DEVELOPMENT OF A NOVEL FERTILIZER-DRAWN FORWARD OSMOSIS AND ANAEROBIC MEMBRANE BIOREACTOR HYBRID SYSTEM FOR HYDROPONICS

by

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CERTIFICATE OF ORIGINAL AUTHORSHIP

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as part of the collaborative doctoral degree and/or fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

This thesis is the result of a research candidature conducted jointly with another University as part of a collaborative Doctoral degree.

Signature of Student: 김 영 지

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Journal Articles Published or Submitted**


system under continuous operation for simultaneous nutrient removal and mitigation of brine discharge, Bioresource Technology, accepted.


** ** Publications made during the PhD candidature including articles not entirely related to the Thesis. * Articles related to the Thesis.

**Conference papers and presentation**


Presentation made during the PhD candidature including proceedings, oral and poster presentations.
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<thead>
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<th>Description</th>
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<tr>
<td>AL-DS</td>
<td>Active layer facing draw solution</td>
</tr>
<tr>
<td>AL-FS</td>
<td>Active layer facing feed solution</td>
</tr>
<tr>
<td>AnFDFOMBR</td>
<td>Anaerobic fertilizer-drawn forward osmosis membrane bioreactor</td>
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<tr>
<td>AnMBR</td>
<td>Anaerobic membrane bioreactor</td>
</tr>
<tr>
<td>BMP</td>
<td>Bio-methane potential</td>
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<tr>
<td>CDI</td>
<td>Capacitive deionization</td>
</tr>
<tr>
<td>CEOP</td>
<td>Cake-enhanced osmotic pressure</td>
</tr>
<tr>
<td>CMC</td>
<td>Critical micellar concentration</td>
</tr>
<tr>
<td>COD</td>
<td>Chemical oxygen demand</td>
</tr>
<tr>
<td>CRCP</td>
<td>Cake-reduced concentration polarization</td>
</tr>
<tr>
<td>CP</td>
<td>Concentration polarization</td>
</tr>
<tr>
<td>CTA</td>
<td>Cellulose triacetate</td>
</tr>
<tr>
<td>DI</td>
<td>Deionised</td>
</tr>
<tr>
<td>DS</td>
<td>Draw solution</td>
</tr>
<tr>
<td>ECP</td>
<td>External concentration polarization</td>
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<tr>
<td>ED</td>
<td>Electrodialysis</td>
</tr>
<tr>
<td>EDGs</td>
<td>Electron donating groups</td>
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<tr>
<td>EDX</td>
<td>Energy dispersive X-ray spectroscopy</td>
</tr>
<tr>
<td>EPS</td>
<td>Extracellular polymeric substances</td>
</tr>
<tr>
<td>EWGs</td>
<td>Electron withdrawing groups</td>
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<tr>
<td>FDFO</td>
<td>Fertilizer drawn forward osmosis</td>
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<tr>
<td>FS</td>
<td>Feed solution</td>
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<td>HA</td>
<td>Humic acid</td>
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HRT  Hydraulic retention time
ICP  Internal concentration polarization
MAP  Mono-ammonium phosphate
MBR  Membrane bioreactor
MD   Membrane distillation
MF   Microfiltration
MKP  Mono-potassium phosphate
MLSS Mixed liquor suspended solids
NF   Nanofiltration
NFT  Nutrient film technique
OLR  Organic loading rate
OMBR Osmotic membrane bioreactor
OMPs Organic micro-pollutants
PA   Polyamide
PAFDO Pressure-assisted fertilizer-drawn forward osmosis
PAO  Pressure-assisted osmosis
PPCPs Pharmaceutical and personal care products
PRO  Pressure retarded osmosis
RO   Reverse osmosis
RSF  Reverse salt flux
RSFS Reverse salt flux selectivity
SEM  Scanning electron microscopy
SOA  Ammonium sulphate
SRB  Sulphate reducing bacteria
SRSF Specific reverse salt flux
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<thead>
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<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>SRT</td>
<td>Solid retention time</td>
</tr>
<tr>
<td>TAN</td>
<td>Total ammonia nitrogen</td>
</tr>
<tr>
<td>TDS</td>
<td>Total dissolved solids</td>
</tr>
<tr>
<td>TFC</td>
<td>Thin-film composite</td>
</tr>
<tr>
<td>TN</td>
<td>Total nitrogen</td>
</tr>
<tr>
<td>TOC</td>
<td>Total organic carbon</td>
</tr>
<tr>
<td>TP</td>
<td>Total phosphorous</td>
</tr>
<tr>
<td>TS</td>
<td>Total solids</td>
</tr>
<tr>
<td>TrOCs</td>
<td>Trace organic contaminants</td>
</tr>
<tr>
<td>UF</td>
<td>Ultrafiltration</td>
</tr>
<tr>
<td>VFAs</td>
<td>Volatile fatty acids</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
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<tr>
<td>ZLD</td>
<td>Zero liquid discharge</td>
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Abstract

A novel fertilizer-drawn forward osmosis (FDFO) – anaerobic membrane bioreactor (AnMBR) hybrid system was proposed for the sustainable hydroponic application as well as wastewater reuse. This system consisted of three parts: (i) FDFO for concentrating municipal wastewater and producing diluted fertilizer solution, (ii) AnMBR-FDFO hybrid system for treating concentrated municipal wastewater and producing biogas as well as diluted fertilizer solution, and (iii) supplying produced fertilizer solution to hydroponics.

The FDFO performance was initially investigated to achieve simultaneous water reuse from wastewater and production of nutrient solution for hydroponic application. Biomethane potential (BMP) measurements, which can be utilized to simulate the anaerobic process in batch mode to assess the bio-methane production potential from different substrates, were carried out to determine the effect of osmotic concentration of wastewater achieved in FDFO on the anaerobic activity. Results showed that 95% water recovery from FDFO was the optimum value for further AnMBR treatment. Nine different fertilizers were then tested based on their forward osmosis (FO) performances (i.e. water flux, water recovery and reverse salt flux (RSF)) and final nutrient concentration. From this initial screening, ammonium phosphate monobasic (MAP), ammonium sulphate (SOA) and mono-potassium phosphate (MKP) were selected for long term experiments to investigate the maximum water recovery achievable. After the experiments, hydraulic membrane cleaning was performed to assess the water flux recovery. SOA showed the highest water recovery rate, up to 76% while MKP showed the highest water flux recovery, up to 75% and finally MAP showed the lowest final nutrient concentration. However, substantial dilution was still necessary to comply with the standards for fertigation even if the recovery rate was increased.
In order to understand and predict the performance behaviour of anaerobic fertilizer-drawn forward osmosis membrane bioreactor (AnFDFOMBR), a protocol for selecting suitable fertilizer draw solute was proposed and evaluated. Among eleven commercial fertilizer candidates, six fertilizers were screened further for their FO performance tests and evaluated in terms of water flux and RSF. Using selected fertilizers, BMP experiments were conducted to examine the effect of fertilizers on anaerobic activity due to reverse diffusion. MAP showed the highest biogas production while other fertilizers exhibited an inhibition effect on anaerobic activity with solute accumulation. Salt accumulation in the bioreactor was also simulated using mass balance simulation models. Results indicated that SOA and MAP were the most appropriate for AnFDFOMBR since they demonstrated less salt accumulation, relatively higher water flux, and higher dilution capacity of draw solution (DS). Given toxicity of sulphate to anaerobic microorganisms, MAP appears to be the most suitable DS for AnFDFOMBR.

Two types of the AnMBR-FDFO hybrid systems were considered for further studies, which are (i) FDFO post-treatment of AnMBR effluent and (ii) AnFDFOMBR. The first was designed to reduce not only the effect of fertilizer DS on the bioreactor but also membrane fouling via microfiltration (MF)/ultrafiltration (UF) as pre-treatment. Besides, contaminants should be treated by three steps: (i) biological treatment, (ii) MF/UF filtration and (iii) FDFO treatment, which can enhance total rejection rate. Therefore, the behaviour of organic micro-pollutants (OMPs) transport including membrane fouling was assessed in FDFO during treatment of AnMBR effluent. The flux decline was negligible when the FO membrane was oriented with active layer facing feed solution (AL-FS) while severe flux decline was observed with active layer facing DS (AL-DS) with di-ammonium phosphate (DAP) fertilizer as DS due to struvite scaling inside the membrane support layer. DAP DS however exhibited the lowest OMPs forward flux or higher OMPs...
rejection rate compared to other two fertilizers (i.e., MAP and KCl). MAP and KCl fertilizer DS had higher water fluxes that induced higher external concentration polarization (ECP) and enhanced OMPs flux through the FO membrane. Under the AL-DS mode of membrane orientation, OMPs transport was further increased with MAP and KCl as DS due to enhanced concentrative internal concentration polarization while with DAP the internal scaling enhanced mass transfer resistance thereby lowering OMPs flux. Physical or hydraulic cleaning could successfully recover water flux for FO membranes operated under the AL-FS mode but only partial flux recovery was observed for membranes operated under AL-DS mode because of internal scaling and fouling in the support layer. Osmotic backwashing could however significantly improve the cleaning efficiency.

A side-stream anaerobic FDFO and UF membrane bioreactor hybrid system was proposed and operated for 55 days. The FDFO performance was first investigated in terms of flux decline with various fertilizers DS. Flux decline was very severe with all fertilizers due to the absence of aeration and the sticky property of sludge. Flux recovery by physical cleaning varied significantly amongst tested fertilizers which seriously affected biofouling in FDFO via RSF. Besides, RSF had a significant impact on nutrient accumulation in the bioreactor. These results indicated that nutrient accumulation negatively influenced anaerobic activity. To elucidate these phenomena, bacterial and archaeal community structures were analysed by pyrosequencing. Results showed that bacterial community structure was affected by fertilizer properties with less impact on archaeal community structure, which resulted in a reduction in biogas production and an increase in nitrogen content.

The sustainable reuse of wastewater using FDFO was investigated through osmotic dilution of commercial nutrient solution for hydroponic application. Results from the
bench-scale experiments showed that the commercial hydroponic nutrient solution exhibited similar performance (i.e. water flux and RSF) with other inorganic DS. The use of hydroponic solution provides all the required or balanced macro- and micronutrients in a single solution. Hydraulic cleaning effectively restored water flux up to 75% while osmotic backwashing restored by more than 95% illustrating the low-fouling potential of FDFO. Pilot-scale studies demonstrated that FDFO can produce the required nutrient concentration and final water quality (i.e. pH and conductivity) suitable for hydroponic applications. Coupling FDFO with pressure assisted osmosis (PAO) in the later stages could help in saving operational costs (i.e. energy and membrane replacement costs). However, a trade-off between the process footprint and energy costs associated with the additional pressure needs to be further investigated. Finally, the test application of nutrient solution produced by the pilot FDFO process to hydroponic lettuce showed similar growth pattern as the control without any signs of toxicity or any mortality.
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