Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2015.1107856

57 (2016) 20470–20475
September

Removal of ibuprofen from municipal sewage by three-dimensional particle electrode combined with a biological aerated filter (TDE-BAF)

Yan Feng[a,b], Xinwei Wang[e], Jingyao Qi[a,*], Yixuan Feng[d], Zhaoyang Wang[a], Bo Wang[a], Xin Li[c]

[a]*School of Municipal & Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China, Tel. +86 053182765848; email: cea_fengy@ujn.edu.cn (Y. Feng), Tel. +86 13604519989; emails: qjy_hit@hotmail.com (J. Qi), 345678@qq.com (Z. Wang), wangbo@163.com (B. Wang)*
[b]*School of Civil Engineering and Architecture, University of Jinan, Jinan 250022, China*
[c]*Department of Chemistry, Harbin Institute of Technology, Harbin 150090, China, email: Lixin@163.com*
[d]*School of Resources and Environment, University of Jinan, Jinan 250022, China, email: 44254810@qq.com*
[e]*China Urban Construction Design & Research Institute Co., Ltd (Shang Dong), Jinan 250022, China, Tel. +86 053189736600; email: wangxinwei@163.com*

Received 5 March 2015; Accepted 8 October 2015

ABSTRACT

The degradation of ibuprofen was investigated in aqueous solution by three-dimensional particle electrode (TDE) technology. When the initial concentration was between 10 and 40 mg/L, almost 80% of the ibuprofen was degraded at 6 h. However, only about 16.64% of ibuprofen was mineralized even at 6 h according to total organic carbon measurements. A combined process of TDE and biological aerated filter (BAF) was therefore developed to enhance the treatment of ibuprofen contaminated municipal sewage. The effluent quality of coupled process was substantially improved by TDE due to the degradation of ibuprofen and related intermediates. The experimental results showed that TDE-BAF is effective for removing ibuprofen from municipal sewage.

Keywords: Ibuprofen; Biological aerated filter (BAF); Three-dimensional particle electrode (TDE); Coupled with

1. Introduction

Pharmaceuticals and personal care products (PPCPs) have been found in surface, ground, sewage, and drinking water samples in different countries [1] and [2]. Although these compounds are typically present in very low concentrations in water compartments, they are considered as a potential hazard for aquatic organisms and human health due to their potential to bio-accumulate and the possibility of causing estrogenic and endocrine effects [3]. At present, conventional full-scale wastewater treatment plants (WWTPs) are generally not equipped to deal with pharmaceutical compounds, as they were designed with the principal aim of removing easily or moderately biodegradable compounds [4]. The biological aerated filter (BAF) is designed with the principal aim of removing easily or moderately biodegradable compounds. A great deal of researchers has conducted

*Corresponding author.

1944-3994/1944-3986 © 2015 Balaban Desalination Publications. All rights reserved.

many studies to enhance the efficiency of biological treatment in the last few years.

Advanced oxidation processes (AOPs), mostly dependent on the generation of hydroxyl radicals (·OH), including ozonation [5], Fenton reaction [6], photolysis [7], sonolysis [8], have been studied as possible alternatives to destroy PPCPs in water [9]. Another way to eliminate persistent pollutants is through the use of the electrochemical method. The three-dimensional particle electrode (TDE) process also belongs to the electrochemical family. Recent studies have been conducted on the electro degradation of PPCPs [10–12]. TDE provides an excellent solution to the disadvantages which limit the application of two-dimensional (2D) electrode [13]. Compared to 2D electrode, the addition of granular activated carbon (GAC) or metal particles may also enhance the conductivity and mass transfer or the adsorption of pollutants [14]. Also, the large specific surface areas of these particles can provide more reactive sites than 2D electrode for pollutants adsorption or even catalytic reactions [15,16], resulting in higher removal efficiency [17–20].

This study focused on ibuprofen, one of the most widely used PPCPs. As previous reports shown, TDE process succeeded to decompose ibuprofen in aqueous solution but it was rather no effective in mineralization [21]. In this regard, TDE combined with a BAF was developed to remove the ibuprofen and its intermediates from wastewater samples. BAF, a type of immobilization reactor, has been demonstrated to be a cost-effective way for the treatment of municipal sewage [22,23]. The main objectives of this study were to investigate TDE-induced degradation of ibuprofen as well as the change of total organic carbon (TOC), CODcr in aqueous solution. The performance of BAF alone in parallel with a TDE-BAF for the treatment of ibuprofen-contaminated municipal sewage was compared.

2. Materials and methods

2.1. Materials

Sodium 2-(4-isobutylphenyl) propionates, known as ibuprofen (Ibu), with a purity of 99.9%, was purchased from Aldrich and used without further purification. A new red mud particle electrode (RMPEs) was prepared with the primary material of red mud.

RMPEs were made up of red mud, zeolite slag, iron powder (Tianjin love Kurt g trade Co., Ltd), and carbon (Tianjin love Kurt g trade Co., Ltd). Red mud was taken from Jinan Mining Development Corporation and zeolite slags were obtained from Weifang Trading Company Ltd, Shandong Province of China

2.2. Reactor description and operation

2.2.1. Reactor description

Biological treatments were performed using two same size reactors, diameter of 80 mm, length of 1,500 mm, and effective volume of 6.0 L. The first column (1#) was tested for the biological treatment alone, while the second column (2#) was tested for the combined process of TDE-BAF. The second column (2#) included BAF and TDE separated by a stainless steel porous cathode plate. The TDE section was crammed with RMPEs and well joined to a power source. BAF was filled with zeolite. First of all, municipal wastewater was poured into the coupled reactor from the bottom, and then, most of effluent outflowed from the top. Moreover, the mixed solution between a proportion of effluent and municipal wastewater reflowed to BAF-TDE by pump, which accomplished the coupling between BAF and TDE. The municipal sewage containing ibuprofen was pumped into the reactors via a peristaltic pump. Air stream was continuously introduced at the bottom of the reactors. The ratio of air to water was fixed at 5:1. For the flow rate of 1.00 L/h, the hydraulic retention time (HRT) of BAF was approximately 6 h.

2.2.2. Wastewater characteristics

In order to investigate the decomposition of ibuprofen and the change of TOC, CODcr, NH_4^+-N in aqueous solution, 10–40 mg/L ibuprofen was prepared with double-distilled water. As for the combined process of TDE-BAF, ibuprofen was dissolved into real surface water to achieve 1–5 mg/L ibuprofen-contained water samples as shown in Table 1.

2.2.3. Start-up of reactors

In order to improve the inoculation rate, activated sludge was introduced to two reactors from wastewater treatment plant. The experimental parameters were the same for them. They were backwashed every 48 h. The two reactors achieved stable running conditions after 7 weeks.

2.3. Analytical methods

During the whole experiments, influent, and effluent samples were taken each day at regular intervals and CODcr and NH_3-N were tested according to conventional national standard methods [24]. The ibuprofen concentration was measured by high-performance liquid chromatography (HPLC, Agilent 1200 Series,

Table 1
Characteristic of ibuprofen-contaminated municipal sewage.

pH	NH_4^+-N (mg/L)	CODcr (mg/L)	TOC (mg/L)	Ibuprofen (mg/L)
6–8	19.14–26.25	101.12–138.30	31.63–45.63	1.22–4.98

Agilent, USA) equipped with an XDB-C18 column, according to standard methods [25]. Probes were used to measure the temperature, pH, and dissolved oxygen during the experimental period. As a whole, influent and effluent samples were taken at the end of the filter run, but occasionally samples were also drawn immediately after backwashing to inspect the recovery of effluent quality. Due to some limitations and because the study was focused on TOC, CODcr, and NH_3-N, some quality parameters (5 d biochemical oxygen demand, nitrate, and nitrite, P) were not measured in all the samples.

2.4. Ibuprofen degradation experiments by TDEs

Performance of the particle electrodes was investigated by their degradation of ibuprofen. The ibuprofen degradation experiment was performed in a TDE cell. Ti electrode was used as anode and stainless steel electrode was used as cathode in the TDE. ADC power was wired between the anode and cathode. The TDE was filled with RMPEs. An air pump was applied in the experiment.

Four groups of degradation experiments were performed in the TDEs. The ibuprofen solution with an initial concentration of 10, 20, 30, 40 mg/L was poured separately into the TDE. In every group of degradation experiment, 0.1 mol/L Na_2SO_4 was served as a supporting electrolyte in the TDEs. The ibuprofen concentration of the solution was measured at 0.5, 1, 2, 3, 4, 5, 6 h, separately in all four groups of experiments. The TOC and CODcr of the solution were also measured at 0.5, 1, 2, 3, 4, 5, 6 h, separately in the second group of degradation experiment.

3. Results and discussion

3.1. Degradation of ibuprofen by TDEs

To establish the removal performance of ibuprofen by TDE, several assays were conducted with initial concentrations of 10.00, 20.00, 30.00, and 40 mg/L, using 0.1 mol/L Na_2SO_4 solutions as the electrolyte. The experiments were conducted at 30 V, for 6 h each. Removals of ibuprofen are presented in Fig. 1. As can be observed, ibuprofen was degraded quickly in

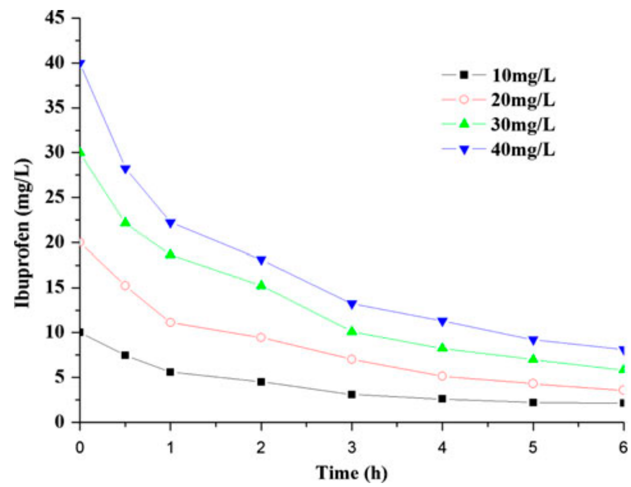

Fig. 1. Effect of time on Ibuprofen degradation.

arrange of 10–40 mg/L at 3 h. The removal efficiency was almost 80% for all the samples at 6 h. Similarly, L. Ciríaco showed that electrochemical oxidation have shown a very good degradation of ibuprofen in 6 h experiments [26]. Our results are consistent with this thesis.

In order to investigate the degradation of ibuprofen as well as the decrease in TOC and CODcr, the initial concentration of 20.21 mg/L ibuprofen was electrolyzed. The results are summarized in Table 2. As is observed on Table 2, Ibuprofen decreased quickly with time increasing. However, even at the time of 6 h, TOC and CODcr removal efficiencies were only 16.64 and 32.69%, respectively.

3.2. Comparing the performance of BAF alone and TDE-BAF

Operational parameters affecting removal efficiency are diverse. In order to study ibuprofen removal characteristics between BAF alone and BAF-TDE reactors, parameters, such as temperature, DO, HRT, and voltage, were maintained constant throughout the study. Temperature is one of the parameters affecting the removal efficiency [27]. The study was performed at 21 ± 2˚C. DO plays a important role on nitrification. If it is <2 mg/L and the biofloccules are

Table 2
Decrease of ibuprofen, TOC, CODcr during TDE

Time (h)	Ibuprofen (mg/L)	CODcr (mg/L)	TOC (mg/L)
0	20.21	54.48	14.84
0.5	15.21	50.12	14.24
1	11.11	48.13	14.00
2	9.44	44.31	13.85
3	7.01	40.22	13.76
4	5.14	38.13	13.02
5	4.29	37.00	12.68
6	3.56	36.67	12.37

bigger than 200 μm, it is not beneficial to nitrification [28]. In TDE reactors, air is sparged for two aims, one is to promote mass transfer and the other is to afford oxygen for some electrochemical reactions, e.g. the cathodic generation of H_2O_2 [29]. So DO was maintained at 4 mg/L in the reactors to prevent DO from being limiting by regulating airflow. For the case of continuous flow electrochemical reactor and BAF, HRT is the corresponding important parameter. HRT was 3 h. Current density/cell voltage is one of the most important variable parameters in TDE system since it influences not only the electrochemical oxidation but also the polarization behavior of particle electrodes [30]. Consequently, it is necessary to look for the balance between removal efficiency and energy consumption by optimization experiments. On the basis of preliminary experiments, the voltage remains 30.0 V and current density is 1.99 mA cm^{-2}.

The ibuprofen and TOC of influent and effluent in two reactors are presented in Fig. 2. From Fig. 2(A), it was found that when the initial ibuprofen concentration was 1.22–4.98 mg/L, the removal efficiency of ibuprofen was only 18.13–25.21% in BAF alone reactor. However, almost average 96.33% decomposition of ibuprofen was obtained after TDE combined with a BAF. As shown in Fig. 2(B), BAF and TDE-BAF reactors had average total TOC removals of 71.96 and 84.89%, respectively. The total TOC removal efficiency was improved 12.93% by using the combined process of TDE-BAF, compared with by means of BAF treatment alone. Obviously, this was mainly due to the degradation of ibuprofen by TDE treatment.

For the performance of BAF alone and TDE-BAF, some comprehensive parameters of the ibuprofen containing municipal sewage, such as CODcr and NH_4^+-N in two reactors are shown in Fig. 3. For the biological treatment alone, the average removal efficiencies of CODcr and NH_4^+-N were 74.99% (66.29–80.21%) and 74.45% (70.27–81.92%), respectively, in BAF reactor. However, for the combined process of TDE-BAF, the removal efficiencies were 88.63% (84.65–93.13%) and 82.76% (78.06–86.92%), respectively.

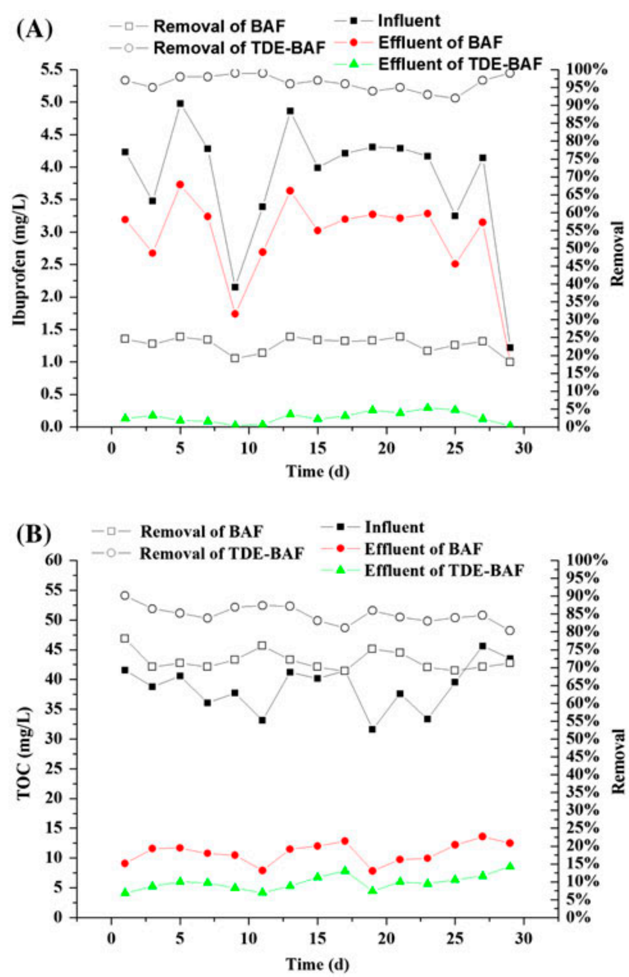

Fig. 2. Ibuprofen and TOC removal in BAF and TDE-BAF reactor.

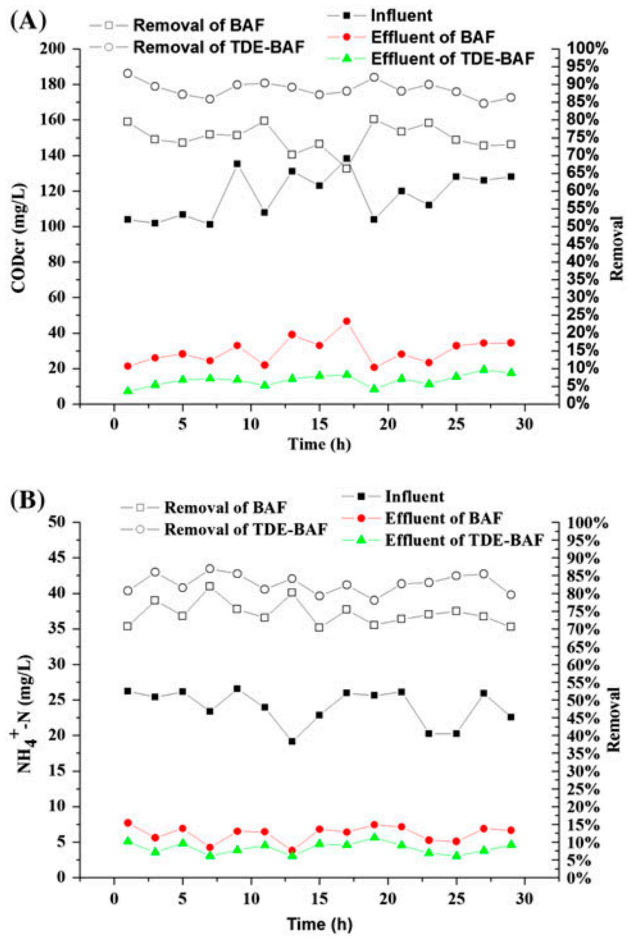

Fig. 3. CODcr removal and NH_4^+-N removal in BAF and TDE-BAF reactor.

4. Conclusions

The experiments demonstrated that ibuprofen can be removed effectively by TDE. In the initial concentration below 40 mg/L, about 80% removal efficiency was achieved at 6 h. However, only about 16.64% ibuprofen was mineralized even at 6 h. For the real ibuprofen contaminated municipal sewage, the average removal efficiencies of ibuprofen, TOC, CODcr, and NH_4^+-N were 23.06, 71.96, 74.99, and 74.45%, respectively, by BAF alone. However, for the combined process of TDE-BAF, the removal efficiencies were improved to 96.33, 84.89, 88.63, and 82.76%, respectively. The removal efficiency was substantially influenced by TDE.

Acknowledgements

This work was funded by the Natural Science Foundation of Shandong Province (ZR2012EEL21, ZR2011EEM003, ZR2011EEM018), National Natural Science Foundation of China (NSFC51508227, NSFC51178207, NSFC50978121), the Special Fund of Harbin Science & Technology Innovation Talents (for outstanding subject leaders, Grant no. RC2013XK015009), A Project of Shandong Province Higher Educational Science and Technology Program (J14LG02), Shandong province science and technology development plan-Policy guidance project (2013YD17003).

References

[1] M. Clara, O. Gans, G. Windhofer, U. Krenn, W. Hartl, K. Braun, S. Scharf, C. Scheffknecht, Occurrence of polycyclic musks in wastewater and receiving water bodies and fate during wastewater treatment, Chemosphere 82 (2011) 1116–1123.

[2] S. Monteiro, A. Boxall, Occurrence and fate of human pharmaceuticals in theenvironment, Rev. Environ. Contam. Toxicol. 202 (2010) 153–154.

[3] J.M. Brausch, G.M. Rand, A review of personal care products in the aquatic environment: Environmental concentrations and toxicity, Chemosphere 82 (2011) 1518–1532.

[4] P. Verlicchi, M.A. Al Aukidy, E. Zambello, Occurrence of pharmaceutical compounds in urban wastewater: Removal, mass load and environmental risk after a secondary treatment—A review, Sci. Total Environ. 429 (2012) 123–155.

[5] R. Andreozzi, M. Canterino, R. Marotta, N. Paxeus, Antibiotic removal from wastewaters: The ozonation of amoxicillin, J. Hazard. Mater. 122 (2005) 243–250.

[6] W. Li, V. Nanaboina, Q.X. Zhou, G.V. Korshin, Effects of Fenton treatment on the properties of effluent organic matter and their relationships with the degradation of pharmaceutical sand personal care products, Water Res. 46 (2012) 403–412.

[7] R. Salgado, V.J. Pereira, G. Carvalho, R. Soeiro, V. Gaffney, C. Almeida, V. Vale Cardoso, E. Ferreira, M.J. Benoliel, T.A. Ternes, A. Oehmen, M.A.M. Reis, J.P. Noronha, Photodegradation kinetics and transformation products of ketoprofen, diclofenac and atenolol in pure water and treated wastewater, J. Hazard. Mater. 244–245 (2013) 516–527.

[8] G.T. Güyer, N.H. Ince, Degradation of diclofenac in water by homogeneous and heterogeneous sonolysis, Ultrason. Sonochem. 18 (2011) 114–119.

[9] X. Yang, R.C. Flowers, H.S. Weinberg, P.C. Singer, Occurrence and removal of Pharmaceuticals and personal care products (PPCPs) in an advanced wastewater reclamation plant, Water Res. 45 (2011) 5218–5228.

[10] C.A. Martínez-Huitle, M.A. Quiroz, Ch Comninellis, S. Ferro, Electrochemical incineration of chloranilic acid using Ti/IrO_2, Pb/PbO_2 and Si/BDD electrodes, Electrochim. Acta 50 (2004) 949–956.

[11] M. Khraisheh, J. Kim, L. Campos, A.H. Al-Muhtaseb, A. Al-Hawari, M. Al Ghouti, G.M. Walker, Removal of pharmaceutical and personal care products (PPCPs) pollutants from water by novel TiO2–Coconut Shell

Powder (TCNSP) composite, J. Ind. Eng. Chem. 20 (2014) 979–987.
[12] C.I. Kosma, D.A. Lambropoulou, T.A. Albanis, Investigation of PPCPs in wastewater treatment plants in Greece: Occurrence, removal and environmental risk assessment, Sci. Total Environ. 466–467 (2014) 421–438.
[13] J.R. Backhurst, J.M. Coulson, F. Goodridge, R.E. Plimley, M. Fleischmann, A preliminary investigation of fluidized bed electrodes, J. Electrochem. Soc. 116 (1969) 1600–1607.
[14] L. Wei, S. Guo, G. Yan, C. Chen, X. Jiang, Electrochemical pretreatment of heavy oil refinery wastewater using a three-dimensional electrode reactor, Electrochim. Acta 55 (2010) 8615–8620.
[15] A. Fortuny, J. Font, A. Fabregat, Wet air oxidation of phenol using active carbon as catalyst, Appl. Catal. B: Environ. 19 (1998) 165–173.
[16] F.L. Cking, H. Koeser, M. Jank, A. Ritter, Iron powder, graphite and activated carbon as catalysts for the oxidation of 4-chlorophenol with hydrogen peroxide in aqueous solution, Water Res. 32 (1998) 2607–2614.
[17] M. Zhou, L. Lei, The role of activated carbon on the removal of p-nitrophenol in an integrated three-phase electrochemical reactor, Chemosphere 65 (2006) 1197–1203.
[18] L. Xu, H. Zhao, S. Shi, G. Zhang, J. Ni, Electrolytic treatment of C.I. Acid Orange 7 in aqueous solution using a three-dimensional electrode reactor, Dyes Pigm. 77 (2008) 158–164.
[19] H.Z. Zhao, Y. Sun, L.N. Xu, J.R. Ni, Removal of Acid Orange 7 in simulated wastewater using a three-dimensional electrode reactor: Removal mechanisms and dye degradation pathway, Chemosphere 78 (2010) 46–51.
[20] L. Yan, H. Ma, B. Wang, Y. Wang, Y. Chen, Electrochemical treatment of petroleum refinery wastewater with three-dimensional multi-phase electrode, Desalination 276 (2011) 397–402.
[21] C. Alfonsín, A. Hospido, F. Omil, M.T. Moreira, G. Feijoo, PPCPs in wastewater-Update and calculation of characterization factors for their inclusion in LCA studies, J. Cleaner Prod. 83 (2014) 245–255.
[22] M. Han, Z. Zhao, F. Cui, W. Gao, J. Liu, Z. Zeng, Pretreatment of contaminated raw water by a novel double-layer biological aerated filter for drinking water treatment, Desalin. Water Treat. 37 (2012) 308–314.
[23] Y. Feng, J. Qi, L. Chi, D. Wang, Z. Wang, K. Li, X. Li, Production of sorption functional media (SFM) from clinoptilolite tailings and its performance investigation in a biological aerated filter (BAF) reactor, J. Hazard. Mater. 246–247 (2013) 61–69.
[24] State Environmental Protection Administration of China, Monitoring and Analysis Methods of Water and Wastewater, fourth ed., China Environmental Science Press, Beijing, 2002.
[25] A. Eaton, L. Clesceri, A. Greenberg, Standard Methods for Examination of Water and Wastewater, 21st ed., American Public Health Association, Washington, 2005.
[26] L. Ciríaco, C. Anjo, J. Correia, M.J. Pacheco, A. Lopes, Electrochemical degradation of Ibuprofen on Ti/Pt/PbO_2 and Si/BDD electrodes, Electrochim. Acta 54 (2009) 1464–1472.
[27] P. Harremoes, A. Haarbo, M. Winthernielsen, C. Thirsing, Six years of pilot plant studies for design of treatment plants for nutrient removal, Water Sci. Technol. 38 (1998) 219–226.
[28] M. Beccari, A.C. Di Pinto, R. Ramadori, M.C. Tomei, Effects of dissolved oxygen and diffusion resistances on nitrification kinetics, Water Res. 26 (1992) 1099–1104.
[29] Y. Hardjono, H. Sun, H. Tian, C.E. Buckley, S. Wang, Synthesis of Co oxide doped carbon aerogel catalyst and catalytic performance in heterogeneous oxidation of phenol in water, Chem. Eng. J. 174 (2011) 376–382.
[30] C.T. Wang, W.L. Chou, Y.M. Kuo, F.L. Chang, Paired removal of color and COD from textile dyeing wastewater by simultaneous anodic and indirect cathodic oxidation, J. Hazard. Mater. 169 (2009) 16–22.