



Article Simultaneous Phosphate Removal and Power Generation by the Aluminum–Air Fuel Cell for Energy Self-Sufficient Electrocoagulation

Xiaoyu Han^{1,2}, Hanlin Qi¹, Youpeng Qu³, Yujie Feng^{2,*} and Xin Zhao^{1,*}

- ¹ Department of Environmental Engineering, School of Resources and Civil Engineering, Northeastern University, Shenyang 110819, China
- ² State Key Laboratory of Urban Water Resource and Environment, School of Environment, Harbin Institute of Technology, Harbin 150090, China
- ³ School of Life Science and Technology, Harbin Institute of Technology, Harbin 150080, China
- * Correspondence: yujief@hit.edu.cn (Y.F.); zhaoxin@mail.neu.edu.cn (X.Z.)

Featured Application: The Al-air fuel cell provides an energy self-sufficient electrocoagulation system for phosphate removal.

Abstract: A self-powered electrocoagulation system with a single-chamber aluminum–air fuel cell was employed for phosphate removal in this study. Electricity production and aluminum hydroxides in solution were also investigated. When the NaCl concentration increased from 2 mmol/L to 10 mmol/L, the phosphate removal increased from 86.9% to 97.8% in 60 min. An electrolyte composed of 10 mmol/L of NaCl was shown to obtain a maximum power density generation of 265.7 mW/m². When the initial solution pH ranged from 5.0 to 9.0, 98.5% phosphate removal and a maximum power density of 338.1 mW/m² were obtained at pH 6.0. Phosphate was mainly removed by aluminum hydroxide adsorption. These results demonstrate that the aluminum–air fuel cell can be applied as electricity-producing electrocoagulation equipment. Aluminum–air fuel cells provide an alternative method to meet the goal of carbon neutrality in wastewater treatment compared with traditional energy-consuming electrocoagulation systems.

Keywords: aluminum-air fuel cell; electrocoagulation; phosphate removal; electricity production

1. Introduction

The discharge of phosphorus into water bodies has caused a serious worldwide water eutrophication problem [1,2]. A concentration of 0.1–0.2 mg/L phosphate can induce incipient eutrophication in running water, while the critical concentration reduces to 0.005-0.01 mg/L of phosphate for still water [3]. The eutrophication of water threatens the health of aquatic creatures, livestock, and even humans. Therefore, the removal of phosphorus from municipal wastewater is essential to avoid water eutrophication. In China, the national limit for total phosphorus (TP) effluent of the Class 1A Discharge Standard is 0.5 mg/L [2]. The TP concentration is approximately 4–9 mg/L in China's municipal wastewater treatment plants [4]. Biological phosphorus removal processes are the conventional and most widely applied technology. However, the biological treatment system occupies a large area of land, requires high energy consumption, and needs to control the operation conditions to reach this standard [5]. For the high discharge standard of phosphate in municipal wastewater treatment, physicochemical treatments, such as chemical precipitation by adding aluminum or iron coagulants, are applied for the further removal of phosphate after biological treatment [6,7]. Chemical coagulation increases the operating costs, produces excessive amounts of chemical sludge, and leads to potential risks of secondary pollution.



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Electrocoagulation (EC) is considered a promising wastewater treatment technology that comprises three aspects: coagulation, flotation, and electrochemistry [8]. In the EC process, a sacrificial metal anode, such as Al or Fe, dissolves and releases metal ions into the solution with an impressed current [8]. Various metal hydroxides are formed by the hydrolysis process. Hydrogen bubbles and OH⁻ ions are discharged at the cathode. Pollutants and impurities are mainly removed by multiple physicochemical reactions with metal hydroxides and generate precipitates simultaneously [9,10]. EC is an effective method for phosphate removal, and the advantages of the EC system compared with chemical coagulation are its easy operation, no additional chemical requirements, and less sludge production [10-12]. However, the electrical energy demands need to be reduced to make the EC process economically viable and eco-friendly. Recently, a new type of EC process using an air cathode has been investigated to reduce EC energy demands [13–15]. At the air cathode, an oxygen reduction reaction proceeds with a related catalyst, and oxygen gas is obtained through air diffusion without aeration. With a minor added operation voltage of 0.5 V, 98% of phosphate was removed in a shortened time of 15 min [16]. The reason for this decrease in the electrical energy requirement of the EC with the air cathode is that the metal anode and the oxygen reduction air cathode exist in a potential gradient.

Metal-air fuel cells, which consist of a metal anode and an air cathode, have been investigated for synchronous contaminant removal and energy generation [17]. Over the last few decades, metal-air fuel cells have been implemented to remove arsenate, chromium, p-arsanilic acid, and humic acid, and for algal recovery [18–24]. Some researchers have also applied metal-air fuel cells for phosphate removal and recovery. When wastewater contained nitrogen and phosphorus together, Mg-air fuel cells were employed to recycle the nitrogen and phosphorus by forming struvite precipitation [25,26]. Iron-air fuel cells are a new option for recovering phosphate from wastewater. Phosphate was recovered by the formation of vivianite precipitation. Within 3 months of continuous operation, the iron-air fuel cell output voltage achieved approximately 400 mV, and 97% of phosphate was removed from synthetic industrial wastewater [27]. Different configurations of iron-air fuel cells were constructed to treat wastewater containing anaerobically digested sludge. The phosphate removal rate of a two-chamber fuel cell obtained a rate of 11.60 mg-P/L/h [28]. Aluminum is also commonly used as an electrode in electrocoagulation for phosphate removal, and aluminum-air fuel cells have been utilized to remove arsenate. Thus, it is important to research the performance of phosphate removal and the influence of the phosphate removal rate on aluminum-air fuel cells.

In this study, an energy self-sufficient single-chamber aluminum-air fuel electrocoagulation system was constructed for phosphate removal. Different electrolyte concentrations, initial pH values of the solution, and phosphate concentrations were investigated to determine their influences on phosphate removal and power generation. The coagulated precipitation in an aluminum-air fuel cell was also selected and analyzed to demonstrate the phosphate removal process in the aluminum-air fuel cell. The self-powered aluminum-air fuel cell provides an energy-friendly electrocoagulation process for nutrient contaminant removal, which is significant in wastewater treatment for achieving the goal of carbon neutrality.

2. Materials and Methods

2.1. Reactor Construction

An aluminum–air fuel cell reactor was constructed using a rectangular plexiglass reactor of 80 mL (length: 4 cm, width: 2 cm, height: 10 cm, Figure 1). The area of the air cathode was 40 cm² (4 cm \times 10 cm). The air cathode, including stainless steel mesh, activated carbon (Xinsen Carbon Co., Ltd., Nanping, Fujian, China), conductive carbon black (Jinqiushi Chemical Co. Ltd., Tianjin, China), and PTFE (60 wt%, Hesen Electrical Co., Ltd., Shanghai, China), was constructed using a rolling process, as described in a previous study [29]. For the diffusion layer, conductive carbon black and an appropriate amount of ethanol were mixed using mechanical and ultrasonic agitation for 15 min. The PTFE was

then added and the solution was further stirred for 15 min. The carbon black was kneaded like dough. Then, it was compacted and shaped between the rollers, and the diffusion layer was pressed to a thickness of 0.4 mm. Finally, the diffusion layer sheet and the stainless-steel mesh were rolled together and calcined in a muffle furnace at 340 °C for 20 min. Activated carbon powder was used as the catalyst to create the catalytic layer. Similar to the operation of the diffusion layer, a certain amount of activated carbon powder, ethanol, and PTFE were mixed and rolled to create a catalytic layer sheet with a thickness of 0.3 mm. The catalytic layer was then rolled together on the other side of the stainless-steel mesh. The anode was a piece of aluminum mesh of 40 cm² (mesh size 1.0 mm \times 1.5 mm), which was placed opposite to the cathode. Before operation, the aluminum mesh was washed with an acetone solution to remove the residual oil and stains on the anode surface. The anode was then immersed in 1.0 mol/L HCl solution for 10 min to remove the oxidation layer on the surface. The anode was used after it was washed with deionized water.



Figure 1. Schematic diagram (a) and photo (b) of the aluminum-air fuel cell for electrocoagulation.

2.2. Solutions and Experimental Procedure

The phosphate solution was dispensed with KH_2PO_4 (99.9%) and NaCl (99.5%), which were prepared to increase the solution's conductivity. The reactor was operated with a resistor of 10 Ω . The concentration of NaCl in the solution varied from 2 mmol/L to 10 mmol/L with 5 mg/L PO₄^{3–}-P to analyze the solution conductivity effect. The initial solution pH of 5.0–9.0 was adjusted by adding 1.0 mol/L HCl or 1.0 mol/L NaOH. The PO₄^{3–}-P concentrations ranged from 1 mg/L to 5 mg/L to compare the performances with different original PO₄^{3–}-P concentrations. Approximately 1.5 mL of solution was removed every 5 min for PO₄^{3–}-P analysis. The self-powered electrocoagulation experiments were conducted at room temperature.

2.3. Measurements and Calculations

The samples extracted at different times were tested after they were filtered through 0.45 µm pore diameter syringe filters. The phosphate concentration was measured using the ammonium molybdate spectrophotometric method (photoLab[®] 7600 UV, WTW, Munich, Germany) [30]. The pH was tested using a pH meter (pH 7110, WTW, Munich, Germany). The voltage across the resistor was monitored at 1 min intervals using a data acquisition system (PISO-813, ICPDAS Co., Ltd, Taiwan, China). The current density (I) was calculated based on Ohm's law (I = U/(RA)), and the power density (P) was calculated as P = IU/A, where U is the voltage (mV), R is the external resistor (Ω), and A is the surface area of the cathode. Polarization curves and electrode potential curves were obtained by recording the voltages under various external resistances (5–1000 Ω). A saturated calomel electrode (SCE, 0.242 V vs. a standard hydrogen electrode) was inserted into the reactor chamber as the reference electrode to test the electrode potential. The theoretical dissolved aluminum ion

concentration was calculated, and the number of aluminum ions was calculated according to Faraday's law. The equation for Faraday's law is as follows:

$$m = \frac{I \times A \times t \times M}{Z \times F}$$
(1)

Here, m is the theoretical dissolved mass of aluminum ions (g), I is the current density (A/cm²), t is the operation time (s), A is the surface area of the cathode (40 cm²), M is the molecular weight of the aluminum material (M = 26.98 g/mol), Z is the number of electrons released from the aluminum anode (the Z of aluminum is 3 eq/mol), and F is Faraday's constant (96,485 C/mol). The volume of the electrolyte solution used for the aluminum ion concentration calculation was V = 80 cm³. The P-precipitants obtained from the 10 mmol/L NaCl solution were collected and vacuum-dried at 40 °C for chemical phase analysis. The precipitates were tested by X-ray diffraction (D8 ADVANCE, Bruker, Karlsruhe, Germany) under operating conditions of 100 mA and 40 kV in the range of 20 = 10–90°. The aluminum anode and precipitates were analyzed using scanning electron microscopy (Carl Zeiss AG, Sigma500, Oberkochen, Germany) and energy-dispersive spectroscopy.

3. Results and Discussion

3.1. Effects of Electrolyte Concentration

The electrolyte concentration of NaCl directly influenced the resistance between the electrodes and changed the output current [30]. In the EC process, increasing the electrolyte concentration could increase the phosphate removal efficiency and reduce the electrolysis operation time. The aluminum–air fuel cell system's phosphate removal performance was monitored with different concentrations of NaCl ranging from 10 mmol/L to 2 mmol/L with a resistor of 10 Ω . With the decrease in the electrolyte concentration, the phosphate removal rate in the aluminum-air fuel cell showed a downward trend, and the required reaction time was prolonged. The phosphate removal in the aluminum-air fuel cell containing 8 mmol/L of NaCl was slightly lower than that of the cell containing 10 mmol/L of NaCl. When the NaCl concentration decreased to 2 mmol/L, the phosphate removal reached 97.8%, 93.3%, 91.8%, and 86.9% at 60 min, respectively (Figure 2a). White flocculent precipitates were observed with different operating concentrations of electrolytes. At 10 mmol/L of NaCl, the phosphate removal was 98.0% at 50 min, and the rate of removal decreased with longer operation times, which demonstrated that the operation time could be reduced with a higher electrolyte concentration. When the NaCl concentration was decreased from 10 mmol/L to 2 mmol/L, the average output current density of the aluminum-air fuel cell decreased from 0.20 mA/cm² to 0.06 mA/cm² (Figure 2b). The low conductivity of the solution led to an increase in the internal resistance of the fuel cell, slowing down the mass transfer in solution and the dissolution rate of the aluminum anode [24]. When the NaCl concentration declined from 10 mmol/L to 2 mmol/L, the calculated dissolutions of aluminum ions were 33.7 mg/L, 32.2 mg/L, 20.3 mg/L, and 9.5 mg/L, respectively. A higher conductivity electrolyte increased the anode dissolution and produced larger amounts of aluminum hydroxides for complex precipitation reactions in the solution; thus, a higher electrolyte concentration was favorable for phosphate removal [20]. In a traditional EC system, an increase in the electrolyte concentration can improve energy consumption, leading to an increase in the EC operating costs. On the contrary, a higher electrolyte concentration will generate more electricity for the aluminum-air fuel cell.



Figure 2. Effects of different electrolyte concentrations on (**a**) phosphate removal and (**b**) current generation in the aluminum–air fuel cell.

To analyze the influence of electrolyte concentrations on the electrochemical performance of the aluminum–air fuel cell, polarization curves and electrode potentials were tested with different electrolyte concentrations. As the electrolyte conductivity decreased, the maximum power density of the aluminum–air fuel cell reduced from 265.7 mW/m² to 76.6 mW/m² (Figure 3a). The electrode potentials of the aluminum anode and the air cathode both decreased with a decline in conductivity (Figure 3b). With a higher electrolyte concentration, more aluminum ions dissolved, generating a higher current output.



Figure 3. (a) Power density and polarization curves and (b) electrode potential curves for the aluminum–air fuel cell with different electrolyte concentrations.

3.2. Effects of Initial pH

The pH value can be a significant factor in the electrocoagulation process. In this work, the initial solution pH ranged from 5.0 to 9.0, the initial phosphate concentration was 5 mg/L, and the NaCl concentration was 10 mmol/L. As shown in Figure 4a, different initial pH values influenced the phosphate removal rate in the aluminum-air fuel cell system. When the solution pH was alkaline, the phosphate removal rate was faster than the rate under acidic conditions in the early stage. Within 50 min, all phosphate removal rates reached values higher than 90% with different initial pH values. At 60 min, as the initial pH 5.0 changed to pH 9.0, the phosphate removal reached 96.3%, 98.5%, 96.3%, 97.0%, and 95.6%. In contrast to the removal rate trend, the highest level of final phosphate removal was obtained at pH 6.0, which was likely due to the higher aluminum dissolution with a higher current output at pH 6.0 [31]. The average current density of the aluminum-air fuel cell ranged between 0.18 and 0.20 mA/cm², which changed slightly during electricity production. At pH values of 5.0 to 9.0, the calculated aluminum ion concentrations were 30.7 mg/L, 33.9 mg/L, 33.1 mg/L, 31.9 mg/L, and 32.2 mg/L. These results also indicated that the self-driven electrocoagulation could adapt to different pH conditions, as shown in previous research [20].



Figure 4. Effects of different initial pH values on (**a**) phosphate removal and (**b**) current generation in the aluminum—air fuel cell.

The power density of the aluminum–air fuel cell varied slightly under different pH conditions (Figure 5a,b). The maximum power densities of the aluminum–air fuel cell were higher at pH 6.0 and pH 7.0, being 338.1 mW/m² and 332.8 mW/m², respectively. Under alkaline conditions, the cell maximum power densities decreased; at pH 9.0, the maximum power density was 292.8 mW/m². The difference in the fuel cell electrode potential under different pH conditions was not obvious.



Figure 5. (a) Power density and polarization curves and (b) electrode potential curves for the aluminum–air fuel cell with different initial pH.

The electrocoagulation performance was influenced by the aluminum dissolution and the final pH. In this experiment, pH variation was also investigated with different initial pH values, as shown in Table 1. After 60 min of operation with the initial pH value ranging from 5.0 to 9.0, the final pH changed in different trends. For the acidic and neutral conditions, the pH value increased, while with initial pH values of 8.0 and 9.0, the final pH value decreased. The variation trends were similar to previously reported trends [20]. The dissolution rate of aluminum was also related to the current density and the pH value. The behaviors of the electrode and aluminum ions in solution also affected the solution pH. For the aluminum–air fuel cell, the electrode and overall reactions were as shown in Equations (2)–(4). At the aluminum anode, electrons were released from the aluminum, and then the aluminum ions reacted with water, as shown in Equation (5) [32,33]:

Anode : Al
$$\to$$
 Al³⁺ + 3e⁻E⁰ = 1.66 V (2)

Cathode :
$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2OE^0 = 1.23 V$$
 (3)

Overall :Al + O₂ + H⁺
$$\rightarrow$$
 Al³⁺ + H₂OE⁰ = 2.89 V (4)

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+$$
(5)

Initial pH	5.0	6.0	7.0	8.0	9.0
Final pH	5.6	6.4	7.3	7.9	8.8

Table 1. The pH value changes of the aluminum–air fuel cell before and after operation for 60 min.

At the air cathode, OH^- ions can also be generated through the four-electron oxygen reduction reaction with water, as shown in Equation (6):

$$O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \tag{6}$$

At pH 5–6, for most of the polymeric species, the ratios of precipitation of Al^{3+} and OH^- were less than 3, which caused the OH^- ions to accumulate in the solution during the operation process and increased the pH [33]. Under alkaline conditions, more OH^- ions could be consumed by aluminum ions to form polymeric aluminum hydroxide, and this process decreased the pH value.

3.3. Effects of Initial Phosphate Concentration

Generally, the removal efficiency of phosphate was negatively correlated with the initial phosphate concentration in the EC process, and the phosphate removal was limited at the start of the operation, as the amount of coagulants produced was relatively small [14, 34,35]. In this study, the phosphate concentration was set to approach the concentration of real domestic wastewater. A 10 mmol/L NaCl electrolyte and a 10 Ω operating condition were used, and the initial concentrations of phosphate in the simulated wastewater were set to 5 mg/L, 4 mg/L, 3 mg/L, 2 mg/L, and 1 mg/L, respectively. As the results in Section 3.1 show, the operation time could be reduced to 50 min with 10 mmol/L of NaCl. The maximum phosphate removal rate was achieved at 50 min, and the level of phosphate removal decreased after 60 min. At lower phosphate concentrations, a higher phosphate removal rate was obtained at the beginning of the operation. With the increase in the phosphate concentration, the phosphate removal rate increased at 50 min, achieving 92.2%, 93.0%, 92.6%, 96.9%, and 98.0%, respectively (Figure 6). In the actual sewage treatment process, the concentration of phosphate in sewage changes greatly. These results indicate that in a relatively low phosphate concentration range, the aluminum–air fuel cell system can achieve a high phosphate removal efficiency. Compared with the existing EC process, which consumes electricity, an electrical-power-generating electrocoagulation system was applied to this work, which provided an energy-friendly and promising electrocoagulation process for phosphate removal.



Figure 6. The relationship between the initial phosphate concentration and phosphate removal in the aluminum–air fuel cell.

3.4. Comparison of Energy Demand and Implications

In traditional EC systems for phosphorous removal using aluminum electrodes or a combination of aluminum and iron electrodes, the energy demand per cubic meter of P-wastewater was approximately 1.1–6.1 kWh, as shown in Table 2. When an air cathode was applied as the electrode, the energy consumption decreased compared with the traditional EC system. In the air cathode EC system which used a titanium inert electrode, only 0.009–0.06 kwh was added, and the operation time required was only 15 min [13]. Photovoltaic solar modules were also applied to power the EC process, and the energy from the solar energy modules was enough for the EC system, although the solar energy conversion efficiency was less than 13% [30]. For the aluminum-air fuel cell, with a 10 mmol/L NaCl electrolyte and a 10 Ω operation condition, the average power density at 60 min was approximately 160 mW/m². As the power density curves show, the aluminum-air fuel cell output power density was related to the output current. The power production was less than that of the solar cell; however, the aluminum-air fuel cell electrocoagulation system did not require an external electricity unit such as a solar cell module. This simple device is more practical in wastewater treatment, and solar energy is also highly dependent on weather. Compared with the traditional EC system, the operating cost for anode consumption is similar, but the cost of the air cathode in the aluminum-air fuel cell is higher than that of the conventional EC cathode [19]. The air cathode cost and lifetime should be one of the future research points considered for scale-up. Therefore, in future studies, the construction of a scale-up system, the electrode lifetime, and overall economic assessment including labor, sludge handling, maintenance, and depreciation costs will be key issues for practical application [18,36–38].

Table 2. Energy consumption of different electrocoagulation systems and the removal rates of phosphorus reported in different studies with Al electrodes.

Electrode Type	Wastewater Type	Energy (kWh/m ³)	Current Density (mA/cm ²)	Phosphate Removal (%)	Reference
Al–Al	urban wastewater	4.5	1	99	[39]
Al–Fe	synthetic solution	1.1-6.1	1–4	97–99	[3]
Al, air cathode	domestic wastewater	0.4–9.9	0.6-1.2	97–99	[14]
Al, air cathode, titanium sheet	domestic wastewater	0.009-0.06	1	71–98	[13]
Al, air cathode, graphite sheet,	domestic wastewater	0.05 - 0.14	1	85–98	[16]
Al–Al, photovoltaic solar	landscape water		1.3-3.8	96–99	[30]
Al-air cathode fuel cell	synthetic solution		0.06-0.2	86–98	This study

Another advantage of the metal–air fuel cell is that the air cathode is capable of generating hydrogen peroxide with a suitable oxygen reduction catalyst, which has the potential to treat wastewater by combining coagulation and oxidization processes. For domestic wastewater treatment, the aluminum–air fuel cell can be used at the pre-treatment segment and the advanced treatment segment to remove phosphate. Organic contaminants such as antibiotics, resistant genes, and persistent organic pollutants which are harmful to human health can coexist with phosphate in wastewater. The multiple physicochemical processes in aluminum–air fuel cells provide promising methods to treat the complex effluents in the advanced treatment stage.

3.5. Mechanisms of Phosphate Removal

The aluminum anode was analyzed before and after fuel cell operation using SEM to clarify the phosphate removal process (Figures 7 and 8). The anode surface formed a corrosion pit after the operation (Figure 7a,b), providing evidence that the anode spontaneously dissolved aluminum ions into the solution. Previous studies have shown that pitting corrosion exists on metal surfaces when they are covered by a compact oxide film [40]. Furthermore, chloride ions in the electrolyte solution are the aggressive ions on the metal surface. Chloride ions can trigger the pitting corrosion reaction, and the passivation layer on the electrode surface will decompose. For the Al anode, a corrosion-protective film consisting of aluminum oxide can easily form on the surface, and the protective film is stable within a pH range of 4–8.5. However, chloride ions can react with the protective film

and expose the anode surface, which will increase the anode dissolution rate [34]. These processes are shown in Equations (7)–(9) [41]:

$$Al_2O_3 + Cl^- + 6H^+ \rightarrow 2AlCl_3 + 3H_2O$$
 (7)

$$Al(OH)_3 + nCl^- \rightarrow Al(OH)_{3-n}Cl_n + nOH^-, n = 1, 2, 3$$
 (8)

$$AlCl_3 + Cl^- \to AlCl_4^+ \tag{9}$$



Figure 7. SEM results of the Al electrode in this study, (**a**) the original electrode, and (**b**) the electrode after the operation.



Figure 8. SEM images of phosphate precipitation in aluminum-air batteries (a) and EDS results (b).

The morphology of the precipitates from 10 mmol/L of NaCl and 5 mg/L of phosphate was observed by SEM, and the elemental compositions were analyzed by EDS, as shown in Figure 8a,b. The precipitates had a blocky structure, and the EDS results showed that the precipitates were mainly composed of P, Al, and O elements, which indicated that the phosphate was removed with the aluminum compounds.

There are two main mechanisms that exist in the aluminum coagulation phosphate removal process, namely co-precipitation by monomeric aluminum and adsorption by aluminum hydroxide [42]. In the EC process with aluminum electrodes, both direct precipitation and adsorption by aluminum hydroxide were observed, and direct precipitation for phosphate removal was more efficient than hydroxide adsorption removal [43,44]. The phosphate removal behavior was related to the water conditions and the aluminum species. AlPO₄ precipitate can be formed by Al³⁺ and phosphate, and the Al₁₃ species can also induce formation of AlPO₄ by complex adsorption [45]. For the coagulation process, pH influences the speciation of aluminum and accordingly affects the phosphorus removal mechanism. When the pH value was below 3.5, the dominant aluminum species was Al³⁺. As the pH value increased from 3.5 to 10.0, the dominant species were complex monomeric and polymeric aluminum hydroxides. Al(OH)₄⁻ became the predominant species when the pH value was above 10.0 [46]. The precipitated products from different initial pH values

were examined by XRD to reveal the crystalline structure (Figure 9). Three diffraction peaks were observed in the flocs. The second peak was in the range of 23–33°, and the third peak was at approximately 41°, which was likely within the spectrum of Al(OH)₃. The crystal plane of AlPO₄ was also found in the second peak range [45–47]. However, the absence of sharp peaks in the XRD results indicated that the flocs had a poor crystalline structure, and that many of the precipitates were in an amorphous state, which was similar to a previous report on aluminum coagulants [47]. Combining the precipitates, XRD results, and the aluminum species constructions with different pH values, it can be concluded aluminum hydroxide adsorption was the main mechanism for removing phosphate. A schematic diagram of the phosphate removal process is summarized in Figure 10.



Figure 9. XRD patterns of floc precipitation in the aluminum-air fuel cell.



Figure 10. Schematic diagram of the phosphate removal process in the aluminum-air fuel cell.

4. Conclusions

A single-chamber aluminum–air fuel cell system was utilized to remove phosphate, and the effects of various factors were examined for phosphate removal. The phosphate removal mechanism was also discussed. An increase in the electrolyte concentration improved the level of phosphate removal and electricity production. With 10 mmol/L of NaCl, 97.8% phosphate removal was achieved, and the maximum power density reached 265.7 mW/m². For different initial pH values, the optimal condition for electricity production was pH 6.0, and 98.5% phosphate removal was achieved in 60 min. The average current density obtained was 0.20 mA/cm², and the maximum power density was 338.1 mW/m². Aluminum hydroxides were synthesized, and phosphate was primarily removed via the aluminum hydroxide adsorption process. The aluminum fuel cell can treat phosphate wastewater and generate energy simultaneously. Compared with conventional

electrocoagulation systems, the aluminum–air fuel cell system can be considered an energy self-sufficient alternative, which is significant for achieving carbon neutrality in the wastewater treatment industry.

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