

Continuous electrocoagulation system for mining wastewater treatment

A. Rodriguez-Prado 

Independent Consultant, Whitehorse, Canada

<https://doi.org/10.1080/19236026.2020.1757985>

ABSTRACT Electrocoagulation is an efficient method for mining, industrial, and municipal wastewater treatment. Investigations have primarily focused on batch systems, while optimizing electrical current and operational time for maximum contaminant removal. Applying the correct amount of current in continuous systems is challenging due to varying water quality and residence time. A continuous laboratory-scale electrocoagulation system was investigated for treatment of ore- and petroleum-processing wastewater. The power to the cell was wired to a control loop feedback system. Preliminary results indicated that contaminants were removed by a minimum of 29% for ore-processing metals and 99% for petroleum hydrocarbons. Additional investigation is granted to study control settings that provide the optimal amount of ions into the electrocoagulation system for specific mining wastewater treatment applications.

■ **KEYWORDS** Electrocoagulation, Kalman filter, Process control, Wastewater treatment

RÉSUMÉ L'électrocoagulation est une méthode efficace pour le traitement des eaux usées minières, industrielles et municipales. Les enquêtes ont principalement porté sur les systèmes par lots, tout en optimisant le courant électrique et le temps de fonctionnement pour l'élimination maximale des contaminants. L'application de la bonne quantité de courant dans les systèmes continus est difficile en raison de la qualité de l'eau variable et du temps de séjour. Un système continu d'électrocoagulation à l'échelle du laboratoire a été étudié pour le traitement des eaux usées des déchets chimiques et pétroliers. L'alimentation de la cellule était reliée à un système de retour de boucle de contrôle. Les résultats préliminaires indiquent que les contaminants ont été éliminés d'au moins 29 % pour les métaux de traitement du minerai et de 99 % pour les hydrocarbures pétroliers. Une étude supplémentaire est accordée pour étudier les paramètres de contrôle qui fournissent la quantité optimale d'ions dans le système d'électrocoagulation, pour des applications spécifiques de traitement des eaux usées minières.

■ **MOTS-CLÉS** contrôle de processus, électrocoagulation, filtre Kalman, traitement des eaux usées

INTRODUCTION

Electrolytic cells have been used in a variety of wastewater treatment applications, including wastewater from almond processing (Valero et al., 2011), pulp mill (Vepsäläinen et al., 2011), poultry slaughterhouses (Bayar et al., 2011), dairy farms (Aoudjehane & Benatallah, 2014), paint manufacturing facilities (Akyol, 2012), metal plating facilities (Akbal & Camcı, 2011), vegetable oil refineries (Un et al., 2009), and petroleum refineries (Yavuz et al., 2010), as well as petroleum hydrocarbon groundwater treatment (Moussavi et al., 2011). The technology has targeted specific contaminants such as nitrates (Lacasa et al., 2011a), phosphates (Lacasa et al., 2011b), hexavalent chromium (Keshmirizadeha et al., 2011), and

organics (Asselin et al., 2008). The process uses sacrificial electrodes that provide metal ions to the wastewater, destabilizing emulsions, agglomerating organic matter and ions, and producing flocculates that reduce and absorb contaminants. The pollutants are further separated and removed from the treated wastewater stream by flotation or sedimentation. As such, electrocoagulation has been efficient at removing oil, organic matter, and suspended solids from wastewater, as well as heavy metals, phosphates, and nitrates from drinking water.

The efficiency of an electrocoagulation system has been shown to depend on a variety of factors such as amount of current added, cell mixing, electrode material (e.g., copper, aluminum, and iron), and chemical composition of the water

to be treated. A number of studies have been carried out in batch mode, applying a fixed amount of current to the electrolytic cell and the concentration of contaminants monitored over time. The optimal time of operation required to remove the highest concentration of contaminants has been determined by offline data analysis. However, few studies have used electrocoagulation systems in continuous mode. The main challenge to implement this type of system in continuous mode has been to adequately measure the contaminants of interest online, so that the appropriate current intensity is applied to the system. Applying too little current would result in a short supply of ions to remove the contaminants of concern, while an excess current would yield extra released ions from the electrodes, which themselves become wastewater contaminants. An additional challenge is related to the temporal variation in wastewater quality, as commonly occurs in full-scale industrial facilities. Finally, scale-up methodologies are underdeveloped, representing a challenge to size up feasibility studies into full-size systems (Hakizimana et al., 2017).

The operation of a continuous mode electrocoagulation system may rely on its ability to measure the concentration of contaminants online and successfully incorporate a control logic for the optimization of the electrode current intensity. The measurement of reduction-oxidation (redox) potential represents a feasible alternative for online data acquisition in many wastewater treatment systems. The redox parameter provides an indication of the ionic speciation in a specific aquatic system and an indirect indication of contaminant concentration. In addition, previous studies have demonstrated that redox measurements were capable of providing feedback to a proportional—integral—derivative (PID) control system for the continuous treatment of hexavalent chromium wastewater (Mustafa et al., 2002).

This paper presents preliminary results of a continuous electrocoagulation system for the treatment of mining wastewater, using

the Kalman filter as a PID controller and redox measurements as feedback. The Kalman filter is an algorithm used in statistics and control theory, in which historical values are used to predict new ones, simultaneously with a noise reduction of the data (filtering). The investigation of a continuous electrocoagulation system is important to provide additional data sets for the development of continuous electrocoagulation systems useful to wastewater treatment in the mining industry.

METHODS

A laboratory-scale continuous electrocoagulation system was built and operated consisting of a feed container, an electrolytic cell, a sedimentation basin, and an effluent collection container (Figure 1). The electrolytic cell consists of a 200 mL, rectangular plastic box holding two copper plates as electrodes, each with an effective area of 52.03 cm² and 0.16 cm thickness. The sedimentation basin consists of a plastic funnel of 1 L capacity, with 3 cm (½") PVC pipes to convey the influent to the bottom of the funnel and the effluent overflowing over the top handle. A DC power supply (Lambda, LL-901) is connected to the

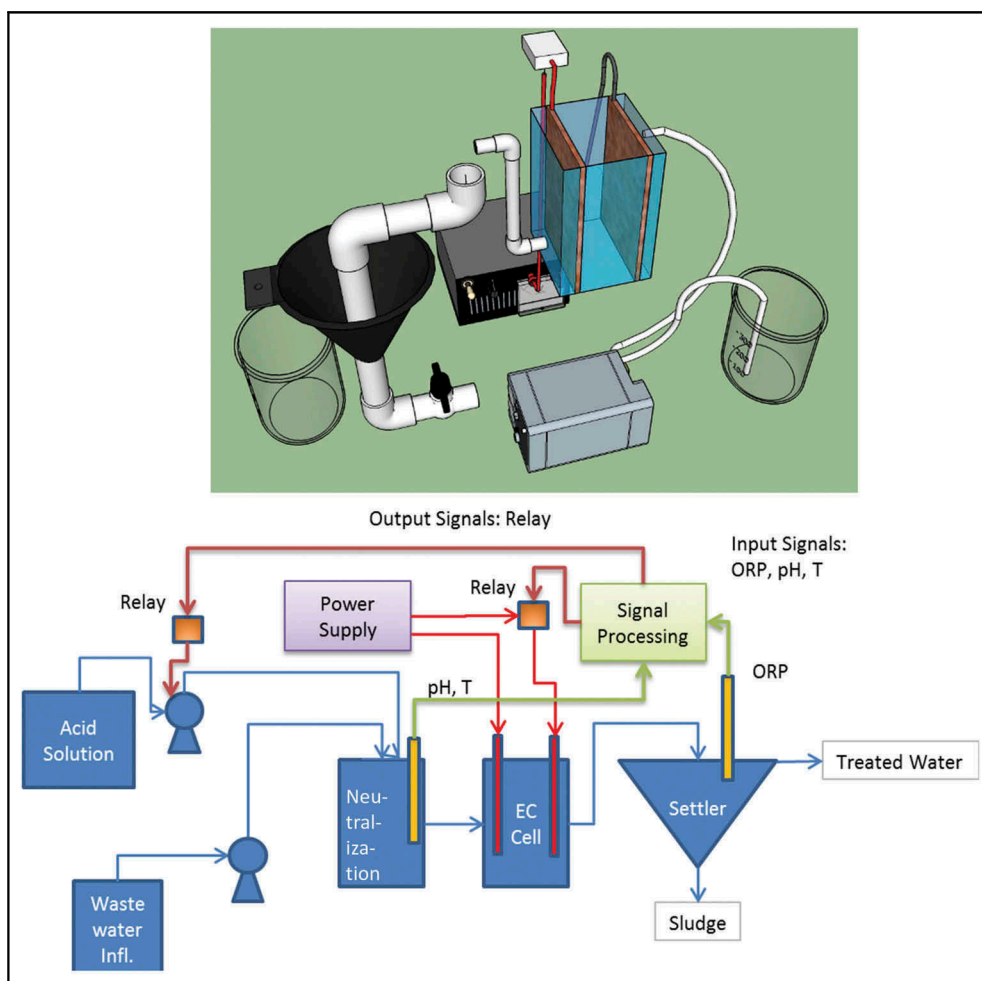


Figure 1. Continuous mode lab-scale electrocoagulation system

electrodes, feeding 4 DCV and 0.05A to provide 36 A/m², similar to Escobar et al. (2006).

The power supply was set up and controlled similarly to previous studies (e.g., Mustafa et al., 2002). An oxidation reduction potential (ORP) probe was submerged into the sedimentation basin and connected to an input/output (I/O) control board (Phidgets InterfaceKit 8/8/8), which in turn was connected to a laptop computer running Microsoft Windows. A Kalman filter algorithm written in Microsoft Excel received inputs from the ORP probe. The value of the filter gradient was used as a decision rule. Once the gradient reached a certain threshold, a signal was sent to the Phidgets application to activate a relay (Phidgets InterfaceKit 0/0/4) connected to power supply wiring, turning it on and off.

Ore processing wastewater treatment

Grab samples of wastewater were collected from an ore processing facility. The pH of the raw wastewater was approximately 12.0. Aliquots were transferred into a 2 L stirred Pyrex beaker serving as an influent container. A peristaltic pump transferred wastewater into a 200 mL neutralization reactor, adding a 10% hydrochloric acid solution. A pH probe submerged into the neutralization basin fed an I/O control to turn on the acid feed at pH > 10.0 and to turn it off at pH < 8.5. The visual characteristics of the wastewater after neutralization were similar to those of the influent, and no precipitates were observed, thus indicating that no treatment was performed by just adjusting the pH of the wastewater alone. The effluent of the neutralization basin overflowed into the electrolytic cell and continued through the system as described above. Influent and effluent samples were collected at different times during continuous treatment and analyzed for metals.

Petroleum processing wastewater treatment

Wastewater aliquots from a petroleum-processing facility were transferred into a 2 L stirred Pyrex beaker. From the beaker, the wastewater was pumped into the electrocoagulation system and treated as described above. Influent and effluent samples were collected at the end of the continuous treatment period and analyzed for benzene, toluene, ethylbenzene, and xylene (BTEX).

Groundwater treatment

In order to test continuous electrocoagulation in a system treating diluted water, groundwater from a supply well known to have arsenic was used. Aliquots were transferred into the stirred influent beaker of the electrocoagulation system. Influent and effluent samples were collected and analyzed for sulfate, arsenic, copper, iron, and total suspended and dissolved solid concentrations. Similarly to ore-processing wastewater treatment, the operational conditions were of interest to investigate the settings that would minimize excess electrode ions. The same control parameters were used as in the previous tests.

RESULTS

Ore processing wastewater

The electrocoagulation system was operated continuously for 150 minutes (4000 mL effluent) treating mining ore-processing wastewater. The values of the Kalman filter parameters used were similar to Mustafa et al. (2002): $\alpha = 1$, $\gamma = 1$, $Q_w = 0.01$, $Q_r = 1$, and $T_s = 1$. The power supply was turned on when the value of the filter gradient (state estimate update in Figure 2) was 0.25 or less and off when greater than 0.25.

The raw wastewater exhibited color and suspended solids by visual inspection, whereas the effluent exhibited a lighter color and less suspended solids (Figure 3). No turbidity or suspended solids analyses were performed on influent and effluent samples. Effluent samples collected at three times of operation were analyzed for metals (Table 1).

Aluminum, iron, nickel, and zinc concentrations decreased to a minimum after 33 minutes of treatment and increased

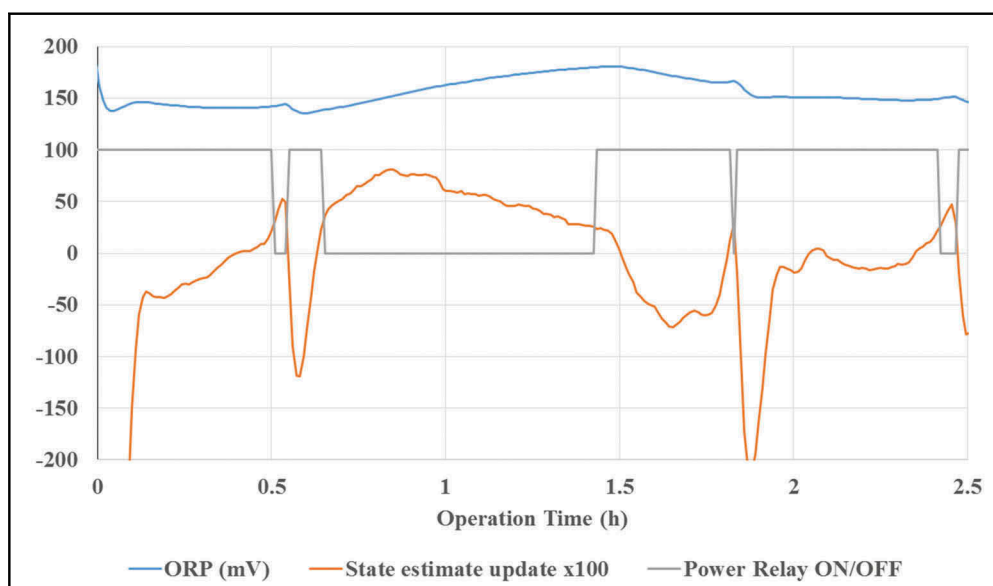


Figure 2. ORP, Kalman filter gradient, and cell powering rule in the electrocoagulation system

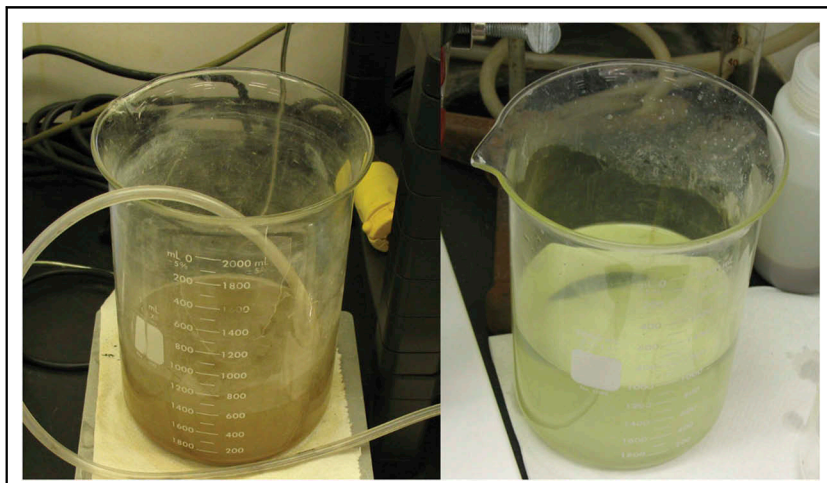


Figure 3. Ore-processing wastewater influent and electrocoagulation treated

Table 1. Metal concentrations in influent and effluent from continuous electrocoagulation system treating ore-processing wastewater

	Concentration in influent (mg/L)	Concentration in effluent (mg/L)			Change at 147 min relative to influent (%)
		33 min	92 min	147 min	
Copper	0.29	0.35	1.1	1.7	486
Cadmium	0.002	0.008	0.008	0.005	150
Chromium	4.9	3.5	3.6	3.5	-29
Arsenic	4.0	2.0	2.0	2.0	-50
Aluminum	43.9	9.4	18.1	15.1	-66
Uranium	15.0	4.0	3.0	3.0	-80
Zinc	0.68	0.038	0.069	0.074	-89
Manganese	0.94	0.036	0.081	0.084	-91
Iron	79.0	3.0	6.5	6.7	-92
Cobalt	0.095	0.003	0.007	0.007	-93
Lead	0.02	0.001	0.002	0.001	-95
Nickel	2.61	0.057	0.12	0.12	-95

again after 92 and 147 minutes of operation (Table 1). Nevertheless, the concentrations of all metals except copper and cadmium were reduced overall compared to their influent concentrations (Table 1). Copper electrodes were used in the electrolytic cell and alligator clips to hold the electrical connections. Although installed above water, the clamps may have been wetted by agitation of water in the electrolytic cell, thus contributing to cadmium dissolution. No samples

were collected between 33 and 92 minutes of operation, therefore no observations were made on whether metal concentrations would continue to decrease or increase with the electrocoagulation cell power off.

The acid addition between pH 8.5 and 10.0 allowed the treated wastewater to be in the range of pH 7–8, which mimics treated wastewater that would be allowed to be discharged without any additional pH adjustment. The ORP of the wastewater was used by the Kalman filter to obtain a state estimate to be subsequently used as a proxy for the cell power philosophy. The filter parameters (not ORP directly) were adequate to operate the electrolytic cell at the performance described by the removal of heavy metals.

The results indicate that electrocoagulation was capable of reducing the concentration of several metals in mining ore-processing wastewater. Copper (electrode metal) was not depleted and rather increased in the wastewater effluent. Further investigation is granted in order to obtain the optimal Kalman filter tuning parameters that minimize copper concentration and maximize the removal of metals of interest. This kind of tuning can be performed to minimize the concentration of specific heavy metals in the wastewater effluent for regulatory or discharge permit purposes.

Petroleum processing wastewater

The electrocoagulation system was operated for the treatment of petroleum processing wastewater. By visual inspection, the influent wastewater exhibited a muddy aspect, with solids in suspension that partially settled. The treated effluent was transparent with no suspended solids and no color (Figure 4).

The petroleum electrocoagulation system used the same Kalman filter tuning parameters as the ore-wastewater system. After 350 minutes of operation, a wastewater

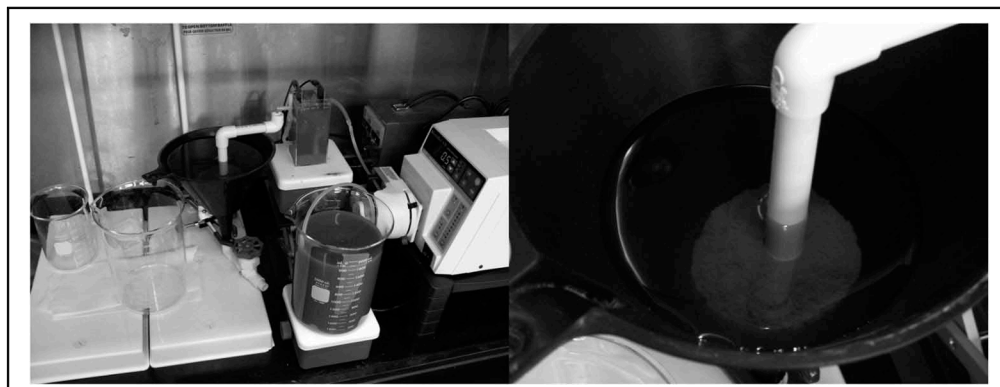


Figure 4. Electrocoagulation system treating petroleum processing wastewater (left) and close-up view of effluent (right)

Table 2. BTEX concentration in petroleum processing wastewater effluent

	Concentration in petroleum processing wastewater influent (µg/L)	Concentration in effluent at 350 minutes (µg/L)
Benzene	780	<0.2
Ethylbenzene	19	<0.2
Toluene	340	<0.2
Xylene	94	<0.2
C6-C10 fraction	210	Not collected

Table 3. Groundwater constituents after electrocoagulation treatment

	SO ₄ (mg/L)	As (µg/L)	Cu (mg/L)	Fe (mg/L)	Total dissolved solids (mg/L)	Total suspended solids (mg/L)
Groundwater influent	870	37	0.027	2.58	2,280	9
Groundwater effluent	920	2.7	0.28	0.0038	2,260	Not collected
Change after treatment (%)	5.7	-92.7	937	-99.9	-0.88	-

effluent sample was collected and analyzed for BTEX. Table 2 shows results for influent and effluent samples, with BTEX fractions reduced to nondetectable levels—representing over 99% removal. The electrocoagulation system was therefore capable of removing the petroleum hydrocarbons of interest from wastewater. The copper ions from the electrodes assisted coagulation and subsequently facilitated the sedimentation of the solids in suspension.

Groundwater

Groundwater was treated using the electrocoagulation system as well. The results presented in Table 3 show that electrocoagulation was capable of reducing arsenic and iron concentrations. However, sulfate and especially copper concentrations increased. Given that copper plates were used, the sulfate increase may be attributed to commercial copper plates, used as electrodes containing a certain amount of anhydrous copper sulfate. The sulfate would dissolve in the water during electrolysis. No additional reagents containing sulfates were added to the treatment system. Total dissolved solid concentrations were similar before and after treatment.

The results indicated that the electrocoagulation system was capable of removing contaminants of concern from groundwater as a dilute treatment system. As similar PID control settings were used, further investigation is warranted to determine the optimal control para-

eters that would produce lower concentration of excess copper ions, while maximizing the removal rate of contaminants of concern. If electrocoagulation was going to be used for drinking water treatment, such as well water, such operation optimization is particularly relevant. In mining wastewater treatment, this aspect is also important, due to the specific discharge limits that the mining companies must meet, including copper, iron, and aluminum.

CONCLUSIONS

A continuous electrocoagulation system was investigated for the treatment of mining ore-processing wastewater, petroleum-processing wastewater, and groundwater containing arsenic. The results presented as preliminary, and showed that the system provided an alternative approach to previously tested batch systems. The Kalman filter adequately controlled the electrolytic cell by means of the efficient removal of contaminants from the wastewater sources investigated. However, the specific set of control parameters yielded excess copper ions for the mining ore-processing wastewater and the groundwater treatment. Further investigation is needed for the investigation of additional sets of control variables to find the combination of parameters that would minimize the amount of dissolved electrode ions and maximize contaminants removal. The electrocoagulation system worked efficiently on petroleum wastewater treatment, as BTEX concentrations were low and the effluent exhibited adequate characteristics by visual inspection.

ACKNOWLEDGMENTS

The experimental work of this investigation was conducted at the Saskatchewan Research Council, of Saskatoon, Canada.

ORCID

A. Rodriguez-Prado  <http://orcid.org/0000-0001-7467-7848>

Paper reviewed and approved for publication by the Environmental and Social Responsibility Society of CIM.

A. Rodriguez-Prado is an environmental engineer and independent consultant with experience in industry, consulting, and research. His areas of specialization include water and wastewater treatment, pollution control facilities, remediation, and contaminant transport and transformation modeling.
arcadio.rodriguez@gmail.com

REFERENCES

- Akbal, F. & Camcı, S. (2011). Copper, chromium and nickel removal from metal plating wastewater by electrocoagulation. *Desalination*, 269(1–3), 214–222. <https://doi.org/10.1016/j.desal.2010.11.001>

- Akyol, A. (2012). Treatment of paint manufacturing wastewater by electrocoagulation. *Desalination*, 285, 91–99. <https://doi.org/10.1016/j.desal.2011.09.039>
- Aoudjehane, M. & Benatallah, M. E. (2014). Treatment of dairy wastewaters by electrocoagulation using iron electrodes. *Water Quality Research Journal of Canada*, 50(2), 198–209. <https://doi.org/10.2166/wqrjc.2014.053>
- Asselin, M., Drogui, P., Brar, S. K., Benmoussab, H., & Blais, J-F. (2008). Organics removal in oily bilgewater by electrocoagulation process. *Journal of Hazardous Materials*, 151(2–3), 446–455. <https://doi.org/10.1016/j.jhazmat.2007.06.008>
- Bayar, S., Yıldız, Y. S., Yılmaz, A.E., & İrdemez, S. (2011). The effect of stirring speed and current density on removal efficiency of poultry slaughterhouse wastewater by electrocoagulation method. *Desalination*, 280(1–3), 103–107. <https://doi.org/10.1016/j.desal.2011.06.061>
- Escobar, C., Soto-Salazar, C., & Tiral, M. I. (2006). Optimization of the electrocoagulation process for the removal of copper, lead and cadmium in natural waters and simulated wastewater. *Journal Environmental Management*, 81(4), 384–391. <https://doi.org/10.1016/j.jenvman.2005.11.012>
- Hakizimana, J. N., Gourich, B., Chafi, M., Stiriba, Y., Vial, C., Drogui, P., & Naja, J. (2017). Electrocoagulation process in water treatment: A review of electrocoagulation modeling approaches. *Desalination*, 404: 1–21. <https://doi.org/10.1016/j.desal.2016.10.011>
- Keshmirizadeha, E., Yousefia, S., & Rofouei, M. K. (2011). An investigation on the new operational parameter effective in Cr(VI) removal efficiency: A study on electrocoagulation by alternating pulse current. *Journal of Hazardous Materials*, 190(1–3), 119–124. <https://doi.org/10.1016/j.jhazmat.2011.03.010>
- Lacasa, E., Cañizares, P., Sáez, C., Fernández, F.J., and Rodrigo, M.A. (2011a). Removal of nitrates from groundwater by electrocoagulation. *Chemical Engineering Journal*, 171(3), 1012–1017. <https://doi.org/10.1016/j.ccej.2011.04.053>
- Lacasa, E., Cañizares, P., Sáez, C., Fernández, F. J., & Rodrigo, M. A. (2011b). Electrochemical phosphates removal using iron and aluminium electrodes. *Chemical Engineering Journal*, 172(1), 137–143. <https://doi.org/10.1016/j.ccej.2011.05.080>
- Moussavi, G., Khosravi, R., & Farzadkia, M. (2011). Removal of petroleum hydrocarbons from contaminated groundwater using an electrocoagulation process: Batch and continuous experiments. *Desalination*, 278(1–3), 288–294. <https://doi.org/10.1016/j.desal.2011.05.039>
- Mustafa, M. M., Rozaimah, S., Abdullah, S., & Rahman, R. A. (2002). Robust on-line control of hexavalent chromium reduction process using a Kalman filter. *Journal of Process Control*, 12(3), 405–412. [https://doi.org/10.1016/S0959-1524\(01\)00039-7](https://doi.org/10.1016/S0959-1524(01)00039-7)
- Un, U. T., Koparal, A. S., & Oğutveren, U. B. (2009). Electrocoagulation of vegetable oil refinery wastewater using aluminium electrodes. *Journal of Environmental Management*, 90(1), 428–433. <https://doi.org/10.1016/j.jenvman.2007.11.007>
- Valero, D., Ortiz, J. M., García, V., Expósito, E., Montiel, V. & Aldaz, A. (2011). Electrocoagulation of wastewater from almond industry. *Chemosphere*, 84(9), 1290–1295. <https://doi.org/10.1016/j.chemosphere.2011.05.032>
- Vepsäläinen, M., Kivisaari, H., Pulliainen, M., Oikari, A., & Sillanpää M. (2011). Removal of toxic pollutants from pulp mill effluents by electrocoagulation. *Separation and Purification Technology*, 81(2), 141–150. <https://doi.org/10.1016/j.seppur.2011.07.017>
- Yavuz, Y., Koparal, A. S., & Öğütveren, U. B. (2010). Treatment of petroleum refinery wastewater by electrochemical methods. *Desalination*, 258(1–3), 201–205. <https://doi.org/10.1016/j.desal.2010.03.013>