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Can electrocoagulation process be an appropriate technology for phosphorus removal from municipal wastewater?

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HIGHLIGHTS

GRAPHICAL ABSTRACT

- New configuration of electrocoagulation (EC) was developed for better TP removal.
- The influenced factors on phosphorus removal in the EC process were investigated.
- EC with higher voltage, initial conductivity and longer electrolysis time is efficient.
- Specific rates of energy and iron consumptions during the EC process were measured.
- Passivation layer formed on the surface of electrodes during operation were overcome.

article info abstract

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This paper evaluated a novel pilot scale electrocoagulation (EC) system for improving total phosphorus (TP) removal from municipal wastewater. This EC system was operated in continuous and batch operating mode under differing conditions (e.g. flow rate, initial concentration, electrolysis time, conductivity, voltage) to evaluate correlative phosphorus and electrical energy consumption. The results demonstrated that the EC system could effectively remove phosphorus to meet current stringent discharge standards of less than 0.2 mg/L within 2 to 5 min. This target was achieved in all ranges of initial TP concentrations studied. It was also found that an increase in conductivity of solution, voltages, or electrolysis time, correlated with improved TP removal efficiency and reduced specific energy consumption. Based on these results, some key economic considerations, such as operating costs, cost-effectiveness, product manufacturing feasibility, facility design and retrofitting, and program implementation are also discussed. This EC process can conclusively be highly efficient in a relatively simple, easily managed, and cost-effective for wastewater treatment system.

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1. Introduction

Phosphorus and nitrogen pollution now pose a serious problem in wastewater being generated today all over the globe and has become a serious issue. Both pollutants have proven to be difficult and expensive to mitigate in the waste stream. Consider phosphorus removal for example: to eliminate phosphorus in municipal wastewater using conventional biological treatment systems is difficult due to inherent limitations of the activated sludge method of mitigation, which is inherently less effective. This treatment method still allows in a residual phosphorus concentration in the effluent which exceeds the wastewater discharge guideline. In a typical biological treatment plant, phosphorus is passed through to the sludge phase, and then removed depending on the amount of excess sludge. Thus, its removal is limited and variable, resulting in low removal efficiency even less than 30% ([Hosni](#page--1-0) [et al., 2007; Sommariva et al., 1997\)](#page--1-0). In this circumstance, addition of chemicals to the process can reduce phosphate levels somewhat, the lack of an efficient process means that any remaining phosphate must be removed via other techniques (Bektaş [et al., 2004; Zeng, 2012](#page--1-0)) or via recombinant chemical dosing. The latter is time consuming, labor intensive, and expensive.

Sources of phosphorus emissions into the environment are mostly effluents from non-point agricultural runoff, and point sources from industrial plants, and municipal wastewater treatment, etc. ([Krishnan](#page--1-0) [Rajeshwar, 1997\)](#page--1-0). Wastewaters containing phosphorus are discharged into receiving waters in excess of the threshold-accepting environmental optimums, which contributes to the acceleration of the eutrophication process in a rapid and unnatural way, overwhelming the natural processes that would otherwise normalize the contaminant load. Normally, phosphorus exists in three main forms in wastewater, namely orthophosphate, polyphosphate, and organic phosphorus. However, the primary phosphorus compounds in wastewaters are generally orthophosphates ([Tran et al., 2012\)](#page--1-0), except in the special case of wastewater.

So far, most large scale biological wastewater treatment systems can meet applicable water quality discharge standards for TN and TP of less than 20 mg TN/L and 2 mgTP/L, respectively. However, only recently have governments realized the catastrophic environmental implications (e.g. eutrophication, red tide) of treated wastewater containing residual phosphorus concentrations above the recommended limits [\(Bui and Yoon, 2011](#page--1-0)). Therefore, governments have urged wastewater treatment plant operators to improve existing treatment systems and encouraged technological innovation to achieve better removal efficiencies, so that the new wastewater discharge standards can be met. Many countries have already implemented new standards for discharge, for instance, 0.5–1.0 mg/L in the USA, 1–2 mg/L in France ([Attour et al.,](#page--1-0) [2014\)](#page--1-0) and less than 0.2 mg/L in South Korea. Even though conventional biological treatment processes are economical, they have long hydraulic retention time and large volume requirements which reduce efficiency. In addition, exponentially greater land and facility requirements sometimes make these technologies less attractive than physicochemical treatments, which can provide for shorter retention time, greater efficiency, easy installation, and simpler operation and maintenance [\(Tran et al., 2012; Asselin et al., 2008; Nguyen et al., 2013; Gatsios](#page--1-0) [et al., 2015\)](#page--1-0). Therefore, there is a need for innovation and applied advanced technology to improve efficiency in phosphorus removal, which requires less space, less capital investment, easier installation, simple equipment, lower operating and maintenance costs, and elimination of additional frequent chemical use (Bektaş [et al., 2004; Markus](#page--1-0) [et al., 2011; Wahab et al., 2011; Oleszkiewicz and Barnard, 2006](#page--1-0)).

Electrocoagulation (EC) is an electrochemical technology that treats drinking water and wastewater which offers many advantages over traditional physicochemical treatments, such as higher efficiency, short retention time, elimination of need to neutralize excess chemicals, reduction of sludge production, and prevention of secondary pollution caused by added chemicals or unintended chemical reactions. Electrodes and equipment also require less space and are simple to operate

[\(Asselin et al., 2008; Linares-Hernández et al., 2009; Hernández-Ortega](#page--1-0) [et al., 2010; Nguyen et al., 2014](#page--1-0)).

EC uses an electrochemical cell with a Direct Current (DC) voltage applied, usually to iron or aluminum electrodes, with water or wastewater as the electrolyte. During EC treatment of wastewaters, several electrochemical, physicochemical and chemical processes take place in the electrolysis device. This process produces the metal ions generation that takes place at the anode, and then creates both iron hydroxides and poly-hydroxides that have significant sorption ability. Meanwhile, hydrogen gas is continuously generated from the cathode, and with the added help of scouring and floating, the pollutants and flocculated particles are formed on the electrodes' surface. Then they flow out of the EC device [\(Krishnan Rajeshwar, 1997; Tran et al., 2012](#page--1-0)).

For a long time, electrochemical technologies in water and wastewater treatment were not widely implemented due to a lack of capital and expensive operating costs. Nowadays, electrocoagulation technologies have advanced to such a stage that they are not only comparable with other technologies in terms of cost, they are also more efficient and more compact ([Bennett et al., 2007; Chen, 2004; Wang et al., 2004;](#page--1-0) [Sadeddin et al., 2011](#page--1-0)).

When iron is used as electrode material, the dominant reactions involved can be summarized as follows [\(Linares-Hernández et al., 2009;](#page--1-0) [Bensadok et al., 2011; Christos Comninellis, 2010; Bernal-Martínez](#page--1-0) [et al., 2013; Linares-Hernández et al., 2010; H.A. Moreno et al., 2009\)](#page--1-0): At the cathode:

$$
2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH_{(eq)}^- \tag{1}
$$

At the anode:

$$
Fe_{(s)} \rightarrow Fe_{(eq)}^{2+} + 2e^- \tag{2}
$$

$$
\text{Fe}_{(s)} \to \text{Fe}_{(eq)}^{3+} + 3\text{e}^{-}. \tag{3}
$$

In the bulk solution, multi-core coordination compounds and $Fe₃(PO₄)₂$, Fe(OH)₂, Fe(OH)₃ precipitates can be formed, with a simplified reactions as follows:

$$
Fe^{2+}_{(eq)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + 2H^+_{(eq)} \tag{4}
$$

$$
4Fe_{(eq)}^{2+} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H_{(eq)}^{+} \tag{5}
$$

$$
3Fe_{(eq)}^{2+} + 2PO_4^{3-} \to Fe_3(PO_4)_{2(s)} \tag{6}
$$

$$
Fe^{3+}_{(eq)} + PO^{3-}_{4\ (eq)} \rightarrow FePO_{4(s)}.\tag{7}
$$

The EC technology used in this study is regarded as a new, urgently needed solution that overcomes the disadvantages of existing EC technologies, for instance, the new EC process can self-cleaning electrode surfaces, enhanced interactivity and reactivity between coagulants (complexes, iron hydroxides, etc.) and contaminants (soluble, colloidal, suspended solid, etc.) in the wastewater, uniform corrosion over the surface of electrodes, short electrolysis times, and lower operating costs.

The main aim of this research is to investigate the performance of the EC process using iron cylindrical electrodes in continuous and batch operating mode for removing phosphorus in municipal wastewater under various experimental conditions. The system was optimized in terms of TP initial concentrations, different voltages, conductivities and electrolysis time, reaction rate constant for TP removal, and sludge production. In addition, the effect of some operating variables such as pH and NaCl concentrations added to wastewater on phosphorus removal of EC process were also evaluated.

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