Membrane Bioreactor (MBR) for the Removal of Emerging Contaminants from Municipal Wastewater and its Viability of Integrating Advanced Oxidation Processes

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Water scarcity

- Global issue!!
- About 2 billion people worldwide experience high water stress
- About 3 billion people experience severe water crisis at least one month of a year
- More than 5 billion people will face water shortage by 2050\(^1\).

(WRI 2015)

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1 World water development report, United Nations 2019
2 Aqueduct Projected Water Stress Country Rankings, World Resources Institute 2015
Municipal wastewater and Emerging contaminants (ECs)

- Comprised of wastewaters generated from various standpoint domains
- About 70-130% of municipal freshwater consumption becomes wastewater¹
- ECs are new chemicals with no regulatory standards and whose effects on environment and human health are still largely unknown
- Pharmaceutically active compounds (PhACs), personal care products (PCPs), stimulants, pesticides, steroid hormones, endocrine disrupting compounds (EDCs)
- To avoid releasing of contaminated wastewaters to natural environment- effective treatment systems are highly needed

¹ Wastewater Treatment Plants: Planning, Design, and Operation, Qasim, 2017
Membrane bioreactor (MBR) as an Advanced wastewater treatment:

- Combination of **biological treatment** and **membrane filtration system**
- **High quality effluent, less** sludge production, low-demand of **tertiary treatment** and less **space** requirement
- The global MBR market was about **2 billion in 2018** and expected to reach **3.8 billion** by **2023**.
- **Dramatic reductions** in the **membrane cost** (1/10) over the last two decades
Membrane fouling: a big challenge in MBR operation

- **Inevitable phenomena** - major bottleneck!
- **Very complex process** - possible deposition of organic, inorganic and biological compounds on/in the membrane surface - deteriorates membrane permeability
- **Fouling mitigation** requires intensive energy and chemicals
- **About 50-70% of OPEX** is attributed to physical fouling controlling in MBRs

Integrated MBR-Advanced oxidation processes (AOPs) concept in wastewater treatment

- For **high grade** effluent quality
- **AOPs** involve aqueous phase oxidation of ECs by *in-situ* generated powerful reactive species, e.g., hydroxyl radicals
- MBRs offer **solids** and **turbidity free** water- gives high technical flexibility to be integrated with AOPs
- Electrochemical, photochemical, ozonation, Fenton or sonochemical processes.

Fig. Decision tree for MBR-AOP integration concept
## Results

### Assessment of MBR performance at different operating conditions

- A pilot-scale aerobic submerged MBR was operated for more than 200 days, including Nordic cold periods (< 10°C) and varying solid retention times

### Table of Parameters and Values

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Avg. Flux (Continuous)</td>
<td>L m⁻² h⁻¹</td>
<td>Paper I: 7.80, Paper II: 4-6, Paper III: 4-6, Paper IV: 4-6</td>
</tr>
<tr>
<td>F/M ratio</td>
<td>kg COD (kg MLSS. d)⁻¹</td>
<td>Paper I: 0.02 – 0.05, Paper II: 0.027 and 0.09, Paper III: 0.02 – 0.09, Paper IV: 0.02 – 0.09</td>
</tr>
<tr>
<td>Aeration intensity</td>
<td>m⁻³ m⁻² h⁻¹</td>
<td>Paper I: 0.4 – 0.6, Paper II: 0.2, Paper III: 0.2 - 0.23, Paper IV: 0.2 - 0.23</td>
</tr>
</tbody>
</table>

![Fig. Schematic diagram of pilot-scale MBR plant](image-url)
Results

- **Assessment of MBR performance**
  - **Nordic cold conditions**
  - Significant membrane permeability reduction (~75%) - at low temperatures (7-10°C). However;
  - Consistently high removal of organics, nutrients and solids.
  - High reductions of pathogens, e.g., human enteric viruses (NoV GII > NoV GI > AdV) and faecal indicators (E-coli and enterococcus)
  - Relatively high heavy metals removal, which meets EU and WHO guidelines

https://doi.org/10.1016/j.scitotenv.2016.11.122
Results

Assessment of the performance of MBR at different solid retention times

- Removal and fate of 23 diverse ECs were studied at different operational solid retention times (SRTs): 21 days and 60 days.
- Large Variations in removal efficiencies of ECs were observed (non-removal to > 99.9%) - MBR is not the optimal solution!
- Physico-chemical (pKa, logD, log $K_d$) and molecular properties of ECs and plant operating conditions greatly influence ECs removal in MBR
- Major mechanism of ECs removal: biotransformation and biosorption

https://doi.org/10.1016/j.scitotenv.2019.02.308
Results

Integration assessment - Electrochemical oxidation (ECO) for treating/polishing MBR effluent:

- Model EC compound: Carbamazepine (CBZ)
- Novel MMO electrode: Ti/Ta$_2$O$_5$-SnO$_2$
- As prepared electrodes were characterized using SEM, AFM, CV etc.

![Fig. Experimental set-up for electrochemical oxidation.](image)
Results

Integration assessment - Electrochemical oxidation (ECO) for treating/polishing MBR effluent:

- Operating parameters, such as current density, initial ECs concentration, pH, temperature - effecting CBZ degradation efficiency, were studied in aqueous solution.

- Optimized condition of current density = 9 mA cm⁻²; pH = 6; T = 11 ± 1 °C was applied to real MBR effluent

Table: Removal of CBZ in real MBR effluent by using Ti/Ta2O5-SnO2 electrode.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Initial concentration in MBR effluent</th>
<th>Final concentration after 8 h of electrolysis</th>
<th>Electrolyte</th>
<th>Removal (%)</th>
<th>EC (kWh m⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbamazepine (µg L⁻¹)</td>
<td>10.75 ± 0.35</td>
<td>&lt; 0.07 (LOD)*</td>
<td>No electrolyte</td>
<td>&gt;99.99</td>
<td>109.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>&lt; 0.07 (LOD)*</td>
<td>0.1 M Na₂SO₄</td>
<td>&gt;99.99</td>
<td>57.2</td>
</tr>
</tbody>
</table>

http://dx.doi.org/10.1016/j.apcatb.2017.09.017
Results

Integration assessment - Photochemical oxidation (PCO) for treating/polishing MBR effluent:

- Model compounds: CBZ and diclofenac (DCF)- not efficiently removed in MBR
- Heterojunction photocatalyst: Ag$_2$O/TiO$_2$(P-25) composite is used
- Operating parameters were optimized both in aqueous (DW) and real MBR effluent (RME) matrices

![Fig. Experimental set-up for photocatalytic oxidation](image)
Results

Integration assessment - Photochemical oxidation (PCO) for treating/polishing MBR effluent:

- About 90% ECs degradation under optimized catalyst dosage in both the matrices
- About 2-fold catalyst dosage was required in RME matrix than in DW matrix to achieve same level of ECs removal
- Mineralization rate of about 55 to 65% for both the compounds in different matrices.

Table: Removal rates (% ±SD) of ECs under the varying catalyst dosages and the extent of mineralization (%) in two different water matrices.

<table>
<thead>
<tr>
<th>Solution Matrices</th>
<th>Target Pollutants</th>
<th>Catalyst concentration (g L⁻¹)</th>
<th>Removal efficiency (%)</th>
<th>Mineralization (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ECs in deionized water (DW)</td>
<td>CBZ</td>
<td>0.2</td>
<td>80.40 ± 0.5</td>
<td>67.90</td>
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<td></td>
<td></td>
<td>0.4</td>
<td>89.10 ± 1.5</td>
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<td></td>
<td></td>
<td>0.6</td>
<td>88.60 ± 1.4</td>
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<td></td>
<td>0.8</td>
<td>91.70 ± 1.5</td>
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<td></td>
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<td>1.0</td>
<td>89.02 ± 4.7</td>
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<tr>
<td>ECs in real MBR effluent (RME)</td>
<td>DCF</td>
<td>0.2</td>
<td>87.60 ± 2.2</td>
<td>64.80</td>
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<td></td>
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<td>0.4</td>
<td>93.50 ± 0.1</td>
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<td>0.6</td>
<td>93.30 ± 0.1</td>
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<td>0.8</td>
<td>93.04 ± 0.1</td>
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<td>1.0</td>
<td>93.40 ± 0.3</td>
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<tr>
<td>ECs in real MBR effluent (RME)</td>
<td>CBZ</td>
<td>0.4</td>
<td>76.60 ± 5.2</td>
<td>60.30</td>
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<tr>
<td></td>
<td></td>
<td>0.6</td>
<td>85.40 ± 6.5</td>
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<td></td>
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<td>0.8</td>
<td>89.74 ± 0.4</td>
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<td>1.0</td>
<td>90.30 ± 0.9</td>
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<tr>
<td></td>
<td></td>
<td>1.2</td>
<td>90.95 ± 2.3</td>
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<tr>
<td>ECs in real MBR effluent (RME)</td>
<td>DCF</td>
<td>0.4</td>
<td>86.60 ± 0.3</td>
<td>55.20</td>
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<td>0.6</td>
<td>90.70 ± 4.5</td>
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<td>0.8</td>
<td>92.00 ± 2.1</td>
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<td>1.0</td>
<td>90.40 ± 0.40</td>
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<td>1.2</td>
<td>92.10 ± 1.10</td>
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https://doi.org/10.1016/j.seppur.2018.12.069
Conclusion

» Severe membrane fouling (permeability drop) was observed in MBRs operation in Nordic real cold-water conditions - needs urgent solution!

» Diverse removal efficiencies of ECs during MBR treatment due to several influencing factors

» MBR is not the optimal solution for complete remediation of many recalcitrant ECs

» AOPs, such as ECO and PCO showed promising integration alternatives to achieve complete removal of highly recalcitrant ECs - i.e., enhanced treatment efficiency

Future research prospects

» Developing novel integrated processes and materials, such as microalgae cultivation, microbial electrolysis cells, engineered nanoparticle coated membranes etc. – for reduced energy consumption and recovery of value-added products (Nutrients, biofuels, electricity etc.)