Poll Res. 39 (3) : 525-530 (2020) Copyright © EM International ISSN 0257–8050

REMOVAL OF ORGANICS FROM HOSPITAL WASTEWATER BY MOVING BED BIOFILM REACTOR (MBBR) WITH POLYURETHANE FOAM MEDIA

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(Received 10 October, 2019; accepted 10 April, 2020)

ABSTRACT

Hospital wastewaters are generally treated using anaerobic-aerobic treatment with conventional activated sludge, which has proven inefficient to reduce the content of antibiotic and antiseptic drug compounds. MBBR application with polyurethane foam media is suitable for treating wastewater with high organic loading (high rate). MBBR-PU Foam can be used for pharmaceutical industry wastewater due to its high durability and ability to decompose chemicals. The general objective of this research is to make a prototype MBBR-PU Foam to eliminate organic substances, especially medicinal compounds (ibuprofen and trimethoprim) which are non biodegradable in hospital wastewater. The long-term goal is that the MBBR-PU Foam can be used in every hospital as a place-saving treatment unit, inexpensive because it saves electricity, is easy to maintain, does not cause odor, has no sludge, and is very efficient in reducing organic waste that is nonbiodegradable. This research was conducted in continuous system (prototype). The wastewater used was artificial wastewaterwith qualities such as the Central Sterile Supply Department CSSD wastewater and hospital laboratories. The MBBR prototype that will be used in this study is an MBBR system consisting of a series of 4 stage reactors, which are expected to provide clear information about the kinetics of removal of organic substances, especially medicinal compounds. The results showed that MBBR with PU-Foam media has significantly reduced the content of organic substances and drug compounds. In 4 hrs HRT the decrease in organic substances was above 67%, while the decrease in ibuprofen was 91% and thrimethropime by 57%.

KEY WORD : MBBR, Waste water, Hospital, Polyurethane foam

INTRODUCTION

Hospitals or health service facilities are social economic institutions that have the function and duty to provide services to the community. In addition to providing benefits to the community at the hospital, it also has a negative impact in the form of pollution due to waste disposal. Wastewater released is one of the potential sources of water pollution because it contains high levels of organic compounds, other harmful chemical compounds and pathogenic microorganisms which are harmful to health. Hospital wastewater is all liquid waste that comes from the entire process of hospital activities that includes liquid domestic waste (bathroom, kitchen, laundry), clinical liquid waste (wound washing water, blood wash), laboratory wastewater. Hospital wastewater that comes from domestic and clinical liquid waste discharges generally contains quite high organic compounds, whereas hospital wastewater from laboratories usually contains toxic compounds.

Hospital wastewater is generally treated using anaerobic-aerobic treatment with conventional activated sludge, which has proven inefficient to reduce the content of antibiotic and antiseptic drug compounds (Joss et al., 2005, 2006; Santos et al., 2009; Verlicchi et al., 2010). Hence some countries have completed their wastewater treatment with MBR (Membrane Bioreactor) (Brepols et al., 2008; Abegglen et al., 2009; Liu et al., 2010), and some advanced technology (post-treatment) such as the use of adsorber activated carbon, ultraviolet photolysis, AOP (advanced oxidation process), osmosis or nanofiltration, to eliminate drug compounds in wastewater (Falås et al., 2013). Another alternative is to use fluidized bioreactors which are effective enough to eliminate drugs in hospital wastewater (Cruz-Morató et al., 2014), and have better efficiency than conventional anaerobicaerobic sludge treatment systems. Biofilm systems such as MBBR (Moving Bed Biofilm Reactors) have also begun to be widely used to eliminate drug compounds directly in hospital wastewater.

The efficiency of MBBR depends on biofilms embedded in media that are immersed and mixed in the reactor. MBBR has been recognized as a compact and powerful reactor for treating wastewater, especially wastewater with a large nitrification process (Rusten et al., 1997; Casas et al., 2015; Tang et al., 2017; Ooi et al., 2018). MBBR technology has also been proven to reduce micro pollutants in wastewater (Karaolia et al., 2017; Stylianou et al., 2018), as shown in research conducted by Falås et al., (2012), MBBR is more effective in eliminating drug compounds that are acidic in the pharmaceutical industry's liquid waste compared to processing using activated sludge. Research Falås et al., (2013) and Escolà Casas et al., (2015) also showed that biofilm in the hybrid biofilm-activated sludge process can eliminate more ibuprofen and trimethoprim compounds than the activated sludge process. MBBR has a fairly good removal efficiency in treating X-ray media washing water, and found that organic compounds that are recalcitrant (very difficult to decompose) can be degraded by using MBBR (Casas et al., 2015; Abtahi et al., 2018; El-taliawy et al., 2018; Liu, Wang and Pang, 2018).

Polyurethane Foam media (PU-Foam), is a media / growth of microorganisms that are used in wastewater treatment using MBBR. PU-Foam has a higher surface area compared to the MBBR media commonly used today such as bioball, kaldness, kaldess, and biochip. Special PU-foam is used in MBBR applications to reduce industrial wastewater with high organic loading (high rate), due to its large surface area (around $15,000 - 20,000 \text{ m}^2/\text{m}^3$ of liquid waste) or 100 x wider than bioball, 40 x compared kaldness / kaldess, and 10x compared to biochips (Jain and Pradeep, 2005).

Moving Bed Biofim Reactors with Poly Urethane Foam media (MBBR-PU) can be used for wastewater from the pharmaceutical industry because of their high durability and ability to decompose chemicals (Moe and Irvine, 2000). Because of its toughness and efficiency in removing chemical compounds, MBBR-PU is considered a good choice for treating hospital wastewater.

The general objective of this study was to examine the overall MBBR-PU in removing organic substances, and drug compounds ibuprofen and trimethoprim in hospital wastewater. Decrease in organic matter is measured by decreasing the number of biochemical oxygen demand (BOD), chemical oxygen demand (COD), concentrations of ibuprofen and trimethoprim, ammonium (NH4 + - N), nitrite (NO₂-N), and nitrate (NO₃-N)

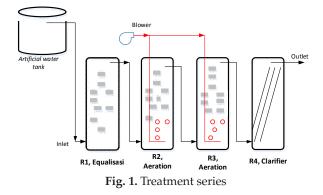
MATERIALS AND METHODS

This research was conducted with a continuing system in the laboratory, using artificial waste water. The series of moving bed biofilm reactor (MBBR) systems that will be used in this study consist of a series of 4 reactors, namely stabilization reactors, 2 (two) aeration reactors, clarifier reactors. The series is expected to provide clear information about removal of organic substances, especially medicinal compounds, including the process of nitrification and denitrification of hospital wastewater. The volume of each reactor is 30 L. The volume of polyurethane foam media in the aeration reactor is 5%. Hospital wastewater used is artificial water that is water that has been spiked with certain drug compounds at certain doses. Tests are carried out periodically according to a predetermined time, in order to obtain the removal performance of organic substances in each reactor, and the performance of the MBBR-PU system in reducing drug compounds.

The chemicals needed for artificial water according to the OECD Guidelines for Testing of Chemicals, Part 303B-Biofilms (Polesel *et al.*, 2017) are: CaCl₂.H₂O, NaCl, K₂HPO₄, MgSO4. 7H₂O, NaHCO₃, KMnO₄, NaOAc, peptone, meat extract, and sucrose. All of these chemicals are analytical grade. Drug compounds that were added to artificial water in this study were ibuprofen, and trimethoprim. Drug concentration varies from 20 and 100 mg/L. Placing the drug in artificial water requires a stirring process so that it is homogeneous. After affixing, artificial water is channeled to each reactor up to a volume of 30 L. The treatment at each reactor lasts for 48 hours. Taking samples at a predetermined sample point using a glass pipette of 10 mL, at 1 minute, 20 minutes, 1 hour, 2 hours, 3 hours, 4 hours, 8 hours, 10 hours, 12 hours, 24 hours, and 48 hours. Every replacement of the affixing of the drug compound, also carried out replacement of the PU-foam media. The concentration of ibuprofen, and trimethoprim (mg/ L) in the sample was analyzed using HPLC-MS / MS.

General parameters that are monitored regularly are COD, and NH_4^+-N ,. Biomass and morphological analysis of biofilm media analyzed after 48 hours of treatment. The biomass analysis is done gravimetically by first taking 10 pieces of media for each reactor and placing them on an aluminum foil cup, drying them at 105 °C overnight, weighing and recording as initial weight. Next wash the media using drops of 2M NaOH solution and rinse with de-ionised water. After the washing process, proceed again with the process of drying the media at 105 °C overnight, weighing and recording as weight after washing. The weight of biomass in the media is the difference in weight before and after washing. Furthermore, the amount of biomass per specific surface area of the media can be known.

Morphological analysis of biofilm media was examined using Scanning Electron Microscopy (SEM). The preparation procedure follows the method used by Abtahi *et al.*, (2018), which is to peel the biofilm which carefully covers the media, and cut it into small pieces. Each piece was immersed in 2 mL of 4% glutaraldehyde, 1 mL of phosphate buffer (pH 7.4) and 1 mL of demineralized water for 20 minutes. Then wash 2 (two) times with 1 mL



phosphate buffer, 2 mL sucrose 0.4 M and 1 mL demineralized water for 15 minutes. The washed biofilm sample is then dried by soaking in a row in 2 mL aceton-water solution (50%: 50%) for 5 minutes, 2 mL aceton-water solution (70%: 30%) for 5 minutes, and 2 mL of acetone-hexamethyldisilazane (HMDS) solution (50%: 50%) for 5 minutes. Then the sample is dried overnight with the HMDS evaporation.

RESULTS AND DISCUSSION

Seeding Microbe on PU-Foam media

MBBR systemwas operated for 48 hours continuously. Before it was used to treat wastewater, seeding microbe was carried out on PU-Foam media. The seeding process was carried out for 7 days, using known aerobic microbial consortiums namely Nitrosomonas sp, Nitrosobacter sp, Aerobacter sp, at a dose of 10-15 mL per mg/L COD inlet. Microbe seeding process is carried out in aerobic process. The seeding procedure was as follows, fill 20 L tub with clean water, and aerate for 24 hours so that the concentration of dissolved oxygen is more than 5 mg/L. Furthermore, into the aeration tub spiked with a nutrient solution, which is a solution with a concentration ratio of C, N and P of 100: 5: 1. The aeration process is carried out continuously during the seeding process (4 days). After 4 days, water in the aeration tub is discharged by 20% per day and replaced with artificial wastewater. This process was carried out until all clean water has been replaced with artificial wastewater, and the process is continued for 2 days.Measurement of biofilm dry weight in PU-Foam media after the seeding process was completed was 1.827 g/L.

Removal organic matter in MBBR continuous system

The concentration of organic matter in this study was measured as a COD parameter. At the beginning of the aeration process, COD artificial water was 798 mg/L, at HRT 4 hours the concentration of organic matter dropped to 100.67 mg/L or 87.39%, and at the end of the experiment (HRT 48 hours) the concentration of organic matter became 2, 3 mg/L. Concentrations of NH_4^+ -N, at the beginning of the aeration process were 1000 mg /L, in HRT 4 hours the concentration of NH_4^+ -N drops to 500 mg/L. At the end of the trial (HRT 48 hours)

the concentrations of NH_4^+ -N, dropped to 100 mg/L. The dry weight of biofilm was 0.343 g / L or during 4 hours, and at the end of the experiment (HRT 48 hours), the dry weight of the biofilm was 4.12 g/L.

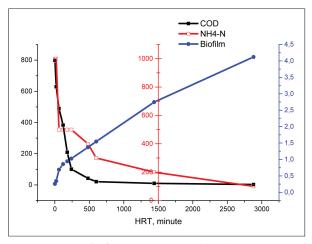


Fig. 2. Removal of organic matter (COD), NH4-N and biofilm formation in 20 mg/L drugs

The results of the Scanning Electron Microscopy (SEM) analysis of PU-Foam biofilm media after 48 hours of aeration were presented in Figure 3 and Figure 5. The SEM analysis shows that the biofim in PU-Foam media looks thick.

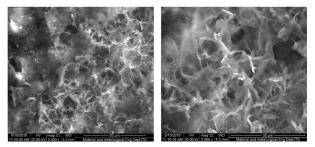


Fig. 3. Scanning Electron Microscopy (SEM) biofilm PU-Foam media after 48 hours aeration at a drug dose of 20 mg / L

In Figure 4. At the beginning of the aeration process, COD artificial water was 797.24 mg/L, at HRT 4 hours the concentration of organic matter dropped to 259.68 mg/L or 67.42%, and at the end of the experiment (HRT 48 hours) the concentration of organic matter became 11.82 mg/L. Concentrations of NH_4^+ -N, at the beginning of the aeration process were 1000 mg/L, after 4 hours HRT the concentration dropped to 500 mg/ L, and at the end of the trial (HRT 48 hours) dropped to 200 mg/L. The dry weight of biofilms was 0.661 g/L or

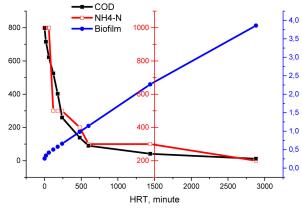


Fig. 4. Removal of organic matter (COD), NH₄-N and biofilm formation in 100 mg/L drugs

during 4 hours, and at the end of the experiment (HRT 48 hours), the dry weight of the biofilm was 3.86 g / L.

The MBBR system with PU-Foam media has the ability to eliminate most of the analyzed pharmaceutical substances (Ibuprofen and Trimethoprim). To get an ideal and simplified data evaluation, all the substances of the analyzed drugs can be mounted on the first level degradation kinetics.

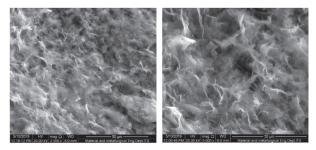


Fig. 5. Scanning Electron Microscopy (SEM) biofilm PU-Foam media after 48 hours aeration at a drug dose of 100 mg / L

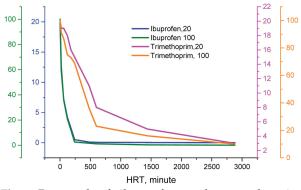


Fig. 6. Removal of ibuprofen and tromethoprim compounds, at doses of 20 mg/L and 100 mg/L

 $C = (Co) e^{(-kt)}$

where k (L/hour.g) was the rate of degradation constant. First-order kinetics are used so that a more valid comparison of the literature can be obtained, because most of the data in the literature is supplemented using first-order degradation kinetics. The kinetic constant values of ibuprofen and trimethoprim degradation were 6.5×10^{-1} and 1.35×10^{-2} , respectively. Trimethoprim was a drug compound that was more difficult to integrate than ibuprofen, this was in accordance with research conducted by Gordon *et al.* (2018).

CONCLUSION

MBBR system with PU-Foam media has been proven to reduce COD compounds by 60.5-77%, NH_4^+-N compounds by 88-90%, ibuprofen compounds by 91% and thrimethoprim by 57%. Although MBBR technology seems promising, further studies are still needed on the ability of MBBR to reduce heavy metal compounds in liquid waste. Further investigation is needed to straighten the degradation path.

REFERENCES

- Abegglen, C., Joss, A., McArdell, C. S., Fink, G., Schlüsener, M. P., Ternes, T. A. and Siegrist, H. 2009. The fate of selected micropollutants in a single-house MBR. *Water Research*. Pergamon. 43 (7) : 2036-2046. doi: 10.1016/ J.WATRES.2009.02.005.
- Abtahi, S. M., Petermann, M., Juppeau Flambard, A., Beaufort, S., Terrisse, F., Trotouin, T., Joannis Cassan, C. and Albasi, C. 2018. Micropollutants removal in tertiary moving bed biofilm reactors (MBBRs): Contribution of the biofilm and suspended biomass. *Science of the Total Environment*. Elsevier B.V., 643, pp. 1464-1480. doi: 10.1016/ j.scitotenv.2018.06.303.
- Brepols, C., Dorgeloh, E., Frechen, F.-B., Fuchs, W., Haider, S., Joss, A., de Korte, K., Ruiken, C., Schier, W., van der Roest, H., Wett, M. and Wozniak, T. 2008. Upgrading and retrofitting of municipal wastewater treatment plants by means of membrane bioreactor (MBR) technology. *Desalination. Elsevier.* 231(1-3) : 20-26. doi: 10.1016/J.DESAL.2007.11.035.
- Casas, M. E., Chhetri, R. K., Ooi, G., Hansen, K. M. S., Litty, K., Christensson, M., Kragelund, C., Andersen, H. R. and Bester, K. 2015. Biodegradation of pharmaceuticals in hospital wastewater by staged Moving Bed Biofilm Reactors

(MBBR). *Water Research*. Elsevier Ltd. 83 : 293-302. doi: 10.1016/j.watres.2015.06.042.

- Cruz-Morató, C., Lucas, D., Llorca, M., Rodriguez-Mozaz, S., Gorga, M., Petrovic, M., Barceló, D., Vicent, T., Sarrà, M. and Marco-Urrea, E. 2014. Hospital wastewater treatment by fungal bioreactor: Removal efficiency for pharmaceuticals and endocrine disruptor compounds. *Science of The Total Environment*. Elsevier, 493 : 365-376. doi: 10.1016/J.SCITOTENV.2014.05.117.
- El-taliawy, H., Casas, M. E. and Bester, K. 2018. Removal of ozonation products of pharmaceuticals in laboratory Moving Bed Biofilm Reactors (MBBRs). *Journal of Hazardous Materials,* Elsevier. 347 : 288-298. doi: 10.1016/J.JHAZMAT.2018.01.002.
- Escolà Casas, M., Chhetri, R. K., Ooi, G., Hansen, K. M. S., Litty, K., Christensson, M., Kragelund, C., Andersen, H. R. and Bester, K. 2015. Biodegradation of pharmaceuticals in hospital wastewater by a hybrid biofilm and activated sludge system (Hybas)', *Science of the Total Environment*. Elsevier B.V., 530-531, pp. 383-392. doi: 10.1016/ j.scitotenv.2015.05.099.
- Falås, P., Baillon-Dhumez, A., Andersen, H. R., Ledin, A. and la Cour Jansen, J. 2012. Suspended biofilm carrier and activated sludge removal of acidic pharmaceuticals. *Water Research. Pergamon.* 46 (4) : 1167-1175. doi: 10.1016/J.WATRES.2011.12.003.
- Falås, P., Longrée, P., la Cour Jansen, J., Siegrist, H., Hollender, J. and Joss, A. 2013. Micropollutant removal by attached and suspended growth in a hybrid biofilm-activated sludge process. *Water Research*. Pergamon, 47 (13) : 4498-4506. doi: 10.1016/J.WATRES.2013.05.010.
- Hari, S. K. M. and Hartaja, D. R. K. 2017. Desain Instalasi Pengolahan Air Limbah Rumah Design of Hospital Waste Water Treatment Instalation Capacity. 40 M 3 / Day. 10 (2) : 99-113.
- Jain, P. and Pradeep, T. 2005. Potential of silver nanoparticle-coated polyurethane foam as an antibacterial water filter. *Biotechnology and Bioengineering*. 90 (1) : 59-63. doi: 10.1002/ bit.20368.
- Jess Krarup, Jakob Søholm, Ole Grønborg, S. Z. 2015. New Standard For Hospital Wastewater Treatment, Filtration + Separation. doi: 10.1016/S0015-1882(15)30141-5.
- Joss, A., Keller, E., Alder, A. C., Göbel, A., McArdell, C. S., Ternes, T. and Siegrist, H. (2005) Removal of pharmaceuticals and fragrances in biological wastewater treatment. *Water Research*. Pergamon, 39(14): 3139-3152. doi: 10.1016/J.WATRES. 2005.05.031.
- Joss, A., Zabczynski, S., Göbel, A., Hoffmann, B., Löffler, D., McArdell, C. S., Ternes, T. A., Thomsen, A. and Siegrist, H. 2006. Biological degradation of

pharmaceuticals in municipal wastewater treatment: Proposing a classification scheme. *Water Research*. Pergamon. 40 (8) : 1686-1696. doi: 10.1016/J.WATRES.2006.02.014.

- Karaolia, P., Michael-Kordatou, I., Hapeshi, E., Alexander, J., Schwartz, T. and Fatta-Kassinos, D. 2017.
 'Investigation of the potential of a Membrane BioReactor followed by solar Fenton oxidation to remove antibiotic-related microcontaminants. *Chemical Engineering Journal*. Elsevier. 310 : 491-502. doi: 10.1016/J.CEJ.2016.04.113.
- Liu, Q., Zhou, Y., Chen, L. and Zheng, X. 2010. Application of MBR for hospital wastewater treatment in China', *Desalination*. Elsevier. 250 (2) : 605-608. doi: 10.1016/J.DESAL.2009.09.033.
- Liu, X., Wang, L. and Pang, L. 2018. Application of a novel strain Corynebacterium pollutisoli SPH6 to improve nitrogen removal in an anaerobic/aerobic-moving bed biofilm reactor (A/O-MBBR)', *Bioresource Technology*. Elsevier, 269, pp. 113-120. doi: 10.1016/J.BIORTECH.2018.08.076.
- Moe, W.M. and Irvine, R. L. 2000. Polyurethane foam medium for biofiltration. I: characterization. *Journal Environmental Eng.* 126 (September) : 815-825.
- Ooi, G. T. H., Tang, K., Chhetri, R. K., Kaarsholm, K. M. S., Sundmark, K., Kragelund, C., Litty, K., Christensen, A., Lindholst, S., Sund, C., Christensson, M., Bester, K. and Andersen, H. R. 2018. Biological removal of pharmaceuticals from hospital wastewater in a pilot-scale staged moving bed biofilm reactor (MBBR) utilising nitrifying and denitrifying processes. *Bioresource Technology*. Elsevier Ltd. 267 : 677-687. doi: 10.1016/ j.biortech.2018.07.077.
- Polesel, F., Torresi, E., Loreggian, L., Casas, M. E., Christensson, M., Bester, K. and Plósz, B. G. 2017. Removal of pharmaceuticals in pre-denitrifying MBBR - Influence of organic substrate availability in single- and three-stage configurations. *Water*

Research. Pergamon. 123 : 408-419. doi: 10.1016/ J. WATRES. 2017.06.068.

- Rusten, B., Kolkinn, O. and Odegaard, H. 1997. Moving bed biofilm reactors and chemical precipitation for high efficiency treatment of wastewater from small communities. *Water Science and Technology*. No longer published by Elsevier, 35(6) : 71-79. doi: 10.1016/S0273-1223(97)00097-8.
- Santos, J. L., Aparicio, I., Callejón, M. and Alonso, E. 2009. Occurrence of pharmaceutically active compounds during 1-year period in wastewaters from four wastewater treatment plants in Seville (Spain). *Journal of Hazardous Materials*. Elsevier. 164 (2-3) : 1509-1516. doi: 10.1016/ J.JHAZMAT.2008.09.073.
- von Sperling, M. 2007. *Basic Principles of Wastewater Treatment, Biological Wastewater Treatment.* doi: 10.5860/CHOICE.45-2632.
- Stylianou, K., Hapeshi, E., Vasquez, M. I., Fatta-Kassinos, D. and Vyrides, I. 2018. Diclofenac biodegradation by newly isolated *Klebsiella* sp. KSC: Microbial intermediates and ecotoxicological assessment. *Journal of Environmental Chemical Engineering*. Elsevier, 6(2) : 3242-3248. doi: 10.1016/ J.JECE.2018.04.052.
- Tang, K., Ooi, G. T. H., Litty, K., Sundmark, K., Kaarsholm, K. M. S., Sund, C., Kragelund, C., Christensson, M., Bester, K. and Andersen, H. R. 2017. Removal of pharmaceuticals in conventionally treated wastewater by a polishing moving bed biofilm reactor (MBBR) with intermittent feeding. *Bioresource Technology*. Elsevier Ltd, 236 : 77-86. doi: 10.1016/j.biortech.2017.03.159.

Veolia (2013) An overview of the technology.

Verlicchi, P., Galletti, A., Petrovic, M. and Barceló, D. 2010. Hospital effluents as a source of emerging pollutants: An overview of micropollutants and sustainable treatment options. *Journal of Hydrology*. Elsevier, 389 (3-4) : 416-428. doi: 10.1016/ J.JHYDROL.2010.06.005.