Statistical Analysis of Anode Efficiency in Electrochemical Treatment of Wastewater and Sludge

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Abstract

Electrochemical processes have proven their potential as effective technologies to treat wastewater from industrial, urban and agricultural activities, and thus, contribute towards a cleaner environment. In this study, we aimed to assess the effectiveness of the leading electrochemical technologies, such as electro-oxidation, electrochemical coagulation and electrochemical advanced oxidation processes (EAOPs), statistically for different types of anodes for the removal of various pollutants from wastewater along with their treatment efficiency. Anode is considered as a source of electron and an essential part of electrochemical processes. So, we have evaluated the relationship between different anode features such as anodic material, surface area versus removal of chemical oxygen demand (COD), dissolved organic carbon (DOC) and colour in various wastewater treatment plants (WWTPs) by IBM SPSS Statistics 26. Apart from that, various process characteristics such as inter-electrode distance, system pH, reactor volume, current density and voltage were also considered in this investigation. From the regression analysis of the electrochemical coagulation system, it was found that the removal efficiency of pollutants is enhanced by the surface area of the electrodes along with the inter-electrode distance. Regarding electro-oxidation, it was seen that COD and colour removal are both dependent on the reaction time of the system, while the DOC removal rate of different EAOPs was strongly related to the reactor volume. Furthermore, the uncertainty of the regression analysis on pollutant removal efficiency prediction was assessed. Finally, sensitivity analysis was done by Monte-Carlo method to check modest changes from input variables.

Keywords Statistical analysis · Electro-coagulation · Anode efficiency · Electro-oxidation · Electrochemical advanced oxidation processes

Article Highlights

- Anode efficiency of different electrochemical methods is studied through statistical analysis.
- Various anode characteristics like surface area and distance between electrodes are also considered.
- Regression equations are developed for different electro-chemical processes.
- Sensitivity analysis is conducted with Monte-Carlo to check correlations of different factors.

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1 Introduction

In the last few decades, many electrochemical technologies have been developed and are used as alternative approaches for the removal of diverse pollutants from wastewater and sludge. Due to their high energy efficiency, amenability to safe operation and cost-effectiveness, electrochemical techniques have considerable potential for application in full-scale plants to remove impurities from liquids, gases and soils (Pulkka et al. 2014). Electrochemical processes, such as electro coagulation (EC), electro-oxidation (EO) and electrochemical flotation (EF), have consequently attracted much attention in recent years (Chen 2004; Feng et al. 2016) since they are simple to operate and can completely remove numerous harmful pollutants before they reach aquatic habitats.

Recently, a number of different electrodes have been developed to enhance electrochemical processes in industrial operations, drinking water disinfection, wastewater and sludge treatment. For high productivity and lower resource consumption, electrochemical technologies can be used either as a pre-treatment step to increase the biodegradability of a pollutant or as an advanced treatment method to further reduce chemical oxygen demand (COD) or colour to meet required effluent standards (Chen 2004; Holt et al. 2005; Mollah et al. 2004; Tuan et al. 2012). Previous work in the field has considered various parameters for pollutant removal and anode types. For example, phenol removal by electrochemical process was studied by Kötzt et al. (1991). A Pt or Ti/Pt anode showed removal efficiency of 30% for total organic carbon (TOC) at pH 12 but 95% efficiency at pH 8.2 for ammonia (Kötzt et al. 1991). Planar graphite can remove 6–17% of COD when phenol is considered as a pollutant and NaOH is used as the electrolyte (Kannan et al. 1995). A Ti/PbO₂ anode showed 40% TOC reduction for phenol at pH 12 (Kötzt et al. 1991), 90% removal of COD from landfill leachate and ≥ 80% COD removal for pollutant 2-chlorophenol (Cossu et al. 1998). A Ti/SnO₂–Sb₂O₅ anode showed COD removal efficiency of about 80–95% when oxalic acid was used as an intermediate and 100% efficiency for COD removal from phenol (Awad and Abuzaid 1997; Marinčić and Leitz 1978; Polcaro et al. 1999; Wang and Farrell 2004). Moreover, 87% removal efficiency of COD was obtained by Fe-Al anode for textile wastewater (Ghanbari et al. 2014). Recently, Myburgh et al. (2019) studied IrO₂-Ta₂O₅/Ti anode for biodiesel wastewater and found 94% COD reduction by an integrated electrochemical process.

The pollutants can be destroyed by either direct or indirect oxidation processes. In direct oxidation, the contaminants are first adsorbed on the anode surface, and then, the transferred electron destroys the pollutant by direct anodic oxidation. Ozone, hypochlorite/chlorine and hydrogen peroxide are generated in situ in indirect electrochemical oxidation processes. The generated oxidant can destroy the pollutant through immediate oxidation (Mahmoud et al. 2018).

The electrochemical generation of hypochlorite/chlorine in a solution containing chloride ions can be described by the following reactions:

The application of an electrochemical oxidation process moderated by hypochlorite/chlorine has been studied by a number of researchers. For example, Rajkumar and Palanivelu (2004) presented results (concentration of COD and TOC) from treatment of the wastewater of phenol-formaldehyde resin manufacturing and an oil refinery using various anodes such as Ti/TiO₂–RuO₂–IrO₂, Ti/Pt or Ti/Pt/Ir.

This paper reviews the effectiveness of different anodes in electrochemical treatment for remediation of diverse synthetic and real wastewaters by anodic oxidation (AO), anodic oxidation with electrogenerated H₂O₂ (AO–H₂O₂) and electro-Fenton (EFe) alone or in combination with other technologies such as electro-coagulation, biological treatment, chemical-coagulation and membrane filtration. Various kinds of synthetic and real wastewaters containing dyes, pesticides and pharmaceuticals have been treated with these processes, and it has been found that colour, TOC and COD can be removed from wastewater and sludge and can be considered as main parameters in regression analysis. In this work, we have collected data of electrochemical processes used in pollutant removal of wastewater and sludge from the published literature and developed correlation equations from regression analysis with the aim to investigate anode-dependent parameters.

2 Fundamentals

2.1 Electrochemical Coagulation

Electrochemical coagulation (EC) is a process in which coagulant is created in situ by anode oxidation. Three main processes occur at the surface of electrodes during electro-coagulation: coagulant formation in the aqueous phase; adsorption of colloidal, soluble pollutants on coagulants; and finally, removal by sedimentation or flotation. The anode can be made of a metal like aluminium or iron and can dissolve as Al³⁺ and Fe²⁺ under application of an electric current. On the cathode, hydrogen gas and hydroxide ions are released at the same time. The hydroxide ions move towards the anode due to electrophoretic motion, and ion pairs are formed through the metal cation. Generally, coagulation agents are from the ion-paired polymeric iron aluminium hydroxide. At the electrodes, the key reactions are:

At the anode:

At the cathode:

Particles that are destabilised then aggregate to form flocs and hydrogen bubbles are formed at the cathode to induce the floatation of most flocs, which separates the particles effectively from the wastewater.

At higher pH, OH[−] chemically attacks the cathode and generates H₂ as follows:

Under acidic conditions:

Table 1 Summary of different electro-coagulation processes using different anode material

No.	Anode	Surface area (cm ²)	Inter-electrode distance (mm)	Current density(mA/cm ²)	Voltage (V)	Monopolar/Bipolar	Anion	Initial conc. (ppm)	pH	Reaction time (min)	Removal efficiency (%)	Reference
1	Fe	45	30	15		Monopolar	CN	300		20	93	(Moussavi et al. 2011)
2	Fe	60	10		40	Bipolar	CN	50		90	90	(Hassani et al. 2011)
3	Al	240	30	11.1		Monopolar	Fluoride	25	7	25	94.5	(Behbahani et al. 2011)
4	Al	80	10		30	Bipolar	Fluoride	20		80	60	(Drouiche et al. 2009)
5	Fe	80	10	12.5	30	Bipolar	Fluoride	25	6	60	60	(Drouiche et al. 2012)
6	Al		5			Monopolar	Fluoride	10	6–8	30	90	(Emanijomeh and Sivakumar 2006)
7	Al		5	3.75		Monopolar	Fluoride	10	6–8		90	(Emanijomeh and Sivakumar 2009)
8	Al		5	62.5		Bipolar	Fluoride	10		30	90	(Ghosh et al. 2008)
9	Al		5	1.875		Monopolar	Fluoride	10	6–8	60	90	(Emanijomeh et al. 2011)
10	Al		10			Monopolar	Fluoride		6–7		80	(Zhu et al. 2007)
11	Al	238		1			Fluoride	5	6	5	80	(Tezcan Un et al. 2013)
12	Al	175	20	6		Monopolar	Fluoride	20	5		92.5	(Essadki et al. 2008)
13	Al		20	12		Monopolar	Fluoride		4–5	30	90	(Essadki et al. 2009)
14	Al	150	15		30	Bipolar	Fluoride	5		5	93	(Khatibikamal et al. 2010)
15	Al	504	10			Bipolar	Fluoride	25			100	(Hu et al. 2005)
16	Al	1500	5	1		Monopolar	Phosphate	50			100	(Irdemez et al. 2006c)
17	Fe	1500	5	1		Monopolar	Phosphate	100	7		86	(Irdemez et al. 2006a)
18	Al	1500	5	1		Monopolar	Phosphate	100	3		100	(Irdemez et al. 2006b)
19	Fe	100	9	3		Monopolar	Phosphate	27		45	99.6	(Lacasa et al. 2011)
20	Al	77	25			Monopolar	Total P	55		20	85	(Zhang et al. 2013)
21	Fe			30					6		92	(Yavuz and Ögütveren 2018)

Table 2 Database for electro-oxidation processes for different types of anodes

No.	Anode	Initial conc. C_0 (mg/L)	Current density(mA/ cm^2)	Electrolyte	COD removal (%)	DOC removal (%)	Colour removal (%)	Reaction time (min)	Power consumption (kWh/m^3)	Reference
1	Ti/TaIrPt alloy	404	26.5	Raw effluent0.5% NaCl0.5% NaCl	10		85	120	5.6	(Chatzisyneon et al. 2006)
2	Graphite rodRuO ₂ /IrO ₂ /- TaO ₂	246.1	5.5	Raw effluent	95.8		100	150	0.26	(Subbarama Raju et al. 2009)
3	Ti/Pt	124	177	Raw effluent			96	30	4.20	(Sala and Gutiérrez-Bouzán 2014)
4	Ti/Pt	1354	80	20 g/L	50		100	240		(Wang et al. 2009)
5	Ti/TiO _x -RuO _x	5800	5	Raw effluent	98		100	360	4.3	(Basha et al. 2012)
6	Ti/TiO _x -RuO _x	560	5	Raw effluent	95		100	360	4.3	(Basha et al. 2012)
7	Si/BDD*	650	40	Raw effluent	100		100	1080	100	(Sales Solano et al. 2013)
8	Si/BDD	470	8	Raw effluent	80		100	180	135	(Tsantaki et al. 2012)
9	Si/BDD	160	40	Raw effluent	99	75	100	180	83	(Abdessamad et al. 2013)
10	Si/BDD	1000	60	Raw effluent	100		100	800	34.8	(Sales Solano et al. 2013)
11	BDD	1800	65	Raw		48.5		180		(Isarain-Chávez et al. 2014)
12	Ti/TiO ₂ -RuO ₂ -IrO ₂	1084	54	Raw				481		(Rajkumar and Palanivelu 2004)
13	Ti/TiO ₂ -RuO ₂ -IrO ₃	602	54	Raw	74.7			401		(Rajkumar and Palanivelu 2004)
14	BDD	246	60	Raw	96.4			240		(Zhu et al. 2009)
15	Ti/RuO _x -TiO _x	17,750	40	4 g/L	75			600		(Ahmed Basha et al. 2010)
16	Ti/RuO ₂	2000	50	2 g/L	75			180		(Valero et al. 2014)

Table 2 (continued)

No.	Anode	Initial conc. C_0 (mg/L)	Current density (mA/ cm^2)	Electrolyte	COD removal (%)	DOC removal (%)	Colour removal (%)	Reaction time (min)	Power consumption (kWh/m^3)	Reference
17	Ti/TiO ₂ -RuO ₂ -IrO ₂	8877	54	Raw	93.2			882		(Rajkumar and Palanivelu 2004)
18	BDD	1600	157	Raw		100		77		(de Vidales et al. 2012a, b)
19	BDD		5.15, 25						95	(Choi et al. 2010)
20	BDD		15–90						100	(Cabeza et al. 2007)
21	BDD		4–20						82	(Lissens et al. 2003)
22	BDD		33–150						100	(Flores et al. 2005)
23	BDD		6–15						99	(Wang and Farrell 2004)
24	BDD		15–100						100	(de Vidales et al. 2012a, b)
25	BDD		15–100						100	(de Vidales et al. 2012a, b)

*BDD: boron-doped diamond

Table 3 Database for electrochemical advanced oxidation processes for different types of anode

No.	Anode	Surface area (cm ²)	Temp. (°C)	Current density (mA/cm ²)	Process	Reactor volume (L)	pH	Power consumption (kW h / g DOC)	Maximum DOC decay (%)	Reference
1	BDD*	6	25	1.7–7.5	EF catalysed by pyrite	0.2	2.9–4.0	2.2	95	(Labadih et al. 2015)
2	Pt	1	25		EF	0.2	3		75	(Daneshtvar et al. 2008)
3	BDD	20	25	1.3–30	AO	0.2	7		41	(Carvalho et al. 2007)
4	Pt or BDD	3	35	8.3–100	AO-H ₂ O ₂ , EF	0.1	3			(García-Segura et al. 2011)
5	Pt or BDD	3	35	17–100	AO-H ₂ O ₂ , EF, PEF-UVA	0.1	3		100	(Florenza et al. 2014)
6	BDD	3	35	17–101	PEF-UVA	0.1	2–6		93	(Almeida et al. 2012)
7	BDD	20	25	25–150	EF, SPEF	2.5	4		93	(Ruiz et al. 2011)
8	Pt or BDD	3	35	33–101	EC + AO, AO-H ₂ O ₂ , EF or PEF-UVA	0.13	3	120	100	(Thiam et al. 2014)
9	BDD	9.4	25	30	AO + MF	2	3			(Juang et al. 2013)
10	Pt or BDD	3	25	33–150	AO-H ₂ O ₂ , EF, PEF-UVA	0.13	3		100	(Fan et al. 2010)
11	Pt or BDD	25	25	50–500	AO-H ₂ O ₂ , EF	0.2	3		90	(Olvera-Vargas et al. 2014)
12	Pt	1	25	19	EF, PEF-Vis, PEF-Vis/oxalate	0.8	3			(Khataee et al. 2010)
13	Pt	11.5	25	9.2–93	EF, PEF-UVC, PEF-UVA or PEF-UVB or PEF-UVC catalysed by ZnO nanoparticles	2	2–6		95	(Iranifard et al. 2011)
14	Sb-SnO ₂	40	24	10–30	AO	1	5.8	0.32	100	(Da Silva et al. 2014)
15	BDD	3	35	50–150	AO-H ₂ O ₂ , EF, PEF-UVA, SPEF	2.5	3	0.4	100	(Solano et al. 2015)
16	Pt	5.5	25	5–1.7	EF		3		98	(Hammami et al. 2007)
17	Pt	11.5	25	19–93	EF, PEF-Vis	2	3			(Khataee et al. 2013)
18	Pt or BDD	90.2	25	33–35	SPEF	10	3		97	(García-Segura and Brillas 2014)
19	BDD	20	35	15–80	EF, SPEF	2.5	2–6	0.15	90	(Salazar et al. 2011)

*BDD: boron-doped diamond

Al^{3+} and OH^- ions are generated from reactions (1) and (2) and can form various monomeric species, which finally transform $Al(OH)_3$ through complex precipitation kinetics (Gürses et al. 2002; Picard et al. 2000). Electrocoagulation has the advantage of removing small colloidal particles, which is not possible by traditional coagulation and flocculation. Short reaction time with a lower amount of sludge production are two further advantages of this process (Alinsafi et al. 2005; Pouet and Grasmick 1995).

2.2 Electro-Oxidation

Electro-oxidation involves either direct or indirect oxidation. It is an electrochemical process that treats the effluents flowing between special anodes and cathodes, as an electric field is applied. At the surface of the electrodes, the dissolved contaminants can be oxidised through direct oxidation.

In indirect mode, pollutants can also be degraded by hydrogen peroxide (Brillas et al. 1996, 1995; Matsue 1981) generated by the electrochemical process. Generally, porous carbon-polytetrafluorethylene (PTFE) is used as a cathode with oxygen supply and Pt, Pb/PbO₂ or Ti/Pt/PbO₂ are considered as the anode where Fe^{2+} might be added or can be formed in situ by dissolution of the iron anode (Brillas et al. 1997) to initiate the Electro-Fenton reaction (Chen 2004). Mixed and harmful wastewater can be treated by mediated electro-oxidation where low and stable valence electrons reach their reactive, high-valence states (Farmer 2006). These ions attack organic content directly and produce hydroxyl free radicals, which can destroy the organic pollutants. Representative mediators include Co^{3+} , Ce^{4+} , Ag^{2+} and Fe^{3+} regenerated on the anode creating a closed loop. These mediators should operate in acidic media, so the system can produce secondary pollution from these heavy metals (Bringmann et al. 1995; Farmer 1992). This pollution is considered as a disadvantage that limits usage of the approach for water treatment.

In direct oxidation, physically adsorbed active oxygen reacts with the organic compounds (R) on the anodes. Organic pollutants, on the other hand, can be oxidised by chemisorbed oxygen (MO_{x+1}) in the selection of selective oxidation products (Chen 2004; Comninellis 1994):

Table 4 Correlations of different parameters of anodes in the electro-coagulation system

		Surface area (cm ²)	Inter-electrode distance (mm)	Removal efficiency (%)
Surface area (cm ²)	Pearson Correlation	1	−0.578*	0.358
	Sig. (2-tailed)		0.039	0.209
	N	14	13	14
Inter-electrode distance (mm)	Pearson Correlation	−0.578*	1	0.049
	Sig. (2-tailed)	0.039		0.842
	N	13	19	19
Removal efficiency (%)	Pearson Correlation	0.358	0.049	1
	Sig. (2-tailed)	0.209	0.842	
	N	14	19	21

*Correlation is significant

The main advantages of anodic-oxidation are that it does not require much chemical for the reaction and there is no tendency to produce secondary pollution. Consequently, anodic oxidation is viewed as more desirable than other electrooxidation routes. On the other hand, anodic material such as Pt-carbon black (Boudenne et al. 1996), MnO_2 (Rajalo and Petrovskaya 1996), Ti/RuO_2 (Naumczyk et al. 1996), carbon felt (Polcaro and Palmas 1997) considered as a significant part of anodic oxidation and already studied; but none of them have same time stability and sufficient activity. Among all anodes, TiO_2 , Pt, IrO_2 etc. were considered as extensively used anode in anodic oxidation (Chen 2004).

Fig. 1 (a) Scatter plot of removal efficiency with surface area, (b) Scatter plot of removal efficiency with reaction time

Fig. 2 Normal P-P plot of linear regression standardised residual

2.3 Electrochemical Advanced Oxidation Processes

Electrochemical advanced oxidation processes (EAOPs) are an encouraging class of advanced oxidation processes (AOPs). Formerly, the AOP process only entailed anodic oxidation (AO) where organic content could oxidise the anode directly through the transfer of electrons or indirectly by $\bullet\text{OH}$ radical at the anode surface. Active chlorine species, ozone, per-sulfates and H_2O_2 at the anode can also induce oxidation (Barrera-Díaz et al. 2014; Brillas et al. 2009; Chaplin 2014; Comninellis et al. 2008).

EAOPs methods are clean and effective techniques, and are becoming increasingly popular. When AO is performed with electrogenerated H_2O_2 , the process is considered anodic oxidation with electrogenerated H_2O_2 (AO- H_2O_2). In anodic oxidation, the radicals are formed by water oxidation on a high O_2 -overvoltage anode such as a Pt, PbO_2 and boron-doped (BDD) electrode. Electrochemical coupling occurs between the Fenton's reagents, where H_2O_2 generated at the cathode can react with Fe^{2+} present in the medium, leading to the formation of a hydroxyl radical from the Fenton reaction. Photoelectron-Fenton (PEF) and solar photoelectric-Fenton (SPEF) processes have also been proposed for water treatment and studied using artificial light and natural sunlight by Comninellis (1994), Belhadj Tahar and Savall (1998) and Brillas et al. (2004):

Table 5 ANOVA model summary for dependent variable removal efficiency (%)

Model	Sum of squares	F	Mean square	Sig.
Regression	984.700		246.175	.
Residual	0.000			
Total	984.700			

Predictors: (Constant), Initial conc. (ppm), pH, Inter-electrode distance (mm), Surface area (cm^2)

Table 6 Estimated regression model coefficients for the dependent variable – removal efficiency

Model	Unstandardised Coefficients, B	Std. error
(Constant)	103.588	0.000
Surface area (cm ²)	0.112	0.000
Inter-electrode distance (mm)	1.003	0.000
pH	−3.500	0.000
Initial conc. (ppm)	−1.664	0.000

Without detailed understanding and systematic analysis, it is difficult to use these processes for real wastewater, although several researchers have used AO, EFe and SPEF for different types of effluent treatment in lab-scale experiments (Oturán and Aaron 2014).

3 Statistical Analyses

The data used in this paper were collected from published peer-reviewed papers. They are summarized in Table 1, Table 2 and Table 3. Once the data were collected, then inaccurate and duplicate data were cleaned up before analysis. The final part can be implemented when applied in real work field. Here, the collected data were organised to check the anode efficiency based on its characteristics, current density, system pH and pollutant removal rate. The database was used to perform regression analysis by IMB SPSS Statistics 26 on anode efficiency, TOC, COD, pH, reaction time and inter-electrode distance in wastewater and sludge treatment. SPSS enabled us to quickly dig deeper into the data and helped to make predictions and draw conclusions (IBM Corp 2018).

4 Results and Discussion

4.1 Efficiency Analysis of Anode Used in Electro-Coagulation

We have considered the value from Table 1 for correlation establishment in SPSS between different variables. The resulting values of surface area, inter-electrode distance and the Pearson correlation are shown in Table 4. The correlation coefficient generally ranges from −1 to +1. The Pearson correlation represents the linear correlation between different variables

Table 7 Model summary for the electro-oxidation process (COD removal percentage considered as a dependent variable)

Model	R	R ²	Adj. R ²	Std. error	F change	Durbin-Watson
1	0.503	0.253	0.029	24.45312	1.132	1.128

Table 8 Regression analysis by ANOVA (COD removal percentage considered as a dependent variable)

Model	Sum of squares	F	Mean square	Sig.
Regression	2029.877	1.132	676.626	0.383
Residual	5979.552		597.955	
Total	8009.429			

Predictors: (Constant), Current density (mA/cm²), Initial conc. (mg/L), Reaction time (min)

and the relationship strength. Table 4 can be used to analyze the degree of relationship for different variables of electro-coagulation. From Table 4, the correlation coefficient of surface area with removal efficiency is 0.358. The value lies between 0.3 and 0.4, which is regarded as a moderate degree of correlation. The sign of the correlation represents the direction of the relationship. The significance value of 0.209 with Pearson correlation of 0.358 indicates that this correlation is valid.

Surface area and inter-electrode distance correlation are -0.578 , but the significance value is 0.039. A significance value below 0.02 is regarded as a significant correlation. The number of cases with non-missing values are signified as N and N is also the probability of obtaining results. A Pearson value below 0.2 is regarded as a very low degree of correlation; so, from this analysis, the removal efficiency of the pollutant is dependent on the surface area of the different anodes (Arkkelin 2014; Landau and Everitt 2004).

Figure 1a and Fig. 1b present the relationship between removal efficiency with reaction time and surface area. Figure 1 shows two variables, which established a relationship over a certain range of variables. If the range of one or both variables is expanded, then the linear relationship observed may or may not continue to exist. Removal efficiency based on surface area for 10 to 230 cm² was almost 85% to 100%. From Fig. 1a, it is shown that by increasing the area even above 230 cm² will not result in further improvement of the removal efficiency.

The relation of removal efficiency with reaction time of the electro-coagulation system is shown in Fig. 1b. It can be seen that lower reaction time enhances the removal efficiency of the pollutant. There is a mild decrease in removal efficiency with time from 30 min to 80 min.

Fig. 3 Regression standardised residual in the histogram

Table 9 Coefficients of variables from the electro-oxidation process

Model	Unstandardised Coefficients, B	Std. error
(Constant)	71.434	15.977
Initial conc. (mg/L)	−0.001	0.001
Reaction time (min)	0.044	0.025
Current density (mA/cm ²)	−0.203	0.305

Different types of anodes are considered here of the electro-coagulation system, so it is not so easy to tell specific performance of an anode because material type and its characteristics need to be considered.

Figure 2 presents the normal probability-probability plot of regression standardized residual where removal efficiency is considered as the dependent variable. The probability plot is used to assess how closely two data sets agree with each other and P-P plots are vastly used to evaluate the skewness of a distribution (IBM Corp 2018). The relationship from the probability plot is:

$$y = 0.9323x + 0.0487 \quad (12)$$

with $R^2 = 0.9556$. Equation (12) shows that the residual value is normally distributed as the points are following the diagonal line. Conversely, residual value would be abnormally distributed if the points don't follow the diagonal value. Moreover, the R^2 value is 0.955, which means that 95.5% is distributed normally, but about 5% is distributed abnormally. Parameters such as current density, reaction time, and removal efficiency dependent on the surface area are considered for regression analysis accordingly (Ambat et al. 2019; Elliott and Woodward 2011).

The F ratio from ANOVA (Table 5) did not indicate anything for the data, and if all variables are forced into the multiple linear regression, the mean square of the regression model is 246.17.

Fig. 4 Simple scatter plot of colour removal with reaction time

Table 10 Model summary of the electro-oxidation process (Dependent variable: colour removal %)

Model	R	R ²	Adj. R ²	Std. error	F change	Durbin-Watson
1	0.408	0.167	-0.250	5.3342	0.400	1.158

Table 6 shows that the final pollutant removal efficiency is positively dependent on the surface area and inter-electrode distance, whereas the pH and initial concentration showed negative coefficients. So, anode surface area and inter-electrode distance could be considered for higher removal efficiency of pollutant but a proper design is necessary to establish this suggestion for future work. So, regression analysis for predicted removal efficiency would be:

Removal efficiency = $103.588 + (0.112 \times \text{surface area}) + (1.003 \times \text{inter electrode distance}) - (3.5 \times \text{pH}) - (1.664 \times \text{initial conc.})$ (13).

4.2 Statistical Analysis of Different Anode Materials in the Electro-Oxidation Process

By following the same way, statistical analysis was carried out by SPSS for different anodes in the electro-oxidation system. Initial concentration, reaction time and current density were considered to generate a model summary and regression analysis. Table 7 shows a summary of the regression model and overall fit statistics for COD removal percentage. The adjusted R² of our model is 0.029 with R² = 0.253, which means that the linear regression explains 25.3% of the variance in the data. Durbin-Watson d = 1.128, which is between the two critical values of $1 < d < 1.5$. Generally, the Durbin-Watson test is one of the regression assumptions where observations are fully independent. Durbin-Watson should show statistics between 1.5 to 2.5 to express no correlation, so the value 1.128 describes data that are correlated with each other (Landau and Everitt 2004; Marshall and Karadimitriou 2015).

The regression analysis by ANOVA is shown in Table 8 for the dependent variable COD removal percentage. This analysis shows how well the model fits with the data. The F value in the regression model is significant if it is less than 0.05; the independent variable can then explain the dependent variable variation. Here, the F value is 1.132, which is larger than 0.05, so the independent variable is unable to explain the dependent ones. The predictor variables are current density, initial concentration and the reaction time of the system. This analysis is used to examine the relationship between two or more predictor variables with a response variable. The standardised residual of the histogram of the regression (Fig. 3) shows that the standard deviation is 0.877 for the dependent variable COD removal percentage. The histogram analysis expresses the data quality to make it more clear by showing its distribution graphically (Arkkelin 2014; Kaper and Engler 2013).

Table 11 ANOVA analysis for colour removal with respect to reaction time, current density and initial concentration

Model	Sum of squares	F	Mean square	Sig.
Regression	34.175	0.400	11.392	0.758
Residual	170.725		28.454	
Total	204.900			

Predictors: (Constant). Current density (mA/cm²), Initial conc. (mg/L), Reaction time (min)

Table 12 Coefficient of variable colour removal with respect to initial concentration, reaction time and current density

Model	Unstandardised Coefficients, B	Std. error
(Constant)	95.069	3.917
Initial conc, (mg/L)	0.001	0.001
Reaction time, min	0.005	0.006
Current density, mA/cm ²	0.009	0.037

The coefficient of different variables like initial concentration, reaction time and current density are considered to predict the dependent variable percent age of COD removal. Table 9 shows that the percentage of COD removal is positively dependent on the reaction time of the system, whereas current density and initial concentration work as statistically insignificant variables. So, the equation for the COD removal percentage is:

COD removal % = 71.434–0.001*(initial conc.) + 0.044*(reaction time) – 0.203*(current density) (14).

To develop regression analysis for percent colour removal, different parameters like reaction time, current density and initial concentration are considered for electro-oxidation. A scatter plot of color removal for different experimental reaction times is shown in Fig. 4.

100% colour removal was observed during the time period of 190 to 300 min and at 800 min of electro-oxidation. A summary of the model and ANOVA analysis for the electro-oxidation process are shown in Tables 10 and 11. The R² value was 0.167, which means that about 16.7% of the variable is explained by the regression. The F value was 0.4, which is greater than 0.05, so again the independent variable cannot clarify effectively the dependent variables.

The regression analysis (Table 12) of the colour removal percentage depends on the initial concentration, reaction time and current density, where all the variables are positively significant. So, the co-efficient of the variable colour removal indicates that it is fully dependent on the initial concentration, reaction time and current density, and the equation for colour removal percentage is:

Table 13 Correlation coefficients of electro-oxidation process variables

		Colour removal, %	Reaction time, min	Initial conc., mg/L	Power consumption, kWh/m ³
Colour removal, %	Pearson	1	0.303	0.198	0.330
	Correlation				
	Sig. (2-tailed)		0.364	0.560	0.386
Reaction time, min	N	11	11	11	9
	Pearson	0.303	1	0.367	0.308
	Correlation				
Initial conc., mg/L	Sig. (2-tailed)	0.364		0.134	0.419
	N	11	18	18	9
	Pearson	0.198	0.367	1	–0.234
Power consumption kWh/m ³	Correlation				
	Sig. (2-tailed)	0.560	0.134		0.545
	N	11	18	18	9
	Pearson	0.330	0.308	–0.234	1
	Correlation				
	Sig. (2-tailed)	0.386	0.419	0.545	
	N	9	9	9	16

Table 14 Model summary for EAOPs with different anodes

Model	R	R ²	Adj. R ²	Std. error	F change	Durbin-Watson
1	0.650	0.423	0.279	12.71336	0.077	1.644

Colour removal, % = $95.069 + 0.001 * (\text{initial conc.}) + 0.005 * (\text{reaction time}) + 0.009 * (\text{current density})$ (15).

The correlation coefficients of the different variables are shown in Table 13. The percentage of colour removal is fully dependent on the time of the reaction, where the significance is 0.364. On the other hand, the correlation of colour removal with respect to initial conditions is 0.198, where the significance is 0.560. The significance value lies between 0.3 and 0.4, which is considered moderate significance, so the colour removal moderately depends on the reaction time and initial concentration.

4.3 Statistical Analysis for EAOPs with Respect to Different Parameters

For EAOP processes, information about different types of anodes with their surface area, pH, and reactor volume for DOC decay rate is presented in the Table 3. A model summary with ANOVA analysis is given in Tables 14 and 15. The R² value is 0.423 and adjusted R² is 0.279, so this model explains 42.3% of the variance of data. From the ANOVA analysis, the F value of the regression analysis is 2.932, which is higher than 0.05, so the independent variable DOC decay rate cannot be explained by the dependent variables reactor volume, pH and surface area.

The coefficients of a number of variables for EAOPs are shown in Table 16. Reactor volume, surface area and pH are considered here and the final equation for DOC decay rate is: $\text{DOC decay rate} = 111.064 + 1.145 * (\text{reactor volume}) - 0.001 * (\text{surface area}) - 6.326 * (\text{pH})$ (16). Where reactor volume is a positively significant factor for the DOC decay rate and the others are negatively significant.

4.4 Sensitivity Analysis of the Regression Analysis

Sensitivity analysis quantifies how a different value of an independent variable can affect the dependent variable. Regression analysis of electro-coagulation, electro-oxidation and EAOPs revealed that removal efficiency, colour removal, and COD or DOC decay rate depend on the various factors of the process. To evaluate the future performance of the electrochemical process for different anodes, sensitivity analysis was done using Monte Carlo simulation. Sensitivity analysis of these results can then guide future research and development of the electro-chemical treatment of wastewater and sludge. Different anodes such as Al, Fe, Si/

Table 15 ANOVA analysis for EAOPs (dependent variable: DOC decay rate)

Model	Sum of squares	F	Mean square	Sig.
Regression	1421.883	2.932	473.961	0.77
Residual	1939.555		161.630	
Total	3361.438			

Predictors: (Constant), pH, Reactor volume (L), Surface area (cm²)

Table 16 Coefficients of variables for DOC decay rate in EAOPs

Model	Unstandardised Coefficients, B	Std. error
(Constant)	111.064	9.106
Reactor volume (L)	1.145	2.459
Surface area (cm ²)	-0.001	0.278
pH	-6.326	2.561

BDD, Ti/TiO₂-RuO₂-IrO₂ and BDD were evaluated in sensitivity (Table 17) and the Monte Carlo simulation was used to determine the uncertainty of the linear equation (Elliott and Woodward 2011; Tang et al. 2009).

Monte Carlo simulation was selected for 10,000 trials as a greater number of trials can increase the convergence of the results. In Fig. 5, Monte Carlo simulation for different electrodes such as Al showed a mean value of 94.25 but Fe had a mean value of 71.35 in the electro-coagulation process. Moreover, it can be seen in Fig. 5C that Si/BDD has a mean value of 91.54, and the mean value of Ti/TiO₂-RuO₂-IrO₂ is 84.73. The normal probability distribution of these simulations simply defines the mean or expected value and a standard deviation refers to the variation about the mean. The values near the middle area are those most likely to occur.

5 Conclusions

A comprehensive database was developed for electro-chemical processes using different anode material. Regression equations between colour removal, DOC or COD reduction with respect to the anode in various electrochemical treatments were developed for sludge and wastewater treatment. Statistical analysis for these processes using appropriate methods was used to quantify the effects of different dependent variables. The efficiency of the electrochemical treatment of wastewater and sludge treatment could be predicted with the established regression equations. The lower R² graphs also showed high variability data with significant trend and this trend provides the prediction for response even though data points are fall further from the regression line. So, future workers can get idea about effect size from these regression equations. It was also found that the efficiency of electrochemical processes is a function of

Table 17 Regression equation for anodes from the electro-chemical treatment processes

No.	Considered anode	Treatment process	Regression equation
	Al	Electro-coagulation	% Removal efficiency = $89.476 + (0.007 \times \text{surface area}) + (0.305 \times \text{current density}) + (3.49\text{E-}17 \times \text{initial conc.})$
	Fe	Electro-coagulation	% Removal efficiency = $41.134 + (0.024 \times \text{surface area}) + (1.692 \times \text{inter-electrode distance})$
	Si/BDD	Electro-oxidation	% Colour removal = $128.805 - (0.024 \times \text{initial conc.}) + (0.008 \times \text{reaction time}) - (0.318 \times \text{power consumption})$
	Ti/TiO ₂ -RuO ₂ -IrO ₂	Electro-oxidation	% COD removal = $31.786 - (0.004 \times \text{initial conc.}) + (0.114 \times \text{reaction time})$
	BDD	EAOP	% DOC decay rate = $138.19 + (0.304 \times \text{surface area}) + (0.018 \times \text{current density}) - (0.126 \times \text{temp.}) + (3.23 \times \text{reactor volume}) - (14.388 \times \text{pH})$

Fig. 5 Monte Carlo simulation for anodes in electro-coagulation, electro-oxidation and EAOPs

surface area, inter-electrode distance and anode, reaction time, initial concentration and electrical density. Monte Carlo simulation for anodes used in electro-coagulation, electro-oxidation and electrochemical advanced oxidation processes quantified the uncertainty and sensitivity of different anode materials, for example, Al showed a mean value of 94.25, Fe had a mean value of 71.35, while Si/BDD showed a mean value of 91.54, and 84.73 was found for Ti/TiO₂-RuO₂-IrO₂. These equations could be used to predict the efficiency of pilot or full-scale processes when all the design parameters are proportionally and properly scaled.

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References

- Abdessamad NEH, Akrouit H, Hamdaoui G, Elghniji K, Ksibi M, Bousselmi L (2013) Evaluation of the efficiency of monopolar and bipolar BDD electrodes for electrochemical oxidation of anthraquinone textile synthetic effluent for reuse. *Chemosphere* 93:1309–1316. <https://doi.org/10.1016/j.chemosphere.2013.07.011>
- Ahmed Basha C, Soloman PA, Velan M, Miranda LR, Balasubramanian N, Siva R (2010) Electrochemical degradation of specialty chemical industry effluent. *J Hazard Mater* 176:154–164. <https://doi.org/10.1016/j.jhazmat.2009.10.131>
- Alinsafi A, Khemis M, Pons MN, Leclerc JP, Yaacoubi A, Benhammou A, Nejmeddine A (2005) Electrocoagulation of reactive textile dyes and textile wastewater. *Chem Eng Process Process Intensif* 44:461–470. <https://doi.org/10.1016/j.ccep.2004.06.010>
- Almeida LC, Garcia-Segura S, Arias C, Bocchi N, Brillas E (2012) Electrochemical mineralization of the azo dye acid red 29 (Chromotrope 2R) by photoelectro-Fenton process. *Chemosphere* 89:751–758. <https://doi.org/10.1016/j.chemosphere.2012.07.007>
- Ambat I, Tang WZ, Sillanpää M (2019) Statistical analysis of sustainable production of algal biomass from wastewater treatment process. *Biomass Bioenergy* 120:471–478. <https://doi.org/10.1016/j.biombioe.2018.10.016>
- Arkkelin D (2014) Using SPSS to understand research and data analysis, in: psychology curricular materials 1. Valparaiso University, Valparaiso, Indiana, p 194
- Awad YM, Abuzaid NS (1997) Electrochemical treatment of phenolic wastewater: efficiency, design considerations and economic evaluation. *J. Environ. Sci. Heal. . Part A Environ. Sci. Eng. Toxicol* 32:1393–1414. <https://doi.org/10.1080/10934529709376617>
- Barrera-Díaz C, Cañizares P, Fernández FJ, Natividad R, Rodrigo MA (2014) Electrochemical advanced oxidation processes: an overview of the current applications to actual industrial effluents. *J Mex Chem Soc*
- Basha CA, Sendhil J, Selvakumar KV, Muniswaran PKA, Lee CW (2012) Electrochemical degradation of textile dyeing industry effluent in batch and flow reactor systems. *Desalination* 285:188–197. <https://doi.org/10.1016/j.desal.2011.09.054>
- Behbahani M, Moghaddam MRA, Arami M (2011) Techno-economical evaluation of fluoride removal by electrocoagulation process: optimization through response surface methodology. *Desalination* 271:209–218. <https://doi.org/10.1016/j.desal.2010.12.033>
- Belhadj Tahar N, Savall A (1998) Mechanistic aspects of phenol electrochemical degradation by oxidation on a Ta/PbO₂ anode. *J Electrochem Soc* 145:3427–3434. <https://doi.org/10.1149/1.1838822>
- Boudenne, J.L., Cerclier, O., Galéa, J., Van der Vlist, E., 1996. Electrochemical oxidation of aqueous phenol at a carbon black slurry electrode. *Appl Catal A Gen* 143, 185–202. [https://doi.org/10.1016/0926-860X\(96\)00027-0](https://doi.org/10.1016/0926-860X(96)00027-0)
- Brillas E, Saulea R, Casado J (1997) Peroxi-coagulation of aniline in acidic medium using an oxygen diffusion cathode. *J Electrochem Soc* 144:2374. <https://doi.org/10.1149/1.1837821>
- Brillas E, Mur E, Casado J (1996) Iron(II) catalysis of the mineralization of aniline using a carbon-PTFE O₂-fed cathode. *J Electrochem Soc* 143:L49. <https://doi.org/10.1149/1.1836528>
- Brillas E, Bastida RM, Lloa E, Casado J (1995) Electrochemical destruction of aniline and 4-Chloroaniline for wastewater treatment using a carbon-PTFE O₂-fed cathode. *J Electrochem Soc* 142:1733. <https://doi.org/10.1149/1.2044186>
- Brillas E, Sires I, Arias C, Cabot PL, Centellas F, Rodriguez RM, Garrido JA (2004) Paracetamol mineralization by advanced electrochemical oxidation processes for wastewater treatment. *Environ Chem* 1:26–28. <https://doi.org/10.1071/En04018>
- Brillas E, Sires I, Oturan MA (2009) Electro-Fenton process and related electrochemical technologies based on Fenton's reaction chemistry. *Chem Rev* 109:6570–6631. <https://doi.org/10.1021/cr900136g>
- Bringmann J, Ebert K, Galla U, Schmieder H (1995) Electrochemical mediators for total oxidation of chlorinated hydrocarbons: formation kinetics of Ag(II), Co(III), and Ce(IV). *J Appl Electrochem* 25:846–851. <https://doi.org/10.1007/BF00233903>
- Cabeza A, Urtiaga A, Rivero MJ, Ortiz I (2007) Ammonium removal from landfill leachate by anodic oxidation. *J Hazard Mater* 144:715–719. <https://doi.org/10.1016/j.jhazmat.2007.01.106>

- Carvalho C, Fernandes A, Lopes A, Pinheiro H, Gonçalves I (2007) Electrochemical degradation applied to the metabolites of acid Orange 7 anaerobic biotreatment. *Chemosphere* 67:1316–1324. <https://doi.org/10.1016/j.chemosphere.2006.10.062>
- Chaplin BP (2014) Critical review of electrochemical advanced oxidation processes for water treatment applications. *Environ Sci Process Impacts* 16:1182–1203. <https://doi.org/10.1039/c3em00679d>
- Chatzisympson E, Xekoukoulotakis NP, Coz A, Kalogerakis N, Mantzavinos D (2006) Electrochemical treatment of textile dyes and dyehouse effluents. *J Hazard Mater* 137:998–1007. <https://doi.org/10.1016/j.jhazmat.2006.03.032>
- Chen G (2004) Electrochemical technologies in wastewater treatment. *Sep Purif Technol* 38:11–41. <https://doi.org/10.1016/j.seppur.2003.10.006>
- Choi JY, Lee YJ, Shin J, Yang JW (2010) Anodic oxidation of 1,4-dioxane on boron-doped diamond electrodes for wastewater treatment. *J Hazard Mater* 179:762–768. <https://doi.org/10.1016/j.jhazmat.2010.03.067>
- Comninellis C (1994) Electrocatalysis in the electrochemical conversion/combustion of organic pollutants for waste water treatment. *Electrochim Acta* 39:1857–1862
- Comninellis C, Kapalka A, Malato S, Parsons SA, Poullos I, Mantzavinos D (2008) Advanced oxidation processes for water treatment: advances and trends for R&D. *J Chem Technol Biotechnol* 83:769–776. <https://doi.org/10.1002/jctb.1873>
- Cossu R, Polcaro AM, Lavagnolo MC, Mascia M, Palmas S, Renoldi F (1998) Electrochemical treatment of landfill leachate: oxidation at Ti/PbO₂ and Ti/SnO₂ anodes. *Environ Sci Technol* 32:3570–3573. <https://doi.org/10.1021/es971094o>
- Da Silva LM, Gonçalves IC, Teles JJS, Franco DV (2014) Application of oxide fine-mesh electrodes composed of Sb-SnO₂ for the electrochemical oxidation of Cibacron marine FG using an SPE filter-press reactor. *Electrochim Acta* 146:714–732. <https://doi.org/10.1016/j.electacta.2014.09.070>
- Daneshvar N, Aber S, Vatanpour V, Rasoulifard MH (2008) Electro-Fenton treatment of dye solution containing Orange II: influence of operational parameters. *J Electroanal Chem* 615:165–174. <https://doi.org/10.1016/j.jelechem.2007.12.005>
- Drouiche N, Aoudj S, Hecini M, Ghaffour N, Lounici H, Mameri N (2009) Study on the treatment of photovoltaic wastewater using electrocoagulation: fluoride removal with aluminium electrodes-characteristics of products. *J Hazard Mater* 169:65–69. <https://doi.org/10.1016/j.jhazmat.2009.03.073>
- Drouiche N, Aoudj S, Lounici H, Drouiche M, Ouslimane T, Ghaffour N (2012) Fluoride removal from pretreated photovoltaic wastewater by electrocoagulation: an investigation of the effect of operational parameters, in: *Procedia engineering*. Pp. 385–391. <https://doi.org/10.1016/j.proeng.2012.01.1218>
- Elliott A, Woodward W (2011) Statistical analysis quick reference guidebook. *Stat Anal Quick Ref Guideb*:191–208. <https://doi.org/10.4135/9781412985949>
- Emamjomeh MM, Sivakumar M (2006) An empirical model for defluoridation by batch monopolar electrocoagulation/flotation (ECF) process. *J Hazard Mater* 131:118–125
- Emamjomeh MM, Sivakumar M (2009) Fluoride removal by a continuous flow electrocoagulation reactor. *J Environ Manag* 90:1204–1212. <https://doi.org/10.1016/j.jenvman.2008.06.001>
- Emamjomeh MM, Sivakumar M, Varyani AS (2011) Analysis and the understanding of fluoride removal mechanisms by an electrocoagulation/flotation (ECF) process. *Desalination* 275:102–106. <https://doi.org/10.1016/j.desal.2011.02.032>
- Essadki AH, Delmas H, Gourich B, Vial C, Bennajah M (2008) Defluoridation of Morocco drinking water by electrocoagulation/electroflotation in an electrochemical external-loop airlift reactor. *Chem Eng J* 148:122–131. <https://doi.org/10.1016/j.cej.2008.08.014>
- Essadki AH, Gourich B, Vial C, Delmas H, Bennajah M (2009) Defluoridation of drinking water by electrocoagulation/electroflotation in a stirred tank reactor with a comparative performance to an external-loop airlift reactor. *J Hazard Mater* 168:1325–1333. <https://doi.org/10.1016/j.jhazmat.2009.03.021>
- Fan Y, Ai Z, Zhang L (2010) Design of an electro-Fenton system with a novel sandwich film cathode for wastewater treatment. *J Hazard Mater* 176:678–684. <https://doi.org/10.1016/j.jhazmat.2009.11.085>
- Farmer JC (2006) Electrochemical treatment of mixed and hazardous wastes: oxidation of ethylene glycol and benzene by silver (II). *J Electrochem Soc* 139:654. <https://doi.org/10.1149/1.2069280>
- Feng Y, Yang L, Liu J, Logan BE (2016) Electrochemical technologies for wastewater treatment and resource reclamation. *Environ Sci Water Res Technol* 2:800–831. <https://doi.org/10.1039/c5ew00289c>
- Florenza X, Solano AMS, Centellas F, Martínez-Huitle CA, Brillas E, García-Segura S (2014) Degradation of the azo dye acid red 1 by anodic oxidation and indirect electrochemical processes based on Fenton's reaction chemistry. Relationship between decolorization, mineralization and products. *Electrochim. Acta* 142:276–288. <https://doi.org/10.1016/j.electacta.2014.07.117>
- Flox C, Garrido JA, Rodríguez RM, Centellas F, Cabot PL, Arias C, Brillas E (2005) Degradation of 4,6-dinitro-o-cresol from water by anodic oxidation with a boron-doped diamond electrode. *Electrochim Acta* 50:3685–3692. <https://doi.org/10.1016/j.electacta.2005.01.015>

- Garcia-Segura S, Brillas E (2014) Advances in solar photoelectro-Fenton: Decolorization and mineralization of the direct yellow 4 diazo dye using an autonomous solar pre-pilot plant. *Electrochim Acta* 140:384–395. <https://doi.org/10.1016/j.electacta.2014.04.009>
- Garcia-Segura S, Centellas F, Arias C, Garrido JA, Rodríguez RM, Cabot PL, Brillas E (2011) Comparative decolorization of monoazo, diazo and triazo dyes by electro-Fenton process. *Electrochim Acta* 58:303–311. <https://doi.org/10.1016/j.electacta.2011.09.049>
- Ghanbari F, Moradi M, Eslami A, Emamjomeh MM (2014) Electrocoagulation/floatation of textile wastewater with simultaneous application of aluminum and Iron as anode. *Environ Process* 1:447–457. <https://doi.org/10.1007/s40710-014-0029-3>
- Ghosh D, Medhi CR, Purkait MK (2008) Treatment of fluoride containing drinking water by electrocoagulation using monopolar and bipolar electrode connections. *Chemosphere* 73:1393–1400. <https://doi.org/10.1016/j.chemosphere.2008.08.041>
- Gürses, A., Yalçın, M., Doar, C., 2002. Electrocoagulation of some reactive dyes: a statistical investigation of some electrochemical variables. *Waste Manag* 22, 491–499. [https://doi.org/10.1016/S0956-053X\(02\)00015-6](https://doi.org/10.1016/S0956-053X(02)00015-6)
- Hammami S, Oturan N, Bellakhal N, Dachraoui M, Oturan MA (2007) Oxidative degradation of direct orange 61 by electro-Fenton process using a carbon felt electrode: application of the experimental design methodology. *J Electroanal Chem* 610:75–84. <https://doi.org/10.1016/j.jelechem.2007.07.004>
- Hassani G, Nasserri S, Gharibi H (2011) Removal of cyanide by electrocoagulation process. *Anal Bioanal Electrochem* 3:625–634
- Holt PK, Barton GW, Mitchell CA (2005) The future for electrocoagulation as a localised water treatment technology. *Chemosphere* 59:355–367. <https://doi.org/10.1016/j.chemosphere.2004.10.023>
- Hu CY, Lo SL, Kuan WH (2005) Effects of the molar ratio of hydroxide and fluoride to Al(III) on fluoride removal by coagulation and electrocoagulation. *J Colloid Interface Sci* 283:472–476. <https://doi.org/10.1016/j.jcis.2004.09.045>
- IBM Corp, 2018. IBM SPSS software [WWW document]. IBM
- Iranifam M, Zarei M, Khataee AR (2011) Decolorization of C.I. basic yellow 28 solution using supported ZnO nanoparticles coupled with photoelectro-Fenton process. *J Electroanal Chem* 659:107–112. <https://doi.org/10.1016/j.jelechem.2011.05.010>
- Irdemez Ş, Demircioğlu N, Yildiz YŞ (2006a) The effects of pH on phosphate removal from wastewater by electrocoagulation with iron plate electrodes. *J Hazard Mater* 137:1231–1235. <https://doi.org/10.1016/j.jhazmat.2006.04.019>
- Irdemez Ş, Demircioğlu N, Yildiz YS, Bingül Z (2006b) The effects of current density and phosphate concentration on phosphate removal from wastewater by electrocoagulation using aluminum and iron plate electrodes. *Sep Purif Technol* 52:218–223. <https://doi.org/10.1016/j.seppur.2006.04.008>
- Irdemez Ş, Yildiz YS, Tosunoğlu V (2006c) Optimization of phosphate removal from wastewater by electrocoagulation with aluminum plate electrodes. *Sep Purif Technol* 52:394–401. <https://doi.org/10.1016/j.seppur.2006.05.020>
- Isarain-Chávez E, De La Rosa C, Godínez LA, Brillas E, Peralta-Hernández JM (2014) Comparative study of electrochemical water treatment processes for a tannery wastewater effluent. *J Electroanal Chem* 713:62–69. <https://doi.org/10.1016/j.jelechem.2013.11.016>
- Juang Y, Nurhayati E, Huang C, Pan JR, Huang S (2013) A hybrid electrochemical advanced oxidation/microfiltration system using BDD/Ti anode for acid yellow 36 dye wastewater treatment. *Sep Purif Technol* 120:289–295. <https://doi.org/10.1016/j.seppur.2013.09.042>
- Kannan, N., Sivadurai, S.N., Berchmans, L.J., Vijayavalli, R., 1995. Removal of phenolic compounds by electrooxidation method. *J. Environ. Sci. Heal. . Part A Environ. Sci. Eng. Toxicol.* 30, 2185–2203. <https://doi.org/10.1080/10934529509376331>
- Kaper H, Engler H (2013) Chapter 9: Regression Analysis. In: Chapter 9: regression analysis, in: mathematics and climate. Society for Industrial and Applied Mathematics, Philadelphia, PA, pp 105–115. <https://doi.org/10.1137/1.9781611972610.ch9>
- Khataee A, Marandizadeh H, Zarei M, Aber S, Vahid B, Hanifehpour YW, Joo S (2013) Treatment of an Azo dye by citrate catalyzed Photoelectro-Fenton process under visible light using carbon nanotube-polytetrafluoroethylene cathode. *Curr Nanosci* 9:387–393. <https://doi.org/10.2174/1573413711309030016>
- Khataee AR, Zarei M, Moradkhannejhad L (2010) Application of response surface methodology for optimization of azo dye removal by oxalate catalyzed photoelectro-Fenton process using carbon nanotube-PTFE cathode. *Desalination* 258:112–119. <https://doi.org/10.1016/j.desal.2010.03.028>
- Khatibikamal V, Torabian A, Janpoor F, Hoshyaripour G (2010) Fluoride removal from industrial wastewater using electrocoagulation and its adsorption kinetics. *J Hazard Mater* 179:276–280. <https://doi.org/10.1016/j.jhazmat.2010.02.089>

- Kötz R, Stucki S, Carcer B (1991) Electrochemical waste water treatment using high overvoltage anodes. Part I: physical and electrochemical properties of SnO₂ anodes. *J Appl Electrochem* 21:14–20. <https://doi.org/10.1007/BF01103823>
- Labiadh L, Oturan MA, Panizza M, Hamadi NB, Ammar S (2015) Complete removal of AHPs synthetic dye from water using new electro-Fenton oxidation catalyzed by natural pyrite as heterogeneous catalyst. *J Hazard Mater* 297:34–41. <https://doi.org/10.1016/j.jhazmat.2015.04.062>
- Lacasa E, Cañizares P, Sáez C, Fernández FJ, Rodrigo MA (2011) Electrochemical phosphates removal using iron and aluminium electrodes. *Chem Eng J* 172:137–143. <https://doi.org/10.1016/j.cej.2011.05.080>
- Landau, S., Everitt, B.S., 2004. A handbook of statistical analyses using SPSS, CHAPMAN & HALL/CRC
- Lissens, G., Pieters, J., Verhaege, M., Pinoy, L., Verstraete, W., 2003. Electrochemical degradation of surfactants by intermediates of water discharge at carbon-based electrodes. *Electrochim Acta* 48, 1655–1663. [https://doi.org/10.1016/S0013-4686\(03\)00084-7](https://doi.org/10.1016/S0013-4686(03)00084-7)
- Mahmoud A, Hoadley AFA, Citeau M, Sorbet JM, Olivier G, Vaxelaire J, Olivier J (2018) A comparative study of electro-dewatering process performance for activated and digested wastewater sludge. *Water Res* 129:66–82. <https://doi.org/10.1016/j.watres.2017.10.063>
- Marinčić L, Leitz FB (1978) Electro-oxidation of ammonia in waste water. *J Appl Electrochem* 8:333–345. <https://doi.org/10.1007/BF00612687>
- Marshall, E., Karadimitriou, S.M., 2015. Outliers, Durbin-Watson and interactions for regression in SPSS, statstutor community project
- de Vidades M, María J, Robles-Molina J, Domínguez-Romero JC, Cañizares P, Sáez C, Molina-Díaz A, Rodrigo MA (2012b) Removal of sulfamethoxazole from waters and wastewaters by conductive-diamond electrochemical oxidation. *J Chem Technol Biotechnol* 87:1441–1449. <https://doi.org/10.1002/jctb.3766>
- de Vidades M, José M, Sáez C, Cañizares P, Rodrigo MA (2012a) Electrolysis of progesterone with conductive-diamond electrodes. *J Chem Technol Biotechnol* 87:1173–1178. <https://doi.org/10.1002/jctb.3742>
- Matsue T (1981) Oxidation of Alkylbenzenes by Electrogenated hydroxyl radical. *J Electrochem Soc* 128: 2565. <https://doi.org/10.1149/1.2127292>
- Mollah MYA, Morkovsky P, Gomes JAG, Kesmez M, Parga J, Cocke DL (2004) Fundamentals, present and future perspectives of electrocoagulation. *J Hazard Mater* 114:199–210. <https://doi.org/10.1016/j.jhazmat.2004.08.009>
- Moussavi G, Majidi F, Farzadkia M (2011) The influence of operational parameters on elimination of cyanide from wastewater using the electrocoagulation process. *Desalination* 280:127–133. <https://doi.org/10.1016/j.desal.2011.06.052>
- Myburgh DP, Aziz M, Roman F, Jardim J, Chakawa S (2019) Removal of COD from industrial biodiesel wastewater using an integrated process: electrochemical-oxidation with IrO₂-Ta₂O₅/Ti anodes and chitosan powder as an adsorbent. *Environ. Process.* 6:819–840. <https://doi.org/10.1007/s40710-019-00401-x>
- Naumczyk, J., Szpyrkowicz, L., Zilio-Grandi, F., 1996. Electrochemical treatment of textile wastewater. *Water Sci Technol* 34. [https://doi.org/10.1016/S0273-1223\(96\)00816-5](https://doi.org/10.1016/S0273-1223(96)00816-5), 17, 24
- Olvera-Vargas H, Oturan N, Aravindakumar CT, Paul MMS, Sharma VK, Oturan MA (2014) Electro-oxidation of the dye azure B: kinetics, mechanism, and by-products. *Environ Sci Pollut Res* 21:8379–8386. <https://doi.org/10.1007/s11356-014-2772-4>
- Oturan MA, Aaron J-J (2014) Advanced oxidation processes in water/wastewater treatment: principles and applications. *A Review Crit Rev Environ Sci Technol* 44:2577–2641. <https://doi.org/10.1080/10643389.2013.829765>
- Picard T, Cathalifaud-Feuillade G, Mazet M, Vandenstendam C (2000) Cathodic dissolution in the electrocoagulation process using aluminium electrodes. *J Environ Monit* 2:77–80. <https://doi.org/10.1039/a908248d>
- Polcaro AM, Palmas S (1997) Electrochemical oxidation of Chlorophenols. *Ind Eng Chem Res* 36:1791–1798. <https://doi.org/10.1021/ie960557g>
- Polcaro AM, Palmas S, Renoldi F, Mascia M (1999) On the performance of Ti/SnO₂ and Ti/PbO₂ anodes in electrochemical degradation of 2-chlorophenol for wastewater treatment. *J Appl Electrochem* 29:147–151. <https://doi.org.ezproxy.cc.lut.fi/10.1023/A:1003411906212>
- Pouet, M.F., Grasmick, A., 1995. Urban wastewater treatment by electrocoagulation and flotation. *Water Sci Technol* 31, 275–283. [https://doi.org/10.1016/0273-1223\(95\)00230-K](https://doi.org/10.1016/0273-1223(95)00230-K)
- Pulkka S, Martikainen M, Bhatnagar A, Sillanpää M (2014) Electrochemical methods for the removal of anionic contaminants from water – a review. *Sep Purif Technol* 132:252–271. <https://doi.org/10.1016/j.seppur.2014.05.021>
- Rajalo G, Petrovskaya T (1996) Selective electrochemical oxidation of sulphides in tannery wastewater. *Environ Technol (United Kingdom)* 17:605–612. <https://doi.org/10.1080/09593331708616424>
- Rajkumar D, Palanivelu K (2004) Electrochemical treatment of industrial wastewater. *J Hazard Mater* 113:123–129. <https://doi.org/10.1016/j.jhazmat.2004.05.039>

- Ruiz EJ, Hernández-Ramírez A, Peralta-Hernández JM, Arias C, Brillas E (2011) Application of solar photoelectro-Fenton technology to azo dyes mineralization: effect of current density, Fe^{2+} and dye concentrations. *Chem Eng J* 171:385–392. <https://doi.org/10.1016/j.cej.2011.03.004>
- Sala M, Gutiérrez-Bouzán MC (2014) Electrochemical treatment of industrial wastewater and effluent reuse at laboratory and semi-industrial scale. *J Clean Prod* 65:458–464. <https://doi.org/10.1016/j.jclepro.2013.08.006>
- Salazar R, García-Segura S, Ureta-Zañartu MS, Brillas E (2011) Degradation of disperse azo dyes from waters by solar photoelectro-Fenton. *Electrochim Acta* 56:6371–6379. <https://doi.org/10.1016/j.electacta.2011.05.021>
- Sales Solano AM, Costa de Araújo CK, Vieira de Melo J, Peralta-Hernandez JM, Ribeiro da Silva D, Martínez-Huitle CA (2013) Decontamination of real textile industrial effluent by strong oxidant species electrogenerated on diamond electrode: viability and disadvantages of this electrochemical technology. *Appl Catal B Environ* 130–131:112–120. <https://doi.org/10.1016/j.apcatb.2012.10.023>
- Solano AMS, García-Segura S, Martínez-Huitle CA, Brillas E (2015) Degradation of acidic aqueous solutions of the diazo dye Congo red by photo-assisted electrochemical processes based on Fenton's reaction chemistry. *Appl Catal B Environ* 168–169:559–571. <https://doi.org/10.1016/j.apcatb.2015.01.019>
- Subbarama Raju P, Sudhakar C, Umamohan C (2009) Chenchus and social transformation: a study of a primitive tribe in Kurnool district of Andhra Pradesh. *Anthropologist* 11:167–172
- Tang, W.Z., Wang, F., Miralles-Wilhelm, F., Darnisse, E., 2009. Uncertainty analysis of rating equations of submerged orifice flow at gated spillway, in: conference on reliability and quality in design. The International Society of Science and Applied Technologies (ISSAT) and the IEEE reliability society, San Francisco, pp. 165–169
- Tezcan Un U, Koparal AS, Bakir Ogutveren U (2013) Fluoride removal from water and wastewater with a batch cylindrical electrode using electrocoagulation. *Chem Eng J* 223:110–115. <https://doi.org/10.1016/j.cej.2013.02.126>
- Thiam A, Zhou M, Brillas E, Sirés I (2014) Two-step mineralization of Tartrazine solutions: study of parameters and by-products during the coupling of electrocoagulation with electrochemical advanced oxidation processes. *Appl Catal B Environ* 150–151:116–125. <https://doi.org/10.1016/j.apcatb.2013.12.011>
- Tsantaki E, Velegraki T, Katsaounis A, Mantzavinos D (2012) Anodic oxidation of textile dyehouse effluents on boron-doped diamond electrode. *J Hazard Mater* 207–208:91–96. <https://doi.org/10.1016/j.jhazmat.2011.03.107>
- Tuan PA, Mika S, Pirjo I (2012) Sewage sludge electro-dewatering treatment-a review. *Dry Technol* 30:691–706. <https://doi.org/10.1080/07373937.2012.654874>
- Valero D, García-García V, Expósito E, Aldaz A, Montiel V (2014) Electrochemical treatment of wastewater from almond industry using DSA-type anodes: direct connection to a PV generator. *Sep Purif Technol* 123: 15–22. <https://doi.org/10.1016/j.seppur.2013.12.023>
- Wang CT, Chou WL, Kuo YM, Chang FL (2009) Paired removal of color and COD from textile dyeing wastewater by simultaneous anodic and indirect cathodic oxidation. *J Hazard Mater* 169:16–22. <https://doi.org/10.1016/j.jhazmat.2009.03.054>
- Wang J, Farrell J (2004) Electrochemical inactivation of triclosan with boron doped diamond film electrodes. *Environ. Sci. Technol.* 38:5232–5237. <https://doi.org/10.1021/es035277o>
- Yavuz Y, Ögütveren B (2018) Treatment of industrial estate wastewater by the application of electrocoagulation process using iron electrodes. *J Environ Manag* 207:151–158. <https://doi.org/10.1016/j.jenvman.2017.11.034>
- Zhang S, Zhang J, Wang W, Li F, Cheng X (2013) Removal of phosphate from landscape water using an electrocoagulation process powered directly by photovoltaic solar modules. *Sol Energy Mater Sol Cells* 117: 73–80. <https://doi.org/10.1016/j.solmat.2013.05.027>
- Zhu J, Zhao H, Ni J (2007) Fluoride distribution in electrocoagulation defluoridation process. *Sep Purif Technol* 56:184–191. <https://doi.org/10.1016/j.seppur.2007.01.030>
- Zhu X, Ni J, Lai P (2009) Advanced treatment of biologically pretreated coking wastewater by electrochemical oxidation using boron-doped diamond electrodes. *Water Res* 43:4347–4355. <https://doi.org/10.1016/j.watres.2009.06.030>

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