

In situ utilization of generated electricity for nutrient recovery in urine treatment using a selective electro dialysis membrane bioreactor



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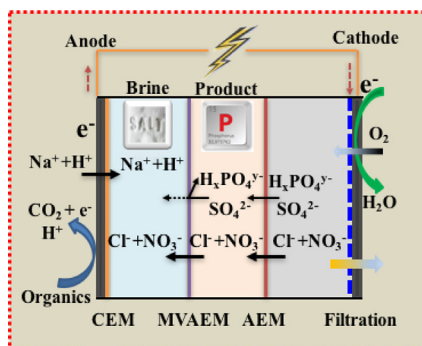
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HIGHLIGHTS

- An electro dialysis membrane bioreactor was developed for urine treatment.
- C, N, P and S in urine could be removed simultaneously in the EDMBR system.
- Phosphate and sulfate separation and selective recovery from urine were achieved.
- The EDMBR showed high potentiality for urine desalination.

GRAPHICAL ABSTRACT



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ABSTRACT

Recovering resources, especially energy and nutrients, from source-separated urine, is attracting increasing interest. By integrating electro dialysis into an electrochemical membrane bioreactor, a novel, selectively electro dialysis membrane bioreactor (EDMBR) was developed for the treatment of source-separated urine and nutrient recovery. Before injection into the EDMBR system, the diluted urine was pretreated by ammonia stripping. The concentration of ammonia in the urine decreased from 1292.2 ± 47.5 mg/L to 235.1 ± 5.7 mg/L upon stripping, denoting an 81.8% ammonia removal efficiency. Phosphate and sulfate recovery from urine were then achieved *in situ*, utilizing the electricity generated by selective electro dialysis in the EDMBR. In this EDMBR system, which was fed with treated urine after stripping, a maximum power density of 23.5 W/m^3 was obtained at an external resistance of 1000Ω . Consequently, 97.4% of the NH_4^+ , 76.7% of the PO_4^{3-} , and 94.5% of the SO_4^{2-} was removed, while 65% of the phosphate and 54.9% of the sulfate (based on their amounts in urine after stripping) were recovered as a concentrated solution. Moreover, other salts, especially NaCl, were selectively retained as brine, indicating that the EDMBR shows promise for urine desalination. This study demonstrates a new strategy for nutrient separation and recovery from source-separated urine by utilizing bioelectricity generated *in situ*.

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1. Introduction

Human urine accounts for only 1% of municipal wastewater by volume but contributes approximately 80% of the nitrogen, 50% of

the phosphate, and 10% of the chemical oxygen demand (COD), as well as considerable quantities of sulfate and potassium (Zang et al., 2012). Therefore, relieving nutrient overloading problems in municipal wastewater treatment plants (WWTPs) by the separate pretreatment of urine has attracted increasing interest (Larsen et al., 2009). Furthermore, these essential nutrients,

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especially mineable phosphorus, are limited resources, which has made them strategic materials (Gilbert, 2009). Thus, the removal and recovery of these nutrients from urine has the benefits of not only minimizing nutrient loading in WWTPs but also contributing to sustainable resource management (Ren and Umble, 2016; Tong and Elimelech, 2016). However, urine cannot be utilized on a large scale owing to its high salinity, prohibitive transport costs, and the potential spread of pathogens and organic micropollutants, as well as the significant odor generation and volatilization of NH_3 during transport and application. In light of these problems, it has been suggested that the most efficient approach for urine utilization is source separation and selective nutrient recovery using a decentralized treatment system (Ledezma et al., 2015). Moreover, desalination of the high-salinity urine should be further considered due to the potential salinization problems during utilization of untreated urine (Bradford-Hartke et al., 2015).

Several technical options are available to effectively recover nutrients from source-separated urine, e.g., nitrogen recovery by stripping or absorption and phosphorous recovery through struvite precipitation, as well as nanofiltration and electrodialysis (ED) (Maurer et al., 2006). Of these techniques, struvite precipitation has attracted much attention for the simultaneous recovery of nitrogen and phosphate (X. Wang et al., 2015). However, the $\text{PO}_4^{3-}\text{-P}$ concentration in urine is much lower than that of $\text{NH}_4^+\text{-N}$, which is a limiting factor for precipitation. Thus, a high $\text{NH}_4^+\text{-N}$ removal efficiency requires the addition of extra phosphate, which results in a high residual $\text{PO}_4^{3-}\text{-P}$ concentration and low phosphate recovery (Zang et al., 2012). In this context, the selective separation of phosphate from urine may increase resource recovery efficiency.

Although a wide variety of technologies may be used to treat source-separated urine, considerable work is required to develop an energy-saving and environmentally friendly urine-processing technique (Maurer et al., 2006). Recently developed bioelectrochemical systems, particularly microbial fuel cells (MFCs) capable of converting waste into bioelectricity, have been used in several complex organic wastewater treatments (Logan and Rabaey, 2012), and MFC-based techniques have been employed for energy and ammonia recovery from source-separated urine (Ieropoulos et al., 2012). By integrating an NH_3 stripping and absorption system into a two-chamber MFC, simultaneous ammonium recovery and energy production have been achieved (Kuntke et al., 2012). By applying a voltage, ammonia removal and hydrogen production have been achieved in microbial electrolysis cell systems (Kuntke et al., 2014; Tice and Kim, 2014). These results show that MFC-based techniques have potential for urine treatment. Furthermore, if the recovery of phosphate and sulfate from the source-separated urine as well as desalination can be achieved in bioelectrochemical systems, this energy-saving technique will be even more appealing.

ED is an electrodriven membrane separation process for the concentration and dilution of salts in the aqueous phase using ion-exchange membranes (IEMs) (Moon and Yun, 2014; Zhang et al., 2012b). With regard to source-separated urine treatment, ED has been investigated for $\text{NH}_4^+\text{-N}$, phosphate, potassium, sodium, sulfate, and chloride recovery and pollutant removal (Dodds et al., 2008; Pronk et al., 2006). However, nutrient recovery has not been achieved by this process. Recently, the ED process has been coupled with a struvite reactor for recovering phosphate from the effluent of an anaerobic bioreactor (Zhang et al., 2013). Continual improvements in the ED process, particularly the development of membrane science, have made the system more reliable and less costly, but the high energy requirement of current ED processes still needs to be lowered to improve its technical and economic feasibility (Xu and Huang, 2008). Integrating IEMs into MFC-based systems (Cao et al., 2009) could make ED technology more applicable and economically effective.

In our previous studies, an electrochemical membrane bioreactor (EMBR), which integrates the advantages of both a membrane bioreactor for nutrient removal and a microbial fuel cell for energy recovery, was developed to recover energy from wastewater while harvesting clean water for reuse (Wang et al., 2011, 2012). Furthermore, it has been demonstrated that it is possible to recover net energy from wastewater and to simultaneously harvest high-quality effluent for reuse and reduce membrane fouling in the EMBR (Wang et al., 2013). The electricity generated from the organics can be used *in situ* for ED or selective ED. Thus, the EMBR system may provide an effective and sustainable energy-recovery strategy for source-separated urine treatment. We hypothesized that by integrating selective ED into the EMBR process, nutrients such as phosphate and sulfate could be selectively recovered while salts such as NaCl could be removed using the *in situ* generated electricity.

In this study, a novel selective electrodialysis membrane bioreactor (EDMBR) is developed for source-separated urine treatment and resource recovery by integrating selective ED into the EMBR. The electricity generation of the EDMBR under different resistances is evaluated, and the phosphate and sulfate separation performances and recoveries, as well as desalination utilizing the generated electricity *in situ*, are investigated. Moreover, the mechanisms of nitrogen, phosphate and sulfate removal were explored. This study provides a novel process for source-separated urine treatment and resource recovery that may be used to reduce the loading of conventional WWTPs.

2. Materials and methods

2.1. Pretreatment of urine

Completely hydrolyzed urine was collected from healthy males and diluted fivefold with deionized water in order to simulate real conditions (Tice and Kim, 2014). To recover ammonia from urine and to avoid inhibition of the microbial activities in the next process by the high ammonia content, aeration was used to strip the ammonia from the hydrolyzed urine (Degermenci et al., 2012). In this study, to recover ammonia from urine and avoid the inhibition of microbial activity in the subsequent process by the high ammonia content, ammonia stripping and absorption processes were used to pretreat the hydrolyzed urine. The phosphate and residual ammonia in the urine were removed by the EDMBR process. The original pH of the hydrolyzed urine was 8.7. The pH of the hydrolyzed urine was adjusted to 11 by adding 1 M NaOH before stripping. Fine air bubbles at a flow rate of 0.5 m³/h were distributed through the bottom of the stripping reactor with an aerator to perform natural mixing for 10 h at 40 °C. The ammonia-air mixture from the stripper was absorbed in 1 M sulfuric acid solution. Then, the residual gas was recycled to the stripper unit by diaphragm vacuum pumps (KNF, LABOPORT, N86 KT.18, Germany). The experiment was carried out in batch mode, and each experiment was repeated twice. After pretreatment, the residual urine was stored at 4 °C before use.

2.2. EDMBR assembly

The EDMBR was constructed using Plexiglas blocks with an interior cubic chamber. The assembled reactor consisted of one anodic and two cathodic chambers, each with a volume of 125 mL (i.e., 5 × 5 × 5 cm), and two IEM stacks, each containing three IEMs, were inserted between them to assemble the ED chamber (Fig. 1). The anodic and cathodic chambers were filled with granular graphite (3–5 mm diameter; Sanye Carbon Co., China). The net efficient working volumes of the anodic and cathodic

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