

Influence of Cell Configuration and Long-Term Operation on Electrochemical Phosphorus Recovery from Domestic Wastewater

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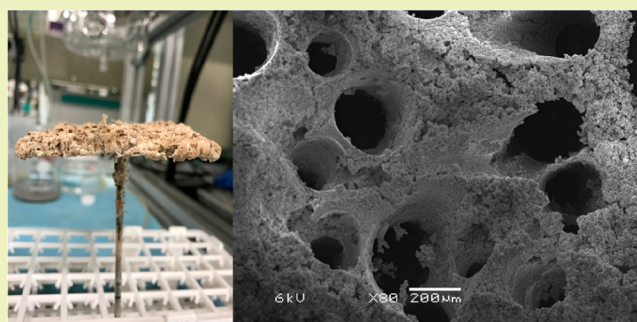
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S Supporting Information

ABSTRACT: Phosphorus (P) is an important, scarce, and irreplaceable element, and therefore its recovery and recycling are essential for the sustainability of the modern world. We previously demonstrated the possibility of P recovery by electrochemically induced calcium phosphate precipitation. In this Article, we further investigated the influence of cell configuration and long-term operation on the removal of P and coremoved calcium (Ca), magnesium (Mg), and inorganic carbon. The results indicated that the relative removal of P was faster than that of Ca, Mg, and inorganic carbon initially, but later, due to decreased P concentration, the removal of Ca and Mg became dominant. A maximum P removal in 4 days is 75% at 1.4 A m^{-2} , 85% at 8.3 A m^{-2} and 92% at 27.8 A m^{-2} . While a higher current density improves the removal of all ions, the relative increased removal of Ca and Mg affects the product quality. While the variation of electrode distance and electrode material have no significant effects on P removal, it has implication for reducing the energy cost. A 16-day continuous-flow test proved calcium phosphate precipitation could continue for 6 days without losing efficiency even when the cathode was covered with precipitates. However, after 6 days, the precipitates need to be collected; otherwise, the removal efficiency dropped for P removal. Economic evaluation indicates that the recovery cost lies in the range of 2.3–201.4 euro/kg P, depending on P concentration in targeted wastewater and electrolysis current. We concluded that a better strategy for producing a product with high P content in an energy-efficient way is to construct the electrochemical cell with cheaper stainless steel cathode, with a shorter electrode distance, and that targets P-rich wastewater.

KEYWORDS: Local high pH, Calcium phosphate, Electrode distance, Current density, Energy consumption



INTRODUCTION

Phosphorus (P) is an essential and irreplaceable element for all life forms, yet the worldwide food production is highly dependent on the finite phosphate rock as a fertilizer.^{1,2} With increasing population and food demand, P scarcity is expected to occur within 50–100 years.³ The geopolitics of the rock will stress prices and availability for fertilizer and consequently food production.⁴ In particular, <1% of phosphate mines are located within the European Union (EU). Therefore, EU listed P as one of critical raw materials because of its importance, prospected scarcity, and irreplaceability.⁵

Phosphate in surface water has significant negative effects on ecosystems by eutrophication.^{2,6} Therefore, many countries have strict legislation on phosphate discharge and usage. In wastewater treatment, the focus has been on the removal of phosphate to prevent eutrophication rather than recovery for reuse. Over the last few decades, there is an increasing interest in recovering phosphate from (municipal) wastewater by precip-

itation and producing valuable products for the fertilizer industry.⁷

A well-known method for phosphorus (P) recovery in wastewater is struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) precipitation, where equal moles of magnesium, ammonium, and phosphate precipitate at alkaline pH.⁷ The struvite process is indeed regarded as a promising way to recover P, as it can be directly used in agriculture as a slow-release fertilizer.⁸ In practice, however, the slow-release fertilizer is rarely used in agriculture and could more effectively be used as a raw material for the fertilizer industry.⁹ Struvite has been researched by many authors, but the process has the disadvantage that Mg addition is needed, which makes the process inefficient and expensive.^{9,10}

Recent studies introduced the possibilities of P recovery by electrochemically induced calcium phosphate precipitation

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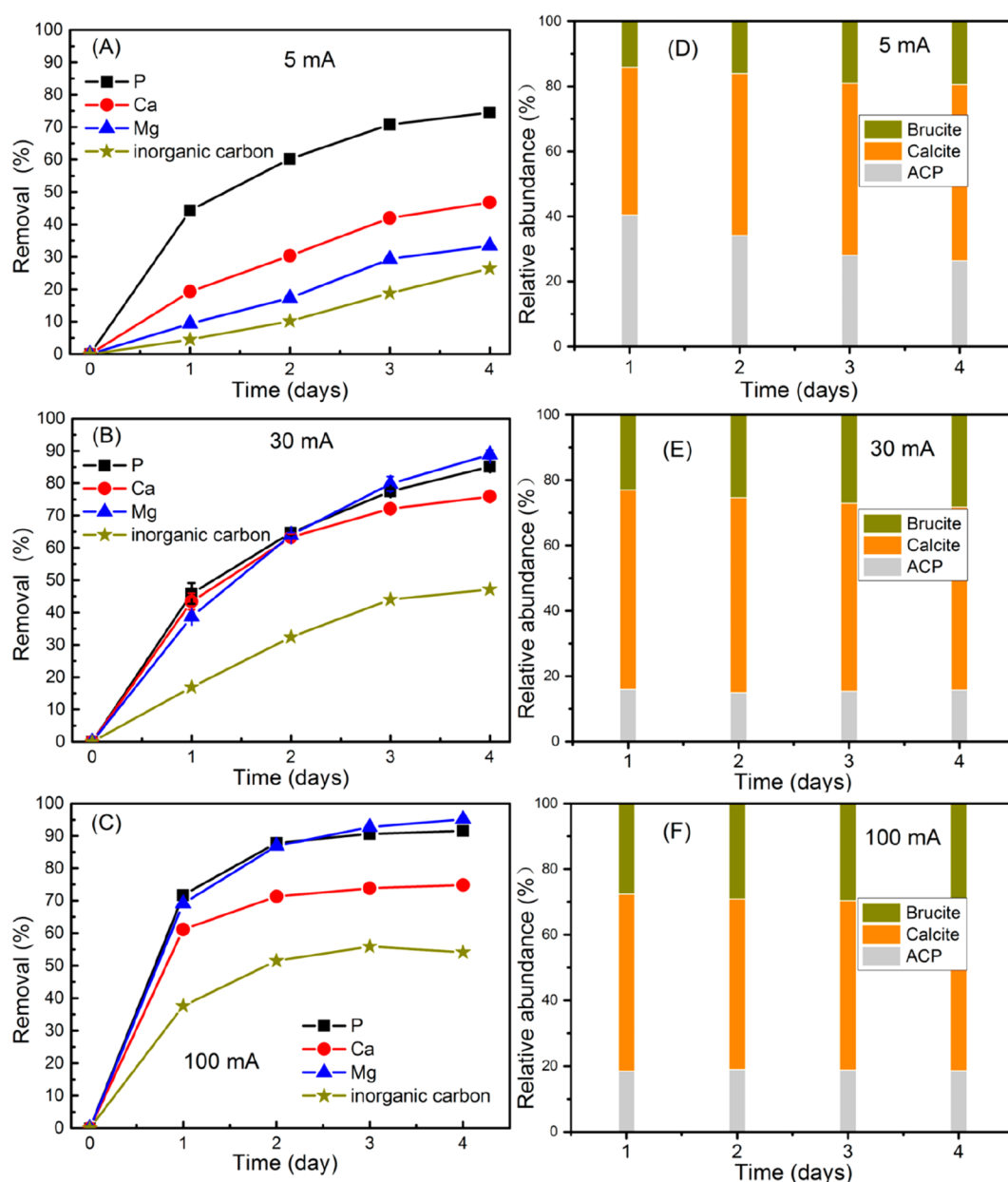


Figure 1. Effects of electrolysis time and current on the removal of P, Ca, Mg, and inorganic carbon, and the corresponding relative abundance of amorphous calcium phosphate (ACP), calcite (CaCO_3), and brucite ($\text{Mg}(\text{OH})_2$) in precipitates. Conditions: electrode distance, 3 cm; anode, Pt-coated Ti disk (\varnothing 8 cm); cathode, grade A Ti 36 cm^2 .

without dosing any chemicals.^{11,12} Calcium phosphate precipitation has the advantage over struvite precipitation that the dosing of calcium is not necessary, as calcium typically occurs in a relatively high concentration in wastewater, in contrast to magnesium.^{9,13} To precipitate hydroxyapatite ($\text{Ca}_5(\text{PO}_4)_3\text{OH}$, HAP), for example, only a Ca/P atomic ratio of 1.7 is needed, while in raw domestic wastewater typically this ratio exceeds 7.¹² Therefore, the addition of chemicals to precipitate P is not needed. Like struvite, calcium phosphate can be used by the fertilizer industry as a raw material. In fact, calcium phosphate is a preferred form as it is the principal component of mined phosphate rock.¹⁴ Electrochemically induced precipitation relies on the in situ production of OH^- ions at the cathode surface, and consequently the pH is increased only locally at the boundary layer.^{11,15} This is another advantage for application in wastewater because the buffering capacity of wastewater hinders

the increase of bulk pH, which otherwise would need high chemical input to reduce.¹⁶ While some authors^{17,18} suggest the use of membranes to separate the cathode and anode solution to reach high P recovery, Lei and co-workers^{11,12,19} developed a membrane-free system to recover P as calcium phosphate, which was also proposed by Wang et al.¹⁵ A membrane-free system omits the issues of membrane fouling and avoids the change of bulk pH, as protons and hydroxide ions are produced simultaneously and equally in the same solution. More importantly, from an engineering point of view, the lack of membranes will simplify the reactor architecture, which could be an important step toward scaling up.

Municipal wastewater was targeted because of its high P recovery potential. The global annual agricultural demand for P-rock is 15–20 Mt.²⁰ The estimated P potential in municipal wastewater is 3–5 Mt per year.¹ Thus, a total load of P in

municipal wastewater lies from 15–25% of the global annual agricultural demand. The possibility of electrochemical P recovery from municipal wastewater was proved recently. The precipitation tendency among calcium phosphate (product), calcite, and brucite (byproducts), and the fate of coremoved calcium, magnesium, and inorganic carbon, had been well-documented,^{12,21} and hence, it will not be discussed in this Article. The goal of this research was to investigate the cell configuration and long-term operation on the removal of P and coexisting ions. Insights in these studies would be useful for further process optimization and scaling up of electrochemical P recovery.

MATERIAL AND METHODS

Electrolysis Cell. The electrolysis cell consisted of a single cylindrical glass reactor with a liquid volume of 1000 mL, where the cathode was placed below the anode. The distance between the electrodes was adjustable. Two inlet connections at the bottom and top allowed a recirculation pump (Masterflex L/S digital economy drive, Germany) for mixing purposes. The reactor was covered by a lid but was open to air, and experiments were carried out at room temperature. For sampling, several inlets were located on the top of the reactor lid. These inlets were also used as feed and effluent points for continuous-flow experiments. The electrochemical cell was run under constant current, and the cell potential was provided by a power supply (ES 030-S, Delta elektronika B.V., The Netherlands). Figure S1 gives an overview about the electrochemical cell.

Electrodes. Electrodes were provided by MAGNETO Special Anodes BV (The Netherlands). The anode is a platinum-coated (20 g/m²) titanium disk (thickness 0.1 cm, Ø 8 cm). A perpendicular 12 cm rod (Ø 0.3 cm) of the same material is welded onto the center to connect the power supply. The cathode has the same design as the anode but is not coated with platinum and is square-shaped (grade A titanium, thickness 0.1 cm, 36 cm²). In addition, a 36 cm² stainless steel (grade A, similarly welded) was used as an alternative cathode material.

Wastewater. The wastewater originated from the influent of the local municipal wastewater treatment plant (Leeuwarden, The Netherlands). Samples were taken from a sampling point after the screening of large particles but before entering the treatment plant. A recent publication discusses processing, storing, and the main wastewater composition.¹²

Analytical Methods. We applied the same analytical methods to quantify the concentrations of P, Ca, Mg, and inorganic carbon before and after treatment.¹² The characterization of solids has been well-addressed previously^{12,21} and is out of the scope of this Article; therefore, it was not performed in this study.

Batch Experiments. We conducted all experiments with a volume of 1000 mL of wastewater without the addition of other chemicals. As a standard, we carried out batch experiments with an electrode distance of 3 cm, titanium cathode (36 cm²), 30 mA (8.3 A m⁻²), and lasting 24 h. In addition to the standard tests, we performed experiments with alternative electrode distance, current (density), material, and longer electrolysis period. We studied each parameter independently from other parameters. For standard batch tests, experiments were carried out in duplicate. For longer period experiments, only the case of 30 mA was repeated.

Continuous-Flow Experiments. Experiments with wastewater entering and leaving the system at a constant flow rate were performed to simulate a continuous-flow system. This configuration simulates to some degree the target application for this technology: inline phosphate recovery in a wastewater treatment plant. The continuous-flow experiments were performed in the same reactor and cell configuration as the standard batch tests. Influent was pumped from a cooled (4 °C) tank into the electrolysis cell at a rate of 1.2 mL/min, which corresponds to an HRT of 14 h. The effluent discharge rate was equal to the influent rate, resulting in a constant electrolyte volume of ~1000 mL. The continuous flow test lasted 16 days.

RESULTS AND DISCUSSION

Influence of Current and Electrolysis Time. Proof of principle of electrochemical P recovery in real domestic wastewater was provided recently by Lei et al.¹² However, the previous tests were performed in a 24 h period, and therefore, the kinetics of the removal of P and coexisting ions for a longer period is not clear. Figure 1 shows the removal trend of four major involved ions in electrochemical wastewater treatment as a function of current and electrolysis time. The overall trend is that the removal efficiencies of P, Ca, Mg, and inorganic carbon all increased with increasing electrolysis time. Moreover, the removal of P is faster than that of Ca and Mg. Furthermore, high current favors the fast removal of all ions but mostly for Ca and Mg.

Specifically, the P removal increased from 75% at 5 mA (1.4 A m⁻²) to 85% at 30 mA (8.3 A m⁻²) and further to 92% at 100 mA (27.8 A m⁻²), all recorded in 4 days. The corresponding removals of Ca were 47%, 76%, and 75%, respectively. The removals of Mg were 33%, 90%, and 95% at 5, 30, and 100 mA, respectively. At 5 mA, while the removal of all ions increased with a longer electrolysis time, the removal of P is always much higher than the rest (Figure 1A). However, at 30 mA, P, Ca, and Mg were removed to almost the same degree, which can be seen from the overlap of the removal trends (Figure 1B). At 100 mA, the removal of Mg becomes dominant and is close or even higher than the removal of P and indeed Ca. While the final removals of P, Ca, Mg, and inorganic carbon were similar between 30 and 100 mA, the increase in the removal of these ions was relatively stable at 30 mA, whereas at 100 mA the removal was accomplished in 2 days. The further increase of electrolysis time to 4 days did not result in an apparent increase in the removal of associated ions (Figure 1C).

Looking at the absolute removal of the four ions (expressed as kg day⁻¹ m⁻², Figure S2), we found that the absolute removal of P is much lower than that Mg, Ca, and inorganic carbon at 30 and 100 mA. This is probably due to the low initial P concentration compared to Mg, Ca, and inorganic carbon. However, at 5 mA, the removal of P is close to that of Mg and lower than that of Ca and inorganic carbon.

Our previous study confirmed the fate of P and coexisting ions.²¹ The removal of P is due to the precipitation of amorphous calcium phosphate (ACP), whereas the removal of Mg is exclusively due to brucite (Mg(OH)₂) formation. Ca was removed mostly as calcite (CaCO₃). In addition, according to the driving force calculation, ACP, as the intermediate phase of HAP, has the highest driving force, followed by calcite and brucite.¹² Indeed, we can see from Figure 1 that the removal efficiency of P is higher than the rest in most cases, although the gap decreased with extended electrolysis time and at increased current. This is probably due to the substantial decrease of P concentration, which results in diffusion limitation and the drop in driving force for ACP formation. For example, 72% removal of P was already achieved in the initial 24 h at 100 mA (Figure 1C). Hence, the further removal of P becomes difficult at a later stage.

However, the removal of Mg is probably limited by the local pH at low current, i.e., 5 mA. Our previous calculation suggested that the precipitation of brucite in the wastewater needs a pH > 10.¹² At high current, however, the removal of Mg is limited by its availability in the vicinity of the cathode due to the fast removal of Mg in the initial 24 h (69%, Figure 1C). Nonetheless, Mg can be removed favorably because Mg²⁺ (positively charged) can be driven to the cathode (negatively charged) by

electromigration even under low concentration, in comparison to phosphate. The removal of Ca and inorganic carbon reaches a plateau in 48 h at 100 mA (Figure 1C). This is due to the diffusion limitation of bicarbonate (negatively charged) to the cathode.²²

The amount of ACP in the recovered precipitates is an important indication of the value of recovered products. On the basis of the calculation approach that we developed previously,²¹ we calculated the relative abundance (RA) of ACP, calcite, and brucite in the recovered precipitates (Figure 1D, E, and F). In general, the RA of ACP is lowered with increased current. In comparison to the RA of ACP at 5 mA (40%), the RA of ACP at 30 and 100 mA is significantly lower (20%). Also, while the RA of ACP is relatively stable at 30 mA (Figure 1E) and 100 mA (Figure 1F) for the whole period, it decreased with increasing electrolysis time at 5 mA (Figure 1D). The RA of brucite increased from 14% (5 mA) to 23% (30 mA) and to 28% (100 mA), which is in line with the conclusion that a high current favors the removal of Mg. However, the precipitates were dominantly calcite. Regardless of the current and electrolysis time, the precipitates consisted of >45% calcite. This is, as explained,²¹ due to the high concentration of inorganic carbon in the wastewater in comparison to phosphate.

In conclusion, at low current (density), the increase of electrolysis time can increase relatively more removal of P. In a short period (i.e., 24 h), the increase of current (density) can drive calcium phosphate formation, but after that, it results in more precipitation of calcite and especially brucite.

Influence of Electrode Distance. Reducing the electrode distance was tested in order to reduce the overall power input, as a smaller electrode gap requires a lower cell potential to reach the required current under same conditions. Also, it was hypothesized that a smaller electrode gap could result in enhanced diffusion of anode-produced protons to the boundary layer of the cathode, reacting with the hydroxide ions to form H₂O.

Figure 2 shows the removal of P, Ca, Mg, and inorganic carbon at different electrode distances. While there is no significant difference for the removal of P, the removal of inorganic carbon is substantially lower with an electrode gap of 0.5 cm, in comparison to 3 and 6 cm. This interesting trend of inorganic carbon removal as a function of electrode distance

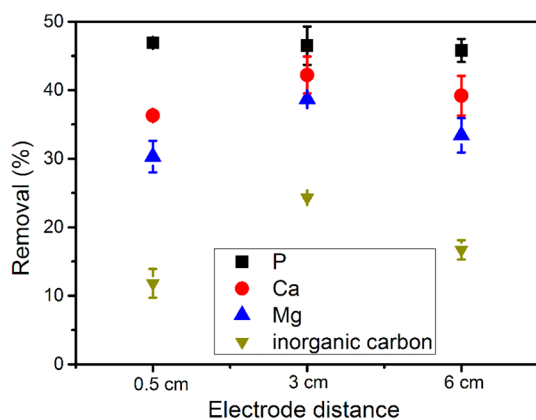


Figure 2. Influence of electrode distance on the removal of P, Ca, Mg, and inorganic carbon. In addition to standard distance (3 cm), electrode distances of 0.5 and 6 cm were included. Conditions: current density, 8.3 A/m²; anode, Pt-coated Ti disk (Ø 8 cm); cathode, grade A Ti 36 cm².

could be explained by the buffer role of inorganic carbon and its precipitation role in calcite formation. In wastewater electrolysis process, protons and hydroxide ions are produced at the anode and cathode, respectively. Given the pH of the wastewater, most of the inorganic carbon was in the form of bicarbonate.²¹ The interaction of bicarbonate with both cathode (OH⁻) and anode (H⁺) is illustrated in Figure 3. Bicarbonate reacts with the

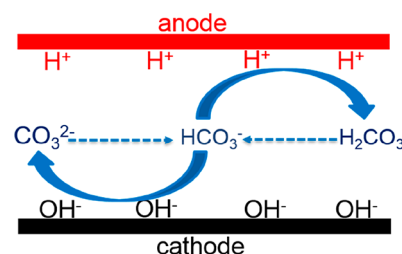


Figure 3. Proposed interaction mechanism of bicarbonate with the anode (generating H⁺) and the cathode (generating OH⁻).

protons and hydroxides ions, forming carbonic acid and carbonate, respectively. Both the formed carbonic acid and carbonate may equilibrate as bicarbonate in the bulk solution. In this way, bicarbonate works as a bridge, achieving indirectly the recombination of H⁺ with OH⁻.²³ The difference is that carbonic acid may decompose as CO₂ and H₂O, whereas carbonate can be removed as calcite. The variation of electrode distance probably influences the local pH for both cathode and anode and therefore influences the precipitation of calcite and the decomposition of carbonic acid. Indeed, we can see that the removal of Mg and Ca shows a similar trend to inorganic carbon. When the electrodes were placed closer, the direct recombination of H⁺ with OH⁻ is likely to be more dominant than the indirect combination through the bridge of bicarbonate. Therefore, the local pH at the cathode might be lower compared to the local pH with a longer electrode distance. This may explain the relatively lower removal of Ca and Mg at 0.5 cm compared to 3 cm. When the electrode distance was increased to 6 cm, the electromigration of bicarbonate (negatively charged) to the anode (positively charged) instead of the cathode (negatively charged) and its reaction with H⁺ may be more favorable, thus both reducing the formation of calcite and the removal of Ca and inorganic carbon. However, it is not clear why the removal of Mg was lower at 6 cm than at 3 cm.

While the studied electrode distance did not affect the performance of electrochemical P recovery, it has implications for cell construction. The required cell potential decreases when the electrodes are placed with a small gap, as the resistance of the system is dependent on the distance between the electrodes. The cell potential decreased from 4.6 V at 6 cm to 4.0 V at 3 cm and further to 3.4 V at 0.5 cm (Table S1). Therefore, the energy consumption was reduced by 15% and increased by 15% when the electrode was placed at 0.5 and 6 cm, respectively, in comparison to the case of 3 cm. However, as was also pointed out in the literature,²⁴ the ohmic drop caused by the electrolyte is only a small part of the needed cell potential. While the difference in applied cell potential is not significantly different, from an engineering point of view, placing the electrodes closer enables the creation of a bigger cathode surface area in a smaller volume. On top of it, the results at different electrode distances also pointed out that the precipitation area only lies in the vicinity of the cathode. Even if the solution is well-mixed and

with an electrode distance of 0.5 cm, a local high pH can be created.

Influence of Electrode Material. A set of indicative experiments were conducted to assess whether electrode material influences the performance of electrochemical P recovery. Stainless steel is much cheaper than titanium, and therefore, the two materials were tested under standard batch conditions. Figure 4 shows that the overall removal using

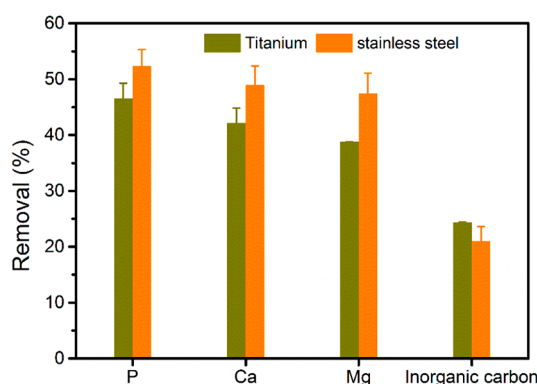


Figure 4. Influence of electrode material on the removal of P, Ca, Mg, and inorganic carbon. Two types of cathode including grade A Ti plate and stainless steel plate were applied. Conditions: current density, 8.3 A/m²; electrode distance, 3 cm; anode, Pt-coated Ti disk (Ø 8 cm).

stainless steel electrode, considering standard deviations, is not significantly different than that with titanium electrode for Ca, P, and inorganic carbon. However, stainless steel appears to achieve at least equal or possibly slightly higher removal

efficiencies, which would make the system economically more feasible because large cathode surfaces are needed for high P removal. The exact explanation for the small difference was not researched in detail, but from a crystallization point of view, it was found by other authors that the surface morphology and material do have a significant role in crystal growth kinetics.^{25,26} Therefore, not only from an economic point of view but also from a process optimization perspective, the cathode material deserves attention.

Performance of Electrochemical P Removal in a Continuous-Flow System. The working mechanism of the electrochemical system relies on the high availability of hydroxide ions in the boundary layer of the cathode. The exact thickness of this layer is unknown. However, it is assumed to be in the range of several μm up to 1 mm.^{11,22} Calcium and phosphate ions need to diffuse into the boundary layer to precipitate, and for this reason, it was predicted that a formed layer on the cathode surface would negatively influence the precipitation rate after a certain period.¹⁷ We performed a 16-day continuous-flow test, and the results are presented in Figure 5A and B. The concentration on day 0 represents the initial concentration.

Removal of P. The plotted figure for P removal (Figure 5A) shows a decrease of concentration within the first day to $\sim 5.3 \text{ mg L}^{-1}$, which corresponds with a removal efficiency of 28%. During the first 6 days, the effluent concentration of P is relatively stable; however, after the sixth day, we saw a clear breaking point where P concentration increased to $\sim 6.7 \text{ mg L}^{-1}$. From this trend, it was concluded that in the first phase of precipitation (0 to sixth day) the layer on the cathode did not inhibit the removal of P and may even enhance the seeding and

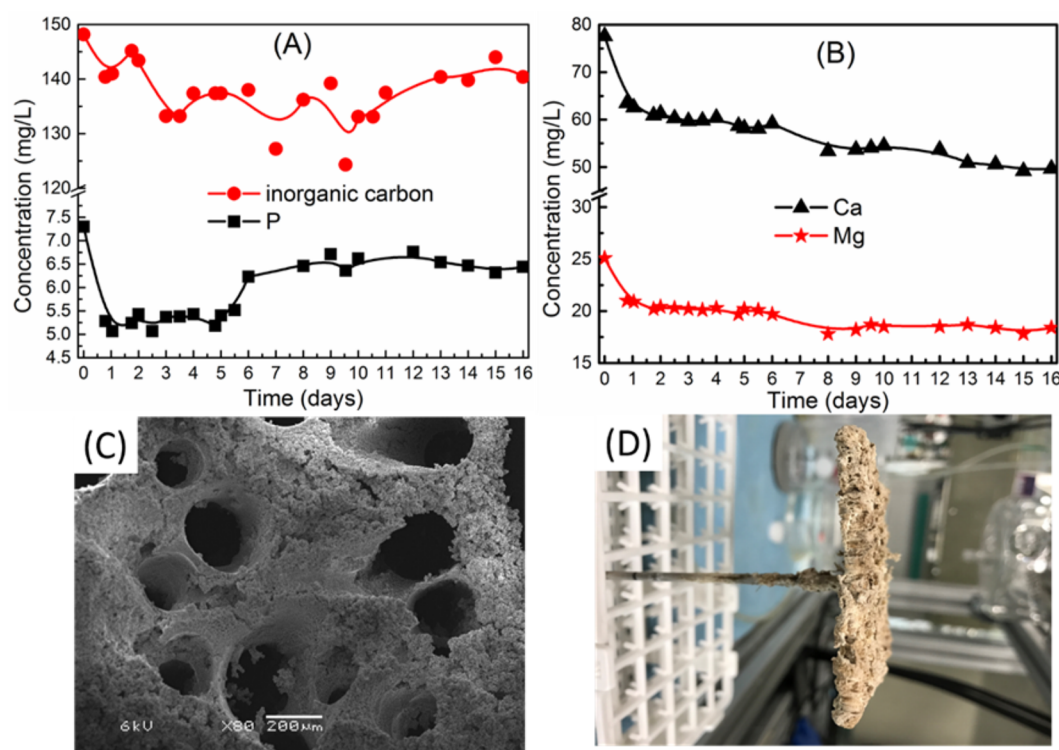


Figure 5. Removal patterns of anions (A) and cations (B) in a 16-day continuous-flow system. (C) Scanning electron microscopy (SEM) image reveals the holes created by H₂ in the precipitates on the cathode surface. (D) Real images of dried precipitates. Day 0 represents the initial concentration at the start of the experiment ($t = 0$). Conditions: current density, 8.3 A/m²; electrode distance, 3 cm; anode, Pt-coated Ti disk (Ø 8 cm); cathode, grade A Ti 36 cm²; flow rate, 1.2 mL min⁻¹; HRT, 14 h.

Table 1. Specific Energy Consumption (kWh/kg) of Electrochemical Phosphorus Recovery under Different Currents; Calculation Based on the Phosphorus Removal and Energy Consumption in 24 h

current	0.005	0.03	0.1	A
current density	1.4	8.3	27.8	A/m ²
cell potential	3.17	3.99	5.56	V
power input	0.01585	0.1197	0.556	W
P removed (%)	44	46	72	%
P removed (mass)	3.44×10^{-6}	3.64×10^{-6}	5.93×10^{-6}	kg P
energy consumption	110	785	2238	kWh/kg P

formation of crystalline phases.²⁵ The formation of hydrogen gas at the cathode surface provides channels in the precipitates, which allows diffusion of ions from the bulk toward the cathode interface. The opposite direction was also proposed: hydrogen gas channels allow hydroxide ions to diffuse from the thin interfacial layer, and therefore crystal growth occurs on top of the existing layer.²² SEM images of the formed products (Figure 5C) show indeed the formation of perfect cylindrical-shaped holes in the precipitates. These holes could provide a contact area with the cathode even after the cathode is completely covered with the precipitated layer. After the breaking point, P removal decreased, which results in a concentration reaching the influent concentration.

Removal of Ca, Mg, and Inorganic Carbon. For Ca and Mg, there was no breaking point observed, as was the case with P removal (Figure 5B). It was doubted if this removal can still be assigned to solely the formation of calcite and brucite or that other processes might be involved. An explanation for the continuous Ca and Mg removal is that part of the removal of the cations might be due to adsorption to the coprecipitated organic substances, which are also present in the wastewater and are negatively charged. The real image of the wet precipitates on the cathode after 16 days operation (Figure S3) in combination with our previous finding on the coprecipitation of organic substances¹⁹ confirmed the presence of organic substances on the precipitation layer. Looking at the removal of inorganic carbon, the data were scattered to such an extent that no clear trend could be identified. During the 16-day continuous-flow test, it was observed that part of the precipitates fell off the cathode or particles were brought into the bulk solution with the formed hydrogen gas bubbles. This could explain the scattered data for inorganic carbon, where carbonate precipitates are dissolved in the bulk solution. Figure 5D shows a picture of the dried cathode after the experiment. It was found that the cathode is completely covered with the precipitation layer. The precipitates occurred on both sides of the cathode and even on the connection rod. On the top side of the cathode, the formed layer was observed to be ~5–10 mm thick.

Economic Feasibility. Table 1 shows the specific energy consumption (kWh/kg P) of electrochemical P recovery. The calculation was based on P removal efficiency and electric consumption in a 24 h period. We can see from Table 1 that the specific energy consumption increased with the increase of current, although a higher current also results in a higher removal efficiency. This indicates that the increase of P removal at high current cannot compensate for the increased energy consumption. The highest specific energy consumption was 2238 kWh/kg P, based on 72% removal of P at 100 mA. The lowest cost was achieved at the lowest current of 5 mA with an energy consumption of 110 kWh/kg P. The high energy consumption of electrochemical P removal is because of the low P concentration in the domestic wastewater. As discussed

earlier, the recovered precipitates consist of a small fraction of calcium phosphate. The fraction of calcite is ~2 times that of ACP (Figure 1D, E, and F). The energy input, especially at high current, was mostly used for calcite and brucite precipitation instead of ACP, due to the low availability of phosphate. Indeed, a calculation based on the published data¹² with 1.15 mM P suggests a cost of 142 kWh/kg P at 30 mA. This experiment was not performed at the lowest current (5 mA). However, if we extrapolate the data from the case of 1.15 mM P and assume a removal efficiency of 40% and the same power input, the specific energy consumption would be only 26 kWh/kg P with 1.15 mM P. If we take the electricity price as 0.09 euro/kWh, the recovery cost lies in the range of 2.3–201.4 euro/kg P, depending on the treatment current and the P concentration in the targeted wastewater.

In comparison to biological and chemical precipitation process, the electrochemical technique seems not that attractive for treating low phosphorus content wastewater. However, if we take the produced O₂ and H₂ into account, the electrochemical technique can be very promising. Our idea is to integrate the electrochemical system with a conventional biological process, which typically includes anaerobic and aerobic processes. The electrochemically produced O₂ can be collected and used for aerobic removal of chemical oxygen demand (COD) and nitrification. The electrochemically produced H₂ can be used as biogas or directly used as electron donor for denitrification. In this way, part of the energy consumption can be compensated by the O₂ and H₂. However, further research is needed to investigate the use of electrochemically produced O₂ and H₂ in biological wastewater treatment process.

CONCLUSIONS

The electrochemical precipitation of calcium phosphate at the cathode surface from raw wastewater was studied for long-term feasibility and efficiency. The production of hydroxide ions at the cathode surface combined with the elevated local pH induce calcium phosphate formation solely on the cathode surface. The coprecipitation of calcite and brucite can be reduced by lowering the current (density) while not affecting the formation of ACP that much. While electrode distance and material have no significant influences on electrochemical P removal, they have important implications for reducing the construction and operation cost and the composition of the product. The evaluation of electrochemical P recovery in a continuous flow system indicated that the initial precipitation of calcium phosphate would not hinder the follow-up precipitation in a certain period. The specific energy consumption of electrochemical P recovery lies in the range of 26–2238 kWh/kg P. The findings of this work can be used as a starting point for a process that can be further optimized to produce a market-feasible technique for removing and recovering phosphate from raw wastewater.

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acssuschemeng.9b00563.

Real image about setup, COD data, cell voltage, absolute removal of ions, and image of cathode after long-term operation (PDF)

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Notes

The authors declare no competing financial interest.

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