



# Nutrient-energy-water recovery from synthetic sidestream centrate using a microbial electrolysis cell - forward osmosis hybrid system



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## ABSTRACT

Recovery of nutrient, water, and energy from high-strength sidestream centrate offers benefits such as reusable resource, minimized discharge and cost-savings in mainstream treatment. Herein, a microbial electrolysis cell - forward osmosis (MEC-FO) hybrid system has been investigated for integrated nutrient-energy-water (NEW) recovery with emphasis on quantified mass balance and energy evaluation. In a closed-loop mode, the hybrid system achieved recovery of  $54.2 \pm 1.9\%$  of water ( $70.4 \pm 2.4$  mL),  $99.7 \pm 13.0\%$  of net ammonium nitrogen ( $8.99 \pm 0.75$  mmol, with extended  $N_2$  stripping), and  $79.5 \pm 0.5\%$  of phosphorus (as struvite,  $0.16 \pm 0.01$  mmol). Ammonium loss primarily from reverse solute flux was fully compensated by the reclaimed ammonium under 6-h extended  $N_2$  stripping to achieve self-sustained FO process. The generated hydrogen gas could potentially cover up to  $28.7 \pm 1.5\%$  of total energy input, rendering a specific energy consumption rate of  $1.73 \pm 0.08$  kWh  $m^{-3}$  treated centrate,  $0.57 \pm 0.04$  kWh  $kg^{-1}$  COD,  $1.10 \pm 0.05$  kWh  $kg^{-1}$  removed  $NH_4^+-N$ ,  $1.17 \pm 0.06$  kWh  $kg^{-1}$  recovered  $NH_4^+-N$ , or  $5.75 \pm 0.54$  kWh  $kg^{-1}$  struvite. Recycling of excess  $Mg^{2+}$  reduced its dosage to  $0.08$  kg  $Mg^{2+}$   $kg^{-1}$  struvite. These results have demonstrated the successful synergy between MEC and FO to achieve multi-resource recovery, and encouraged further investigation to address the challenges such as enhancing hydrogen production, reducing nutrient loss, and optimizing MEC-FO coordination towards an energy-efficient NEW recovery process.

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## 1. Introduction

Wastewater has been considered as a resource rather than a waste in the 21st century to sustainably address some major challenges, i.e. water scarcity, nutrient depletion with increasing food demands and energy shortage (McCarty et al., 2011). Conventional wastewater treatment generates a considerable amount of treated water while producing excess mixed liquor (high sludge content) as a sidestream. Subsequent sidestream treatment by anaerobic digestion (AD) enables degradation and stabilization of sludge featuring significant reduction of solid volume (up to 77%) (Kim et al., 2012). The concomitant resource recovery accredits to anaerobic conversion to biogas and nutrients reclamation (N and P) from digested sludge centrate (referred as “centrate” unless otherwise stated) as struvite ( $MgNH_4PO_4 \cdot 6H_2O$ ) (De-Bashan and Bashan, 2004; Qiu et al., 2015). However, the resource recovery

capabilities of AD are greatly impaired by its incomplete organics removal ( $1000$ – $2000$  mg  $L^{-1}$  remaining COD) (Arango et al., 2016; Uggetti et al., 2014) and unsatisfying struvite crystallization under an imbalanced N/P ratio with a low P level ( $\sim 56$  mg  $L^{-1}$ ) as a major limiting factor (Xie et al., 2016), rendering an abundant amount of  $NH_4^+-N$  ( $>1000$  mg  $L^{-1}$ ) that is not recovered. Recycling the nutrient-rich centrate back to the mainstream, though contributing less than 1% of total wastewater flow, leads to notable increase of nutrient loading rate (up to 30%), additional energy demand, and elevated environmental footprint of mainstream treatment facilities (Lackner et al., 2008).

Insufficient resource recovery from sidestream centrate urges substantial process reformation with emphasis on comprehensive reclamation of nutrients (both N and P), bioenergy derived from remaining organics, and high-quality water towards zero-liquid discharge (Tong and Elimelech, 2016). Physicochemical approaches deliver unique advantages for simultaneous nutrient concentration and centrate desalination. For example, clean water could be extracted from centrate in an electro dialysis-reverse osmosis (ED-RO) coupled system (Mondor et al., 2008), with

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ammonium being separated through air stripping (Ippersiel et al., 2012). The concentrate with an elevated P level ( $>100 \text{ mg L}^{-1}$ ) could facilitate subsequent struvite precipitation (recovery rate  $>95\%$ ) (Xie et al., 2016). RO could be replaced by microfiltration (MF) for enrichment of volatile fatty acids (VFAs) towards biogas fermentation (Tao et al., 2016), or membrane distillation (MD) for generating contaminant-free fresh water (Kim et al., 2016). However, these electricity-, pressure-, and thermal-driven membrane processes require substantial energy investment, weakening their potential for broader applications.

The path forward lies in energy-efficient innovations for integrated nutrient-energy-water (NEW) recovery from sidestream centrate, such as bioelectrochemical systems (BES) powered by bioenergy and osmotically-driven membrane processes (OMDPs). BES has been deemed as an attractive treatment method due to continuous nutrient enrichment and energy recovery as electricity (microbial fuel cell, MFC) (Logan et al., 2006) or value-added products such as hydrogen gas (microbial electrolysis cell, MEC) (Kelly and He, 2014). OMDPs, especially forward osmosis (FO), take advantage of osmotic pressure gradient across a semi-permeable membrane to spontaneously harvest high-quality product water (Lutchmiah et al., 2014). Integrating BES with OMDP may accomplish multiple tasks including nutrient recovery, bioenergy conversion, and wastewater treatment towards further reuse (Hou et al., 2016; Lu et al., 2014).

Our previous study has demonstrated the feasibility of a hybrid MEC-FO system using recovered ammonium as a draw solute for subsequent water recovery (Qin and He, 2014). However, phosphorus recovery was not addressed, and this warranted synergistic coordination between MEC and FO towards multi-nutrient reclamation. Exact energy performance (i.e. energy output and recovery efficiency) was not quantified to enable better comparison with existing technologies aiming at similar targets. In addition, lacking thorough mass/energy balance on a system level would jeopardize the scaling-up potential of this hybrid system. Herein, we have comprehensively evaluated the feasibility of the aforementioned NEW recovery from synthetic centrate by using this hybrid MEC-FO system (linked to a struvite precipitation process). The specific objectives of this study were to (1) enhance the effectiveness of phosphorus recovery via struvite from low-P synthetic centrate; (2) analyze NEW mass balance on a system level with emphasis on  $\text{NH}_4^+\text{-N}$  loss and self-compensation, closed-loop struvite precipitation yield, continuous water extraction capability, and bioenergy recovery as hydrogen gas; and (3) quantify specific energy consumption of the hybrid system to identify the challenges for future development.

## 2. Materials and methods

### 2.1. MEC-FO hybrid system

The MEC-FO hybrid system was operated either in an open-loop mode for start-up or in a closed-loop mode to minimize liquid discharge (Fig. 1). In the open-loop mode, a synthetic centrate was continuously pumped into the MEC anode chamber, where exoelectrogens (electrochemically active microorganisms) converted the organics into electricity to power ammonium migration across the cation exchange membrane (CEM,  $12 \text{ cm} \times 6 \text{ cm}$ ), rendering continuous enrichment of ammonium and energy recovery as hydrogen gas in the cathode. Upon utilization of biodegradable COD (indicated by diminished electricity), the anolyte was replaced by fresh synthetic centrate (for next batch cycle), and the discharged anolyte was transferred to the FO feed chamber for water recovery and phosphorus concentration. Finally, the concentrated feed solution was filtered, and the phosphorus was reclaimed from filtrate as struvite ( $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ ) via chemical precipitation. In a closed-loop mode, in addition to the above processes, the supernatant was then recycled back to the anode of the MEC mixing with fresh synthetic centrate as the influent. For example, the struvite precipitation supernatant from the cycle 1 was mixed with fresh synthetic digestate to serve as the MEC anode influent in the cycle 3 with a mixing ratio shown in Table S1 (Appendix A. Supplementary Materials). The whole system was operated in a temperature-controlled lab ( $20 \pm 2 \text{ }^\circ\text{C}$ ).

### 2.2. MEC setup and operation

A flat-plate MEC ( $20 \text{ cm} \times 10 \text{ cm}$ ) was constructed with two identical chambers (90 mL each). A 12-cm carbon brush was inserted into the anode chamber as an anode electrode. Two pieces of carbon cloth ( $12 \text{ cm} \times 4 \text{ cm}$ ) coated with Pt/C as catalyst ( $0.5 \text{ mg Pt cm}^{-2}$ ) were bound on two sides of a stainless-steel mesh ( $12 \text{ cm} \times 4 \text{ cm}$ ) and served as the cathode electrode. The synthetic centrate contained (per liter DI water): 2.60 g NaAc ( $2000 \text{ mg L}^{-1}$  COD), 3.82 g  $\text{NH}_4\text{Cl}$  ( $1000 \text{ mg L}^{-1}$   $\text{NH}_4^+\text{-N}$ ), 0.31 g  $\text{KH}_2\text{PO}_4$  ( $70 \text{ mg L}^{-1}$   $\text{PO}_4^{3-}\text{-P}$ ), 0.10 g  $\text{MgSO}_4$ , 0.05 g  $\text{CaCl}_2$ , and 1.0 mL trace elements, to mimic digestion centrate without any buffer agents (Münch and Barr, 2001). Each of the anolyte and the catholyte (0.01 M NaCl) was connected with a 200-mL reservoir bottle (Table S1), and recirculated at  $10 \text{ mL min}^{-1}$  ( $0.85 \text{ cm s}^{-1}$  for 0.5 cm pipe). The MEC was first operated in an MFC mode (detailed information in Supplementary Materials) for two weeks and then switched to the MEC mode with 0.8 V external voltage. The MEC was operated in a

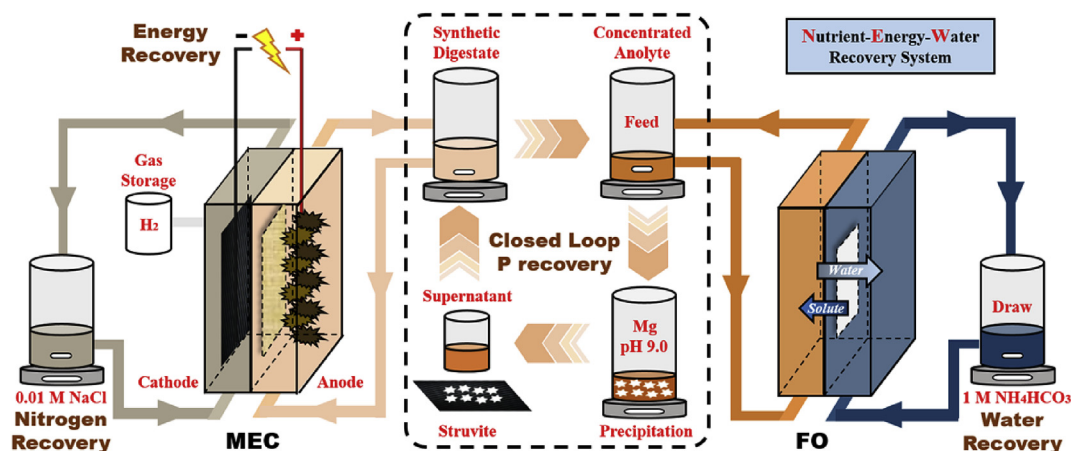


Fig. 1. Schematic of the hybrid MEC-FO system for nutrient-energy-water (NEW) recovery.

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