devices for wastewater depth treatment: DC power (1), electrolysis cell (2), inflow (3), outflow (4), and flow meter (5)



Table 1 The physicochemical characterization of wastewater

Water characteristics	Initial value	Electrolysis 30.0 min	Outlet criteria	Requirement to reused	Units
Chemical oxygen demand (COD)	130~160	40~60	60	q	mg L^{-1}
Color	160~220	16~32	30	30	times
pH	7.0-8.0	7.0-8.0	6–9	6.5-8.5	_
Fecal coliforms	1.0×10^5	<20	10^{4}	_	CFU mL ⁻¹
Chloridion	800~1000	800~1000	-	_	$mg L^{-1}$
Conductivity	7.50~8.00	7.50~8.00	_	-	$mS \cdot cm^{-1}$

the wastewater flow direction. Five current densities (60, 80, 100, 120, and 140 A m⁻²) and five flow velocities (2.0, 4.0, 7.0, 14.0, and 21.0 m h^{-1}) were applied, consistent with the pilot-scale study, to explore the effect of operating parameters (current density and flow velocity) on the current efficiency, which are directly related to the operation cost. The removal of chemical oxygen demand (COD) and color were selected as the two main indicators to evaluate the results.

To explore the influence of different flow velocities at the same electrolysis time, three identical sets of the abovementioned electrolysis cells were designed and installed in series or in parallel.

All electrolysis procedures were conducted under galvanostatic conditions. No chemicals were used during the electrolytic process.

Analytical methods

All of the water samples were taken from the electrolysis cells at fixed time spans every day.

The COD concentration was determined by the fast digestion-spectrophotometric method (HJ/T 399-2007), employing 0.24 g mL⁻¹ HgSO₄ solution to eliminate chlorine



Fig. 2 The effect of current density on COD removal. Current density 60 Am^{-2} (squares), 80 Am^{-2} (circles), 100 Am^{-2} (triangles), 120 Am^{-2} (inverted triangles), and 140 A m⁻² (stars); flow velocity 7.0 m h^{-1}

interference. The conductivity was ensured with a HACH LA-EC20 conductivity meter, while the pH value was measured with a Sartorius PB-21 pH meter. The color in sample was determined by the platinum-cobalt method in accordance with the water quality-determination of colority (GB 11903-89). The concentration of chlorine species was measured with the water quality-determination of chloride-silver nitration method (GB 11896-89), employing 50 g L^{-1} K₂CrO₄ solution as indicator. Fecal coliforms were determined by manifold zymotechnics (HJ/T 347-2007).

Results and discussion

Wastewater characteristics

The effluent came from secondary sedimentation tank of an industrial park wastewater treatment plan, still contained residual pollutants, most of which are non-degradable. The main physicochemical characteristics (pH, COD, color, and coliforms) of this effluent were measured, and the monitoring data were listed in Table 1. The results for wastewater treatment within 30.0-min electrolysis under the operating conditions (natural pH value, current density 100 A m^{-2} , flow velocity 14.0 m h^{-1}) were also listed in Table 1, compared with emission standards and reclaimed-water quality standard values.

The average values of COD and coliforms for the effluent were 141 mg L^{-1} and 1.0×10^5 CFU m L^{-1} , respectively. The average value of color was 180 times, and the colored organic matters in this wastewater were acid azo dye (Echt Brown, Acid Orange, Azorubine, etc.) and diazo dye (diazogen color, diazol fast orange, diazol violet, etc.) which were difficult to be degraded directly by microbial and contribute to COD value in wastewater. The conductivity of the effluent, 7.50 \sim 8.00 mS cm⁻², showed effluent good conductive and was suitable for electro-oxidation treatment without requiring addition of extra electrolytes (Sala and Gutiérrez-Bouzán 2014). Table 1 showed that the electro-oxidation process had an excellent capability to remove the residues from the effluent. The water qualities after an electrolysis time of 30 min met water quality standard for green space irrigation (GB/T

 Table 2
 The fitted curves of

 COD removal at different current
 densities

Current density	60 A m ⁻²	80 A m ⁻²	100 A m^{-2}	120 A m ⁻²	140 A m ⁻²
$-100 \times k(\min^{-1})$ R^2	2.00	2.40	2.71	2.83	2.85
	0.999	0.999	0.997	0.997	0.996

25499-2010), water quality standard for scenic environment use (GB/T 18921-2001), etc.

The influence of current density

Current density, which is defined as the electric current per unit cross-section area of the electrodes, is related to the reaction rate and influences the reaction energy consumption. A lower current density may be economic and save energy but may also prolong the electrolysis time of wastewater, which will enlarge the size and capital costs of the facility. On the contrary, a higher current density could shorten the electrolysis time and reduce the facility size but results in higher operation costs due to higher proportion of wasted electricity (Garcia-Segura et al. 2015). Thus, it is crucial to select a suitable current density after balancing the capital and operation costs.

Five applied current densities (60, 80, 100, 120, and 140 A m⁻²) were designed according to the pilot-scale study results and the construction scale of the project. As shown in Fig. 2, COD of the wastewater dropped rapidly with electrolysis time. The natural logarithm of COD_t/COD_0 had a strong linear relationship with electrolysis time for the correlation coefficients (R^2) that were all above 0.99 (as listed in Table 2). So, the removal of COD had an exponential relationship with electrolysis time. From the slopes of lines, the apparent reaction rate constant k for the five current densities were 2.00×10^{-2} , 2.40×10^{-2} , 2.71×10^{-2} , 2.83×10^{-2} , and



Fig. 3 The effect of current density on apparent reaction rate. Flow velocity 7.0 m h^{-1}

 2.81×10^{-2} min⁻¹ (as listed in Table 2). The current density made a great effect on the apparent reaction rate.

The relationship between the apparent reaction rate constant k and the current density j was described in Fig. 3. As shown in Fig. 3, the apparent reaction rate constant k increased obviously with the current density from 60 to 100 A m^{-2} while slowed down as the current density from 100 to 140 A m^{-2} . As known, organic pollutant degradation in electro-oxidation process was controlled by the reaction rate and mass transfer behavior. Several previous studies explored that the removal rate is under current control while the applied current density was less than the limiting current density, whereas the removal rate is under mass transport control (Venczel et al. 1997; Wang et al. 2015). It meant that too high current density could not always improve the apparent reaction rate but only increase the energy consumption. The variation in the apparent reaction rate constants indicated that the current density of 100 A m^{-2} was the optimized value.

The effect of flow velocity

As above described, the removal rate is under mass transport control when the applied current density is more than the limiting value. The efficient method for improving the removal rate is to enhance the mass transfer. In the industrial-scale devices, five flow velocities (2.0, 4.0, 7.0, 14.0, and 21.0 m h⁻¹) were applied to explore the influence of flow velocity on pollutant removal.



Fig. 4 The effect of flow velocity on COD removal. Flow velocity 2.0 m h^{-1} (*squares*), 4.0 m h^{-1} (*circles*), 7.0 m h^{-1} (*triangles*), 14.0 m h^{-1} (*inverted triangles*), and 21 m h^{-1} (stars); current density 100 A m⁻²

 Table 3
 The apparent reaction

 rate constant of COD removal
 under different flow velocities

Flow velocity	2.0 m h^{-1}	4.0 m h^{-1}	7.0 m h^{-1}	14.0 m h ⁻¹	21.0 m h ⁻¹
$\frac{1}{100 \times k(\min^{-1})}$ R^{2}	1.51	2.00	2.71	2.99	3.12
	0.998	0.999	0.996	0.999	0.997

As shown in Fig. 4, the flow velocity had an effect on COD removal, and the removal rate increased with flow velocity. The value of $\ln(\text{COD}_t/\text{COD}_0)$ had a great linear relationship with electrolysis time, and the correlation coefficients (R^2) were all above 0.99. The apparent reaction rate constant k for the five flow velocities were 1.51×10^{-2} , 2.00×10^{-2} , 2.71×10^{-2} , 2.99×10^{-2} , and 3.12×10^{-2} min⁻¹ (as listed in Table 3). It showed that flow velocity enhanced the apparent reaction rate constant k and then improved the rate of COD removal.

Similar to the current density, the relationship between apparent reaction rate and the flow velocity was represented in Fig. 5. As shown in Fig. 5, the fitting formula for the relationship was given as

$$k = 3.05 - 2.72 \exp(-0.26u) \tag{1}$$

where k is the apparent reaction rate constant, min⁻¹, and u is flow velocity, m h⁻¹. The flow velocity affected the diffusion field of matter, which related to the mass transfer rate. In our previous research, the mass transfer coefficient was the power function of the flow velocity (Díaz et al. 2011). It indicated that higher flow velocity could enhance the mass transfer. However, too high flow velocity also increased the pressure loss of the reactor, which was unfavorable to the system operation. Considering the apparent reaction rate, the scale of this construction, and the mean amount of influent, the flow velocity of 14 m h⁻¹ would be optimal and reasonable.



Fig. 5 The effect of flow velocity on apparent reaction rate. Current density 100 A m^{-2}

Environ Sci Pollut Res (2016) 23:18288-18295

The performance of PFER on effluent treatment

Pilot-scale experimental results previously identified that the electro-oxidation process has the ability to effectively decompose these pollutants. Based on the above information on current density and flow velocity, the industrial-scale electro-oxidation devices with capacity of 500 t day⁻¹ had been built at the industrial park wastewater treatment plant in 2014, to test the stability and reliability of the novel electro-oxidation reactor. The performance information of these devices on effluent treatment were shown in Figs. 6 and 7.

As shown in Fig. 6, the initial COD concentration of the influent fluctuated between 131 and 158 mg L⁻¹. At an optimized current density of 100 A m⁻² and a suitable flow velocity of 14.0 m h⁻¹, the industrial-scale PFER removed these pollutants efficiently, similar to the pilot-scale tests. The COD values in the effluent were reduced to 40–58 mg L⁻¹ within a 30-min electrolysis period. The removal efficiency for COD was over 60.0 %, and an average value of 65.2 % was maintained during continuous operation for 60 days.

The electro-oxidation method also removed color with high efficiency. As shown in Fig. 7, the color of the influent changed from 160 to 220 times. With the same optimized current density of 100 A m^{-2} and the suitable flow velocity of 14.0 m h^{-1} , the color in the effluent was reduced below 30 times within a 30-min electrolysis period, and the decolorizing efficiency was all over 84.0 %.



Fig. 6 COD removal of wastewater by the industrial-scale plunger flow electro-oxidation device. Current density 100 A m^{-2} ; flow velocity 14.0 m h^{-1}



Fig. 7 Color removal over the electrolysis time. Current density 100 Am^{-2} ; flow velocity 14.0 m h⁻¹

Disinfection

Wastewater requires sterilization to eliminate organisms and bacteria before it is discharged into the environment. Conventional disinfectant methods include addition of chlorine, monochloramine, chlorine dioxide, ozone, hydrogen peroxide, UV irradiation, and electrochemical treatment. The results from the pilot-scale study indicated that electro-oxidation had an excellent capability for wastewater disinfection, even for *Cryptosporidium parvum* oocysts and *Clostridium perfringens* spores, which are resistant to chlorine treatment (Cañizares et al. 2005; Zhu et al. 2015).

Figure 8 shows a comparison between electro-oxidation and conventional methods for fecal coliform disinfection of the effluent. The initial concentration of fecal coliforms in the effluent was determined to be about 1×10^5 CFU mL⁻¹ by manifold zymotechnics (HJ/T 347-2007). Ozone and electrooxidation had the best disinfection effect on fecal coliforms.



Fig. 8 Comparison of four disinfection methods in fecal coliform elimination. Electrochemical process, current density 100 A m⁻², time 30 min (*a*); ozone (3.0 mg L⁻¹), time 10 min (*b*); chlorine (0.5 mg L⁻¹), time 30 min (*c*); monochloramine (0.5 mg L⁻¹), time 30 min (*d*)



Fig. 9 Current efficiency and energy consumption. Current density 100 A m^{-2} ; flow velocity 14.0 m h^{-1}

The disinfection efficiencies were almost 100 % using ozone within 10 min and chlorine and electro-oxidation within 30 min, while it was only 94 % using monochloramine within 30 min. As mentioned above, 30 min was selected for COD removal by electro-oxidation. That meant that an electrolysis time of 30 min could fulfill the requirement for wastewater disinfection before discharge.

Economic evaluation

The operation cost is a vital indicator for evaluating and selecting a new wastewater treatment technology. For the electro-oxidation process, the electric energy or power consumption and the electrode maintenance are the main operating cost items. In addition, labor costs for staff to operate the wastewater treatment device should be taken into account.

Power consumption is usually expressed by the specific energy consumption (SEC), which is defined as the amount of electric energy consumed per unit mass of organics (e.g., COD) removed or per cubic meter wastewater treated. Some elementary information on the SEC of the electro-oxidation process was reported in the previous pilot-scale study (Zhu et al. 2015). Clearly, differences in absolute values existed between the pilot and industrial scales.

As shown in Fig. 9, the SEC increased with electrolysis time, while the current efficiency (CE) decreased. At the optimal current density of 100 A m⁻² and a suitable flow velocity of 14.0 m h⁻¹, CE and SEC were 32.8 % and 43.5 kWh kg COD^{-1} , 1.64 and 0.50 times of the pilot-scale values, respectively, within an electrolysis time of 30 min. This information indicates that the industrial-scale electrolysis cell had a superior capability because of the fluid flow regime and improved mass transfer properties compared with the pilot-scale device.

At present, Fenton agent, ozonation, and membrane filtration are also feasible processes for wastewater depth treatment. For comparison, these three processes were selected to treat the same effluent in field tests, but detailed information **Table 4**Comparison of electro-
oxidation with three other
conventional wastewater depth
treatment processes

Methods	Operation cost \$ m ⁻³						
	Electro-oxidation	0.44	_	_	0.44		
Fenton	0.02	0.51	0.12	0.65			
Ozonation	0.21	0.7	_	0.91			
Reverse osmosis	0.09	0.09	0.32	0.56			

The secondary pollutant was sludge for Fenton and concentrated wastewater for RO

The operation conditions for Fenton are 30 % H_2O_2 (1.5 L m⁻³), FeSO₄·7 H_2O (0.25 kg m⁻³); for ozonation: O_3 (0.3 kg m⁻³); for reverse osmosis: operating pressure (1.5 Mpa)

on these tests of other three processes are not presented in this paper. The Fenton agent process requires regulation of the pH values through acid and alkali addition plus hydrogen peroxide and ferrous irons to oxidize the pollutants, while the chemical sludge produced must also be treated as solid waste. Meanwhile, the neutralization reaction between acid and alkali increases the total dissolved solids (TDS) in the effluent, which negatively impacts wastewater recycling. Membrane filtration can separate pollutants from wastewater and produce freshwater for reuse, but the concentrated wastewater must then be treated by other advanced oxidation processes (e.g., electro-oxidation). Ozonation is considered to be a green wastewater treatment method that oxidizes pollutants without any secondary pollutants produced.

The operation costs of the four depth treatment processes mentioned above are presented in Table 4. All operation costs contained electric energy, chemical agents, and secondary pollutant handling but not staff expenses and equipment depreciation. The prices of electricity and agents are H₂O₂ (0.25 kg^{-1}), FeSO₄·7H₂O (0.10 kg^{-1}), 98 % H₂SO₄ (0.25 kg^{-1}), NaOH (0.20 kg^{-1}), pure oxygen (\$0.77 Nm⁻¹), and electric energy (\$0.11 per kWh). The cost of sludge handling and disposal was about \$0.25 kg⁻¹. The consumption of electric energy and chemical agents were calculated according to the optimized operating parameters. For the same discharge standard (e.g., 60 mg L^{-1} , a limited COD value for municipal wastewater treatment plant in China), the electro-oxidation process was the most reasonable and efficient among these four conventional wastewater depth treatment processes. The operation cost of the electro-oxidation process was the lowest, \$0.44 per ton effluent and \$4.78 kg COD^{-1} at a price of \$0.11 per kWh, at the optimal operating conditions of current density 100 A m⁻² and flow velocity 14.0 m h^{-1} .

Conclusions

The industrial-scale application of a plunger flow electrooxidation reactor showed that electro-oxidation is a green, efficient technology for purifying secondary biochemical effluent. At an optimized current density of 100 A m^{-2} and a suitable cross-flow velocity of 14.0 m h⁻¹, the industrial-scale PFER could remove pollutants efficiently, similar to pilotscale tests. The removal efficiencies for COD and color in effluent were over 60.0 and 84.0 %, respectively. The average COD value of the effluent was reduced from 141 to 49 mg L⁻ within an electrolysis time of 30 min. Meanwhile, the electrooxidation process had excellent disinfect ability. The SEC value of this industrial-scale devices was 43.5 kWh kg COD^{-1} with a current efficiency of 32.8 %, which is superior to the pilot-scale efficiency. To meet the requirements for emission or reuse, the energy consumption was 4.67 kWh and the operation cost was \$0.44 per ton of effluent at the local average price for electricity of \$0.11 kWh⁻¹. Compared with the Fenton agent process, ozonation, and membrane filtration, the electro-oxidation process comprised a high-efficiency and low-power, environmentally friendly technology.

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