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Enhancement of Antibiotic Removal in Membrane Permeate by Ozonation

Vo Thi Kim Quyen[1], Cao Ngoc Dan Thanh[2], Luu Vinh Phuc[2], Vo Thi Dieu Hien[2], Nguyen Nhu Sang[2], Tran Thanh Dai[3], Bui Xuan Thanh[2,*]

[1]Ho Chi Minh City University of Food Industry

[2]University of Technology, Vietnam National University–Ho Chi Minh City

[3]Dong Nai University of Technology, Bien Hoa city, Vietnam

ABSTRACT

In this study, the performance of ozone treatment was evaluated for removing antibiotics from permeate of membrane bioreactor (MBR) in hospital wastewater treatment. Four widely used of antibiotics were targeted including Sulfa-methoxazole (SMX), Norfloxacin (NOR), Ciprofloxacin (CIP) and Ofloxacin (OFL). Before treatment, CIP was found to be the dominant antibiotic due to the highest concentration ranging from 12.462 to 13.542 µg/L. Lower concentrations were observed for NOR (8.643-10.928 µg/L), OFL (9.164-9.864 µg/L) and SMX (1.864-3.247 µg/L) in influent samples. The operational condition of ozonation was found to be ideal parametersat pH of 8.5 and contact time of 10 minutes for this study. In details, SMX, NOR, CIP and OFL were mostly removed by 70% at this condition. In conclusion, the removal efficiency of those antibiotics by ozone treatment was depending on their contacting time with ozone and pH condition.

Keywords: Antibiotic; ozonation; hospital; wastewater

1. INTRODUCTION

Nowadays, several research conducted in environmental field has been extended beyond matters such as polychlorinated biphenyls (PCBs), dioxins and pesticides to some emerging pollutants which are potential to cause serious issues for our environment and risk for human health. Among them, an important group of pharmaceuticals so-called antibiotics which were utilized as human and animal medicine. They might be supposed to be dangerous due to their continual input into aquatic environment.

Antibiotics are used to treat health problems for human and veterinary. After being used, they are excreted with urine and feces. In addition, expired antibiotics are inadequately discharged. The residue of individual antibiotics are detected at concentration of the range from 0.4 ng/L and 35.5 µg/L in natural water, wastewater and drinking water (Mompelat et al., 2009; Snyder et al., 2007). NOR and OFL were detected 120 ng/L and 20 ng/L in surface water, respectively (Christian et al., 2003; Kolpin et al., 2002). In the effluents of sewage treatment plants (STPs), OFL and SMX were detected up to 82 ng/L and 370 ng/L, respectively (Alexy et al., 2006). Although the concentrations of antibiotics in aquatic environment are quite low, its continuous inputs may cause the dangerous

[*]Corresponding to: bxthanh@hcmut.edu.vn

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accumulation and also potential risks for living creatures in both aquatic and terrestrial ecosystems. Moreover, antibiotics also induce resistance in strains of bacteria and upset the ecological balance (Lanzky and Halting-Sørensen, 1997). There are some studies about *Daphnia*, algae and bacteria being chronically affected by antibiotics (Boxall et al., 2003; Halling-Sørensen, 2000; Kümmerer et al., 2000; Yamashita et al., 2006).

Based on found report, there are just some kinds of available antibiotics in markets belong to different classes have been detected in wastewater mainly from East Asia, North America, Europe and Australia. So far, there was no report on the presence of antibiotics in Vietnam except one study conducted in Hanoi-Vietnam by Duong and colleagues. In particular, they studied the antibiotics discharged from six hospitals in Hanoi and the concentrations of CIP and NOR were 1.1-44.0 µg/L and 0.9-17.0 µg/L, respectively (Duong et al., 2008).

The major removal mechanisms of antibiotics in wastewater treatment methods include adsorption, biodegradation, disinfection as well as membrane separation. Besides, hydrolysis, photolysis and volatilization also play minor roles for elimiting antibiotics. However, depending on the class of antibiotics, they will have dominant pathways to be removed by biological processes. Many previous studies have demonstrated that conventional biological treatment system cannot remove antibiotics completely and they will subject into the environment via effluent or sludge (Batt et al., 2006, 2007; Golet et al., 2002, 2003; Ternes et al., 2004). On the other hand, the removal efficiencies and biodegradation rates reported for some antibiotics varied greatly among different studies. For instance, quinilones could be removed by adsorption rather than biodegradation. In some studies, antibiotics belong to quinilones class as Norfloxacin (NOR), Ciprofloxacin (CIP) and Ofloxacin (OFL) were reduced by 87-100% (Lindberg et al., 2006; Vieno et al., 2006), 85% (Batt et al., 2007) and 75-77% (Brown et al., 2006; Radjenović et al., 2009). Moreover, there are some studies on the transformation of sulfamethoxazole (SMX) demonstrating that it was very low adsorption of SMX on sludge in biological treatment as activated sludge (Batt et al., 2007; Pérez et al., 2005). At the same point, Watkinson et al. (2007) also indicated that SMX could be removed with an average elimination efficiency of 25%. One more report about poor SMX removal efficiency of 20% was also introduced in biological treatment process (Brown et al., 2006).

Therefore, it is necessary to treat antibiotics adequately so that advanced process asozonationis recommended to solve this issue. Ozone (O_3) has been known as a strong oxidizer and due to its high oxidation capacity, ozone is used for disinfection, color removal, increasing biodegradability for refractory organic compounds, and especially eliminating antibiotics. Ozone reacts with organic compounds in two different ways: by direct oxidation as molecular ozone or by indirect reaction through formation of secondary oxidants like hydroxyl radical OH• (Baig and Liechti, 2001) produced by the decomposition of ozone. Hydroxyl free radical (OH•) is known as a strong oxidant to destroy compounds that cannot be oxidized by conventional oxidant (Carey, 1992) due to hydroxyl radicals react non-selectively with organic compounds present in wastewater, including antibiotics. This is the reason why ozonation can remove multiple cleasses of antibiotics in effluent. There were many researches under taken on antibiotics removal by ozone treatment along with the studies of contacting time. For examples, SMX, Sulfapyridine (SUL), Azithromycin (AZI), Clarithromycin (CLA) and Roxithromycine (ROX) were removed by 87.4, 93.9, 92.6, 84.6

and 90.9%, respectively, with the contacttime of 27 minutes and pH of 7.95 (Nakada et al., 2007). On the other hand, the removal efficiency of SMX, CIP, Enrofloxacin (ENR) and Tylosin (TYL) were more than 99% at contact time of 1 minute and pH of 7.7 was reported (Dodd et al., 2006). Huber also reported about the SMX removal efficiency of 90-99% at contacting time of 8.4 ± 0.4 minutes and pH varying from 7-7.5 (Huber et al., 2005). In a German municipal sewage treatment plant (STP), SMX was removed about 92% by onzone-UV disinfection at contacting time of 18 minutes and pH of 7.2 (Ternes et al., 2003). On the other hand, the mechanism of ozonation in micropollutants removal was found to be depended on the trasnformation of N-nitrosamoines along with the increase of ozone dosage of 45 mg/L (Fujioka et al., 2014).

The purpose of this study is to evaluate removal efficiencies of SMX, NOR, CIP and OFL after ozone treatment with the under-standing of the effects of contact time and pH. Achieved results would be used as background to conduct further studies.

2. MATERIALS AND METHODS

2.1 Experimental setup

In this study, ozonation process occurred in the ozone reactor was made from glass material and designed to have working volume of 2 L and dimension of WxH as 8 cm x 42 cm. Ozone was generated from oxygen in the ambient air by using FD-3000 II model ozone generator. The diffuser system was placed at the bottom of the reactor to subjected ozone at flow rate of 20-40 mg O_3/h. The released ozone was blowed into gas absorption bottle containing 2% KI solution.

2.2 Operational conditions

Hospital wastewater was collected in the equalization tank of the wastewater treatment plant of a hospital in HCMC. After that, hospital wastewater was treated by a sponge membrane bioreactor (SMBR) in order to reduce the concentration of pollutants. The operating conditions of sponge membrane bioreactor was simimar and mentioned in Nguyen et al. (2016). Membrane permeate was flowed to ozone reactor to enhance the antibiotics removal efficiency. Initially, in order to make clear about the ozone behavior in reaction, the concentrations of ozone (which was reacted or still remaining in tank or lost) after accomplishing treatment were evaluate to find out the ozone balance in this study. Similarly to major objectives of this study, experiments were conducted at different pH and reaction time.

After preliminary study, system were operated at fed-batch mode with variation of pH and contact time. Firstly, during treatment by ozonation, the optimization of pH was carried out by varying pH values at different points of 7, 8, 8.5, 9 and 10 with contact time of 10 mins. Secondly, the suitable concentration of supplied ozone, the contact times were varied from 5, 10, 20 to 40 mins. The input and output were sampled before and after process to analyze and determine the adequate concentrations of antibiotics. All experiments were performed at ambient temperature (25 ± 2˚C).

2.3 Characteristics of membrane perme-ate

Before entering to ozonation tank, raw wastewater was treated by Sponge membrane bioreactor (SMBR) to remove proportion of organic matters and nitrogen. Membrane permeate was subjected into ozonation process with remaining antibiotics concentration of SMX, NOR, CIP and OFL were 1.864-3.247 μg/L, 8.643-10.928 μg/L, 12.462-13.542 μg/L and 9.164-9.864 μg/L, respectively.

2.4 Analytical methods

In this study, the method used for determining the concentration of antibiotics was described previously (Dinh et al., 2011; Siddiqui et al., 2013). Antibiotics were analyzed byhigh performance liquid chromatography-mass spectrometry (HPLC-MS/MS). Ozone consumption was evaluated under Ozone Demand/Requirement-Semibatch Method 2350E (Apha, 1998). Ozone balance was determined as Eq.1 below:

$$\frac{M_c}{V_s} = \frac{M_t - M_a - M_g}{V_s} \quad (1)$$

Where M_c = transferredozone (mg); M_t = applied ozone (mg); M_a = ozone residual (mg); M_g = unreacted ozone absorbed by KI solution (mg) and V_s = volume of wastewater (L).

3. RESULTS AND DISCUSSION

3.1 Optimization of pH

To elucidate the effect of pH on ozonation process to treat antibiotics, different pH values of 7, 8, 8.5, 9 and 10 were examined. Table 1 displays the concentration of antibiotics in the output.

Table 1 showed that the ability to eliminate antiniotics of ozone oxidation process increased slightly at neutral to alkaline pH and there is no significant difference in antibiotics removal efficienciesat various pH values. The results are similar to those of other study. Ozone reacts with organic compounds in two different ways: by direct oxidation as molecular ozone or by indirect oxidation by hydroxyl radicals, and type reaction depending on pH. At acidic environment, direct oxidation by molecular ozone is the major oxidant, whereas indirect oxidation by faster and less selective hydroxyl radicals becomes dominant at pH greater than 7 (Langlais et al., 1991). Low pH decreases the formation of hydroxyl radicals from ozone (Beltran et al., 1999). As a result, Balcioğlu and Ötker (2003) reported that the removal of human antibiotic in wastewater increased from 24% at pH 3 to 69% at pH 7 and remained almost unchanged of 71% at pH 11 (Balcıoğlu and Ötker, 2003).

Fig. 1 shows that ozone consumption levels were different at pH points. For instance, at pH 8.5, the highest ozone consumption of 1.5 mg was recorded. At other pH values such as 7, 8, 9 and 10, ozone consumption levels were 0.3; 1.2; 1.0 and 0.8 mg/L. Although ozone consumption was the highest one at pH 8.5, the removal efficiencies of examined antibiotics were not significantly different in comparision with other pH values. It is assumed that ozone reacts not only with antibiotics but also with other organic compounds in hospital wastewater (e.g. treating COD, colorants, etc.). For this reason, the removal efficiencies of COD and color at pH 8.5 were 23% and 38%, respectively, were higher than others (Fig. 2). Other previous researches also reported that the removal efficiencies of organic compounds were higher at pH from 8.5 to 9 (Azbar et al., 2004; Thanh et al., 2011). Under those circumstances, pH 8.5 was selected as optimal value for system operation since targeted matter as antibiotics and organic matters could be treated well.

3.2 Effect of the ozone contact time

The concentration of supplied ozone also affects to the removal efficiency of antibiotics. Increasing supplied ozone concentration lead to increase ozone consumption, hence, more hydroxyl radicals formed and the removal efficiency was higher. In this research, the ozone generator firstly supplied ozone at the flow rate of 40 mg O_3/h, then, in order to change the concentration of supplied ozone, the time was varied from 5, 10, 20 to 40 mins, corresponding with 1.67 mg O_3/L, 3.34 mg

O_3/L, 6.67 mg O_3/L and 13.34 mg O_3/L, respectively. The results revealed only within 10 mins of reaction since over 70% of SMX, NOR, CIP and OFL were removed.

Antibiotics were removed rapidly when contact time was increased from 0 to 10 min. Although ozone was supplied to increase the concentration corresponding with increasing time of process, antibiotic concentrations were not eliminated more at 20 and 40 min (Fig. 3). The reason might relate to 10 min assumed as abundant time for antibiotics reacted with ozone. Another reason might due to types of tested antibiotics in this study. Dodd et al. (2006) reported that only four types of antibiotics (Penicilin G, Cephalexin, Amikacin and N(4)-acetylsulfamethaxazole) among 14 antibiotics examined were oxidized predominantly by ozone. However, as basic knowledge about ozone process, ozone usually attacks some special functional groups such as C=C double bond or aromatic structure (Huber et al., 2005; Nakada et al., 2007). Fig. 4 shows that the reacted concentration and loss of ozone kept inclearingalong with the increased time while remaing concentration was constant. Since influent of ozonation process did not contain only those targeted antibiotics, ozone reacted favorably with those components which are more attractive than antibiotics at the same time of reaction. Consequently, there is still no clear answer for this phenomenon so that some additional works have to be conducted to find out the mechanism.

Table 1 Concentrations of antibiotics at various pH values (Unit: µg/L)

Antibiotics	**SMX**	**NOR**	**CIP**	**OFL**
MBR permeate	3.247	10.928	13.542	9.864
7	1.353	5.673	6.854	3.546
8	1.258	5.544	6.543	3.325
8.5	1.296	5.343	6.124	3.426
9	1.164	5.237	6.439	3.126
10	1.034	4.784	5.742	3.085

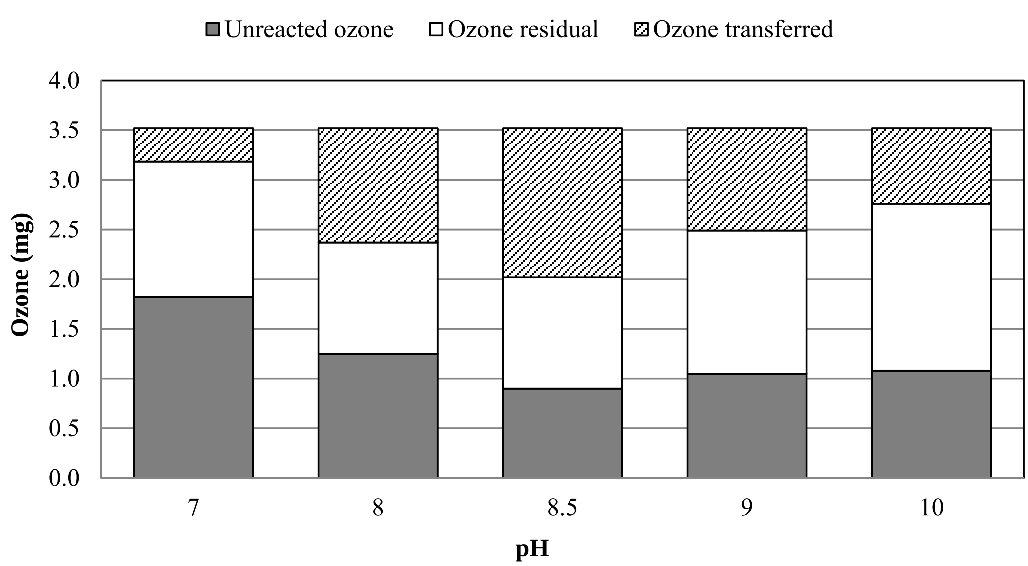

Figure 1 Ozone balance at various pH values

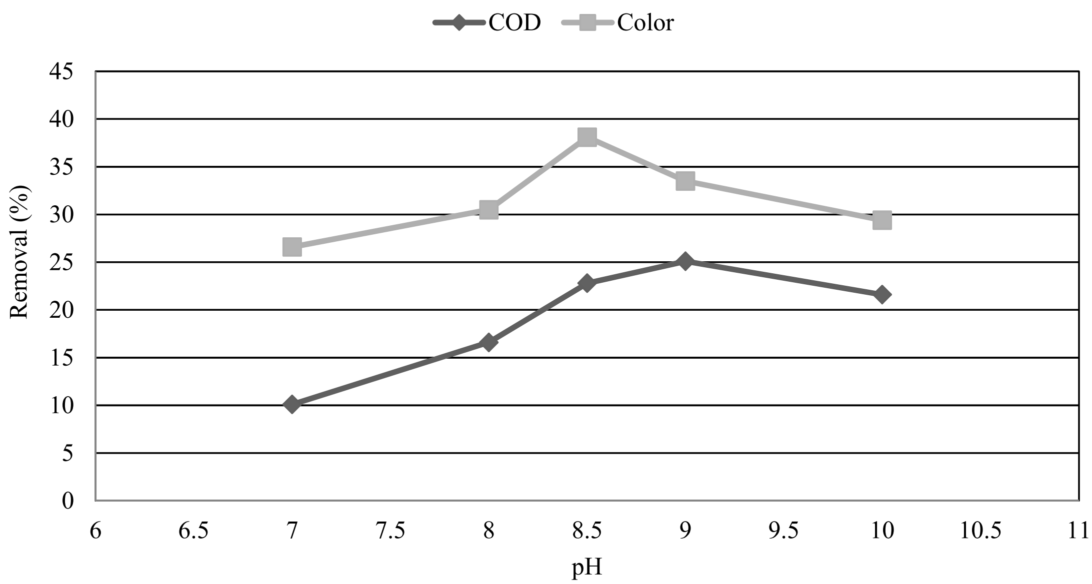

Figure 2 Removal of COD and color at different pH values

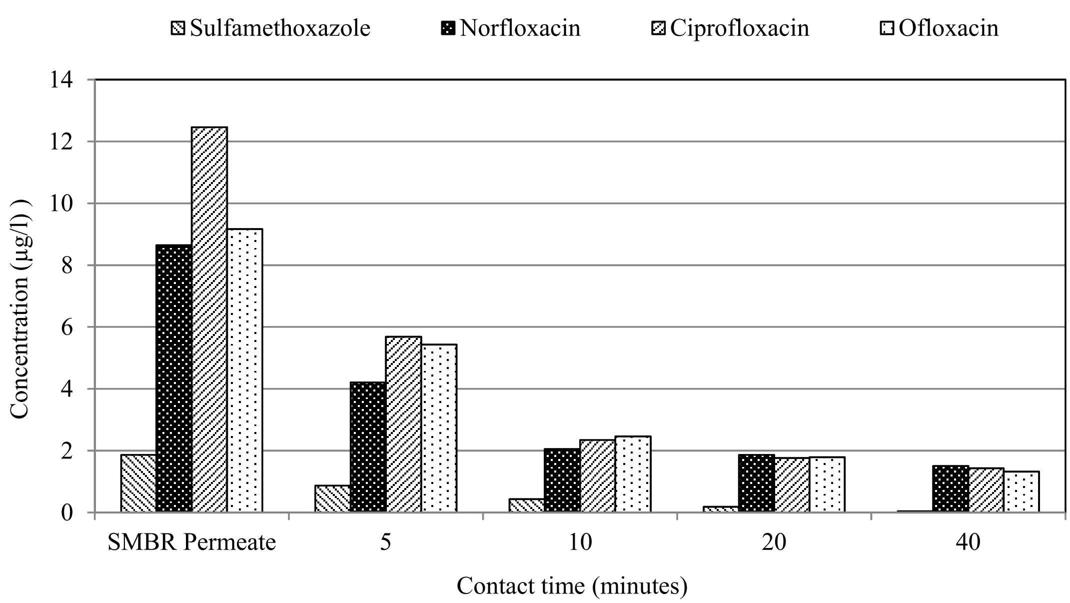

Figure 3 Removal of antibiotics removal at various contact time

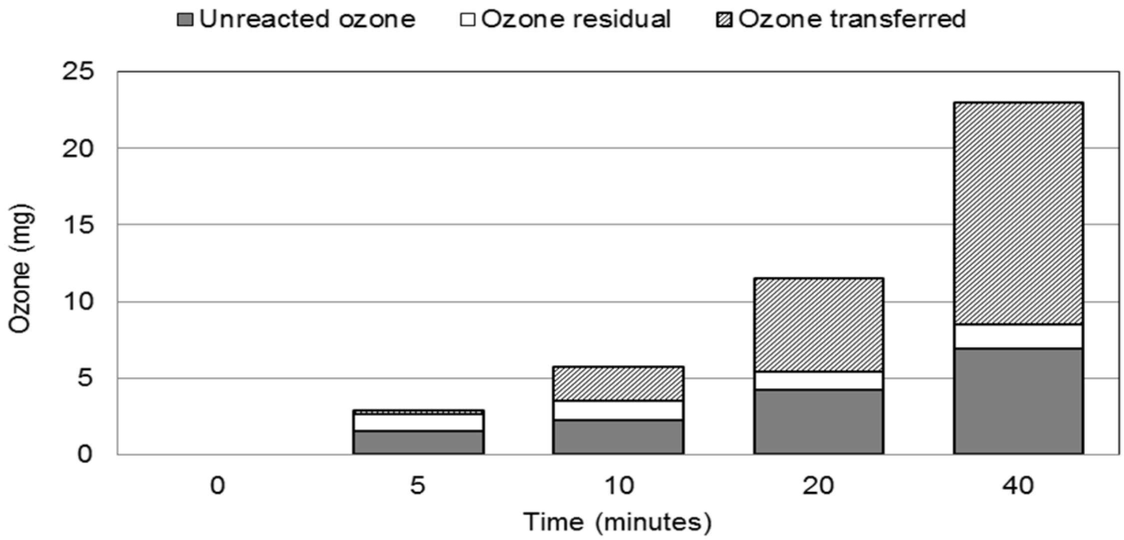

Figure 4 Ozone balance at variation of contact time

CONCLUSIONS

In conclusion, this study found that ozonation process could perform well in terms of antibiotics removal from permeate of membrane bioreactor. Significantly, ideal operational conditions were determined to have pH of 8.5 and contact time of 10 minutes to produce potential ozone concentration of 3.34 mg O_3/L. Only within 10 minutes of reaction since over 70% of Sulfamethoxazole (SMX), Norfloxacin (NOR), Ciprofloxacin (CIP) and Ofloxacin (OFL) were removed from membrane permeate.

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