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Review

When Bioelectrochemical Systems Meet Forward Osmosis: Accomplishing Wastewater Treatment and Reuse through Synergy

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Abstract: Bioelectrochemical systems (BES) and forward osmosis (FO) are two emerging technologies with great potential for energy-efficient water/wastewater treatment. BES takes advantage of microbial interaction with a solid electron acceptor/donor to accomplish bioenergy recovery from organic compounds, and FO can extract high-quality water driven by an osmotic pressure. The strong synergy between those two technologies may complement each other and collaboratively address water-energy nexus. FO can assist BES with achieving water recovery (for future reuse), enhancing electricity generation, and supplying energy for accomplishing the cathode reactions; while BES may help FO with degrading organic contaminants, providing sustainable draw solute, and stabilizing water flux. This work has reviewed the recent development that focuses on the synergy between BES and FO, analyzed the advantages of each combination, and provided perspectives for future research. The findings encourage further investigation and development for efficient coordination between BES and FO towards an integrated system for wastewater treatment and reuse.

Keywords: bioelectrochemical systems; microbial fuel cells; microbial electrolysis cells; microbial desalination cells; forward osmosis; wastewater treatment and reuse; bioenergy; draw solution

1. Introduction

Water and energy are two crucial factors for human and sustainable socio-economic development. According to a recent report [1], 768 million people lack access to fresh water, and another 2.5 billion suffer from sanitation problems. In addition to the deterioration in ecosystems and hydrological cycle, global water consumption is estimated to increase by 55% by year 2050, resulting in significantly increased freshwater demand [1]. On the other hand, energy especially electricity is not available to more than 1.3 billion people, and roughly 2.6 billion of the population are using fossil fuels, which causes various environmental issues such as global warming and environmental pollution. It is estimated that global electricity demand will grow by 70% over the period to year 2035 [1]. A continuous increase in energy demand and serious problems regarding water shortage have called for alternative renewable energy sources and new water treatment technologies with less energy demand. Various technologies have been proposed and investigated [2–5], and among them, bioelectrochemical systems (BES) and forward osmosis (FO) are two state-of-the-art technologies that may play important roles in addressing the issues of water-energy nexus.

As a novel concept for sustainable energy production and energy-efficient wastewater treatment, BES has attracted great attention in the past decade [6]. Assisted by the exoelectrogens (electrochemically-active microorganisms) growing on the anode electrode of a BES, the organic pollutant in wastewater can be degraded anaerobically and the electrons released by exoelectrogens flow spontaneously from the anode electrode to the cathode electrode through an external circuit to produce electrical current (Figure 1A). The first concept of BES was proposed in 1931 [7], and in the past decade the BES has been greatly advanced from both fundamental research and technological development. For wastewater treatment, BES is studied mainly in three aspects [8]: (1) capturing the electrical power directly from organic contaminants in microbial fuel cells (MFCs) [9,10]; (2) harvesting the value-added products (e.g., H_2, CH_4, high-quality water) in microbial electrolysis cells (MECs), microbial electrosynthesis cells (MESs), or microbial desalination cells (MDCs) [11–14]; and (3) removing specific contaminants (heavy metal, perchlorate, *etc.*), for instance, microbial remediation cells (MRCs) [15,16]. Despite the rapid development, there are still some challenges for BES that need to be addressed before practical applications. For example, as an anaerobic wastewater treatment process, the anode effluent of a conventional BES, which contains microbes, organics, nitrogen, phosphorus, *etc.*, cannot be directly reused or discharged without further treatment. In addition, to drive some reactions in MEC or MES, extra energy is required, thereby increasing the energy demand of the whole system.

Forward osmosis (FO) is based on the natural phenomenon of osmotic processes, and can extract clean water and energy from wastewater (e.g., in pressure retarded osmosis—PRO) [17]. FO refers to the water molecule movement across a semi-permeable membrane driven by osmotic pressure difference between the solutions with high (draw) and low (feed) solute concentrations (Figure 1B). FO can achieve

effective rejection of a wide range of contaminants due to the small pore radius (0.25–0.37 nm) [18–20], and is cost-saving and energy-efficient resulted from the low membrane fouling of the naturally spontaneous process; thus, it is advantageous over conventional wastewater treatment technologies, especially hydraulic pressure-driven membrane processes [21–23]. FO can be engineered for producing potable water from natural waters, extracting reusable water from complex wastewater (industrial streams, landfill leachate, digester centrate, *etc.*) [24–28], and dewatering of waste activated sludge [29–31]. However, it should be noted that FO is actually concentrating wastewater instead of degrading contaminants. The remaining concentrates from the feed side of FO still require post-treatment and the energy stored in organic contaminants are not recovered [32]. Moreover, pure water cannot be obtained directly from FO processes because the product water needs to be separated from the draw solutes, which consumes a large amount of energy. Therefore, the selections of draw solutes and the regeneration method are of essential importance to FO processes [30,33]. Because of inherent problems, FO membrane cannot prevent all dissolved draw solute moving from draw solution into feed solution [34], which is called reverse solute leakage [35], another critical shortcoming of FO processes that may decrease the flux and exert negative effects to the post-treatment of wastewater. PRO is a modified FO process with a hydraulic pressure applied in the draw side, and generates the osmotic energy stored in the salinity gradient between saline and fresh waters by releasing the pressurized, diluted salt water through a hydro-turbine [17,36–38]. As estimated by the Norwegian state power company, the amount of osmotic energy may reach 2000 TWh worldwide [36].

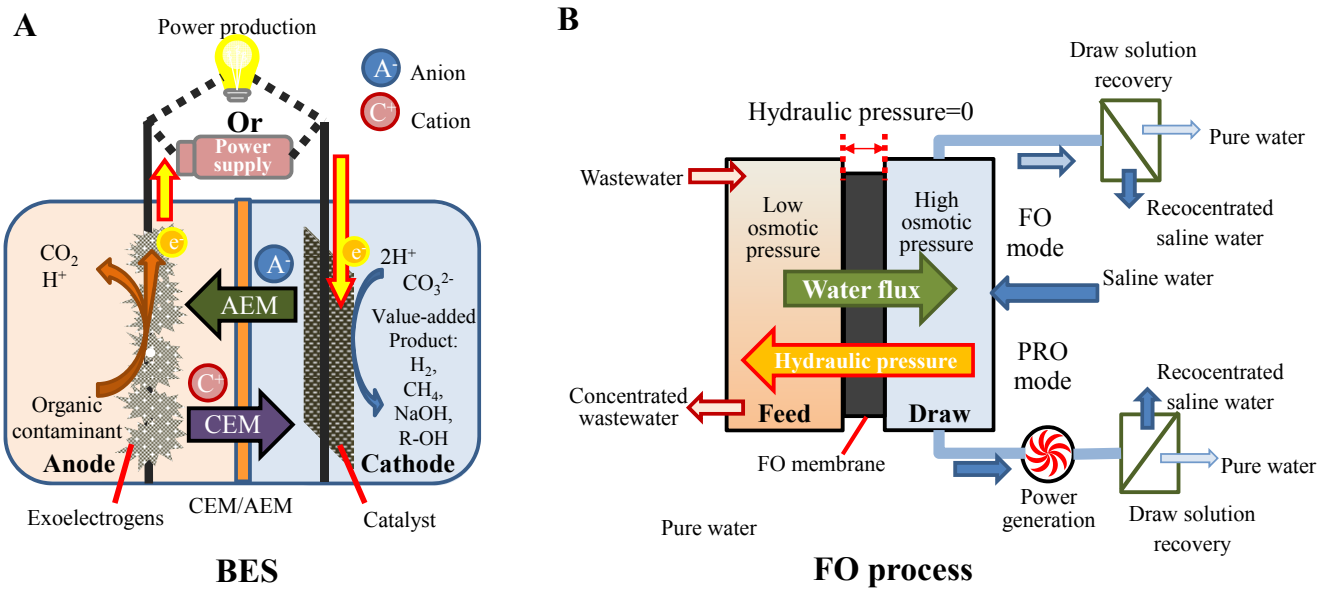

Figure 1. Schematics of bioelectrochemical system (BES) (**A**) and forward osmosis (FO) process (**B**). CEM: cation exchange membrane; AEM: anion exchange membrane.

BES and FO, two distinctly different technologies, have some features that may complement each other to achieve improved wastewater treatment and reuse. Since the first introduction of an osmotic MFC that integrated FO into an MFC in 2011 [39], there has been some advancement in this new synergy. This paper has reviewed the work that links BES to FO in the past four years and provided perspectives on how the synergy between those two technologies can be further developed.

2. Synergy between BES and FO

The synergy between those two technologies lies in the complementing functions in energy recovery and water/wastewater treatment. With the advantage of effective water extraction, an FO process can improve water recovery from the treatment by BES. PRO process may provide BES with sufficient and sustainable energy via water flux. In return, BES can achieve degradation of contaminants in wastewater, provide source of treated wastewater for recovery, and harvest valuable products to improve the sustainability of the whole system. A proper combination of the two may also solve some key issues in each process, such as the supply of draw solute, reverse solute leakage in the FO, and energy requirement for carrying out certain reactions in the BES. By far, there have been two approaches to achieve the synergy between BES and FO, direct integration of FO into BES (osmotic bioelectrochemical systems, OsBES) and external linkage between the two (BES-FO/PRO). In an OsBES, FO membrane is used in place of ion exchange membrane as a separator between different compartments, for example osmotic microbial fuel cells (OsMFCs) [39,40] and osmotic microbial desalination cells (OsMDCs) [41]. For the BES-FO/PRO combined systems, the FO and BES can be integrated through hydraulic and/or electrical connection [42,43]. Those approaches result in two types of functional benefits, FO assisting BES and BES helping FO.

2.1. FO Assisting BES

2.1.1. Extracting High-Quality Water

Extraction of high-quality water via forward osmosis is a distinct feature of an OsBES compared with a conventional BES. Because of the higher water permeability coefficient of FO membrane compared with AEM [44], in the presence of an FO membrane and high salinity catholyte, high-quality water can move from the anode (wastewater) into the cathode in an OsBES. For example, an OsMFC (Figure 2) achieved water flux of 3.94 ± 0.22 liters per square meter per hour (LMH) with a catholyte containing 116 g NaCl/L, while there was no obvious water flux in a conventional MFC [39]. The extracted water can be recovered after further purification to remove draw solute in a process such as reverse osmosis (RO), electrodialysis (ED) or MDC. In this regard, the catholyte of the OsMFC functions as a "catalyst" for clean water extraction. Water flux also results in the dilution of the catholyte; the conductivity of the OsMFC catholyte decreased by 8%–35% after 10–12 h operation [39,40], implying that this system could act as a desalination system. Based on that finding, an OsMDC was developed, in which the conductivity of the artificial seawater was reduced by more than 60% during three-day treatment through combined water dilution and salt removal [41]. Unlike an AEM that allows chloride to move through, an FO membrane in the OsMDC can reject the chloride [41], which may potentially reduce the inhibition of chloride accumulation to microbes [45]. It should be noted that the water flux in an OsBES may gradually decrease due to membrane fouling and reduction of osmotic driving force, resulted from the dilution of catholyte and the concentration of anolyte by water transportation and reverse salt permeation [40,46]. In addition, the increase of salt concentration in the anode due to a concentrating effect may inhibit the microbial growth and reduce the anode performance of OsBES [32,47,48], and this adverse effect could be reduced through increasing anolyte flow rate or adding additional desalination chambers (e.g., concentrating chambers to avoid anion moving into the anode). The periodic

cleaning of FO membrane, reconcentration of catholyte and replacement of anolyte are necessary for achieving continuing water extraction. Because an FO process is generally faster than biological treatment process in a BES, the unbalanced treatment capacity between the two can result in a short hydraulic retention time (HRT) in the BES anode, which will lead to low treatment efficiency for contaminants. Proper coordination between FO and BES treatment capacity (e.g., increasing the size of BES) may accommodate a longer HRT and improved treatment performance in the BES.

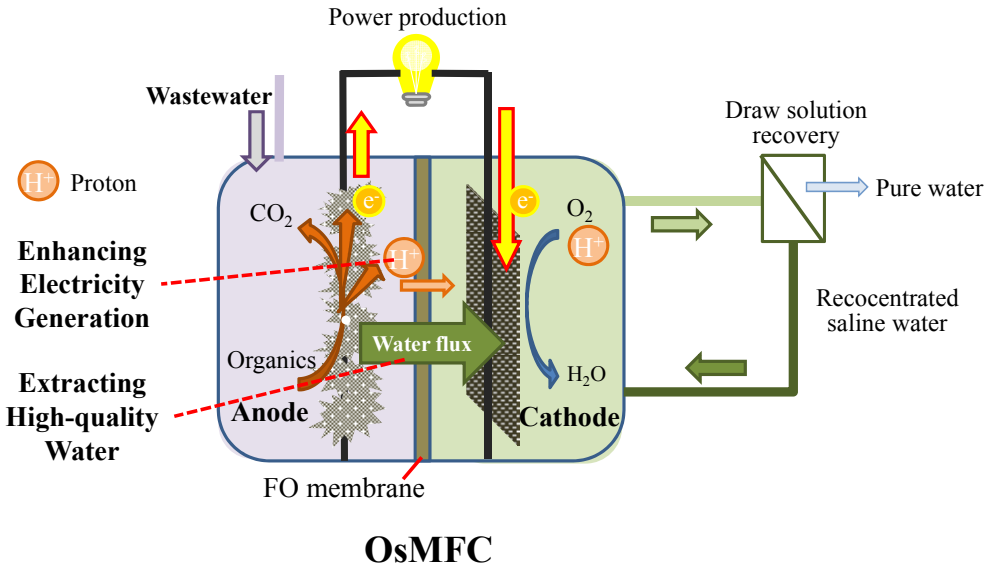

Figure 2. Schematic of an osmotic microbial fuel cell (OsMFC).

2.1.2. Enhancing Electricity Generation

It is found that by replacing ion exchange membrane with FO membrane, greater power production can be achieved in the OsBES compared to the conventional BES. According to polarization curves, the maximum power density of an OsMFC with an aeration-cathode (catholyte: 58 g/L NaCl) was 4.74 W/m^3, 36% higher than that of the MFC with cation exchange membrane (CEM) [39]. In an air-cathode OsMFC, the power density with a catholyte of 35 g/L NaCl was 8% and 87% higher than that of the MFCs containing anion exchange membrane (AEM) and CEM, respectively [40].

The reasons for this enhancement are still under investigation, and there are some preliminary thoughts/hypotheses. In general, the performance of MFCs can be assessed in terms of open circuit voltage and internal losses (including ohmic loss, activation loss, bacterial metabolic loss and concentration loss) [9]. With the same reactor configuration and electrolyte composition, the open circuit voltage of the OsMFC is comparable to that of the conventional MFCs. It is thus reasonable to hypothesize that the enhancement of electricity generation in the OsMFC can be mainly attributed to the reduction in internal losses, e.g., lower membrane resistance, less variation of pH in catholyte and/or anolyte, and higher oxygen rejection performance. It was reported that the air-cathode OsMFC had a relatively lower internal resistance (54 Ω), and the resistance to the flow of ions through the FO membrane was 9 Ω and 19 Ω lower than that through AEM and CEM, which might be due to the acceleration of ion transport [40]. Promoting proton diffusion by water flux may also contribute to the enhanced performance in the OsMFC. The efficient proton transportation supports the reactions on the

electrodes and buffers the increased pH of the catholyte. In an air-cathode OsMFC the decreased anolyte pH (0.86 pH unit) was 24% of the theoretical value and 57% of that in the AEM-equipped MFC, whilst the increased catholyte pH (4.06 pH unit) was only 63% of the theoretical value and 70% of that in a regular MFC [40]. Similar results were obtained in another OsMFC, in which the pH of the catholyte reached 9.76 after 10-h operation, lower than 10.90 in a conventional MFC [39]. These results indicate that the OsMFC may exert positive effect on pH stabilization and thus effectively reduce overpotential [49–51]. Additionally, the use of the high-conductivity catholyte can also help reduce the resistance of the catholyte, resulting in greater power production [46,52].

2.1.3. Supplying Renewable Energy

When a BES is used for harvesting value-added products (e.g., hydrogen or synthesized organic compounds), an extra voltage (thus energy) was usually required to overcome thermodynamic barriers [53]. For example, an external voltage >0.2 V will be required to drive the hydrogen evolution reaction (HER, −0.41 V *vs.* normal hydrogen electrode, pH = 7) with a cathode electrode, when acetate is used as an electron donor in an MEC [9,12,13]. Thus, energy consumption by an external power source is one of the major challenges for the practical application of this type of BES. To make the BES technology economically viable, several sustainable electricity sources (e.g., MFC, solar, wind, waste heat, and geothermal) have been investigated as alternative power supply for BES cathode reactions [53–57]. PRO, a pressurized FO process, may also provide energy to drive BES reaction, and this has been demonstrated in one of our recent studies, in which we applied osmotic energy produced by a PRO to drive hydrogen production in an MEC [43]. In this system, wastewater firstly flowed through the feed side of the PRO for generating osmotic energy via water flux, and then into the anode of the MEC for degradation of organic contaminants. The diluted saline water from the draw side of the PRO functioned as a catholyte in the MEC. Based on the simulation of PRO model, a maximal energy of 106.3 J obtained during the 2-h PRO process was applied to the MEC, which removed 24.9% of COD and produced 11.5 ± 0.8 mL of hydrogen gas with a production rate of 0.031 ± 0.002 $m^3 \cdot m^{-2} \cdot d^{-1}$ [43]. Those results demonstrate the promise of linking PRO to BES via energy supply for addressing water and energy issues.

2.2. BES Helping FO

2.2.1. Enhancing Water Treatment

BES can help FO with improving the treatment of contaminant in water/wastewater. The FO processes generate concentrated feed solution (wastewater) and diluted draw solution (saline water), both of which need further treatment to meet the standard for discharge and reuse. BES can achieve organics degradation in the anode or salt removal driven by electricity. The studies of the OsMFCs showed that overall organics removal in the anode could reach 80%–90% [40,58]. In a PRO-MEC system, the concentrated feed solution from the PRO was treated in the MEC as the anolyte, which removed 20%–60% of chemical oxygen demand (COD) and the organics removal efficiency was improved by increasing the salt concentration in the draw solution of the PRO [43]. With the advantage in low energy consumption [59], MDCs can be applied to reconcentrate the draw solution and/or recover desalinated

water. For example, an OsMDC was developed to recover the desalinated draw solution through both FO and salt removal processes and achieved salt removal of 17%–66% [41]. In another study, an OsMFC was hydraulically coupled with an MDC via sharing the anolyte (wastewater) and salt solution [60]. This coupled system achieved about 85% of total COD removal and more than 95% of conductivity reduction in saline water through dilution in the cathode chamber of the OsMFC and then desalination in the middle chamber of the MDC.

2.2.2. Providing Suitable Draw Solute

A key element in an FO process is draw solution, and the features of an ideal draw solution include high osmotic pressure, easy regeneration, non-toxicity and low cost [33]. The commonly used draw solution based on inorganic salts requires external supply (due to reverse salt flux) and energy-intensive regeneration processes. Ammonium bicarbonate has been demonstrated to be an efficient and recyclable draw solute because of its high osmotic efficiency, high circulation capacity and relatively simple recovery by using moderate heat [21,61–63]. Considering that some wastewaters such as digester centrate and landfill leachate contain a high concentration of ammonium [64], recovering ammonia from wastewater may provide a sustainable source of draw solute for FO treatment. This concept was presented in a recent study that linked an ammonium-recovery MEC to an FO process (Figure 3) [42]. In this system, the MEC was used for ammonia recovery driven by electricity generated from organics oxidation, and the recovered ammonia (0.8 M) was then used for preparing the draw solution of the following FO treatment. Carbon dioxide produced from the MEC anode was also collected to make ammonium bicarbonate. The prepared ammonium bicarbonate draw solute was successfully applied to the FO process and achieved the extraction of half of the MEC anode effluent with a maximum water flux of 2.9 ± 0.1 LMH. The diluted draw solution could be regenerated using the waste heat of combustion processes [65], and resupplied with continuing MEC operation.

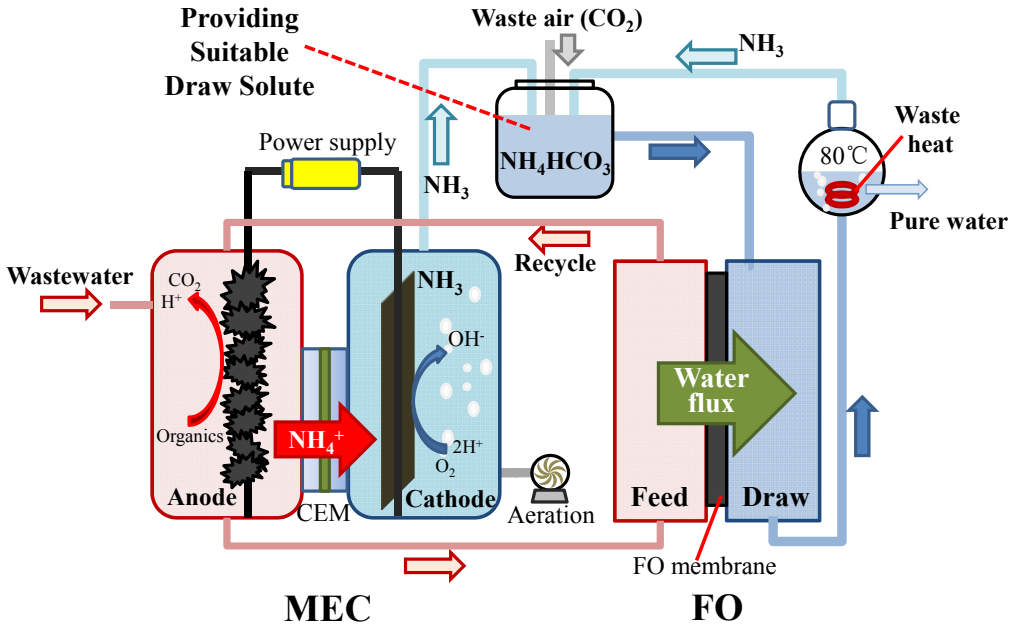

Figure 3. A coupled system consisting of an MEC providing ammonium bicarbonate draw solute for the FO.

2.2.3. Stabilizing Water Flux

Water flux is a key performance parameter for FO and it decreases with FO operation because of the decreased osmotic pressure due to draw solution dilution/feed solution concentration and reverse salt flux [66]. This is especially important to wastewater treatment when FO is applied in the form of osmotic membrane bioreactor (OsMBR) [67,68], as decreased water flux lowers the treatment capacity and increased salt concentration in wastewater inhibits biodegradation [32,47,48]. To alleviate the salt effects on wastewater treatment and maintain a low salinity in wastewater, an MDC can be linked to an OsMBR as a desalination unit. In such a system, the energy in the organic contaminants in wastewater can be extracted by the MDC, and is applied as the driving force for desalination. The desalinated effluent and biodegraded wastewater of the MDC are further treated in an OsMBR, thereby maintaining a stable water flux through decreasing salt concentration.

3. Challenge and Perspectives

The synergy between BES and FO has created great opportunities for achieving sustainable energy production, and efficient wastewater treatment and reuse simultaneously. This is a cutting-edge concept and warrants further investigation of system optimization and identifying proper application niches. To advance the research in this area, we must also understand the challenges associated with system development:

(1) As a membrane-based process, the BES-FO system involves both ion exchange membranes and FO membranes, and thus membrane fouling will be a critical and inevitable problem, which is related to the operational cost [40,46]. More interestingly, the interaction between the fouling of different membranes, which is related to the interaction between BES and FO, is worth further investigation.

(2) Reverse salt flux during the FO and how the leaked salt affects wastewater treatment should be further studied. The leakage of draw solute into the feed solution can decline the water quality of the treated effluent, and on the other hand, the movement of organic contaminants from feed solution to draw solution may decline the quality of recovered water.

(3) BES and FO have different treating speeds; therefore, it is necessary to coordinate the treatment capacity between the two. In general, an FO is faster, compared with biological treatment in a BES. As a result, to maintain a coordinated treatment, the liquid volume of the FO will be much smaller than that of the BES. Practically, there could be multiple BES reactors linked to a single FO.

(4) The energy issues should be better understood. Both BES and FO are considered as low-energy processes; however, the exact energy benefits through coupling those two are not clear and require detailed analysis through comparing with the existing technologies that can achieve similar treatment goals.

(5) System scaling up remains as a great challenge, mainly for BES, as FO has been scaled up to a pilot scale [69,70]. BES scaling up involves the issues such as low-cost materials, scalability of the bench-scale systems, and operation of a scaled system. We are currently working on BES at a scale >100 L and expect to obtain experiences that are helpful with further scaling up.

4. Conclusions

The research has discovered strong synergy between BES and FO that can be used to achieve an integrated system for simultaneous wastewater treatment and sustainable resource recovery. The two processes are functionally complementary. With appropriate integration through hydraulic and electrical connection, BES and FO may gain the additional function/benefits for recovering high-quality water and improving biological wastewater treatment. Future research and development of the integrated BES-FO systems must address several key challenges including membrane fouling, reverse salt flux, scaling up, and coordination for operation capacity between the two technologies.

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Conflicts of Interest

The authors declare no conflict of interest.

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