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Research Article

Electrochemical Oxidation Using BDD Anodes Combined with Biological Aerated Filter for Biotreated Coking Wastewater Treatment

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Coking wastewater is characterized by poor biodegradability and high microorganism toxicity. Thus, it is difficult to meet Grade I of Integrated Wastewater Discharge Standard of China by biological treatment technology; specifically, COD cannot meet above standard due to containing refractory organics. A novel coupling reactor, electrochemical oxidation using BDD anodes and biological aerated filter (BAF), has been developed for carbon and nitrogen removal from biotreated coking wastewater, focusing on COD, BOD_5, NH_4^+-N, and NO_3^--N removal on operation over 90 days with average effluent value of 91.3, 9.73, 0.62, and 13.34 mgL^{-1}, respectively. Average value of BOD_5/COD and BOD_5/NO_3^--N was enhanced from 0.05 to 0.27 and from 0.45 to 1.21 by electrochemical oxidation, respectively, with average energy consumption of 67.9 kWh kg^{-1} COD. In addition, the refractory organics also were evidently mineralized in the unit based on the data of the three-dimensional fluorescence spectra. Meanwhile, its effluent provided excellent substrate for biological denitrification in BAF. At hydraulic retention time (HRT) of 13.08 h, about 12 mgL^{-1} NO_3^--N was depleted through denitrification, and it mainly occurred at top of 0.25 m height of BAF. Therefore, it is feasible to apply the coupling reactor for biotreated coking wastewater treatment and achieve desirable effluent quality.

1. Introduction

The biologically treated coking wastewater usually cannot meet the stringent effluent standard in China, namely, Grade I of Integrated Wastewater Discharge Standard of China [1–6], especially, for COD less than 100 mg L^{-1}. And the effluent of biotreated coking wastewater showed the characteristics of poor biodegradability and lower carbon-nitrogen ratio. Therefore, upgrading of biotreated coking wastewater is first imperative.

At present, advanced oxidation processes (AOPs), such as ozone oxidation [7], wet air oxidation [8], Fenton oxidation [9], and electrochemical oxidation [4, 10, 11], are good alternative technologies for coking wastewater treatment, in which electrochemical oxidation technology using BDD anodes has been proposed due to its advantages regarding extremely wide potential window, very low background current, and robust oxidation capacity [12–15]. Zhu et al. [4] investigated the electrochemical oxidation using BDD anodes for advanced treatment of coking wastewater and under the condition of current density of 20–60 mA cm^{-2}, pH of 3–11, and temperature of 20–60°C, the complete mineralization of organic pollutants was almost achieved with energy consumption of 64 kWh kg^{-1} COD. Haidar et al. [16] studied degradation of antibiotic sulfachloropyridazine using an electrochemical oxidation system with BDD anode, and the results showed that the system could efficiently remove the antibiotic from water and the higher mineralization rate can be obtained at increasing applied current. Urtiaga et al. [17] studied the remediation of wastewater containing tetrahydrofuran by electrochemical oxidation using BDD anodes, and the results showed that tetrahydrofuran can be totally mineralized by BDD anodes.

What is more, our group found that during the oxidation process for biotreated coking wastewater by BDD anodes in different electrolysis time [10] the ratio of BOD_5 to COD increased from an initial value of 0.05 to 0.65 at electrolysis time of 90 min and the BOD_5/NO_3^--N ratio increased

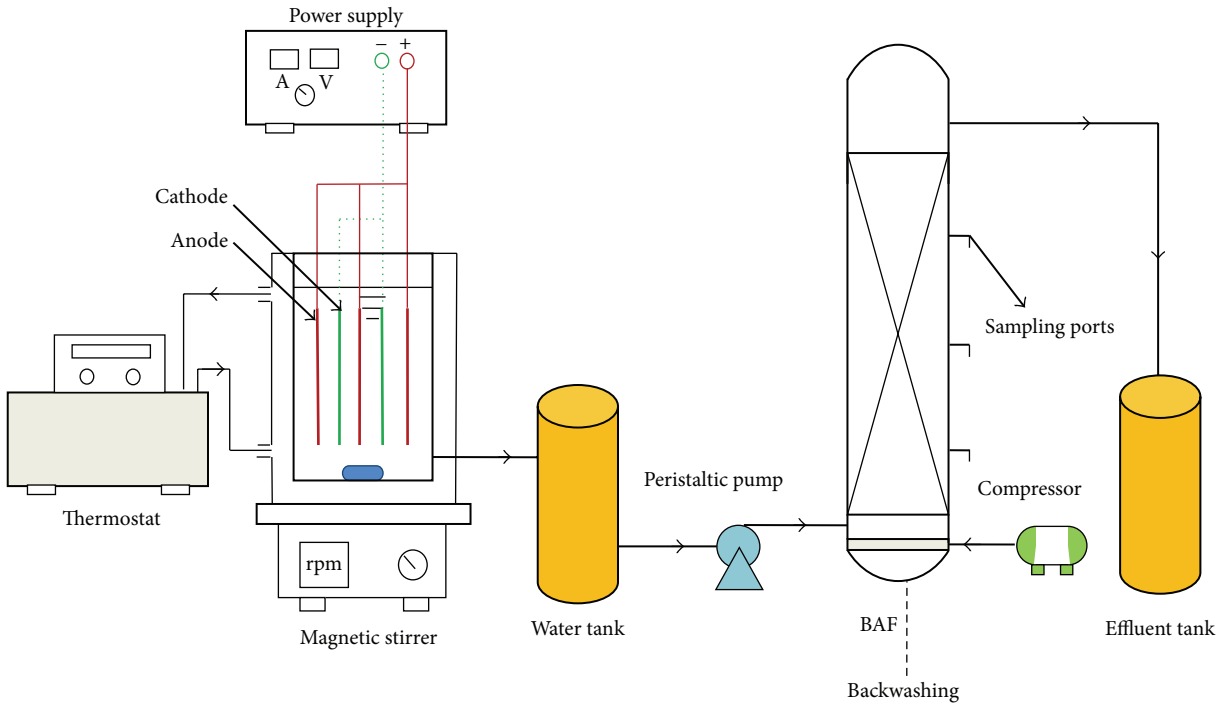

Figure 1: Experimental system.

from an initial value of 0.48 to 1.25 at 45 min and then gradually dropped to 0.69 at 90 min. Moreover, the NO_3^--N concentration in electrochemical oxidation effluent was evidently increased at electrolysis time of 90 min with energy consumption of 116.0 kWh kg^{-1} COD. Therefore, based on the consideration of energy consumption and effluent characteristics in different electrolysis time, it is strongly suggested that electrochemical technology using BDD anodes should be combined with biological denitrification technology for the advanced treatment of biotreated coking wastewater to better remove organics and nitrogenous compounds.

Nowadays, tertiary biological technology for wastewater treatment and reclamation mainly was biological aerated filter (BAF). For examples, Chen et al. [18] adopted anaerobic/anoxic/aerobic and BAF system for wastewater treatment and mainly discussed the effects of COD/TN ratio and nitrate recycling ratio on nitrogen and phosphorus removal. Cui et al. [19] combined ozone oxidation and BAF processes for treatment of cyanide containing electroplating wastewater, and the results showed that BAF can tolerate higher cyanide toxicity than some other bioreactor and obtain high removal efficiency with low influent BOD_5/TN in BAF. Moreover, the cyanide compounds could be used as the nitrogen source for the microbes. In addition, regarding BAF's features of simultaneously nitrification and denitrification, without settling tank and excellent effluent quality [20–22], it was selected as biological denitrification technology for advanced biotreated coking wastewater.

Accordingly, based on the effluent characteristics of electrochemical oxidation, combining the advantages of electrochemical oxidation using BDD anodes with BAF, the object of this study is to develop a novel coupling system for simultaneous removal of carbon and nitrogen from biotreated coking wastewater, and its performances for COD, BOD_5, NH_4^+-N, and NO_3^--N removal in different periods were focused on. Meanwhile, improvement of biodegradability and carbon-nitrogen ratio, its energy consumption in EO unit, and removal characteristics of COD, NH_4^+-N, and NO_3^--N as special distribution in BAF unit were also investigated.

2. Materials and Methods

2.1. Experimental Equipment. The experiment was carried out in a coupling reactor, consisting of electrochemical oxidation (EO) unit and BAF unit (in Figure 1), in which EO unit was made of Plexiglas with wok volume of 3 L. The electrodes constituted of 3 pieces of BDD anodes and 2 pieces of stainless steel cathodes at spacing of 1 cm. The anodes were purchased from CONDIAS GmbH (Germany) with size of 195 mm × 26 mm × 2 mm (length × width × thickness) [10]. The dimension of BAF was diameter of 10 cm, work height of 1.0 m, mainly packed with lava (diameter of 0.5 cm) as media.

2.2. Biotreated Coking Wastewater Quality and Effluent Standard. The biotreated coking wastewater in this paper was derived from a coking plant of the Capital Iron and Steel Group in Beijing, China; its main parameters of influent quality were shown in Table 1, with poor biodegradability (BOD_5/COD of 0.054–0.06) and low carbon-nitrogen ratio (BOD_5/NO_3^--N of 0.48–0.52), while effluent quality was required to meet Grade I of Integrated Wastewater Discharge Standard of China (GB 8978-1996), in which several main parameters were listed in Table 1.

Table 1: Main parameters of influent quality and effluent standard.

Items	COD ($mg\,L^{-1}$)	BOD_5 ($mg\,L^{-1}$)	Volatile phenol ($mg\,L^{-1}$)	Cyanide ($mg\,L^{-1}$)	NH_4^+-N ($mg\,L^{-1}$)	NO_3^--N ($mg\,L^{-1}$)	Conductivity ($\mu S\,cm^{-1}$)	pH
Influent	190.2–255.8	10.28–15.23	0.56–0.78	2.52–3.80	5.2–7.6	21.12–29.38	1850–1920	7.2–7.9
Effluent	≤100	≤20	≤0.5	≤0.5	≤15	—	—	6–9

Table 2: Operation parameters of coupling system in different period.

Items		R_1	R_2	R_3
Electrochemical oxidation	Current density ($mA\,cm^{-2}$)	75	75	75
	HRT (min)	45	45	45
BAF	HRT (hours)	13.08	8.72	6.54
	Ratio of air to water	1 : 1	1 : 1	1 : 1
Run days		30	30	30

In addition, conductivity in biotreated coking wastewater was in range of 1850–1920 $\mu S\,cm^{-1}$.

2.3. Operating Conditions. Based on previous results from our group [10], conditions of EO were selected. It lasted for 90 days; three different long-term steady-state experimental runs (R_1–R_3) were carried out with HRT of 45 min and current density of 75 $mA\,cm^{-2}$ for EO unit. Meanwhile, HRT of BAF was kept at 13.08 h, 8.72 h, and 6.54 h with the same air/water ratio of 1 : 1 throughout the whole experimental period (see Table 2), in which, EO unit was intermittently operated and run 3.75 to 6.75 hours every day with treatment capacity of $4\,L\,h^{-1}$. BAF was continuously operated with treatment capacity of $14.4\,L\,d^{-1}$ to $28.8\,L\,d^{-1}$.

2.4. Analytical Instruments and Methods. COD, BOD_5, NH_4^+-N, and NO_3^--N in influent and effluent of each reactor were analyzed daily, and COD was measured by a COD meter (ET99722, Hanna Corporation, Italy). BOD_5 was determined by dilution inoculation method. Ammonia (NH_4^+-N) concentration was obtained via Nessler's reaction using UV2100 spectrophotometer (Shanghai Unico Instruments Co., Ltd., China). Nitrate (NO_3^--N) concentration was detected by disulfonic acid phenol reaction using UV2100 spectrophotometer (Shanghai Unico Instruments Co., Ltd., China). Conductivity was tested by conductivity meter (CDC40103) (HACH, USA).

The fluorescence spectra were recorded using a spectrophotometer (F7000 fluorescence spectrophotometer, Hitachi, Japan) equipped with a 700-W xenon arc-lamp. Excitation scans were performed in the range 200–450 nm at fixed intervals of 10 nm, and the emission was recorded at wavelengths ranging from 260 to 550 nm in 10 nm steps with integration time of 0.1 s. The results file of each three-dimensional spectrum was saved in CSV format and then they were output by origin 8.0.

Energy consumption (EC) was calculated using the following equation [4, 23]:

$$\mathrm{EC} = \frac{1000UIt}{(C_i - C_e)V}, \quad (1)$$

where EC is energy consumption (in $kWh\,kgCOD^{-1}$), U is voltage (in V), I is current (in A), t is electrolysis time (in h), C_i is initial COD concentration of influent (in $mg\,L^{-1}$), C_e is COD concentration of effluent (in $mg\,L^{-1}$), and V is volume of treated wastewater (in L).

3. Results and Discussion

3.1. Overall Performance of EO + BAF Coupling Reactor. Figures 2(a) and 2(b) show profiles of COD and BOD_5 concentration in influent and effluent of EO and BAF unit during experimental periods, respectively. As shown in Figures 2(a) and 2(b), COD and BOD_5 concentration in final effluent from the coupling system was maintained below 100 $mg\,L^{-1}$ and 20 $mg\,L^{-1}$ with the average value of 91.3 $mg\,L^{-1}$ and 9.73 $mg\,L^{-1}$, respectively, which met Grade I of Integrated Wastewater Discharge Standard of China (GB 8978-1996). And average value of BOD_5/COD ratio and BOD_5/NO_3^--N ratio was enhanced by electrochemical oxidation from 0.05 to 0.27 and from 0.45 to 1.21, respectively, which was helpful for microorganism denitrification in BAF because of enough carbon resource, which also could be confirmed by data of nitrate removal (Figure 2(c)).

Figures 2(c) and 2(d) show profiles of NH_4^+-N and NO_3^--N concentration in influent and effluent of EO and BAF unit during experimental periods, respectively. As shown in Figures 2(c) and 2(d), NH_4^+-N and NO_3^--N concentration in final effluent from the coupling system was maintained below 15 $mg\,L^{-1}$ and 20 $mg\,L^{-1}$ with the average value of 0.62 $mg\,L^{-1}$ and 13.34 $mg\,L^{-1}$, respectively, which met Grade I of Integrated Wastewater Discharge Standard of China (GB 8978-1996) and NO_3^--N concentration below 20 $mg\,L^{-1}$. It also can be found that nitrogen removal mainly occurred at BAF unit, and the impact of HRT of BAF on nitrate removal was much more evident than on ammonia and organics removal. Generally, nitrate removal efficiency increased with the increase of HRT. As influent ammonia concentration was lower, its variation tendency as HRT also was not obvious. In addition, due to the fact that BAF was run at air to water ratio of 1 : 1, the simultaneous nitrification and denitrification took place in BAF unit; thus organics may be partly removed

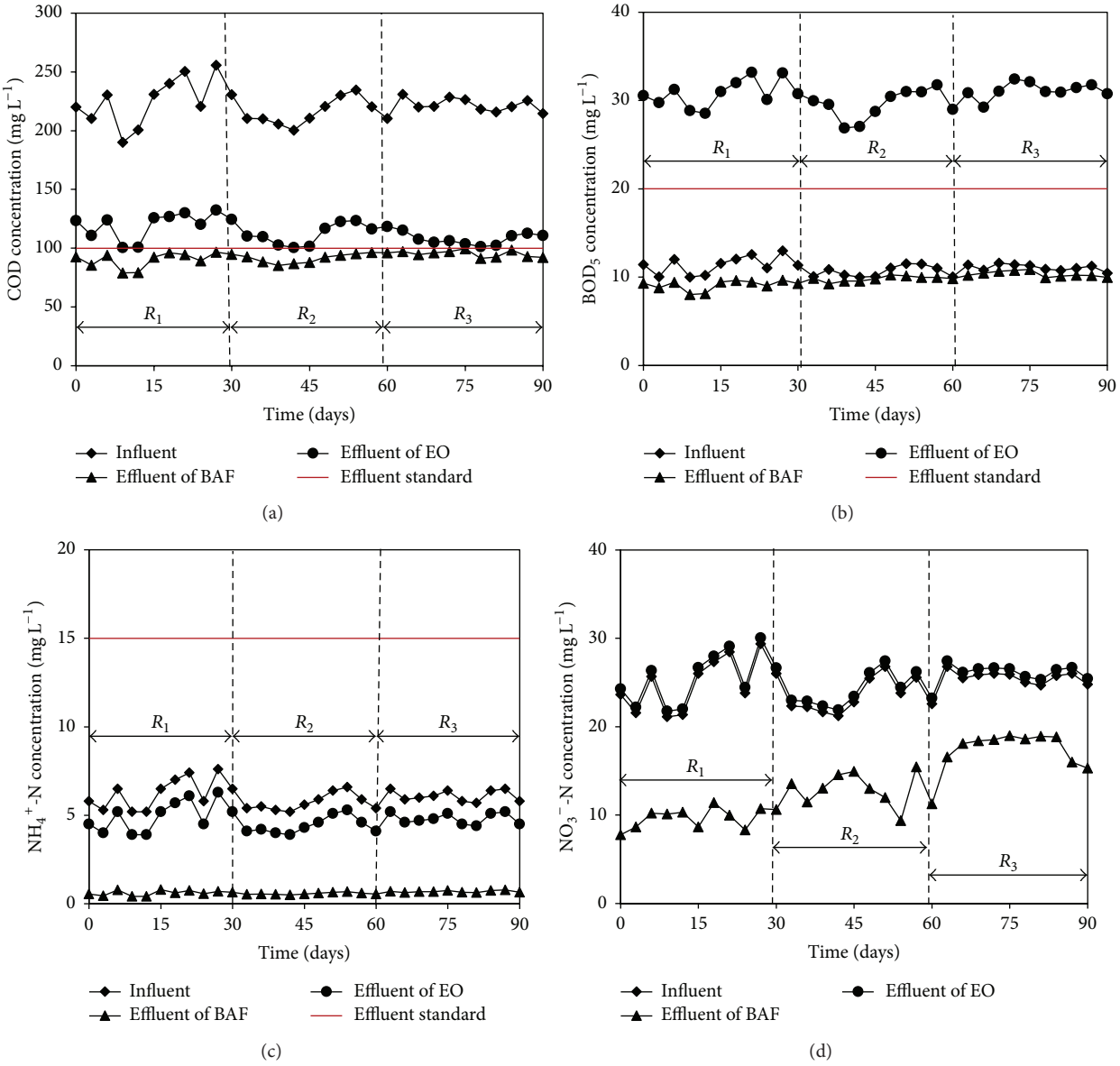

Figure 2: COD (a), BOD_5 (b), NH_4^+-N (c), and NO_3^--N (d) concentrations of influent and effluent of each unit in coupling system during different period.

by heterotrophic bacteria and partly removed by denitrifiers, which led to the fact that organics removal was not much more evident with the increase of HRT.

3.2. Pollutants Removal Efficiency and Energy Consumption in EO Unit. To clearly identify the performance in EO unit, COD, BOD_5, NH_4^+-N, and NO_3^--N, removal efficiencies during different operating periods are presented in Figure 3. Average COD, BOD_5, NH_4^+-N, and NO_3^--N removal efficiency by EO was 48.74%, −63.82%, 21.15%, and −2.57%, respectively.

Notably, the reason for BOD_5 concentration increase instead of decrease was that the biodegradability of biotreated coking wastewater was poor and the organics mainly were refractory organics, which can be confirmed from the three-dimensional fluorescence spectra (Figure 4(a)); there were two fluorescence peaks detected at $\lambda_{ex}/\lambda_{em}$ = 250/370 and $\lambda_{ex}/\lambda_{em}$ = 320/370, respectively, from raw biotreated coking wastewater. The former peak was confirmed as aromatic-protein-like substance II and can be referred to as refractory biodegraded dissolved organic matter [24]. The latter peak represents humic acid-like substance. After the EO unit, the refractory organics were partly mineralized into CO_2 and H_2O and partly transferred into biodegradable organics, which can be found from the three-dimensional fluorescence spectra of effluent (Figure 4(b)). Compared with Figures 4(a) and 4(b), it can be found that the fluorescence intensity of aromatic-protein-like substance II has been greatly reduced from about 6100 to less than 150.

In addition, the increase of nitrate concentration after EO unit was mainly attributed to the nitrogen heterocyclic

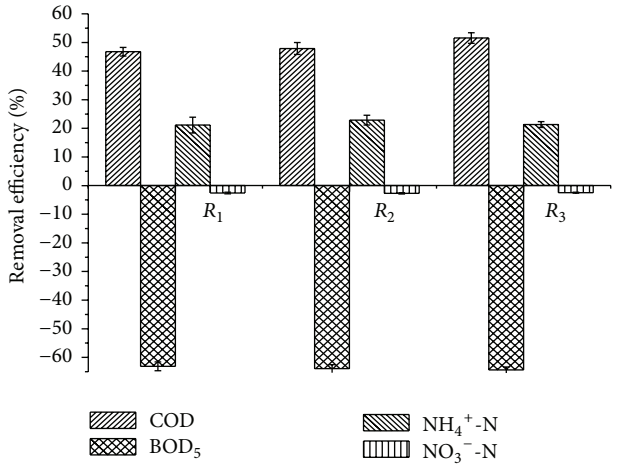

Figure 3: COD, BOD_5, NH_4^+-N, and NO_3^--N removal efficiency in EO unit during operation periods.

organic compounds and NH_4^+-N being oxidized by oxidant into NO_3^--N [10], thus resulting in the increase in the NO_3^--N concentration.

Based on (1), at the current density of 75 mA cm^{-2} and HRT of 45 min, the mean voltage and current was 2.6 V and 15 A in EO unit, respectively, so when mean COD was removed from 221.2 mg L^{-1} to 113.5 mg L^{-1}, the mean energy consumption was calculated as 67.9 kWh kg^{-1} COD. Compared with the results from Zhu et al. [4], who studied advanced treatment of biologically pretreated coking wastewater by electrochemical oxidation using BDD electrodes with energy consumption of 64 kWh kg^{-1} COD, Serikawa et al. [25] treated a wastewater with an initial COD concentration of 14000 mg L^{-1} at a BDD pilot plant with energy consumption of 30 kWh kg^{-1} COD and Anglada et al. [26] carried on a study on the electrochemical oxidation of landfill leachate by BDD anode with energy consumption of 53–94 kWh kg^{-1} COD at different current density. The energy consumption of kg COD removal in this BDD anode was inexpensive.

3.3. Pollutants Removal Efficiency and Its Concentration Profile in BAF Unit. In BAF unit, COD, BOD_5, NH_4^+-N, and NO_3^--N removal efficiencies during different operating periods also are presented in Figure 5. As HRT was decreased from 13.08 h to 8.72 h and 6.54 h, average COD removal efficiency by BAF was reduced from 24.45% to 18.40% and 14.55%, BOD_5 removal efficiency was reduced from 70.50% to 66.71% and 66.79%, and NH_4^+-N and NO_3^--N removal efficiency was reduced from 87.69% to 86.88% and 85.55% and from 61.80% to 46.29% and 32.13%, respectively. It can be found that as HRT decreased, above pollutants removal efficiencies all were decreased in BAF unit, but all pollutants' removal by BAF during different operating period was relatively stable.

Figure 6 shows profiles of COD, NH_4^+-N, and NO_3^--N along the height of BAF at HRT of 13.08 h. It is evident that sudden drops in both COD and NO_3^--N concentration occurred at top 0.25 m height of BAF due to denitrification

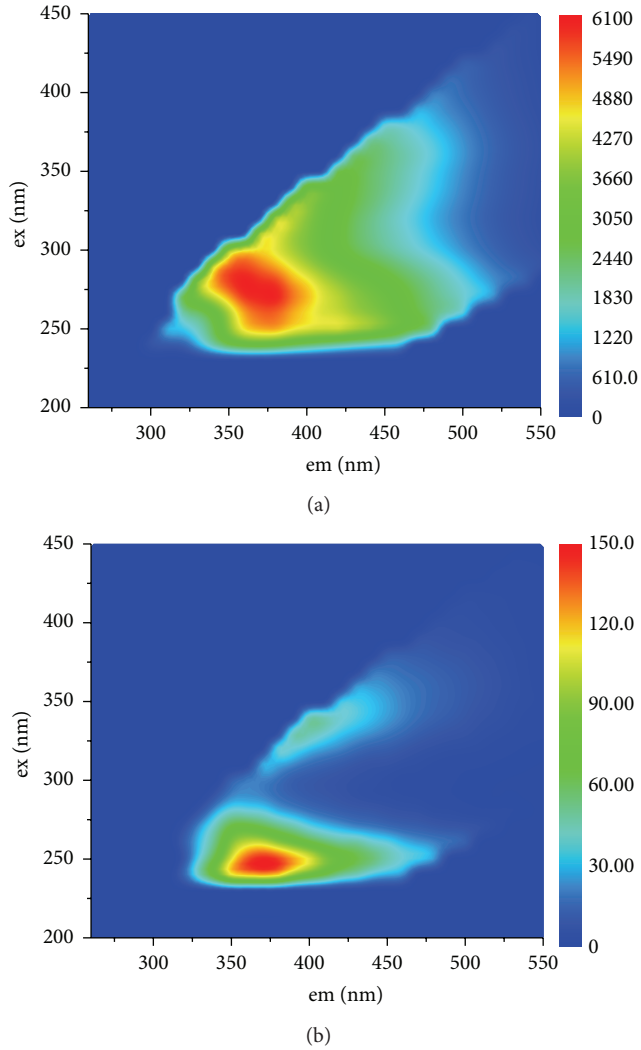

Figure 4: Three-dimensional fluorescence spectra of (a) influent and (b) effluent in EO unit.

quickly performed at the top of the BAF in the presence of abundant NO_3^--N and biodegradable organics, while their concentrations were slowly varied at height of 0.5 m, 0.75 m, and 1.0 m. About 12 mg L^{-1} NO_3^--N was depleted through denitrification. Comparatively, NH_4^+-N concentration was slowly changed at different height of BAF with total 4.1 mg L^{-1} ammonia removed.

4. Conclusions

A coupling system, electrochemical oxidation using BDD anodes and BAF, for biotreated coking wastewater treatment was investigated to study feasibility of simultaneous removal of carbon and nitrogen. Long-term operating results over 90 days showed desirable effluent quality and steady process performance with the average effluent concentration of COD, BOD_5, NH_4^+-N, and NO_3^--N of 91.3, 9.73, 0.62, and 13.34 mg L^{-1}, respectively, reaching the Grade I of Integrated Wastewater Discharge Standard of China. Average value of

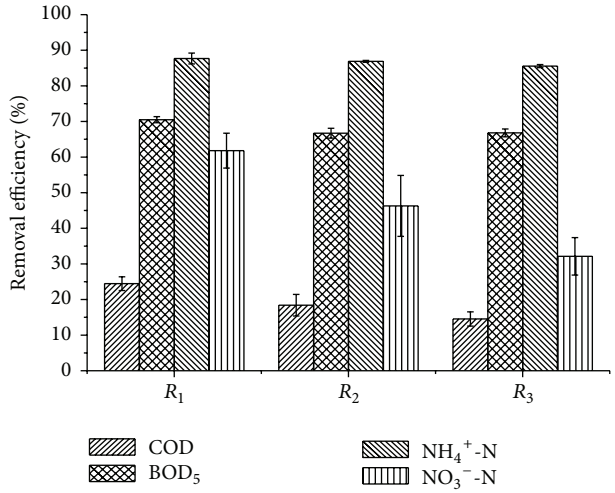

Figure 5: COD, BOD_5, NH_4^+-N, and NO_3^--N removal efficiency in BAF unit during operation periods.

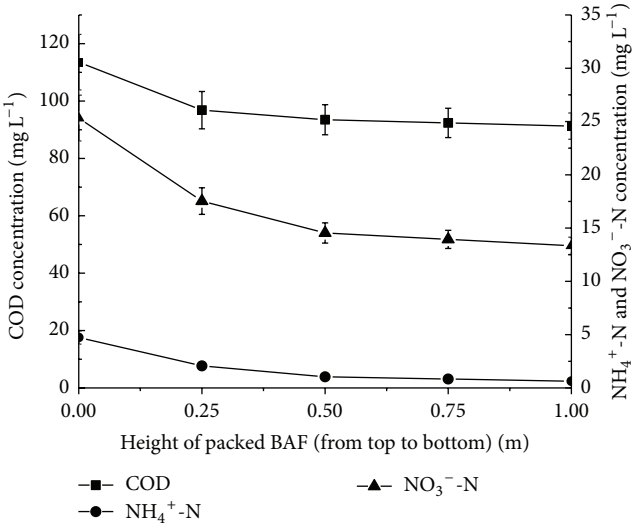

Figure 6: Profile of COD, NH_4^+-N, and NO_3^--N along the height of media layer in BAF unit.

BOD_5/COD and BOD_5/NO_3^--N was improved from 0.05 to 0.27 and from 0.45 to 1.21, respectively, in electrochemical oxidation unit. Meanwhile, the data of three-dimensional fluorescence spectra in the unit confirmed that the refractory organics were evidently mineralized. At current density of 75 mA cm^{-2} and HRT of 45 min, the mean energy consumption of EO unit was calculated as 67.9 kWh kg^{-1}COD and the corresponding COD, BOD_5, NH_4^+-N, and NO_3^--N removal efficiency was 48.74%, −63.82%, 21.15%, and −2.57%, respectively. In addition, its effluent provided excellent substrate for biological denitrification in BAF. As its HRT was decreased from 13.08 h to 8.72 h and 6.54 h, all pollutants removal efficiencies were decreased. Based on the data at HRT of 13.08 h, the biological denitrification mainly occurred at top 0.25 m height of BAF, and about 12 mg L^{-1} NO_3^--N was depleted through denitrification in BAF unit. Therefore, it is feasible to apply the coupling system for biotreated coking wastewater treatment.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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