



Phosphorus removal from wastewater: energy demand in electrochemical process

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ABSTRACT

The electrochemical process (EP) as an advanced treatment method has been widely applied to the removal of pollutants from wastewater streams. Several studies have investigated the efficiency and optimization of the electrochemical method for removing phosphorus compounds from aqueous solutions. However, the energy demand for this method has only been reported in limited studies. Therefore, this research aimed to determine the energy consumed during an electrochemical process to remove organic phosphorus from wastewater. This experimental bench-scale study was carried out in a batch system on 180 samples of phosphorus contaminated wastewater. In order to prepare synthetic wastewater samples, containing 2–32 mg/L organic phosphorus, adenosine monophosphate salt was added to the dechlorinated tap water. The wastewater samples were then subjected to treatment in an electrolytic cell which consisted of steel electrodes. Electricity power was immobile at 0.6 A DC (1.15 mA/cm²). The remaining phosphorus was examined by 4500P.D method described in 21st Edition of Standard Methods for Examination of Water and Wastewater. A mixed design analysis of variance test with repeated measurements by SPSS16 software was applied for data analyzes. The results showed that, the EP using steel electrodes is capable of removing organic phosphorus up to 98%. For the diminution of phosphorus from samples to less than 1 mg/L, the mean of maximum energy demands was about 0.59 kWh/g of removed organic phosphorus. The energy demand can be calculated by the suggested graphs depending on the initial concentration of organic phosphorus.

Keywords: Energy; Electrochemical process; Organic phosphorus

1. Introduction

Excess phosphorus concentrations in effluents from municipal and industrial wastewater treatment facilities result in eutrophication, a phenomenon that upsets the balance of living organisms and affects water quality, mostly

owing to the loss of dissolved oxygen caused by algal decomposition in many natural water bodies [1–3].

Total phosphorus in wastewater is classified into two operationally distinct fractions based on colorimetric detection techniques [4]. These fractions are referred to as reactive (inorganic fraction) and non-reactive phosphorus (condensed phosphates and organic phosphorus) [1].

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Although organic phosphorus has been reported as the minor component of total phosphorus in raw wastewater, a large portion can pass through conventional treatment methods unaffected [5]. The majority of total phosphorus in treated wastewater has been found to be in the form of dissolved organic phosphorus, which may threaten sensitive receiving waters [2,5].

Municipal wastewater contains 5–20 mg/L total phosphorus, of which 1–5 mg/L of which is organic, and derived from human excretions and detergents used. In addition to municipal wastewater, industrial wastewater may also contain phosphorus compounds [6]. It has been documented that agriculture and animal husbandry waste streams lead to the accumulation of phosphorus in soil and water bodies [7], particularly in regions where these activities are prevalent. Moreover, the increasing demand for organic phosphorus compounds in industries like pesticides, detergents, and pharmaceuticals would result in a high concentration of these compounds in the effluent [2]. Effective and efficient technologies for organic phosphorus removal from both municipal and industrial wastewater streams are required.

The electrochemical process (EP) as an advanced wastewater treatment method has been widely applied to the treatment of organic and inorganic pollutants [8,9]. The major EP in wastewater treatment are electrocoagulation, electro flotation, electro-reduction, and electro-oxidation [10].

Ease of use, simple equipment, shorter hydraulic retention time, no need or reduction of chemical feeder equipment, less sludge volume, environmental compatibility, adaptability, energy savings, more safety, compatibility with fluctuations, no corrosion and environmental pollution, and relatively low costs are among the advantages of the electrochemical method [10–12].

Using the EP, phosphorus compounds (depending on the medium composition and pH) could be eliminated by different processes, including coagulation, complexation, adsorption on metal hydroxide surfaces, or direct precipitation with metal cations discharged into the water [8,10]. The combination of many processes in the electrochemical method results in a high efficiency of the method at wastewater treatment [13]. However, energy consumption for removal of pollutants from water and wastewater is a major issue that should be considered in the EP. Although several studies have investigated the efficiency and optimization of the EP in removing phosphorus compounds from aqueous solutions [3,8,9,12,14–17], the energy demand has only been reported in limited studies [3,9,15,16]. Irdemez et al. [16] studied the effects of current density and concentration of phosphate on phosphorus removal from wastewater using an electro-coagulation process. The phosphorus removal efficiency in the reaction time of 8 min for the initial concentrations of 25, 50, 100, and 150 mg/L of phosphorus has been reported to be 100%, 98%, 61%, and 42%, respectively. Furthermore, the amounts of energy consumed for the aforementioned time and concentrations have been reported at 0.81, 0.506, 0.444, and 0.323 kWh/g, respectively. In another study, Irdemez et al. [9] investigated the effect of pH on phosphate removal from wastewater by an electro-coagulation process using iron electrodes. They found that in the reaction time of 20 min, in a solution with a pH

of 3, the phosphate removal efficiency and energy consumption were 40% and 0.5 kWh/g, respectively. When the pH was raised to 7, these parameters changed to 93% and 0.64 kWh/g, respectively. While the efficiency and energy consumption for a pH of 9 were reported to be 61% and 0.56 kWh/g, respectively.

Since there are some contradictions in energy consumption in the removal of phosphorus by the EP, the present study was conducted with the specific aim of investigating the energy needs of the EP for the removal of organic phosphorus from artificial wastewater.

2. Materials and methods

In the present study, organic phosphorus was removed from synthetic wastewater using an electrolytic cell in a bench-scale batch system and the energy demand was estimated. In the first step, the characteristics of the tap water that was used further for the preparation of synthetic wastewater were determined as follows:

Electrical conductivity (EC) = 2,070 micro-Siemens per centimeter, pH = 7.2, total dissolved solids (TDS) = 1,242 mg/L, temperature = 21°C, total hardness = 516 mg/L as CaCO₃, calcium hardness = 318 mg/L as CaCO₃. Also, the measured phosphate concentration was 0.049 mg/L.

In the second step, the phosphorus-contaminated wastewater was synthesized by adding adenosine monophosphate salt to the dechlorinated tap water. The initial pH of solution was adjusted using HCl and NaOH (1 N, 5 N) before experiments. For preparing a complete mixed solution a magnetic stirrer was used. In this study, we prepared six different concentrations of phosphorus (2, 4, 8, 16, 24, and 32 mg/L).

The electrolytic cell was composed of eight pieces of steel plate with dimensions of 15 cm × 2.5 cm × 0.02 cm (length × width × thickness) as anode and cathode which were fixed 1.5 cm apart from each other. Prior to the initiation of the experiments, electrodes were washed by hydrochloric acid (1:6). Then, 2,000 mL of the synthetic wastewater sample was poured into the electrolytic cell. The electrodes were submerged into the sample to the extent of 13 cm of their length. The contents of the electrolytic cell were mixed at 300 rpm using a magnetic stirrer. It should be noted that the experiments were repeated five times for each of the aforementioned concentrations of phosphorus. The input power was measured using an ammeter and a voltmeter. All experiments were conducted at a laboratory temperature of about 21°C. The current intensity of 0.6 A was applied to the reactor contents, which was selected based on the optimal results of the previous studies conducted by the authors [18,19]. Progress of phosphorus removal from wastewater was monitored by taking samples from the reactor at 10 min intervals during 1 h. Therefore, for each concentration, the efficiency of phosphorus removal was investigated at 10, 20, 30, 40, 50, 60 min from the beginning of the experiments. A total of 180 samples were analyzed (6 concentrations, 6 reaction times, and 5 replications for each). Samples were filtered through cellulose acetate membrane filters with a pore diameter of 0.45 μm to separate unwanted sludge and then analyzed. To prevent passivation of electrodes,

cathode and anode were changed every 10 min during experiments.

Subsequently, the residual phosphorus in the taken samples was changed into orthophosphate using the persulfate digestion method (Standard Methods for Examination of Water and Wastewater 4500-P.B.5) and the concentration of formed orthophosphate was determined using the tin chloride method (Standard Methods for Examination of Water and Wastewater 4500 P.D.) [4]. The concentration of remaining phosphorus was measured at a wavelength of 690 nm using a spectrophotometer model DR/2010. In this study, experiments that reduced residual phosphorus to less than 1 mg/L were subjected to energy demand calculation, subsequently [20].

The following equation was used to determine the energy consumed:

$$EC = \frac{UIt}{1,000 \times V [C_0 - C_t]}$$

where EC = kWh/g of phosphorus removed, U = the average voltage (in Volts) during the EP, I = the average electric current intensity (A) during the EP, t = the time of the electrolysis process (h), V = the volume of under processed wastewater (m^3), C_0 = the initial phosphorus concentration (g/m^3), C_t = the phosphorus concentration after t reaction time (g/m^3).

For statistical analysis, SPSS 16 software and a mixed analysis of variance test with repeated measures were used.

3. Results

The study investigated energy consumption in the EP for the removal of organic phosphorus from synthetic wastewater at a constant 0.6 A electric current (current density = 1.15 mA/cm^2). The reaction time ranged from 10 to 60 min, and the initial organic phosphorus concentration was in the range of 2–32 mg/L. The process removed organic phosphorus efficiently, up to 98%.

In Fig. 1, the energy consumption of electrochemically removing phosphorus for different initial concentrations of organic phosphorus at different time points was compared.

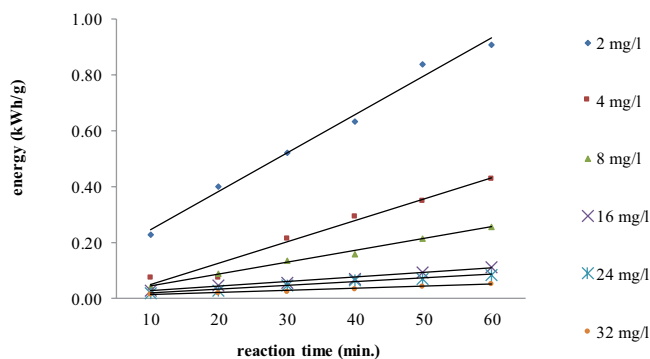


Fig. 1. Comparison of energy consumption for different initial concentrations of organic phosphorus as a function of reaction time.

Statistical analysis showed significant differences in reaction time and also between different initial concentrations of organic phosphorus in energy demand (P -value < 0.05). The lowest energy consumption was observed in samples in an initial concentration of 16 mg/L and 10 min reaction time. On the other hand, samples with 2 mg/L organic phosphorus initial concentration which were treated for 60 min consumed more energy.

The average energy consumption based on reaction time vs. initial concentration of organic phosphorus is depicted in Fig. 2. In this case, energy consumption decreased from 0.59 to 0.03 kWh/g as the initial concentration of organic phosphorus in samples increased from 2 to 32 mg/L.

As is shown in Fig. 3 with increase in the organic phosphorus removal efficiency, the average amount of energy consumed increased. Exponential equations presented in Figs. 2 and 3 can be used for estimation of energy demand in similar EP for organic phosphorus removal from aquatic samples at different concentrations and times, respectively.

4. Discussion

The results showed that the EP using steel electrodes is capable of removing organic phosphorus up to 98%. This finding is compatible with the results reported by Mesdaghinia et al. [18] and Rabbani et al. [19] in which EP resulted in the removal of 93% of phosphorus compounds

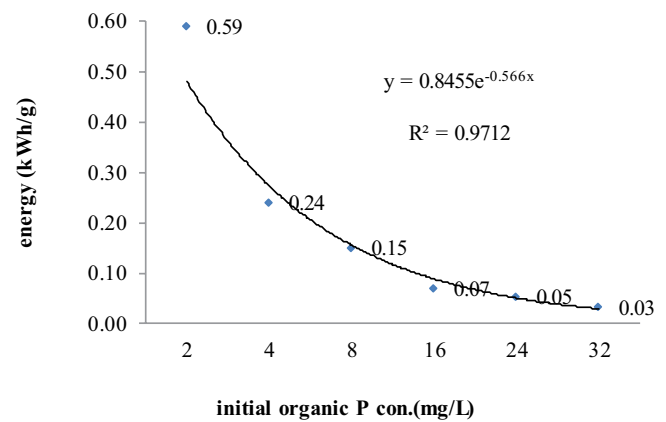


Fig. 2. Average energy consumption vs. initial concentration for organic phosphorus removal.

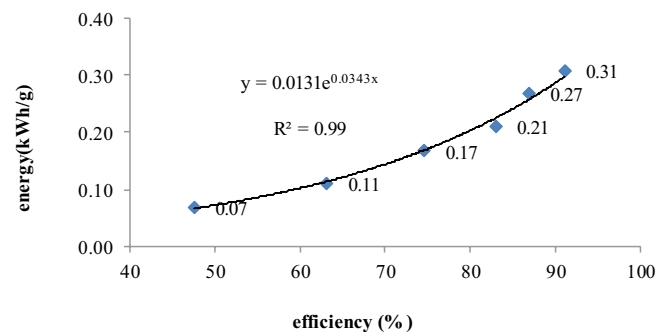


Fig. 3. Average energy consumption vs. organic phosphorus removal efficiency.

from activated sludge effluent. In a study conducted in Spain, complete removal of phosphate by electrocoagulation using iron and aluminium electrodes was reported [17]. Another study achieved up to 99% removal of phosphate from water using a new baffle plates aluminum-based electrochemical cell within 60 min at an initial concentration of phosphate of 100 mg/L [21]. However, lower efficiency (80.74%) for phosphate removal (at initial concentration of 15–75 mg/L) using iron electrode was reported in the Ano et al. study [8].

Since the main goal in our study was to investigate the variation in energy consumption during the removal of various concentrations of organic phosphorus from wastewater, parameters including pH, temperature, and current density were retained constantly during experiments. Optimized conditions for efficient removal of organic phosphorus were reported in the previous studies performed by the authors [18,19].

In the current study, pH of bulk wastewater subjected to electrochemical degradation was neutral (~7) and the removal efficiency was achieved up to 98%. Irdemez et al. [9] investigated the effect of pH on phosphate removal from wastewater in the electro-coagulation process using iron electrodes. They showed that in the reaction time of 20 min, with a pH of 7, the phosphate removal efficiency and energy consumption were 93% and 0.64 kWh/g, respectively. In the electrochemical systems, reduction of water molecules at the cathode resulted in the production of OH⁻, increase in local pH, and promotion of phosphorus crystal formation. It has been proven in other studies that bulk pH is not critical in electrochemical systems [22,23]. Lei et al. [22] reported that the local pH near the cathode can reach as high as 13.2, which is significantly higher than the bulk pH. In a study conducted on the electrochemical removal of phosphorus from domestic wastewater, removal efficiency at bulk pH of 7.5 was reported to be 55%, and at acidic pH (3.8) it was not significantly different (about 49%).

It is well known that the applied current density plays an important role in the electrochemical degradation of organic compounds because the amounts of ions and complexes are related to the applied current density [24]. In our study, the current density was kept constant at 1.15 mA/cm². Lei et al. [22] reported that the phosphorus removal efficiency was increased from 0.05 to 0.13 mg/L when the current density was increased from 26 to 150 A/m² (batch operation, 18.6 mg/L phosphorus). Recently, Wang et al. [23] reviewed studies on the application of EP in phosphorus removal and revealed that energy demands in these processes ranged from 2 to 2,000 kWh/kg of phosphorus using different cathode materials at different current densities and initial concentrations of phosphorus. According to Al Aji et al. [25], the energy consumption for heavy metal removal was reported at 49 kWh/m³, and the efficiency and current density were 25 mA/cm² and 96%, respectively. In the study of Lacasa et al. [17], the current densities were in a range of 0.1–5 mA/cm², and higher electrocoagulation rates were observed at higher current densities. However, energy consumption increases with increasing current density. The low current densities preserved the raw metal and reduced the energy consumption [17,26].

For the EP, energy demand is an important factor that determines the economic feasibility of the process. The major operating cost is associated with electrical energy consumption during the electrochemical degradation process [24,27]. Therefore, optimizing energy consumption and process efficiency is critical. The higher the initial concentration of organic phosphorus, the greater the possibility of collisions between the hydroxides produced during the EP, resulting in higher process efficiency and lower energy consumption. The maximum energy required to reduce organic phosphorus from solutions containing 2–32 mg/L to less than 1 mg/L (as the Iranian Standard for Phosphorus Concentration in Wastewater) was on average 0.59 kWh/g [20]. As depicted in Fig. 1, the lowest energy consumed was observed at the initial concentration of 16 mg/L, and at higher concentrations, no significant decrease in energy consumption was observed. In a previous study, 98% removal of phosphorus compounds from filtered effluent of activated sludge samples was achieved with energy consumption of 0.45 kWh/m³ [3]. These findings are compatible with whatever may have been declared by Irdemez et al. [16], that reported energy consumption for phosphorus removal from solutions with concentrations of 25, 50, 100 and 150 mg/L as 0.81, 0.506, 0.444 and, 0.323 kWh/g, respectively [16]. Their study showed that at the reaction time of 8 min, the removal efficiencies for the mentioned initial phosphorus concentrations were 100%, 98%, 61% and 42%, respectively [16]. In a study conducted on phosphate removal from rinse water, the energy consumptions were 0.18–11.29 kWh/m³ for aluminum electrode and 0.24–8.47 kWh/m³ for iron electrode in the same current density range [26].

As is mentioned earlier, in our study, by increasing the initial concentration of organic phosphorus beyond 16 mg/L, the energy consumption did not decrease significantly. An excessive increase in the initial concentration of the contaminant increases the possibility of the formation and accumulation of intermediates during the treatment process [28]. These intermediates could compete with the degradation of organic compounds and thus decrease energy efficiency [24,29].

5. Conclusion

The result of the study reveals that an EP using steel electrodes can efficiently remove organic phosphorus from wastewater, and energy consumption for economic assessment can be calculated using the equations obtained from the presented figures. In practice, depending on the initial concentration of organic phosphorus, the energy demand can be determined by Figs. 2 and 3, and to find the suitable reaction time, Fig. 1 can be used. The main design parameters that have been represented in this work are useful for the design of an EP to remove phosphorus from wastewater in future studies. Based on the results of our study, EP in comparison with conventional phosphorus removal methods can be more cost-effective. However, further studies should be conducted on system stability and efficiency under long-term operation and for different wastewater compositions and energy demands of large-scale reactors.

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Competing interests

The authors of this article declare that they have no conflict of interests.

References

- [1] K. Venkiteshwaran, P.J. McNamara, B.K. Mayer, Meta-analysis of non-reactive phosphorus in water, wastewater, and sludge, and strategies to convert it for enhanced phosphorus removal and recovery, *Sci. Total Environ.*, 644 (2018) 661–674.
- [2] H.E. Gray, T. Powell, S. Choi, D.S. Smith, W.J. Parker, Organic phosphorus removal using an integrated advanced oxidation-ultrafiltration process, *Water Res.*, 182 (2020) 115968, doi: 10.1016/j.watres.2020.115968.
- [3] D. Rabbani, F. Rashidipour, S. Nasser, S.G. Abas Mousavi, M. Shaterian, High-efficiency removal of phosphorous from filtered activated sludge effluent using electrochemical process, *J. Cleaner Prod.*, 263 (2020) 121444, doi: 10.1016/j.jclepro.2020.121444.
- [4] APHA, Standard Methods for the Examination of Water and Wastewater, 2012. Available at: <https://doi.org/ISBN9780875532356>.
- [5] C. Qin, H. Liu, L. Liu, S. Smith, D.L. Sedlak, A.Z. Gu, Bioavailability and characterization of dissolved organic nitrogen and dissolved organic phosphorus in wastewater effluents, *Sci. Total Environ.*, 511 (2015) 47–53.
- [6] A. Metcalf & Eddy Inc., *Wastewater Engineering: Treatment and Reuse*, International Edition, Revisada por Tchobanoglous, McGraw-Hill Education, New York, 2003, p. 819.
- [7] A.E. Johnston, C.J. Dawson, *Phosphorus in Agriculture and in Relation to Water Quality*, Agricultural Industries Confederation Peterborough, 2005.
- [8] J. Ano, A.S. Assémian, Y.A. Yobouet, K. Adouby, P. Drogui, Electrochemical removal of phosphate from synthetic effluent: a comparative study between iron and aluminum by using experimental design methodology, *Process Saf. Environ. Prot.*, 129 (2019) 184–195.
- [9] Ş. Irdemez, N. Demircioğlu, Y.Ş. Yildiz, The effects of pH on phosphate removal from wastewater by electrocoagulation with iron plate electrodes, *J. Hazard. Mater.*, 137 (2006) 1231–1235.
- [10] G. Chen, Electrochemical technologies in wastewater treatment, *Sep. Purif. Technol.*, 38 (2004) 11–41.
- [11] Y. Yavuz, A.S. Kopal, Ü.B. Ögütveren, Treatment of petroleum refinery wastewater by electrochemical methods, *Desalination*, 258 (2010) 201–205.
- [12] S. Dehghan, M.B. Miranzadeh, D. Rabbani, Electrochemical process efficiency for the removal of organic phosphorus from synthetic wastewater, *Feyz J. Kashan Univ. Med. Sci.*, 16 (2012).
- [13] M. Asselin, P. Drogui, H. Benmoussa, J.-F. Blais, Effectiveness of electrocoagulation process in removing organic compounds from slaughterhouse wastewater using monopolar and bipolar electrolytic cells, *Chemosphere*, 72 (2008) 1727–1733.
- [14] S. Vasudevan, J. Lakshmi, J. Jayaraj, G. Sozhan, Remediation of phosphate-contaminated water by electrocoagulation with aluminium, aluminium alloy and mild steel anodes, *J. Hazard. Mater.*, 164 (2009) 1480–1486.
- [15] Ş. Irdemez, Y.Ş. Yildiz, V. Tosunoğlu, Optimization of phosphate removal from wastewater by electrocoagulation with aluminum plate electrodes, *Sep. Purif. Technol.*, 52 (2006) 394–401.
- [16] Ş. Irdemez, N. Demircioğlu, Y.Ş. Yildiz, Z. Bingül, The effects of current density and phosphate concentration on phosphate removal from wastewater by electrocoagulation using aluminum and iron plate electrodes, *Sep. Purif. Technol.*, 52 (2006) 218–223.
- [17] E. Lacasa, P. Cañizares, C. Sáez, F.J. Fernández, M.A. Rodrigo, Electrochemical phosphates removal using iron and aluminium electrodes, *Chem. Eng. J.*, 172 (2011) 137–143.
- [18] A.R. Mesdaghinia, D. Rabbani, S. Nasser, F. Vaezi, Effect of coagulants on electrochemical process for phosphorus removal from activated sludge effluent, *Iran. J. Public Health*, 32 (2003) 1–7.
- [19] D. Rabbani, A.R. Mesdaghinia, S. Naseri, K. Naddafi, Effect of electrochemical process on phosphorous removal from activated sludge effluent, *J. Kashan Univ. Med. Sci.*, 7 (2003) 21–29.
- [20] Institute of Standards and Industrial Research of Iran, *Drinking Water-Physical and Chemical Specifications*, 2011 (in Persian).
- [21] K.S. Hashim, R. Al Khaddar, N. Jasim, A. Shaw, D. Phipps, P. Kot, M.O. Pedrola, A.W. Alattabi, M. Abdulredha, R. Alawsh, Electrocoagulation as a green technology for phosphate removal from river water, *Sep. Purif. Technol.*, 210 (2019) 135–144.
- [22] Y. Lei, B. Song, R.D. van der Weijden, M. Saakes, C.J.N. Buisman, Electrochemical induced calcium phosphate precipitation: importance of local pH, *Environ. Sci. Technol.*, 51 (2017) 11156–11164.
- [23] Y. Wang, P. Kuntke, M. Saakes, R.D. van der Weijden, C.J.N. Buisman, Y. Lei, Electrochemically mediated precipitation of phosphate minerals for phosphorus removal and recovery: progress and perspective, *Water Res.*, 209 (2022) 117891, doi: 10.1016/j.watres.2021.117891.
- [24] Y. Wang, C. Shen, M. Zhang, B.T. Zhang, Y.G. Yu, The electrochemical degradation of ciprofloxacin using a SnO₂-Sb/Ti anode: influencing factors, reaction pathways and energy demand, *Chem. Eng. J.*, 296 (2016) 79–89.
- [25] B. Al Aji, Y. Yavuz, A.S. Kopal, Electrocoagulation of heavy metals containing model wastewater using monopolar iron electrodes, *Sep. Purif. Technol.*, 86 (2012) 248–254.
- [26] M. Kobya, E. Demirbas, A. Dedeli, M.T. Sensoy, Treatment of rinse water from zinc phosphate coating by batch and continuous electrocoagulation processes, *J. Hazard. Mater.*, 173 (2010) 326–334.
- [27] K. Venkiteshwaran, P.J. McNamara, B.K. Mayer, Meta-analysis of non-reactive phosphorus in water, wastewater, and sludge, and strategies to convert it for enhanced phosphorus removal and recovery, *Sci. Total Environ.*, 644 (2018) 661–674.
- [28] X. Chen, G. Chen, P.L. Yue, Investigation on the electrolysis voltage of electrocoagulation, *Chem. Eng. Sci.*, 57 (2002) 2449–2455.
- [29] M. Malakootian, A. Shahesmaeili, M. Faraji, H. Amiri, S. Silva Martinez, Advanced oxidation processes for the removal of organophosphorus pesticides in aqueous matrices: a systematic review and meta-analysis, *Process Saf. Environ. Prot.*, 134 (2020) 292–307.