

Ammonium and phosphorus recovery and electricity generation from mariculture wastewater by the seawater battery

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ABSTRACT

A magnesium anode seawater battery system was built for recovering $\text{NH}_4\text{-N}$ (ammonium) and P (phosphorus) from mariculture wastewater for the first time. The Mg^{2+} needed for struvite formation was provided by the anode corrosion. The performances of the system were investigated by the struvite crystallization and electricity generation at different conditions. The results show that the pH value of the wastewater was rapidly increased to pH 9 and stable at about pH 10 by the seawater battery without adjusting pH. The pH range was suitable for struvite crystallization. Satisfied P recovery was achieved and it was about 99% at different conditions. The struvite formation was increased with increasing the P:N molar ratio and 1:1 was found to be suitable for enhancing the struvite crystallization. The recovery ratios of the $\text{NH}_4\text{-N}$ and P were 37.37% and 98.99%, and the residual concentrations of them were 14.09 and 0.55 mg/L, respectively. The optimum pH value was pH 9 and the $\text{NH}_4\text{-N}$ recovery was 48.07% at this pH condition. Increasing temperature could shorten the time needed for recoveries in the range from 15 °C to 35 °C. Expanding the magnesium anode area was benefit for the recovery. Increasing temperature or expanding the anode area could improve the electricity generation. The seawater battery could supply relatively stable electricity, and it was 20 mW during the process treating 0.9 L mariculture wastewater. Our results suggest that the seawater battery system is a possible approach for simultaneous $\text{NH}_4\text{-N}$ and P recovery and electricity production from wastewater.

1. Introduction

With the rapid expansion of the mariculture industry, the mariculture wastewater has brought severe pollution to the offshore waters. It is only 20–30% of the N(nitrogen) and even smaller percentage of the P(phosphorus) from the feed that is retained by the fish and the remainder is released into the water [1]. High concentration of nitrogen and phosphorus induces water eutrophication, decreases dissolved oxygen and kills fish [2]. Various chemical and biological processes of N and P removals have been developed by many researchers, such as precipitation[3,4], absorption[5], constructed wetland systems [6], periphyton biofilters [7], and the struvite crystallization process[8]. Among them, the biological processes are the best cost-effective ones. However, they always need long time treatment. For absorption, the absorbent required regeneration. Struvite crystallization is a perfect technique because it can realize N and P recovery from wastewater. Especial for P recovery, it was reported that all reserves of P rock will be exhausted by 2090 [9]. Recovering P from wastewater is an essential and sustainable way for assuring long-term and economical P supply.

Struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) is a white crystalline compound consisting of equal molar concentrations of magnesium, ammonium and phosphorus. The solubility of the struvite was decreased as increasing the pH. It crystallizes under alkaline condition. The struvite forms according to the reaction shown as follows [10]:



Since the concentration of magnesium is relatively lower than that of ammonia and phosphate in many wastewaters, adding magnesium salts is necessary. However, it would increase the cost of the struvite precipitation [11]. Magnesium chloride (MgCl_2) is a quick dissociative substance and widely used as the Mg source for the precipitation [12]. In order to reduce the cost, various low-cost materials have been studied as the alternative sources of magnesium ions. Gunay et al. [13] used magnesit (MgCO_3) as the Mg source in the struvite precipitation from landfill leachate. The performance of magnesium oxide (MgO) was also performed, however, it needed longer reaction time [14]. Lee et al. [11] evaluated the feasibility of bittern and found it was equivalent for phosphate removal and inferior for ammonia removal compared with

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MgCl₂. Because adding magnesium salts is an important part of the struvite precipitation cost, finding a suitable Mg source is essential for the struvite precipitation.

Struvite precipitation is affected mainly by the molar ratio of Mg:NH₄:P and the pH value of the wastewater. Because of the presence of other species, the actual optimum ratio was usually different. It had been confirmed that increasing Mg²⁺ or PO₄³⁻ dosages could increase NH₄-N removal [15]. Struvite is sparingly soluble in neutral or alkaline media and the solubility of it is as low as 0.023 g/L, but readily soluble in acids [16]. On the other hand, Most of the NH₄⁺ was converted to NH₃ at pH > 10.7 [17]. The appropriate pH is range from 8 to 10.7. The variation of different wastewater characteristics led to different appropriate pH. Adding NaOH, Ca(OH)₂ or Mg(OH)₂ is the most common method for increasing the pH value of the wastewater. Liu et al. [8] used aeration to adjust pH. The pH value increased to 8.65.

The seawater batteries have been developed for almost one hundred years and it used seawater as a source for electrolyte. The electricity generation of the battery mainly relied on the corrosion of an activated metal anode in seawater and the reduction of oxygen or water in inert cathode [18]. Magnesium often was applied as the promising anode material due to its various favorable characteristics. Its negative standard electrode potential is −2.37 V (vs SHE), which is closer to aluminum (−2.31 V (vs SHE)) and more negative than zinc (−1.25 V (vs SHE)) [19,20]. In addition, it has high energy density, high current capacity and is environment-friendly [21]. Different types of magnesium alloy also had been developed and studied to enhance the performance of the seawater batteries [22]. As the magnesium anode corrosion, the concentration of the magnesium ions in electrolyte increased. If the magnesium ions were used for other appropriate purposes, the magnesium would be used more sufficiently.

Except wastewater, the marine farms have to face the problem of lacking electricity. As they always located in remote areas, it was difficult and expensive to build large infrastructures there. The solar irradiation had been used for water treatment and electricity generation by hybrid systems that integrated photovoltaic panels with water treatment reactors [23,24]. However, the photovoltaic electricity is influenced by the weather condition and unstable. Seawater batteries can generate stable electricity. It is a good choice for the marine farms.

The objective of this work is to demonstrate the use of magnesium anode seawater batteries for recovering NH₄-N and P and generating electricity from the mariculture wastewater simultaneously. The Mg²⁺ produced by the corrosion of magnesium anode could form struvite crystallization with NH₄-N and P from mariculture wastewater and realize resource utilization. In addition, the battery could provide electricity for the marine farms. The effects of P:N molar ratios, the pH value, the temperature and the anode areas on the struvite crystallization in mariculture wastewater were performed. Further, the electricity performance of the battery was also investigated during the experiments.

2. Materials and methods

2.1. Materials

The magnesium plate used as magnesium anode was purchased from Huayu Company China. Its purity was 99.8%. The titanium plate used as cathode was obtained from Guangrui Company China. NH₄Cl, KH₂PO₄, HCl, NaOH and other reagents used for adjusting pH and determining NH₄-N and P concentrations were supplied by Sinopharm Chemical Reagent Company China. All chemicals used during this study were analytical grade.

The mariculture wastewater used in this study was collected from a seawater farm in Fujian province China and stored in a 10 L container at 4 °C and prepared for the experiments. The characteristics of the wastewater were showed in Table 1.

Table 1
Characteristics of the mariculture wastewater.

pH	NH ₄ -N (mg/L)	TP (mg/L)	SS (mg/L)	Salinity
7.70 ± 0.35	24.13 ± 2.78	7.06 ± 1.24	71 ± 1.28	3.1 ± 0.1%

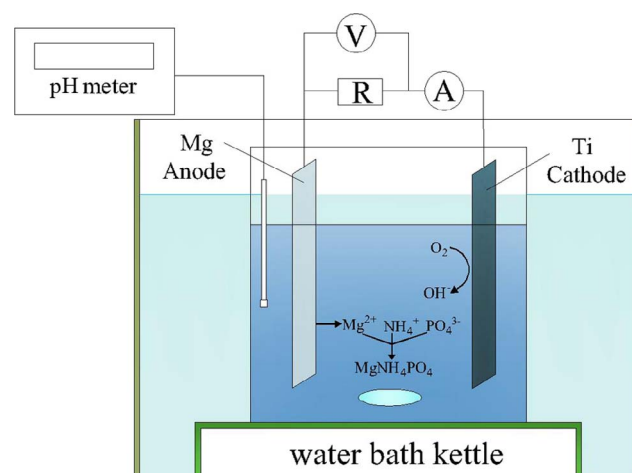


Fig. 1. Schematic of the lab-scale device.

2.2. The experimental device

The experiments were performed with the device as Fig. 1 shows. The reactor was made of borosilicate glass. The seawater battery was consisting of a magnesium anode (252 cm²) and a Ti cathode (34 cm²). The anode and cathode were connected by a resistor (2 Ω) and formed circuit. The electrode distance was 3 cm. The mariculture wastewater was used as electrolyte. A water bath kettle (DF-101S Yuhua company, Gongyi city, China) was used for controlling the temperature of the wastewater. The pH value of the wastewater was detected and recorded by the pH meter (PHS-3C, Shanghai INESA science instrument company, China). Two digital multimeters (Fluke Corporation America) were used to determining the current and the voltage of the device by serial and parallel connection as shown in Fig. 1. 0.9 L wastewater was agitated with a magnetic stirrer at a constant speed during the experiments.

2.3. Analysis

The wastewater samples was taken and filtrated by a 0.45 μm filter membrane (Haining shenghua filtration equipment company, Zhejiang, China) and prepared for determining NH₄-N and P concentrations. The molybdenum antimony anti spectrophotometric method was used for detecting the concentration of phosphate. Its maximum absorption wavelength is 700 nm [25]. The NH₄-N concentration was detected by the Nessler's reagent spectrophotometric method, which was determined from the absorbance at its maximum absorption wavelength (λ_{max}: 420 nm) [25]. The absorbance was detected by a UV–vis spectrophotometer (Cary5000, Agilent, US, with resolution ≤ 0.005 nm) during this study. The recovery ratios of NH₄-N and P were definite as follows:

$$N_R (\%) = \frac{N_0 - N_t}{N_0} \times 100\% \quad (2)$$

$$P_R (\%) = \frac{P_0 - P_t}{P_0} \times 100\% \quad (3)$$

where the N₀ (mg/L) and P₀ (mg/L) are the initial concentrations of NH₄-N and P, respectively. N_t (mg/L) and P_t (mg/L) are the concentrations of NH₄-N and P at time t (s), respectively.

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