Sustainable Bioelectrical Approaches for the Treatment of Endocrine Contaminants in Wastewater

Tanaka H.R.1 & Volkov A.M.2
***1 Department of Neurology, Tokyo Medical University, Tokyo, Japan**
2 Department of Clinical Medicine, Moscow Health Academy, Moscow, Russia

ABSTRACT

EDCs or Endocrine disrupting compounds are a class of emerging micropollutants that are known to disturb the endocrine system of animals and man. Increase in the level of these micropollutants in water; wastewater etc. has raised the concern for research on the causes and effects of these EDCs and suitable technologies for their degradation. Wastewater treatment plants (WWTPs), employing only conventional processes for treatment, are considered as one of the primary source of various micropollutants. The removal efficiency of WWTPs varies according to the physiochemical properties of the pollutants and the treatment processes involved for them. The present paper reviews the anaerobic methods for the EDC treatment with particular emphasis on Microbial fuel cell technology that generates electricity by utilizing the wastewater components.

Keywords: *Biodegradation; EDC; Bioelectrical System; MFC; Micropollutants; Wastewater treatment.*

I. INTRODUCTION

Micropollutants or emerging contaminants are a group of organic substances which have a negative impact on the aquatic environment, wildlife and humans due to their toxic, persistent and bioaccumulative nature (Yashas and Murthy, 2017). The EDCs comprise pharmaceuticals, personal care products, surfactants, various industrial additives and multiple chemicals intended to be an endocrine disrupter that has become a threat to our water supply network (Bolong *et al.*, 2009). Effects of EDCs on the human health include increased risk of breast, testicular, and prostate cancers, reproductive disorders such as impaired fertility, irregularities of the menstrual cycle, and infertility(C and Klemenc, 2011). Immune and hormonal disorders, obesity, fewer male offspring, diabetes, metabolic disorders, and cardiovascular disease. Therefore, the detection of endocrine disrupters is an essential area of research for environmental monitoring and control (Yashas and Murthy, 2017). The contamination of estrogenic compounds also causes an adverse impact on the aquatic environment, which creates an imbalance in the ecosystem (Zaharin *et al.*, 2014). According to,(Vidal and Diez, 2005) the proportion of EDC degradation by techniques such as primary settling, aerating volatilization, sludge absorption, and chemical precipitation is relatively small as compared to biodegradation. The biodegradation by anaerobic treatment is strongly dependent on the properties of the wastewater (Liu *et al.*, 2008). This study aims to review the literature for the occurrence, environmental and health implications and techniques for the degradation of the EDC in the ecological facility with a significant focus on energy generating wastewater treatment methods (Yashas and Murthy, 2017).

The occurrence and fate of micropollutants in the environment have been recognized since long as issues of public health and environmental concern. A wide variety of organic micropollutants have been detected and identified in sewage and effluent-impacted water bodies, including surface waters and groundwater (Al-rifai *et al*., 2011). Among EDCs, estrogen is the main contributor of estrogenic activity, and their pathway to the environment is mainly through wastewater effluent from municipal treatment plants, hospital effluent and from livestock activities (Ying *et al.*, 2002).The synthetic estrogen 17a-ethinylestradiol (EE2), the main ingredient of commonly used oral contraceptive pills, has been detected in wastewaters and surface waters at ng/L levels (Comninellis *et al.*, 2008). The current wastewater treatment system is not effective in elimination of these emerging contaminants as these have not been monitored due to the absence of stringent regulation specific to these contaminants (Bolong *et al.*, 2009).

II. ADVANCES IN ENERGY-PRODUCING ANAEROBIC BIOTECHNOLOGIES FOR WASTEWATER TREATMENT

Anaerobic processes when compared with the activated sludge process avoids the consumption of energy for aeration and instead are responsible for the production of electricity whereas activated sludge process have the limitation of being energy intensive (Lier, 2008). Moreover in anaerobic processes, the nutrients in wastewater can be preserved for further reuse or recovery (Loosdrecht and Brdjanovic, 2014) thereby further elevating energy and economic benefits (Li and Yu, 2016). The anaerobic biotechnologies for treatment of wastewater can be divided into following-

Anaerobic membrane bioreactors (AnMBR)
An AnMBR is a firmly packed bioreactor that plays a dual role in the removal of contaminant
and separation of sludge (Pretel *et al.*, 2015). Its excellent capacity for retention of sludge and particulate organic matter gives it a much higher effective treatment than other anaerobic bioreactors (Smith *et al.*, 2013).

Bioelectrochemical systems (BES)
Bioelectrochemical systems (BES) are developing technologies that convert organic materials into usable forms of energy, such as electricity or hydrogen gas. The formation of electroactive biofilms is the result of the mutualistic interaction between microorganisms which acts as a critical element to longevity and success in bioelectrochemical systems (Islam *et al.*, 2017). BES technologies applied in the given configurations of microbial fuel cells (MFCs) and microbial electrolysis cells (MECs) as shown in Fig.1. In both systems, a consortium has grown in an anaerobic anode chamber where the incoming organic material is oxidized during respiration resulting in the production of electrons and protons. That gets transported to the cathode where they combine to form either water, in aerobic systems, or hydrogen gas, in anaerobic systems (Beegle, 2017).

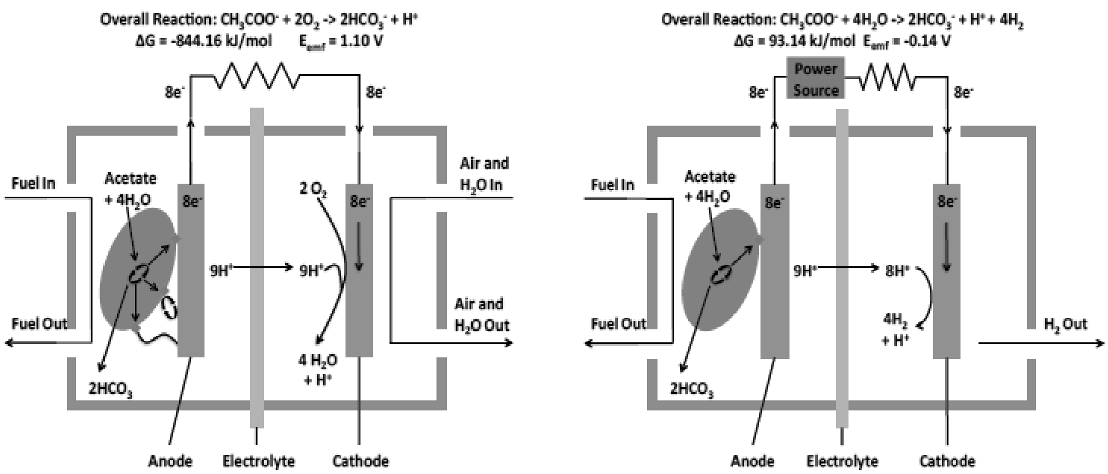

Figure. 1: Schematic diagram of Microbial Fuel Cell (MFC) (Left) and Microbial Electrolysis Cell (MEC) (Right)

III. ENERGY GENERATING ANAEROBIC TECHNOLOGIES FOR TREATMENT OF WASTEWATERS FROM DIFFERENT INDUSTRIES

Wastewater consists of energy-rich sources that are salutary for the growth of various anaerobic and facultative bacterial species. These bacteria have the potential to transfer electrons to an anode which acts as a terminal electron acceptor and hence are classified as electrogenic bacteria (Blackall *et al.*, 2007;Rabaey *et al.*, 2004).
Bacterial composition and performance of anodic biofilms in MFC by using dairy wastewater was investigated. Anaerobic mixed-sludge was used as an initial source of electrogenic bacteria. *Commamonas*, *Bacillus*, and *Sphingobacterium*were the genus used for the culturing of anodic biofilm. The highest power density, volumetric power, and COD removal efficiency of MFC operation were of $131mWm^{-2}$, $2.4Wm^{-3}$, and 76%, respectively (Retnaningrum and Wilopo, 2017). Double-chambered MFCs were employed for analyzing the electrogenic activity of microbial consortium. The MFCs was operated with microalgalbiocathode, the anode chamber being inoculated with anaerobic microbial consortiums and the cathode chamber with cultures of the microalga, *C. vulgaris*. By operating the MFCs for 32 days an increase in power density (from $23.17mW/m^2$ to $327.67\ mW/m^2$) and in the potential (from 200 to 954 mV) was observed(Huarachi-olivera *et al.*, 2018). Microbial fuel cells were operated with different anode and cathode compositions and salt bridges in a study carried out by (Naureen *et al.*, 2016**)** using wastewater samples from University of NizwaWWTP. After 7 and 21 days of incubation, an increase in the Open Cathode Voltage (OCV) was observed for all bacterial isolates indicating a high flow of electrons through salt bridge with increase in time. Isolate *Amantichitinumursilacus* (Z2) generated an OCV of 645 mV. This entails that the bacterial isolate *Amantichitinumursilacus* (Z2) might have electrogenic properties due to C-type cytochromes or

conductive nano-wires (Gorby *et al.*, 2006) on its cell membrane (Environ *et al.*, 2011). In a study aimed at examining the electrogenic property of *B. cereus* and its ability to inhibit the methanogenesis in MFC (Islam *et al.*, 2017),a strong redox peak in the Cyclic Voltammetry (CV) of MFC with *B. cereus* was obtained portraying that it contains electrogenic properties. Further, the incorporation of *B. cereus* in AS showed the rise in power generation (4.83 W/m^3) and Coulombic Efficiency (22%) of MFC compared to the MFC solely inoculated with AS (1.82 W/m^3, 12%).

Anaerobic technologies for degradation of EDCs

To investigate the removal and fate of estrogens such as 17β-estradiol (E2) and 17α-ethinylestradiol (EE2), a laboratory-scale anaerobic anoxic-oxic (AAO) activated sludge system was established by (Miran *et al.*, 2015).Microbial community structure in a dual chamber MFC was fed with brewery waste for the degradation of azo dye and generation of electricity. A stable voltage generated (0.41V and 0.38V) with a maximum power density of 305 and 269mWm-^{2} for brewery waste alone (2000 mg L-^{1}) and after the azo dye (200 mg L-^{1}) addition, respectively. Microbial community, *Desulfovibrio* was supposed to play a significant role in electron transfer to the anode since its outer membrane contains c-type cytochromes (Kang *et al*., 2006). Several studies employing the AAO process for removal of EDCs are reported (Li *et al.*, 2011**)**; (Publishing and Science, 2011).

For analyzing the role of disruptive endocrine estrogens in electron transfer, compounds like Estriol (E3) and 17α-ethynylestradiol (EE2) used as model estrogen compounds in the study carried out by (Kumar *et al.*, 2012). Anaerobic consortia from operating full-scale UASB reactor treating wastewater was used as a biocatalyst.Both the EDC showed removal under electrogenesis. However, E3 showed higher removal efficiency than that of EE2. A decrease in the removal of E3 observed with increase in the concentrations from C1 to C3 supporting the toxic effects of estrogens on the biocatalyst at higher concentrations. E3C1 (Conc. 500ug/L) showed higher removal (50.28%) followed by E3C2 (Conc. 1000ug/L) (41.04%) and E3C3 (Conc. 2000ug/L) (16.25%). Maximum power density was found with EE2C2 (40.02 mW/m^2) at followed by E3C2 (34.56 mW/m^2), E3C2-PP (31.84 mW/m^2), control (24.32 mW/m^2), E3C1 (22.26 mW/m^2) and E3C3 (20.19 mW/m^2). (Yoshimoto *et al.*, 2004)studied the degradation of estrogens by isolates taken from activated sludge of wastewater treatment plants. Four strains of *Rhodococcus*,*R. zopfii*Y 50158 and *R. equi*Y 50155, Y 50156, and Y 50157were capable of degrading estrogens E2, E1, E3, and EE2. E2 and E1 were degraded completely in 24 h, and E3 and EE2 degraded by about 80% of their concentrations in 24 h. The effect of the Fe(III) an oxidant and facultative anaerobic strain of iron-reducing bacteria on the anaerobic degradation of estrogens in reject water was investigated by (Ivanov *et al.*, 2010). The percentage removal in the natural estrogens such as 17β-estradiol, estriol, and estrone was found to be 92%, 60%, and 27%, respectively. White rot fungi (WRF) and their lignin-modifying enzymes (LMEs), i.e., laccase and lignin and manganese-dependent peroxidase possess the ability to treat EDCs. Numerous authors reported a high efficiency of LF and LMEs in biodegradation or transformation of EE2 (Cajthaml, 2015; Cabana *et.al,* 2007). Several bacterias, are also found to be effective in the biodegradation of estrogenic compounds at STPs and ETPs (Roh and Chu, 2010); (Yoshimoto *et al.*, 2004); (Yu *et al.*, 2011).

IV. RESULT AND DISCUSSION

In an endeavour to reduce global dependence on fossil fuels and to minimise damage caused to the environment with the release of carbon dioxide, the world faces the challenge to develop alternative sustainable sources of energy to meet its escalating energy demands.This paper highlilghted the possibility of using MFC as sustainable source of energy. MFC is a wonderful technology in converting the chemical energy inside the wide varieties of the waste organic matter with the help of the microorganism into bio-electricity. Despite the fact that the current MFC technology is capable of producing very low power output but it is suitable for small telemetry and wireless sensor system with a small power requirement in the remote areas. Moreover, removal of EDC’s is extremely important to reduce the potential risk caused by them in treated wastewaters. Thus bioelectrical system could be suitable alternatives to current expensive physico-chemical technologies for removal of these micropollutants.

V. CONCLUSION

The goal of achieving energy self-sufficiency in wastewater treatment has spurred tremendous research efforts to develop more efficient energy-producing anaerobic biotechnologies. Microbial fuel cells are evolving to become a simple, robust technology. The potential electrogenic bacteria could be readily isolated from wastewater sediments and employed in MFCs for an eco-friendly and economically feasible method of electricity generation.Also, they could be utilized for the degradation of several Endocrine disrupting compounds.

VI. ACKNOWLEDGEMENTS

This work was supported by Science and Engineering Board (SERB), providing with financial assistance and appointing first author as a Research Associate. Also the authors wants to thank management of Manipal University Jaipur for providing us with laboratory facilities.

REFERENCES

1. *Al-rifai J. H., Khabbaz H. and Schäfer A. I. 2011 Removal of pharmaceuticals and endocrine disrupting compounds in a water recycling process using reverse osmosis systems. Separation and Purification Technology. Elsevier B.V., 77(1), pp. 60–67.doi: 10.1016/j.seppur.2010.11.020.*
2. *Beegle J. R. 2017 A Bug's Life : Integration of Anaerobic Digestion and Bioelectrochemical Systems for Enhanced Energy Recovery from Wastewater Solids and Other Waste Substrates. Master's Thesis, University of Tennessee.http://trace.tennessee.edu/utk_gradthes/4724*
3. *Blackall L. L., Keller J., Gross P., Rabaey K., Rodrı J., Batstone D., Verstraete W. and Nealson K. H. 2007 Microbial ecology meets electrochemistry : electricity-driven and driving communities, pp. 9–18. doi: 10.1038/ismej.2007.4.*
4. *Bolong N., Ismail A. F., Salim M. R. and Matsuura T. 2009 A review of the effects of emerging contaminants in wastewater and options for their removal, DES. Elsevier B.V., 239(1–3), pp. 229–246. doi: 10.1016/j.desal.2008.03.020.*
5. *C M. R. and Klemenc A. K. 2011 Negative impact of endocrine-disrupting compounds on human reproductive health, pp. 403–416. doi: 10.1071/RD09300*
6. *Cabana H., Jones J. P. and Agathos S. N. 2007 Elimination of Endocrine Disrupting Chemicals using White Rot Fungi and their Lignin Modifying Enzymes : A Review, (5), pp. 429–456. doi: 10.1002/elsc.200700017.*
7. *Cajthaml T. 2015 Minireview Biodegradation of endocrine-disrupting compounds by ligninolytic fungi : mechanisms involved in the degradation,17, pp. 4822–4834. doi: 10.1111/1462-2920.12460.*
8. *Comninellis C., Kapalka A., Malato S., Parsons S. A., Poulios I. and Mantzavinos D. 2008 Advanced oxidation processes for water treatment : advances and trends for R & D, 776(February), pp. 769–776. doi: 10.1002/jctb.*
9. *Environ E., Strycharz-glaven S. M., Snider R. M., Guiseppi-elie A. and Tender L. M. 2011 Environmental Science On the electrical conductivity of microbial nanowires and biofilms, pp. 4366–4379. doi: 10.1039/c1ee01753e.*
10. *Gorby Y. A., Yanina S., Mclean J. S., Rosso K. M., Moyle, D., Dohnalkova A., Beveridge T. J., Chang I. S., Kim B. H., Kim K. S., Culley D. E., Reed S. B., Romine M. F., Saffarini D. A., Hill E. A., Shi L., Elias D. A., Kennedy D. W., Pinchuk G., Watanabe K., Ishii S., Logan B., Nealson K. H. and Fredrickson J. K. 2006 Electrically conductive bacterial nanowires produced by Shewanella oneidensis strain MR-1 and other microorganisms, 103(30).* https://doi.org/10.1073/pnas.0604517103
11. *Huarachi-olivera R., Dueñas-gonza A., Yapo-pari U., Vega P., Romero-ugarte M., Tapia J., Molina L., Lazarte-rivera A., Pacheco-salazar D. G. and Esparza M. 2018 Bioelectrogenesis with microbial fuel cells (MFCs) using the microalga Chlorella vulgaris and bacterial communities, Electronic Journal of Biotechnology. Elsevier España, S.L.U., 31, pp. 34–43. doi: 10.1016/j.ejbt.2017.10.013.*
12. *Islam M. A., Ethiraj B., Cheng C. K., Yousuf A. and Khan M. R. 2017 Electrogenic and anti-methanogenic properties of Bacillus cereus for enhanced power generation in anaerobic sludge driven microbial fuel cell.***doi:** *10.1021/acs.energyfuels.7b00434*
13. *Ivanov V., Lim J. J., Stabnikova O. and Gin K. Y. 2010 Biodegradation of estrogens by facultative anaerobic iron-reducing bacteria,45,pp. 284–287. doi: 10.1016/j.procbio.2009.09.017.*
14. *Kang J., Katayama Y. and Kondo F. 2006 Biodegradation or metabolism of bisphenol A : From microorganisms to mammals, 217, pp. 81–90. doi: 10.1016/j.tox.2005.10.001.*
15. *Kumar A. K., Reddy M. V., Chandrasekhar K., Srikanth S. and Mohan S. V. 2012 Bioresource Technology Endocrine disruptive estrogens role in electron transfer : Bio-electrochemical remediation with microbial mediated electrogenesis, Bioresource Technology. Elsevier Ltd, 104, pp. 547–556. doi: 10.1016/j.biortech.2011.10.037.*
16. *Li W. and Yu H. 2016 Advances in Energy-Producing Anaerobic Biotechnologies for Municipal*

Wastewater Treatment, Engineering. Elsevier LTD on behalf of Chinese Academy of Engineering and Higher Education Press Limited Company, 2(4), pp. 438–446. doi: 10.1016/J.ENG.2016.04.017.

17. *Li Y. M., Zeng Q. L. and Yang S. J. 2011 Removal and fate of estrogens in an anaerobic-anoxic-oxic activated sludge system, pp. 51–56. doi: 10.2166/wst.2011.008.*
18. *Lier J. B. Van 2008 High-rate anaerobic wastewater treatment : diversifying from end-of-the-pipe treatment to resource-oriented conversion techniques, pp. 1137–1149. doi: 10.2166/wst.2008.040.*
19. *Liu Z., Kanjo Y. and Mizutani S. 2008 Removal mechanisms for endocrine disrupting compounds (EDCs) in wastewater treatment — physical means , biodegradation , and chemical advanced oxidation : A review, Science of the Total Environment, The. Elsevier B.V., 407(2), pp. 731–748. doi: 10.1016/j.scitotenv.2008.08.039.*
20. *Loosdrecht B. M. C. M. Van and Brdjanovic D. 2014 Anticipating the next century of wastewater treatment, 344(6191), pp. 2009–2011.doi: 10.1126/science.1255183*
21. *Miran W., Nawaz M., Kadam A., Shin S. and Heo J. 2015 Microbial community structure in a dual chamber microbial fuel cell fed with brewery waste for azo dye degradation and electricity generation. doi: 10.1007/s11356-015-4582-8.*
22. *Naureen Z., Ali Z., Al R., Nasser M., Jabri A., Khalfan S., Housni A., Gilani S. A., Mabood F., Farooq S., Hussain J. and Harrasi A. Al 2016 Generation of Electricity by Electrogenic Bacteria in a Microbial Fuel Cell Powered by Waste Water, (July), pp. 329–335.doi: 10.4236/abb.2016.77031*
23. *Pretel R., Durán F., Robles A., Ruano M. V, Ribes J., Serralta J. and Ferrer J. 2015 Designing an AnMBR-based WWTP for energy recovery from urban wastewater : The role of primary settling and anaerobic digestion, SEPARATION AND PURIFICATION TECHNOLOGY. Elsevier B.V. doi: 10.1016/j.seppur.2015.09.047.*
24. *Publishing I. W. A. and Science W. 2011 Removal of endocrine disrupting chemicals in a large scale membrane bioreactor plant combined with anaerobic-anoxic-oxic process for municipal wastewater reclamation Chunying Wu , Wenchao Xue , Haidong Zhou , Xia Huang and Xianghua Wen, pp. 1511–1518. doi: 10.2166/wst.2011.140.*
25. *Rabaey K., Boon N., Siciliano S. D., Verstraete W., Rabaey K., Boon N., Siciliano S. D., Verhaege M. and Verstraete W. 2004 Biofuel Cells Select for Microbial Consortia That Self-Mediate Electron Transfer Biofuel Cells Select for Microbial Consortia That Self-Mediate Electron Transfer, 70(9). doi: 10.1128/AEM.70.9.5373.*
26. *Retnaningrum E. and Wilopo W. 2017 Performance and Bacterial Composition of Anodic Biofilms in Microbial Fuel Cell Using Dairy Wastewater, 20018(2016). doi: 10.1063/1.4953492.*
27. *Roh H. and Chu K. 2010 A 17 -Estradiol-utilizing Bacterium , Sphingomonas Strain KC8 : Part I - Characterization and Abundance in Wastewater Treatment Plants, 44(13), pp. 4943–4950.***doi:** *10.1021/es060923f*
28. *Smith A. L., Skerlos S. J. and Raskin L. 2013 Psychrophilic anaerobic membrane bioreactor treatment of domestic wastewater, Water Research. Elsevier Ltd, 47(4), pp. 1655–1665. doi: 10.1016/j.watres.2012.12.028.*
29. *Vidal G. and Diez M. C. 2005 Methanogenic toxicity and continuous anaerobic treatment of wood processing effluents, 74, pp. 317–325. doi: 10.1016/j.jenvman.2004.09.008.*
30. *Yashas S. R. and Murthy B. M. S. 2017 Synergistic Impact of Endocrine Disrupting Compounds (EDC's) On Environmental Facilities : A Mini Review, 52(1), pp. 17–21.doi:10.14445/22315381/IJETT-V52P204*
31. *Ying G., Kookana R. S. and Ru Y. 2002 Occurrence and fate of hormone steroids in the environment, 28, pp. 545–551.https://doi.org/10.1016/S0160-4120(02)00075-2*
32. *Yoshimoto T., Nagai F., Fujimoto J., Watanabe K., Mizukoshi H., Makino T., Kimura K., Saino H., Sawada H. and Omura H. 2004 Isolates from Activated Sludge in Wastewater Treatment Plants, 70(9), pp. 5283–5289. doi: 10.1128/AEM.70.9.5283.*
33. *Yu Y., Huang Q., Cui J. and Zhang K. 2011 Determination of pharmaceuticals, steroid hormones, and endocrine-disrupting personal care products in sewage sludge by ultra-high-performance liquid chromatography–tandem mass spectrometry, pp. 891–902. doi: 10.1007/s00216-010-4295-2.*
34. *Zaharin A., Soraya A. and Mangala S. 2014 Occurrence of 17 α -ethynylestradiol (EE2) in the environment and effect on exposed biota : a review, Environment International. Elsevier Ltd, 69, pp. 104–119. doi: 10.1016/j.envint.2014.04.011.*