

# Phosphorus removal from domestic wastewater in electrocoagulation reactor using aluminium and iron plate hybrid anodes



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## ABSTRACT

The aim of this study was to investigate the effects of initial pH ( $pH_i = 4.0\text{--}7.0$ ), current density ( $j = 10\text{--}40\text{ A/m}^2$ ), initial phosphorus (P) concentration ( $C_i$  of  $5.01\text{--}52.13\text{ mg/L}$ ) and electrocoagulation (EC) time ( $t_{EC} = 10\text{--}100\text{ min}$ ) on phosphorus removal from domestic wastewater by a batch EC reactor using hybrid aluminium (Al)-iron (Fe) anodes and titanium cathode. Phosphorus removal from domestic wastewater containing  $52.13\text{ mg/L PO}_4\text{-P}$  was obtained to be  $99.99\%$  at optimum conditions ( $pH_i = 4$ ,  $j = 20\text{ A/m}^2$  and  $EC\text{ time} = 80\text{ min}$ ). The energy and electrode consumptions at optimum conditions were calculated as  $3.422\text{ kWh/m}^3$  and  $0.328\text{ kg/m}^3$ , respectively. The amount of removed P per electrochemically dissolved total metal electrode ( $q_e$ ) was calculated as  $55.69\text{ mg P/g}$ , while the dissolved metal to removed phosphorus ratio ( $M_e/P$ , mol/mol) was  $4.40$  ( $Fe/P = 2.63$  and  $Al/P = 1.77$ ) at optimum conditions. It can be concluded that phosphorus removal by hybrid Al-Fe anodes is as effective as using only Fe or Al anodes as per the results present in literature. In addition, the effluent pH after EC treatment process at optimum conditions was  $8.8$ , hence requiring no pH adjustment before discharge.

## 1. Introduction

The release of phosphorus from wastewaters into watercourses can cause severe pollution problems, such as eutrophication (Roy, 2017; Park et al., 2016; Ramasahayam et al., 2014). Problems associated with eutrophication include profuse algal blooms, excessive growth of nuisance aquatic plants, negative aesthetic aspects, deoxygenation, and problems relating to water purification for potable use (Dunne et al., 2015; de Haas et al., 2000). According to the European Environment Agency, the highest phosphorus concentration in European rivers is  $0.91\text{ mg/L}$ , and this value was reported in Turkey (EEA, 2015). The critical concentrations for incipient eutrophication are about  $0.1\text{--}0.2\text{ mg/L PO}_4\text{-P}$  in running water and  $0.005\text{--}0.01\text{ mg/L PO}_4\text{-P}$  in still water. In view of the potential hazard to surface waters, EU Directive 91/271/EEC specifies limit values for discharge of phosphate compounds into receiving waters. Depending on the size of the sewage treatment plant, these values are  $2\text{ mg/L P}_{tot}$  for  $10,000\text{--}100,000\text{ PE}$  (population equivalents) or  $1\text{ mg/L P}_{tot}$  for  $> 100,000\text{ PE}$  (EEC, 1998).

The need for efficient removal of P from wastewaters before discharge into watercourses will help reduce eutrophication of receiving surface water bodies. Various technologies for phosphate removal from wastewater have been investigated, these include; adsorption (Paradelo

et al., 2016; Karageorgiou et al., 2006), biological processes (Zhang et al., 2013; Oehmen et al., 2007), chemical precipitation by adding aluminium, iron or calcium salts (Park et al., 2016; Yang et al., 2010; Georgantas and Grigoropoulou, 2007; Fytianos et al., 1998), ion-exchange (Choi et al., 2011) and membrane processes (Ensano et al., 2016; Wei et al., 2009). Chemical precipitation and adsorption are currently the most effective and well-established methods. For low concentrations, biological treatment can remove up to  $97\%$  of the total phosphorus at a low-cost, but the variability in chemical composition, high phosphorus concentration and temperature of wastewater would make the implementation of this process impracticable for wastewater treatment. Furthermore, many of the above methods are disadvantaged by long operation time, removal inefficiencies and are expensive. Therefore, electrocoagulation (EC) for phosphorous removal has been attempted as an alternative process (to especially chemical precipitation) to overcome these disadvantages (Wysocka and Sokolowska, 2016). Moreover, this technique has advantages such as simple equipment, easy to operate, less retention time, reduction or absence of adding chemicals, rapid sedimentation of the electro-generated flocs, less sludge production and environmental compatibility, when compared to conventional methods (Can et al., 2003; Chen et al., 2000). However, in any EC process, the electrode type is regarded as a

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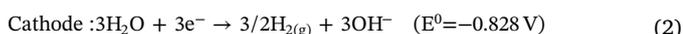
E-mail address: [omwene@gtu.edu.tr](mailto:omwene@gtu.edu.tr) (P.I. Omwene).

significant factor affecting the process performance. Aluminium and iron are the most common sacrificial electrodes used in the EC process for phosphorus removal, and up to 51–100% removals have been reported in the literature (Nguyen et al., 2016; Inan and Alaydin, 2014; Attour et al., 2014; Bouamra et al., 2012; Gharibi et al., 2010; Irdemez et al., 2006a, 2006b; Vasudevan et al., 2008, 2009; Zheng et al., 2009). Aluminium plate anode has been well documented to demonstrate higher performance compared to iron plate anodes. This is ascribed the lower co-precipitation and adsorption capacity of hydrous ferric oxides for phosphate ions compared to hydrous aluminium oxide (Behbahani et al., 2011; Irdemez et al., 2006b; Bektas et al., 2004). Phosphorus removal studies by EC process have investigated the influence of the operational parameters such as initial pH, type of supporting electrolyte and its concentration, and current density. However, just as for the case of chemical coagulation, phosphate removal efficiency is greatly affected by the molar ratios of dissolved metal coagulant (Fe or Al) to the removed phosphorus during EC (Stafford et al., 2014). In addition, appropriate selection of the electrode pair (Al and Fe hybrid electrodes) at different operating parameters (i.e., initial pH, current density, reaction time and initial phosphorus concentration) is important. Investigations on phosphorus removal using hybrid electrode in view of the present literature is still inadequate (Kuokkanen et al., 2015; Chen et al., 2014; Gao et al., 2012). Most of the existing studies focused on analyses of metal to phosphorous ( $M_e/P$ ) ratio of a single electrode. In the present study the  $M_e/P$  ratio is provided as combination of Al and Fe hybrid electrodes and the results discussed in relation to those available in the literature. In this regard, we present phosphorous removal from synthetic domestic wastewater using hybrid Al and Fe plate electrodes in a batch EC reactor. To optimize the conditions, different parameters like effect of the initial pH, effect of current density, and effect of initial phosphate concentration were studied. We hope that the findings of this study shall be of interest to many researchers involved in the development of electrochemical treatment technologies for domestic wastewater.

## 2. Mechanism of phosphate removal by EC

EC involves the generation of coagulants in situ by electrochemically dissolving either aluminium or iron ions from the respective electrodes. The metal ions generation takes place at the anode, whereas hydrogen gas is simultaneously released from the cathode. The generated hydrogen gas also helps in floatation of the flocculated particles out of the water (Moussa et al., 2017; Can et al., 2003).

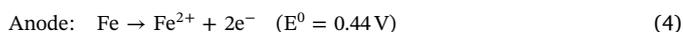
Anode and cathodes reactions for Al electrodes:



In the case of Al electrodes,  $\text{Al}^{3+}$  and ions generated by anode and cathode reactions (1) and (2) form various monomeric and polymeric species, which transform finally into  $\text{Al}(\text{OH})_{3(\text{s})}$  according to complex precipitation kinetics. At 25 °C, the solubility product of  $\text{Al}(\text{OH})_{3(\text{s})}$  is  $5 \times 10^{-33}$  (mol/L).



Anode and cathodes reactions for Fe electrodes:



Ferric ions ( $\text{Fe}^{3+}$ ) generated by electrochemical oxidation of Fe electrodes used in EC process may form monomeric and polymeric species depending on the pH of the aqueous medium, and which transform finally into  $\text{Fe}(\text{OH})_{3(\text{s})}$ . At 25 °C, the solubility product of  $\text{Fe}(\text{OH})_{3(\text{s})}$  is  $4 \times 10^{-38}$  (mol/L).

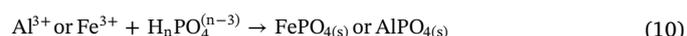


The rate of the oxidation of  $\text{Fe}^{2+}$  depends on the availability of dissolved oxygen (Chen et al., 2000).



$\text{Al}^{3+}$ ,  $\text{Fe}^{3+}$  and  $\text{Fe}^{2+}$  ions also react with hydroxide to form amorphous aluminium and iron hydroxide flocs. Consequently, the removal mechanism of pollutants from wastewaters with both electrodes is related to forming of  $\text{Fe}(\text{OH})_{3(\text{s})}$ ,  $\text{Al}(\text{OH})_{3(\text{s})}$ , monomeric and polymeric iron and aluminium species due to coagulation, precipitation, co-precipitation, and electro-oxidation (Chen et al., 2000). Furthermore, freshly formed amorphous  $\text{Al}(\text{OH})_{3(\text{s})}$  and  $\text{Fe}(\text{OH})_{3(\text{s})}$  “sweep flocs” in the EC process have large surface areas which are beneficial for a rapid adsorption of soluble organic compounds such as phosphate ions and trapping of colloidal particles.

Phosphate ion has strong affinity for metal ions like  $\text{Al}^{3+}$  and  $\text{Fe}^{3+}$ . Oxides of Al and Fe in aqueous medium will have surface OH groups. Phosphate ions in solution undergo ligand exchange with the  $\text{OH}^-$ . Adsorption of phosphate ions on aluminium and iron oxide surfaces leads to inner surface complexation. When  $\text{H}_2\text{O}$  molecules are present between the adsorbed phosphate ions and oxide surface, it's referred as outer surface complexation. The electrolysis of the electrode produces not only precipitates such as ferrous, ferric, and aluminium phosphate or hydroxyl-phosphate:  $\text{Al}_{1.4}\text{PO}_4(\text{OH})_{1.2(\text{s})}$ ,  $\text{Fe}_{1.6}\text{H}_2\text{PO}_4(\text{OH})_{3.8(\text{s})}$ , and  $\text{Fe}_{2.5}\text{PO}_4(\text{OH})_{4.5(\text{s})}$ , as shown in Equations (9), (10), and (11), but also hydroxides such as  $\text{Fe}(\text{OH})_{2(\text{s})}$ ,  $\text{Fe}(\text{OH})_{3(\text{s})}$ ,  $\text{FeOOH}(\text{s})$  and  $\text{Al}(\text{OH})_{3(\text{s})}$  (Zheng et al., 2009; Karageorgiou et al., 2006; Fytianos et al., 1998). It was found that when iron and aluminium are present in the water,  $\text{FePO}_4(\text{s})$  and  $\text{AlPO}_4(\text{s})$  forms at low pH range (< 6.5) and at higher pH range (> 6.5) iron and aluminium increasingly convert to oxides and hydroxides.  $\text{FePO}_4(\text{s})$  has minimum solubility within pH range of 4.5–5.5, but solubility of the  $\text{FePO}_4(\text{s})$  increases with increasing pH (Nguyen et al., 2016; Zhang et al., 2010; Irdemez et al., 2006a). Moreover, the optimum pH for phosphate precipitation with ferric iron is the range of pH 4.0 to 5.0, while that for ferrous iron is close to pH 8.0.



In general, two major mechanisms have been linked to the coagulation of  $\text{PO}_4^{3-}$  with Al/Fe(III) salts; (i) Formation of Al/Fe-hydroxyl-phosphate complexes;  $\text{Me}(\text{OH})_{3-x}(\text{PO}_4)_x(\text{s})$ . These complexes either adsorb onto positively charged Al/Fe(III) hydrolysis species or act as centres of precipitation for Al/Fe(III) hydrolysis products. (ii) Adsorption of  $\text{PO}_4^{3-}$  ions on to the Al/Fe(III) hydrolysis species. Such hydroxides are partially transformed into hydroxyl-complexes depending on the pH of the solution, but these can remove phosphate by adsorption (Nguyen et al., 2016; Lacasa et al., 2011; Zhang et al., 2010; Szabo et al., 2008; Thistleton et al., 2002; Jiang and Graham, 1998).  $\text{FePO}_4(\text{s})$  (strengite),  $\text{AlPO}_4(\text{s})$  (variscite) and  $[\text{Al}_6(\text{OH}_{15})]\text{PO}_4(\text{s})$  are the stable solid phases if phosphate is precipitated in the pH range of 5.0–7.0 (Ensano et al., 2016). Minimum solubility of  $\text{AlPO}_4(\text{s})$  occurs at pH 6.0 which is one unit higher than that of  $\text{FePO}_4(\text{s})$ . The stoichiometric molar mass ratios ( $r$ ) of Fe/P for  $\text{FePO}_4(\text{s})$  and  $\text{Fe}_3(\text{PO}_4)_2(\text{s})$  are 1.8/1 and 2.7/1, respectively. The solubility products of some precipitant phosphate compounds such as  $\text{AlPO}_4(\text{s})$ ,  $\text{Al}_{1.4}\text{PO}_4(\text{OH})_{1.2(\text{s})}$ ,  $\text{FePO}_4(\text{s})$  (for  $r = 1$ ),  $\text{Fe}_3(\text{PO}_4)_2(\text{s})$ , and  $\text{Fe}_{2.5}\text{PO}_4(\text{OH})_{4.5(\text{s})}$  (for  $r = 2.5$ ) at the  $\text{pH} > 9$  at 25 °C are  $6.3 \times 10^{-19}$ ,  $1.3 \times 10^{-22}$ ,  $1.3 \times 10^{-30}$ , and  $1 \times 10^{-97}$ , respectively (Zheng et al., 2009; Fytianos et al., 1998). Iron salt in either ferrous or ferric form is commonly used in phosphorus removal from municipal wastewater, and the influent with total

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