



Phosphorus recovery from digested sludge centrate using seawater-driven forward osmosis



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ABSTRACT

In this study, we demonstrate a novel seawater-driven forward osmosis (FO) process to recover calcium phosphate precipitates from digested sludge centrate without any chemical addition and draw solute regeneration. The FO process effectively pre-concentrated phosphate and calcium in the digested sludge centrate. Spontaneous precipitation of calcium phosphate minerals in the digested sludge centrate was achieved by the sustained concentrative action of the FO process and the gradual pH increase due to the diffusion of protons to the draw solution. Pre-concentrating digested sludge centrate by three-fold resulted in a 92% recovery of phosphate via precipitation. The phosphate precipitate only constituted 3% of the total inorganic solids recovered, therefore subsequent treatment steps would be required to recover phosphorus in a useable form. A water flux decline of 30% from the initial value was observed as the digested sludge was concentrated by three-fold. This observed water flux decline was mostly attributed to the decrease in the effective osmotic driving force due to the increasingly concentrated feed solution and diluted draw solution. It is also noteworthy that membrane fouling was readily reversible. By flushing the membrane with deionised water and subjecting the membrane to feed and draw solutions with the same osmotic pressure as the initial conditions, complete water flux recovery could be achieved.

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1. Introduction

Phosphorus is a key element for all life on earth. Without the phosphorus in biological molecules such as ATP (or adenosine triphosphate) and DNA (or deoxyribonucleic acid), life would not be possible. Phosphorus can also be found in the minerals in bones and teeth. Thus, phosphorus is an essential part of the human diet and a vital element for plants. Indeed, food security is increasingly dependent on the availability of phosphate fertilisers. As natural phosphorus reserves continue to deplete, it is necessary to improve resource efficiency by investing in the recycling and recovery of phosphorus [1]. A considerable fraction of the phosphorus consumed by society ends up in municipal wastewater. In wastewater, phosphorus is a pollutant. When discharged to the environment, phosphorus can cause the widespread eutrophication of receiving waters. This has motivated the implementation of regulatory standards for phosphorus removal at wastewater treatment plants. Overall, the environmental and regulatory need for phosphorus removal, together with the non-renewable nature of phosphorus,

give significant incentive for the wastewater treatment sector to recover phosphorus from wastewater [2–4].

A pragmatic option to supplement phosphorus resources is to recover it from wastewater, or more precisely from anaerobically digested sludge centrate. In a typical wastewater treatment plant, influent phosphorus is biologically accumulated in sludge. When sludge is anaerobically digested, orthophosphate is released and remains dissolved in the sludge centrate (i.e. supernatant). Digested sludge centrate can contain phosphate concentrations in the range of 75–300 mg/L, compared with about 8 mg/L present in influent wastewater [2]. The elevated phosphate concentration in digested sludge centrate presents significant opportunities for phosphorus recovery techniques to be readily integrated into current wastewater treatment infrastructure. Implementing phosphorus recovery can improve nutrient management at wastewater treatment plants. Nutrient rich digested sludge centrate is commonly returned to the head of the treatment plant, leading to the gradual build-up of phosphorus in the plant, decreasing the efficiency of wastewater treatment with respect to phosphorus removal [5]. More importantly, the build-up of phosphorus can also result in gradual struvite precipitation causing blockages and equipment scaling [6]. Thus, by recovering phosphorus from

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digested sludge centrate, phosphorus removal to comply with effluent discharge standards can be improved and costly maintenance due to blockages can be avoided. At the same time, phosphorus fertilisers can be produced.

Despite the benefits of phosphorus recovery from digested sludge centrate, there are several challenges to developing techniques that are both economically viable and environmentally friendly. Conventional techniques to precipitate agronomically suitable phosphate minerals are expensive and chemically intensive. For example, controlled precipitation of the slow-release fertiliser struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) requires the addition of magnesium in a significant quantity to exceed the stoichiometric ratio. The high cost of magnesium salts makes the conventional struvite recovery process uneconomical and consumes more resources than are produced [7]. Indeed, the primary drivers for most recently installed plants are to prevent struvite blockages and to enhance phosphorus removal. Calcium phosphate precipitates have gained recent attention as an alternative phosphate mineral to be recovered from wastewater, attributed to the simplicity, lower cost and easy acceptance into industrial fertiliser production [8]. Additionally, the initial phosphate concentration is the most important parameter that dictates the efficiency, thus cost effectiveness of a phosphorus recovery process. Therefore, phosphorus recovery can also be enhanced by firstly pre-concentrating the phosphate in digested sludge centrate [9,10].

Pre-concentrating phosphate in digested sludge centrate can increase the precipitation kinetics of phosphorus mineral recovery. There have been several membrane-based techniques (e.g. membrane distillation and reverse osmosis) for mineral pre-concentration and subsequent recovery from saline industrial wastewaters [11,12]. However, most of them are not suitable for digested sludge centrate given its high fouling propensity. One key technology with significant potential to perform this function is the osmotically driven membrane filtration process forward osmosis (FO). FO has a number of advantages when applied for the treatment of complex solutions including digested sludge centrate [10,13], fracking fluid [14,15], reverse osmosis brine [16], and landfill leachate [14]. FO membranes can retain more than 97% phosphate in digested sludge centrate [10,13]. Furthermore, fouling in FO is mostly reversible, even with complex feed solutions [17]. The bi-directional transport phenomenon of FO is another important advantage. This leads to an increase in pH of the feed solution [10,18], which is optimal for phosphate mineral precipitation. Additionally, the back diffusion of draw solutes can be utilised, thus, seawater can be applied as a draw solution as a potential additional source of calcium for calcium phosphate precipitation. These key attributes make FO possibly the most superior process to pre-concentrate digested sludge centrate for subsequent phosphorus recovery.

Recent demonstrations of FO to pre-concentrate nutrients prior to phosphate mineral precipitation have shown excellent potential to lower chemical requirements, increase precipitation kinetics, and improve the efficiency of phosphorus recovery from wastewater [4,10,13,19–21]. Xie et al. [10] demonstrated struvite recovery from digested sludge centrate using a hybrid forward osmosis – membrane distillation system with MgCl_2 as the draw solution. In addition to providing favourable conditions for struvite precipitation (i.e. elevated ammonium and orthophosphate concentrations, and an elevated pH), the FO system supplied additional magnesium required for struvite precipitation by reverse draw magnesium flux. However, as noted above, a major drawback with the current approach for phosphorus recovery from digested sludge centrate via struvite precipitation is the need for costly magnesium addition. Furthermore, FO usually requires draw solution regeneration, thus, adding another significant cost component to the overall process.

Here, we demonstrate a novel seawater-driven FO technique to recover phosphorus from digested sludge centrate without any chemical addition and draw solute recovery. In this process, phosphate is retained in the digested sludge centrate by the FO membrane, and water is transferred to the seawater draw solution. The enrichment of phosphate, calcium, and magnesium ions, as well as a slightly alkaline condition in the digested sludge centrate allow phosphate minerals to spontaneously precipitate in the feed solution during the filtration process. This avoids the costs associated with calcium or magnesium addition, as well as pH adjustment – a critical requirement of conventional precipitation processes. In addition, the use of seawater as the draw solution eliminates the need for draw solute regeneration, thus providing an energetically favourable system. The process was evaluated in terms of nutrient pre-concentration efficiency, facilitation of phosphate mineral recovery, water flux dynamics, and membrane fouling.

2. Materials and methods

2.1. Forward osmosis system

A lab-scale, cross-flow FO membrane system with an effective membrane area of 123.5 cm^2 was used for all filtration experiments [22]. The FO membrane cell consisted of two symmetric flow channels each with a length, width, and height of 130, 95, and 2 mm, respectively. The feed and draw solutions were circulated through each flow channel by two variable speed gear pumps (Micropump, Vancouver, Washington, USA). Two rotameters regulated the circulation flow rate at 1 L/min, which corresponds to a cross-flow velocity of 9 cm/s. The feed solution reservoir was positioned on a digital balance (Mettler-Toledo Inc., Hightstown, New Jersey, USA) and weight changes were recorded to determine the permeate water flux during experiments.

A cellulose triacetate (CTA) membrane with embedded polyester screen support was used in this study, and was acquired from Hydration Technologies Innovation (HTI) (Albany, Oregon, USA). The membrane was operated in FO mode (i.e. active layer facing the feed solution).

2.2. Experimental protocol

The FO system was used to process digested sludge centrate until 80% water recovery had been achieved (approximately 3 days). The feed and draw (seawater) solutions had initial volumes of 3 and 10 L, respectively, and the system was operated in a closed loop arrangement. A large draw solution to feed solution volume ratio (i.e. V_{DS}/V_{FS}) was selected to minimise the effects of draw solution dilution, and feed solution concentration (i.e. approach of osmotic equilibrium) on water flux decline during experiments.

Water recovery (Rec) was used to represent the water extraction rate of the FO process for each filtration cycle and is defined by Eq. (1). This was calculated based on the ratio of the cumulative permeate volume and the initial feed solution volume ($V_{f,0}$). Where A_m is the effective membrane area and J_w is the observed water flux at time t .

$$\text{Rec} = \frac{A_m \int_0^t J_w dt}{V_{f,0}} \quad (1)$$

Solution temperature, pH, and electrical conductivity were monitored throughout the duration of experiments. Samples of 20 mL were withdrawn from the feed and draw solutions at specific intervals for analysis. The rejection of nutrients by the FO

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