

Removal of perfluorinated compounds in wastewater treatment plant effluents by electrochemical oxidation

Chunhui Zhang, Liangliang Wang, Juan Li, Peidong Su and Chen Peng

ABSTRACT

The presence of perfluorinated compounds (PFCs) in the effluents of a wastewater treatment plant (WWTP) in Beijing was investigated in the current study. Perfluorooctanoate acid and perfluorooctane sulfonate were the predominant PFCs in wastewater, accounting for 19–27% and 18–31%, respectively. The concentrations of PFCs with long chains were much lower than those PFCs with short chains ($\leq C8$). An electrochemical oxidation reactor was employed for advanced treatment of PFCs in WWTP effluents using stainless steel plates as anode and cathode electrodes. It was concluded that the removal efficiency of PFCs was improved accordingly with the increasing applied current density. The removal efficiencies of target PFCs ranged from 23.53 to 51.79% with a reaction time of 30 minutes, current density of 20 mA/cm², electrode plate distance of 1.0 cm and electrode plate amounts of five pairs.

Key words | defluorination, electrochemical oxidation, perfluorinated compounds, persistent organic pollutants, wastewater treatment plant

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INTRODUCTION

Perfluorinated compounds (PFCs), with the capacity to form smooth surfaces resisting water, oil, stain, etc., have been applied in a great variety of industrial products, such as paints, surfactants, coatings, emulsifiers, fire retardants and polymers in the past several decades (Prevedouros *et al.* 2006; Sun *et al.*, 2012). PFCs can be released into the environment during use and disposal. Previous studies have found that PFCs have been detected in the atmosphere, water, soil environment and biological organisms all over the world (Martin *et al.* 2004; Conder *et al.* 2008; Huset *et al.* 2008). Many of PFCs are long-term persistence which have potential hazards, i.e. toxic or bioaccumulated in the environment. The contaminated wastewaters, especially wastewater treatment plant (WWTP) effluents, are considered as a major source of PFCs to natural water (Boulanger *et al.* 2005; Pan *et al.* 2010). For PFCs released from WWTP effluents, there is an increasing concern on their potential for contaminating surface and underground water (Huset *et al.* 2011).

In urban areas of China, most wastewaters are treated in WWTPs before discharge or reusing. However, the existing traditional and advanced treatment processes mainly aim at the remediation of chemical oxygen demand (COD),

ammonia nitrogen, total nitrogen, total phosphorus and other conventional organic pollutants. Micro pollutants treatment, including PFCs and other persistent organic pollutants (POPs), are not considered in the design of WWTPs (Zhang *et al.* 2013b). It is necessary to find an effective and efficient method to remove PFCs in wastewater.

Recently, some techniques, including adsorption, photo-electrocatalysis, ultrasonic irradiation, zero-valent iron and so on have been investigated to remove PFCs in wastewater. However, these technologies exist the common shortages, such as short service life, high construction and running costs, and unstable removal efficiencies (Wang & Shih 2005). Compared with the above methods, electrochemical oxidation has caused wide attention as one of the economical- and environmental-friendly technologies in the field of wastewater treatment (Zhu *et al.* 2009). It has been successfully applied in the remediation of coking, dye, tannery wastewater and landfill leachate (Ma *et al.* 2009). The electrochemical oxidation mechanism is supposed to generate H₂O₂ at cathode with the dissolved oxygen and obtained at anode in wastewater (Li *et al.* 2005; Shi *et al.* 2009).





Another course of chemical reaction may possibly happen,



Therefore, the HO· is generated by H₂O₂.



Organic compounds can be removed after the oxidation by HO·, as interpreted as follows:

Organic pollutants



Stainless steel, aluminum, diamond and titanium, etc., are often used as electrodes in electrochemical oxidation processes (Zhu *et al.* 2008). Stainless steel electrodes, especially, are dissolved and form Fe²⁺ (aq) ions and Fe³⁺ (aq) ions. The organic pollutants can be co-precipitated as sediments besides electrochemical oxidation (Golder *et al.* 2007). Compared with other electrode materials, the stainless steel is the cheapest. For the above views, an effective electrochemical oxidation method along with stainless steel electrodes for the removal of PFCs in wastewater effluents is proposed in this paper.

METHODS

Chemicals

All chemicals were of analytical grade and used as received without any further purification treatments. Organic chemicals such as perfluorobutanoic acid (PFBA, 99.5%), perfluoropentanoic acid (PFPeA, 95%), perfluorohexanoic acid (PFHxA, 98%), perfluoroheptanoic acid (PFHpA, 98%), perfluorooctanoate acid (PFOA, 98%), perfluorooctane sulfonate (PFOS, 99%), perfluorononanoic acid (PFNA, 98%), perfluorodecanoic acid (PFDA, 98%), perfluoroundecanoic acid (PFUnA, 96%), perfluorododecanoic

acid (PFDoA, 95%), potassium perfluorobutanesulfonate acid (PFBS, 98%) and sodium perfluorohexanesulfonate acid (PFHxS, ≥98%), were purchased from Alfa Aesar Co., Ltd, Ward Hill, MA, USA. The internal standard organics like ¹³C₄-PFOS (MPFOS, >98%) and ¹³C₈-PFOA (MPFOA, 99%) were mainly obtained from Wellington Laboratories (Guelph, ON, Canada), while the standards and stock solutions were cautiously prepared using HPLC grade methanol (99%) and then reserved at 4 °C in polypropylene (PP) tubes. Other chemicals such as ammonium acetate (NH₄OAc, 97%), ammonium hydroxide (50%), sodium hydroxide (>98%) and acetic acid (≥99.5%) were purchased from Alfa Aesar Co., Ltd, Ward Hill, MA, USA and Beijing Chemical Reagent Factory (Beijing, China), respectively. During the experiment, all solutions were prepared using Milli-Q ultrapure water.

Collection of wastewater samples

The wastewater effluent samples were systematically collected in high-density polyethylene bottles (1,000 mL) from effluents in Miyun Tanzhou WWTP (founded in 1991, Beijing, China; Treatment process: SBR + MBR + O₃; Treatment capacity: 80,000 m³/d) during the summer and autumn of 2013. The wastewater samples were collected four times within 24 hours and were well mixed with equal volume because of the relatively constant emission of wastewater effluents. The above sampling bottles were immediately transported to the laboratory within four hours by an ice chamber and then the bottles were preserved in a refrigerator at 4 °C for further analysis.

Preparation of wastewater sample

The wastewater samples were extracted using solid-phase extraction (SPE) as described by Li *et al.* (2011) with slight modifications. Two-hundred mL filtrated water samples were briefly spiked with 5 ng of ¹³C₄-PFOS and ¹³C₈-PFOA as internal standards and extracted with Oasis HLB cartridges (6 cc, 200 mg; Waters Corp., Milford, MA, USA) (Becker *et al.* 2008). The final elution was concentrated to 1 mL for injection.

Chemical analysis

Concentrations of PFCs in wastewater samples were measured by using high performance liquid chromatography (HPLC) coupled with an electrospray ionization tandem mass spectrometer (HPLC-ESI/MS/MS, API 3200; Applied

Biosystems, Foster City, CA, USA) operated in negative mode. The HPLC separation was carried out by a Dionex Acclaim 120 C18 column (4.6 mm × 100 mm, 5 μm) using a gradient composition of methanol and 50 mM NH₄OAc. More details concerning the method have been given by Shi *et al.* (2010). The pH, total organic carbon (TOC), suspended solids (SS), COD and 5-day biochemical oxygen demand (BOD₅) were analyzed in our laboratory according to standard methods of China (Xi *et al.* 2008).

Quality assurance and quality control

The procedure of quality assurance and quality control (QA/QC) was described by Pan *et al.* (2010). Milli-Q water samples (0.02 L, $n = 10$) were spiked with 1 ng target standards (PFCs) and treated following the same procedures as the wastewater sample. The calibration curve was prepared with a series of concentrations (1–50 μg/L) which showed strong linearity with correlation coefficients of >0.99. The PFCs were extracted with the recoveries ranging between 89.5 and 116.2%, indicating that the SPE extraction was sufficient to support quantitative extraction. Meanwhile, duplicate matrix spikes were prepared for each sample. More details concerning QA/QC have been given in the study of Shi *et al.* (2010). Ultrapure Milli-Q water was used as blank solution with wastewater samples. The concentrations of the samples were calibrated by the blank concentration.

Electrochemical oxidation procedure

The electrolytic cell was designed as a parallelepipedic plexiglas reactor (Figure 1). The valid working volume, length,

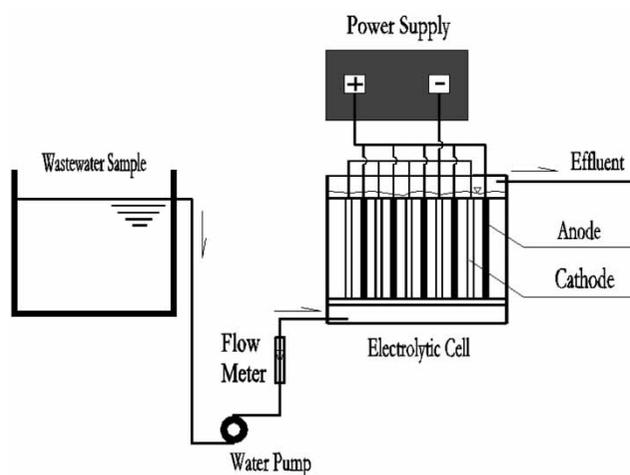


Figure 1 | Schematics of electrolytic reactor cell.

width and height of reactor were 6,000 mL, 30 cm, 20 cm and 15 cm, respectively. In all cases, stainless steel electrode plates (dimensions: 20 cm × 10 cm; thickness: 1 mm) were used as anode/cathode pair. The cathode and anode electrodes were alternately arranged and placed in the middle of the reactor. The supplied power was a MPS 702 DC power generator in this experiment, while cell potential was controlled by a voltmeter.

RESULTS AND DISCUSSION

PFCs in wastewaters

The characteristics of the initial wastewater effluent samples used in the experiment are listed in Table 1. The concentrations of the target PFCs detected in the initial wastewater effluent samples are summarized in Table 2.

From all the quantitatively determined PFCs, PFOA and PFOS were the predominant PFCs in wastewater, accounting for 19–27% and 18–31%, respectively. The concentrations of PFDoA were lower than the limit of detection both in influents and effluents. The PFOA in influents and effluents were proved to contain approximately equal concentration. The concentrations of PFDA, PFOS and PFBS were generally higher in effluents than in influents. The increase of PFDA, PFOS and PFBS in effluents may be due to the wastewater treatment processes that might cause biodegradation of some precursors or the potential additional sources. This phenomenon has also been reported in some former studies (Sinclair & Kannan 2006; Loganathan *et al.* 2007; Pan *et al.* 2010). For other PFCs, the concentrations were decreased in different extent after treatment of WWTP. In all, the concentrations of PFCs with long chains, PFDA, PFNA, PFDoA and PFUnA, were much lower than those of PFOA, PFOS, PFPeA, PFHpA and PFHxA with short chains ($\leq C8$). In contrast, the PFCs concentrations in influent and effluent

Table 1 | Characteristics in the initial wastewater effluent samples*

Parameter	Unit	Mean
pH	/	7.23
COD	mg/L	68.6
TOC	mg/L	27.5
BOD ₅	mg/L	23.4
SS	mg/L	18.2

*The quantity of samples analyzed: 4.

Table 2 | PFCs concentrations in the initial wastewater effluent samples* (ng/L)

Parameters	Mean ± S.D.			
	09 June		22 September	
	Influent	Effluent	Influent	Effluent
PFD _o A	ND	ND	ND	ND
PFUnA	0.093 ± 0.0091	0.025 ± 0.0066	0.086 ± 0.0063	0.032 ± 0.0055
PFDA	0.055 ± 0.0051	0.078 ± 0.0061	0.061 ± 0.0058	0.065 ± 0.0088
PFNA	0.046 ± 0.0037	0.034 ± 0.0058	0.051 ± 0.0045	0.018 ± 0.0042
PFOA	5.43 ± 0.0529	5.20 ± 0.0497	5.03 ± 0.0952	5.28 ± 0.0622
PFHpA	3.52 ± 0.0716	2.75 ± 0.0668	3.85 ± 0.0622	2.55 ± 0.0683
PFHxA	2.77 ± 0.0606	1.45 ± 0.0698	2.86 ± 0.0548	1.58 ± 0.0469
PFPeA	2.73 ± 0.0762	1.92 ± 0.0605	2.85 ± 0.0535	1.74 ± 0.0770
PFBA	2.95 ± 0.0622	0.82 ± 0.0476	2.88 ± 0.0424	0.75 ± 0.0424
PFOS	4.74 ± 0.0483	6.38 ± 0.0476	4.93 ± 0.0787	5.41 ± 0.0316
PFHxS	3.64 ± 0.0392	1.80 ± 0.0440	3.38 ± 0.0548	2.12 ± 0.0476
PFBS	0.86 ± 0.0424	0.95 ± 0.0337	0.93 ± 0.0516	1.05 ± 0.0622
ΣPFCs (ΣMean)	26.83	21.41	26.91	20.60

*The quantity of samples analyzed: 4.
ND: not detected.

wastewater of WWTP, it could be seen that PFCs concentrations did not change much. This indicated that the existing WWTP processes had no apparent effects on removal of PFCs. Hence, some advanced treatment methods must be applied to remove PFCs in wastewaters. The PFCs values measured in this study were comparable to those reported by Pan *et al.* (2010), which were quite lower than some other former studies (Loganathan *et al.* 2007; Wang *et al.* 2009). It has been assumed that the industrial and municipal wastewaters may be a likely source of PFCs (Schultz *et al.* 2006). The differences in PFC concentrations in different WWTPs are most likely due to the diversity of treatment processes, operating processes and the source of collected wastewater.

Removal of PFCs by electrochemical oxidation

Effect of current density and reaction time

Previous studies indicated that reaction time, applied current density, electrode distance and amounts of electrode plate played major roles in the removal efficiency of pollutants in wastewater (Matamoros *et al.* 2009). According to the results of our former research, the selected electrode plates were set as five pairs and the electrode distance was uniformly set as 1.0 cm (Zhang *et al.* 2013a). The capability of electron transfer

and hydroxyl radical generation may control the efficiency of pollutants in the electrochemical oxidation process. However, the above abilities, especially hydroxyl radical generation capability, depended on the applied current density and reaction time (Wang *et al.* 2008). Hence, the effect of applied current values on the removal efficiency of PFOA in wastewater effluent samples was investigated by setting the current density to 10, 15, 20 and 25 mA/cm², and the reaction time was set from 0 to 60 minutes. The initial concentration of PFOA was 5.23 ng/L.

Figure 2 shows that the degradation efficiency of PFOA increased accordingly with an increase in the applied current density. After 30 minutes reaction time, the highest average PFOA removal ratio reached 25 mA/cm². As expected, the PFOA removal ratio increased with increasing reaction time. For example, the average PFOA removal ratio was 28.4% after 20 minutes electrolysis, while it increased to 43.9% after 30 minutes electrolysis with applied current density of 20 mA/cm². A possible explanation is that the capability of electron transfer and hydroxyl radical generation may govern the degradation and defluorination efficiency of PFOA in aqueous solution. The above abilities, especially hydroxyl radical generation capability, depends on the applied current density (Zhuo *et al.* 2011; Lin *et al.* 2012). The trends of PFOA degradation efficiencies of our work were consistent with former studies (Ochiai *et al.* 2011; Zhuo *et al.* 2012). The removal efficiency of PFOA

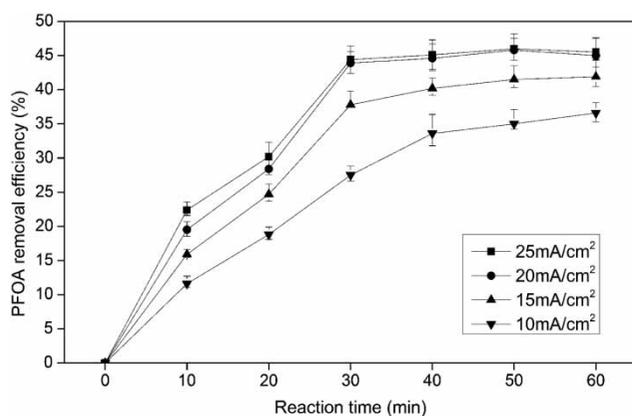


Figure 2 | Removal efficiencies for PFOA in wastewater effluent samples.

changed slightly after the reaction time was 30–60 minutes. However, the prolonged reaction time will increase the energy cost. For applied current density, the removal efficiencies of PFOA were almost the same at 20 and 25 mA/cm². To save energy, we selected an applied current of 20 mA/cm² and reaction time of 30 minutes as the reaction parameters of electrochemical oxidation during the following experiments.

Removal efficiency of PFCs

The electrochemical reactor was operated under the above reaction parameters, which included reaction time (30 minutes), current density (20 mA/cm²), plate distance (1.0 cm) and electrode plate amounts (five pairs). The removal efficiencies for PFCs in wastewater effluents are summarized in Table 3.

It can be seen from Table 3 that the removal efficiencies of target PFCs ranged from 23.53 to 51.79% under the condition as following: current density of 20 mA/cm², electrode plate distance of 1.0 cm, electrode plate amounts of five pairs and reaction time of 30 minutes. The removal efficiencies of PFCs with long chains were much lower than those with short chains ($\leq C8$), such as PFOA and PFOS. Moreover, the PFCs with higher initial concentrations (PFOA, PFOS, PFHpA) can achieve higher removal efficiencies. The initial concentration is one of the most important affecting parameters on the removal efficiencies. The higher initial PFC concentrations, the better removal efficiencies of PFCs would be achieved with longer reaction time. Some former data on the remediation of PFOA and PFOS had been researched, along with the more limited information currently available on other PFCs (Lau *et al.* 2007). For example, Lin *et al.* (2012) used Ti/SnO₂-Sb electrodes

Table 3 | The removal effect of PFCs after electrochemical oxidation*

Parameters	Influent		Effluent		Removal efficiencies (%)
	Mean (ng/L)	S.D.	Mean (ng/L)	S.D.	
PFDoA	ND		ND		-
PFUnA	0.034	0.0059	0.026	0.0071	23.53
PFDA	0.098	0.0094	0.045	0.0069	54.08
PFNA	0.032	0.0078	0.021	0.0037	34.38
PFOA	5.46	0.0829	2.93	0.0559	46.34
PFHpA	3.22	0.0535	1.86	0.0516	42.24
PFHxA	1.56	0.0316	0.93	0.0605	40.38
PFPeA	1.72	0.0516	1.09	0.1080	36.63
PFBA	0.95	0.0182	0.68	0.0392	28.42
PFOS	6.16	0.0432	2.97	0.0787	51.79
PFHxA	2.05	0.0622	1.21	0.0408	40.98
PFBS	1.14	0.0365	0.85	0.0529	25.44

*The number of samples analyzed: 4.
ND: not detected.

to remediate PFOA with an initial concentration of 100 mg/L, the results indicated that the defluorination rate could achieve above 73.9% after 90 minutes electrolysis. Zhuo *et al.* (2012) applied boron-doped diamond electrodes for PFOA degradation at different initial concentrations (20, 30, 50 mg/L) with current density of 23.24 mA/cm² after 30 minutes electrolysis. The results showed that the removal ratios were 79.33%, 70.99% and 68.54%, respectively. For PFCs remediation, most of the previous studies focussed on degradation mechanism research and the initial PFCs concentrations which were very high (generally several hundreds of ng/L or above 1.0 mg/L). According to other authors the low organic pollutants in water, the rapid decreasing of the reaction rate and low removal efficiency is achieved by electrochemical oxidation (Liao & James 2009). With regard to the low PFCs concentrations as shown in this study, so far, the electrochemical oxidation method has never been reported.

However, PFCs are extremely resistant to environmental degradation processes and thus persist indefinitely. Unlike most other persistent and bioaccumulative organic pollutants, PFCs are water-soluble and bioaccumulate in serum rather in fat (Post *et al.* 2012). And PFOA has been classified as likely to be carcinogenic to humans by the USEPA Science Advisory Board (USEPA 2006). Hence, it is important and urgent to find some feasible and economical technologies to degrade PFCs to harmless species. Conventional technologies, such as biological degradation,

oxidation and reduction, are difficult to destruct PFCs in water due to their stable properties. Although some special techniques like ultrasonic irradiation, zero-valent iron, ultraviolet irradiation may remediate PFCs in solution, the special conditions and high energy consumption are required (Ochoa-Herrera et al. 2008). The electrochemical oxidation method has advantages for its simple equipment, less land demand and easy to be controlled and operated automatically. More important, it will not bring second pollution and produce poisonous intermediate products (Xu et al. 2008). The electrochemical oxidation can be a promising method for the advanced treatment of PFCs and other POPs in WWTP effluents.

CONCLUSIONS

An electrochemical oxidation reactor, using stainless steel plates as anode and cathode electrode, for advanced treatment of PFCs in WWTP effluents was investigated. In the investigated conditions, the removal efficiencies of target PFCs ranged from 23.53 to 51.79%. The reaction time and applied current density were the major operating factors affecting on the removal efficiency of PFCs in wastewater. PFCs are harmful for human health. They are very stable organics, which made them resistant to reduction and oxidation degradation and were environmental persistent. It is necessary to find an effective and efficient method to remove PFCs in wastewater. Although we used a simple reactor with electrochemical oxidation method, it would be attractive to develop a similar continuous electrochemical oxidation reactor system for the advanced treatment of PFCs in WWTP effluents.

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