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COUPLING OF MEMBRANE BIOREACTOR AND OZONATION FOR REMOVAL OF ANTIBIOTICS FROM HOSPITAL WASTEWATER

Vo Thi Kim Quyen[1], Luu Vinh Phuc[2], Nguyen Thanh Tin[2], Vo Thi Dieu Hien[2], Cao Ngoc Dan Thanh[2], Dinh Quoc Tuc[2] & Bui Xuan Thanh[2,*]

[1]*Ho Chi Minh City University of Food Industry*
[2]*University of Technology, Vietnam National University – Ho Chi Minh City*
*Email: bxthanh@hcmut.edu.vn

Abstract - *Antibiotic residues in the environment and their potential toxic effects have been considered as one of the emerging research area in the environmental field. Their continuous introduction in our environment may increase their negative impacts on human health. In this study, the eliminations of antibiotic such as Norfloxacin (NOR), Ciprofloxacin (CIP), Ofloxacin (OFL) and Sulfamethoxazole (SMZ)in wastewaterof hospital were processed by membrane bioreactor (MBR) coupled with ozonation process. In particular, the MBR was applied for the antibiotic removals followed by ozonation process as apost-treatment stage to create an adequate integration to enhance removal efficiency. Achieved results after MBR treatment showed that the removal efficiency of NOR, CIP,OFLand SMZwere90 ± 4.0% , 83 ± 13% , 81 ± 13 % and 39 ± 6%, respectivley. In addition, those antibiotic matters were continously removed by ozonation process with the removal efficiency of 87±9.0% , 83±1.0% , 81±2.3% and 66±2.3% for NOR, CIP,OFL and SMZ, respectively. In summary, antibiotics could be basically limited by the combination of MBR and ozonation before discharging in aquatic environment.*

Keywords: *antibiotic, membrane bioreactor, ozonation, hospital, wastewater.*

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1. INTRODUCTION

Antibiotics are defined as organic compounds that eradicate or inhibit the growth of microorganism which have been used worldwide to treat human and animal health problems. In reality, antibiotic residues are discharged from different sources, including production and manufacturing pharmaceuticals (PCs), therapeutically use of PCs for human and animals, aquaculture and plant agriculture, etc. (Duong et al., 2008)investigated the hospital wastewater in Hanoi, Vietnam and the results showed that most of hospital wastewater was not treated before discharging directly into aquatic environment. Among five antibiotics were evaluated in this study such as CIP, NOR, Levofloxacin (LEV), OFL and Lomefloxacin (LOME), the concentrations of CIP and NOR were varied in range of 1.1-44 and 0.9-17 µg L^{-1}, respectively.The presence of antibiotics in surface and ground water has gained the attention in last decades. In other studies, NOR was 120 ng L^{-1} detected in surface water(Kolpin et al., 2002). OFL was 82 ng L^{-1}existing in sewage treatment plant effluent (Alexy et al., 2006) and 20 ng L^{-1} in surface water(Christian et al., 2003). SMZ was 370, 163, 410 ng L^{-1} in sewage treatment plant effluent, surface water and ground water, respectively(Alexy et al., 2006; Sacher et al., 2002).

The most remarkable effect of antibiotics on environment is the potential of adversely affecting organisms in both aquatic and terrestrial ecosystems that would upset the ecological balance and antibiotics also induce resistant strains in bacteria(Lanzky & Halting-Sørensen, 1997). However, the numbers of studies that are available on the effects on organisms are increasing but still too low(Fent et al., 2006). These studies examined the chronic effects of antibiotics on *Daphnia*, algae and bacteria (Halling-Sørensen, 2000; Kümmerer et al., 2000; Yamashita et al., 2006)and antibiotics such as diclofenac, ibuprofen, carbamazepine and the antidepressant fluoxetine. Moreover, when antibiotics are transferred within the food web, they may effect detrimentally on organisms.Oaks et al., studied the effect on the oriental white-backed vulture between 2000 and 2003, the results showed that high annual adult and sub-adult mortality (5-86%) and the declines in population (34-95%) were associated with renal failure and visceral gout(Oaks et al., 2004).

None of the technologies can remove all of the antibiotics and the efficiency dependson the type of compound (Kümmerer et al., 2000).Moreover, antibiotics have not been treated effectively by conventional wastewater treatment techniques,soadvanced treatment technologiesare recommended to solve this issue.The application of MBR technology in wastewater treatment becomes more and more popular all over the world due to its advantages compared with conventional activated sludge (CAS). MBR system with benefits of space saving, high MLSS concentration, high sludge retention time can operate flexibly (Visvanathan et al., 2000). In addition, MBR technology can remove Pharmaceutical Active Compounds (PhACs) existing in wastewater especially in hospital wastewater which cannot be eliminated by CAS(Sipma et al., 2010). According to(Li & Zhang, 2010), the antibiotics removal in MBR could be classified into four groups: (1) very easy biodegraded (95% of ibuprofen); (2) easy biodegraded (90% of bezafibrate); (3) moderately biodegraded (50-80 % of Naproxen, Ketoprofen, Gemfibrozil, Erythromycin; Trimethoprim and SMZ); (4) poorly biodegraded (40 % of Diclofenac and Carbamazepine).

Besides, ozone has been known as a strong oxidizer. Ozone reacts with organic compounds in two different ways: direct oxidation as molecular ozone and indirect reaction through the formation of secondary oxidants like hydroxyl radical OH•(Baig & Liechti, 2001). Molecular O_3 reacts selectively with amines, phenols and double bonds in aliphatic compounds, while hydroxyl free radical (OH•) is known as a strong oxidant to react non-selectively with compounds that cannot be oxidized by conventional oxidant (Carey, 1992). Ozone is capable attacking different therapeutic pharmaceuticals (Balcıoğlu & Ötker, 2003).(Huber et al., 2005) studied the ozonation of pharmaceuticals in municipal wastewater, the results showed that 10 of 11 pharmaceuticals were oxidized at ozone dose more than 2 mg L^{-1}. (Andreozzi et al., 2002) reported that at pH 5.5 and ozone dose 1 mg L^{-1}, Carbamazepine (CAR) was completely degraded after 4 mins and 30% mineralized after 60 mins, the effluent is not toxic to algae. However, (Hua et al., 2006) indicated that CAR was 66-100 % degraded after 20 mins at pH 7.5 and ozone dose 1.5-2 mg L^{-1}, the conventional treatment alone fails to remove pharmaceutical residues.(Andreozzi et al., 2005) also reported that at pH of 2 to 7 and ozone dose of 1.6 10^{-4} M, amoxicillin was 90% removal and 18% mineralized after 4 and 20 mins, respectively.

The aim of this study is to demonstrate the combination of MBR and ozonation in order to remove antibiotics in a hospital wastewater. In principle, SMZ, NOR, CIP and OFL were evaluated to introduce the potential of MBR integrated with ozonation process in terms of high antibiotic removal efficiency.

2. MATERIALS AND METHODS

2.1. Experimental setup

Membrane bioreactor (MBR) reactor was constructed by a glass bioreactor with volume of 8L and dimension of L×W×H = 0.28m × 0.08 m × 0.60 m. A hollow-fiber submerged membrane module (W×H = 200 mm × 310 mm) purchased from Mitsubishi, Japan with surface area of 0.1 m^2 and pore size of 0.4 μm was installed in the reactor. The cube sponges made from polyester-urethanewith porosity of 98% and dimension of 1 cm × 1 cm × 1 cm was added to reactor with the amount of 20% serving as the reactor volume.

Raw hospital wastewater was collected from Trung Vuong hospital, Ho Chi Minh city (HCMC), Vietnam and fed directly into the reactor using a feeding pump in order to control the feeding rate while the effluent flow rate was controlled by a suction pump. The special and innovative advantage of MBR system is automatic operation using timers, solenoid valves and digital pressure gauges. Air diffuser was installed in MBR not only for aeration to maintain dissolved oxygen value of 4 mg/L but also for air scouring to decrease membrane fouling. The MBRs' permeate pump was operated in a cyclic mode (8 min on/2 min off). For every new operation, the membrane was externally cleaned by chemicals (0.5% NaOCl) for 4 h. The digital pressure gauges were used to observed and recorded the trans-membrane pressure (TMP) daily.

The seed of activated sludge was collectedfrom another MBR system operating for long time in Universitywhich had similar characteristics of inlet. The mixed liquor suspended solids (MLSS) was approximately 5,000 mg/L. This is one of advantages of MBR system that MLSS concentration can obtain the highest value of 12,000 mg/L compared with very low MLSS concentration in conventional activated sludge. Other operational parameters such as Food/Micro-organism (F/M), organic loading rate (OLR), hydraulic retention time (HRT) and sludge retention time (SRT) were 0.19±0.05kg COD/kg MLSS.day, 0.64±0.16 kg COD/m^3.day, 10h and 20 days, respectively.

For ozonation application as next stage, the MBR permeate flowed to the glass reactor with volume of 2L and dimension of D× H = 8 cm × 42 cm. The flow rate of 20-40 mg O_3/h was supplied by FD-3000 II model ozone generator. Air diffuser was also installed at the bottom of the reactor.

2.2. Characteristics of influent

Hospital wastewater was collected in the equalization tank of the wastewater treatment plant of a hospital located in HCMC. Initially, wastewater characterictics such as pH, COD, TSS, NH_4^+-N, NO_3^-N, TKN and TP were 7.24, 320 mg/L, 75 mg/L, 25 mg/L, 0.075 mg/L, 32.3 mg/L and 5.2 mg/L, respectively. In terms of antibiotics matters, SMZ, NOR, CIP and OFL were determined as 1.439, 16.118, 7.083 and 23.476 (μg/L), respectively.

2.2. Analytical methods

In this research, in order to evaluate the wastewater treatment efficiency, parameters such as pH, COD, TSS, NH_4^+-N, NO_3^--N, TKN,TP were determined under standard methods(Apha, 1998). On the other hand, in terms of antibiotic removals, samples were pre-treated and analyzed in HCMU laboratory. Firstly, the sample pH was adjusted at 6.5 to 7.5 prior to filtration through 0.45 μm glass fiber filter to remove SS. Then, in solid phase extraction-SPE stage,the C18 HD cartridges (HySphere, Spark Holland, 2mm×10mm) were conditioned with 3mL of MeOH to activate cartridges and 3mL of distilled water to prevent cartridges from drying and then samples were loaded onto cartridges. In order to clean cartridges after loading, 2mL solvent mixture including 5% of MeOH and 95% of distilled water were loaded onto cartridges with 5 mins to wash and cartridges werecovered by biofilms after washing accomplishmented. Samples were eluted with 5 ml solvent of MeOH (1L) + phormic acid (1ml) to release residues. Additionally, samples were standardized by 1 ml of MeOH after being dried due to evaporation cause by nitrogen supply. At last, final samples had to be stored at 4° C prior to be examined by HPLC-MS/MS(Dinh et al., 2011).

3. RESULTS AND DISCUSSION

3.1. Elimination by MBR

The mechanisms of antibiotics removed by MBRmay include physical retention of membrane, biotransformation, air stripping, sorption and photo-transformation (Pomiès et al., 2013; Sipma et al., 2010; Verlicchi et al., 2012). However, since the micro-ultrafiltration membrane have the pore size between 100 and 1000 times bigger than the physical size of pharmaceutical compounds, no direct physical retention by membrane bioreactor can be expected (Larsen et al., 2004). On the other hand, sorption or biological degradation can be considered as reason of reducing those matter combing with solid retention in system. Most of antibiotics have a Henry coefficient (dimensionless, μg L^{-1} air/ μg L^{-1} wastewater) smaller than 10^{-5} (e.g., SMZ of 3.91 10^{-11}), consequently removal by air stripping is negligible(Li et al., 2015).

Theoretically, sorption of antibiotics to activated sludge of MBR depends on hydrophobicity and the presence of positively charged groups of a compound (Ternes et al., 2004). Since the sludge-water partition coefficient K_d of a compound is less than 500 L kg_{SS}^{-1}, indicating that it is not absorbed to activated sludge (Li et al., 2015). The biotransformation of antibiotics in MBR varies in the large range (from zero to complete) (Radjenović et al., 2009) and depends on the degradation constant K_{biol}. Antibiotics with K_{biol}< 0.1 L g_{SS}^{-1} d^{-1} are not removed, with K_{biol}> 10 L g_{SS}^{-1} d^{-1} are transformed higher than 90%, and with K_{biol} in between are moderate removal(Joss et al., 2006).

In this research, the results showed that the removal efficiencies were 39±6%, 90±4%, 83±13% and 81±13% for SMZ, NOR, CIP and OFL, respectively (Figure 1). Comparetively, the low removal efficiency of SMZ was caused by low K_d values lower than 500 L kg_{SS}^{-1} (260 L kg_{SS}^{-1}) so its sorption was not significant (Batt et al., 2007; Göbel et al., 2005).Also regarding to $K_d$values as previous studies, CIP and NOR have K_d values higher than 15,000 L kg_{SS}^{-1} so the major removal mechanisms was sorption to activated sludge (Golet et al., 2003; Li & Zhang, 2010).

Although they are lower than others ,these achivements are reasonable according to authors knowledge and cited references. In constrat, there is an article reported that SMZ and OLF could be removed about 73±11% and 93.5±2% by MBR, respectively(Radjenović et al., 2009) due to wide-range input antibiotic concentrationswhich might be the cause of this.

3.2. Elimination by MBR –ozonation

The MBR's permeate flowed into the ozone reactor, hydroxyl free radicals that generated by ozone oxidized antibiotics in wastewater to CO_2 and H_2O or simple organic compounds. The average antibiotics concentrations and removal efficiencies after ozonation were shown in Figure 2. The high removal efficiencies were 87±9.0%, 83±1.0% and 81±2.3% for NOR, CIP and OFL, respectively;however, SMZ was only treated 66±2.3% at pH 8.5 within 10 mins. These obtained results are not much different comparing to other researchs. For instance, Liu et al. (2014),reported that 99% of NOR and OFL were removed by coupling nanofiltration andozonation.Similarity, according to Vasconcelos et al., efficiency of ozonation process for simultaneous degradation was shown to reduce CIP concentration by 90% (Vasconcelos et al., 2009). The SMZ removal efficiency went against with Dantas et al. (2008), since they indicated that ozone hada ability to remove 95-99% of SMZ while only 66±2.3% could be removed in this study. After treating by MBR and ozonation, the antibiotics concentration of SMZ, NOR, CIP and OFL in the effluent were reduced to 1.137±0.138 μg L^{-1}, 2.116±0.312 μg L^{-1}, 2.132±0.288 μg L^{-1} and 1.826±0.250 μg L^{-1}, respectively. This reduces the adverse effects of antibiotics on the environment.

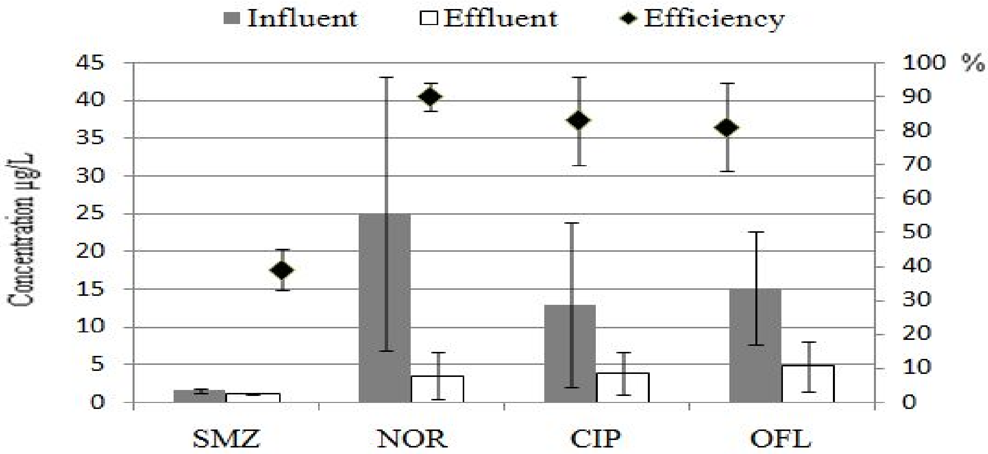

Figure 1. Demonstration of antibiotic removal in terms of concentration (μgL^{-1}) and efficiency (%) treated by membrane bioreactor process (MBR)

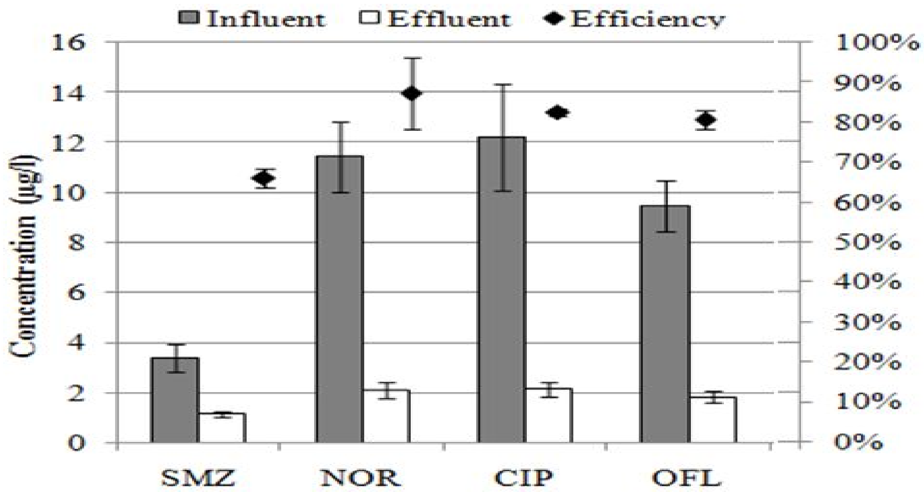

Figure 2. Demonstration of antibiotic removal in terms of concentration (μgL^{-1}) and efficiency (%) treated by ozonation after MBR process

4. CONCLUSIONS

The presence of antibiotics in wastewater has posed a remarkable problem for environment and caused serious risks on human health.Under those circumstances,the elimination of those antibiotic could be solved well by membrane bioreactor (MBR) combining with Ozonation process for NOR, CIP and OFL due to high efficiency over than 80%. Conversely, SMZ was not removed effectively in both processes. The reason might relate to its K_d values of 500 Lkg_{SS}^{-1} or adsorption on membrane material. However, there is still some lack of answers for the mechanism without determination of quantifying antibiotics sorbed into sludge phase and sponge cubes, distinguishing between sorption and biodegradation, indicating microorganism that favor removal of antibiotics, etc. For further concerns, the activated sludge within antibiotics must be strictly managed in order to avoid effects on terrestrial environment, but knowledge about the interaction of antibiotics with sludge is still too sparse.

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