Design and Evaluation of Electrocoagulation System for the Treatment of Effluent from Recycled Paper Production

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Effluent found in the pulp and paper industry can cause considerable damage if it is discharged untreated, because of the high biochemical and chemical oxygen demands. Electrocoagulation is a physicochemical process widely used in industrial wastewater treatment. The removal of different pollutants depends on the sample type and operating conditions. The aim of this research was to evaluate the efficiency of an electrocoagulation system for COD removal from recycled paper production effluent *via* aluminum and iron electrodes. Different operational parameters, such as the electrolysis time (5 min to 15 min), current density (7 A/m^2 to 11 A/m^2), and distance between each electrode (5 mm to 20 mm), were evaluated. The turbidity, total suspended solids, chlorides, sulfates, and COD had removal efficiencies of 92.7%, 91.3%, 70.4%, 66.6%, and 64%, respectively. A polynomial model was generated to estimate the optimum conditions for COD removal. The optimum times for the current densities 7 A/m^2, 8 A/m^2, 9 A/m^2, 10 A/m^2, and 11 A/m^2 were 39.5 min, 39.5 min, 35.7 min, 34.1 min, and 32.8 min, respectively, with a 15-mm electrode gap.

Keywords: Electrocoagulation; Prototype; Recycled paper production; Wastewater

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INTRODUCTION

In paper mills, paper is made from wood, pulp, or recycled paper (Latorre *et al.* 2005). Recycled paper is an important raw material in the paper production, making it the second most common ingredient (Bajpai 2017). Recycled paper contains chemicals from additives, inks, glues, *etc*. or by cross-contamination from other waste materials during collection, which are eliminated through the wastewater (Pivnenko *et al.* 2015). Therefore, there are significant difference in the composition of the wastewaters depending on the raw material used.

The right choice of treatment can be difficult (Young and Akhtar 1998). In the past, the most used physicochemical treatments were sedimentation-flotation, coagulation-precipitation, filtration, reverse osmosis, adsorption, and ozonation (Kamali and Khodaparast 2015; Ordaz-Diaz *et al.* 2017).

Soloman *et al.* (2009) mentioned that electrochemical methods are more technically and economically feasible for a large-scale operation compared with other physicochemical treatment techniques. The cost of chemical coagulation has been found to be 3.2 times as high as the operating cost of electrocoagulation (Kobya *et al.* 2007).

Electrocoagulation is more effective when dealing with high molecular weight dissolved organic matter than with low molecular weight compounds (Lewis *et al.* 2013). Furthermore, electrocoagulation does not require the addition of reagents, which can save on operational and environmental costs (Khansorthong and Hunsom 2009). Aluminum and iron electrodes release Al^{+3} and Fe^{+2} ions, respectively, which promotes coagulation and flotation/precipitation (Fu and Wang 2011).

Aluminum anode, produces the cationic monomeric species according to the following reactions (Modirshahla *et al.* 2007):

$$Al \leftrightharpoons Al^{3+}_{(aq)} + 3e^- \quad (1)$$

$$Al^{3+}_{(ac)} + 3H_2O \rightleftharpoons Al(OH)_3 + 3H^+ \quad (2)$$

$$n\ Al(OH)_3 \rightarrow Al_n(OH)_{3n} \quad (3)$$

When the Fe^{2+} is dissolved in wastewater by Fe oxidation at the anode, the following reaction is carried out (Daneshvar *et al*. 2003; Zodi *et al.* 2009),

$$Fe \rightarrow Fe^{2+} + 2e^- \quad (4)$$

And the hydroxide ion and H_2 gas are generated at the cathode:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_{2(g)} \quad (5)$$

The production of hydroxide causes an increase in pH during electrolysis, and the formation of insoluble $Fe(OH)_2$ favors the coagulant precipitation (Brillas and Martínez-Huitle 2015).

Simultaneous application of Al-Fe anode has been successfully employed in textile wastewater (Ghanbari *et al.* 2014a) and for nitrate removal (Ghanbari *et al.* 2014b). Aluminum and iron compared with other ions, favors the coagulation process with lower coagulant concentration (Garcia-Segura *et al.* 2017).

Electrocoagulation has been employed previously in the treatment of paper industry wastewaters from the pulping of wood fibers (Sharma *et al.* 2014; Asaithambi 2016; Buchanan 2017; Chen *et al.* 2017) and from recycled fibers (Behrooz *et al.* 2011; Izadi *et al.* 2018).

The removal of different pollutants with these methods is strongly dependent on the operational conditions (Kamali and Khodaparast 2015) and sample type (Al-Shannag *et al.* 2012). Hart *et al.* (2012) note that models are used to make predictions and explain phenomena under different conditions. Hence, it is very important to estimate the optimum conditions for COD removal on recycled paper production effluent owing there is no studies have reported.

The aim of this work was to evaluate the efficiency of an electrocoagulation prototype process for the internal treatment of wastewater from the recycled papermaking process via aluminum and iron electrodes. Electrode gaps, current density, and reaction times on the COD removal efficiencies were studied to determine the optimum process conditions. To estimate the optimum conditions for COD removal, a predictive model was generated.

EXPERIMENTAL

Electrocoagulation System Design, Construction, and Operation

The electrocoagulation system was a BATCH-type reactor (2-L capacity). It consisted of 10 electrolytic cells, and the electrodes were square with a rectangular flange. The dimensions of the electrodes are given in Fig. 1. The electrodes had a thickness of 5 mm. The cathode was made of aluminum and iron oxide. The cells consisted of four areas: sedimentation, flotation, reaction, and circulation (Fig. 1b). The electrodes were arranged in parallel (10-mm spacing) to lower the potential difference between the electrodes (Groterud and Smoczyński 1986), and they were connected in series. All of the anodes were connected at a single point, and all of the cathodes were connected at another point (Fig. 1c). The reaction area was where the electrical transfer between the electrodes and solution occurred. The sedimentation area allowed flocs to precipitate and accumulate without clogging the reaction area. Bubbling occurred in the circulation area and caused water circulation between the electrodes. The reactor consisted of 10 electrodes in total, where five were aluminum and five were iron (Fig. 1c).

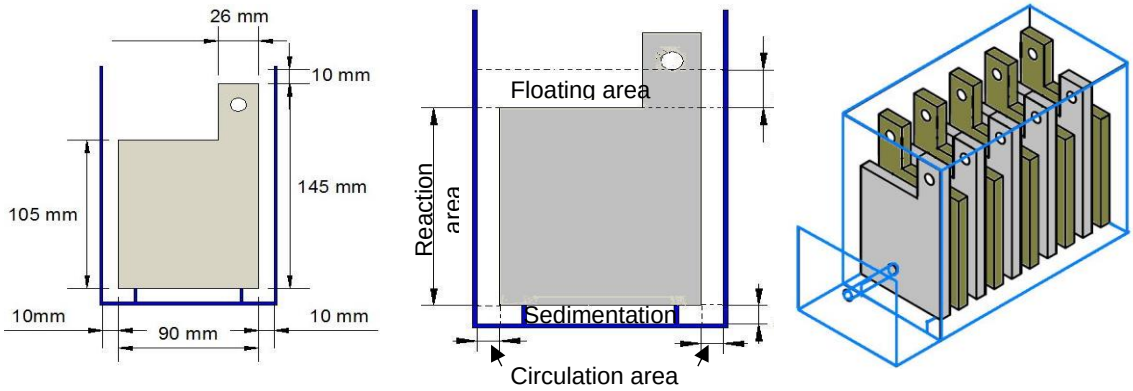

Fig. 1. a) Electrode dimensions; b) cell areas: reaction, sedimentation, flotation, and circulation; and c) electrode distribution inside of the reactor and configuration

Laboratory-scale experiments were conducted with a rectangular vessel using a laboratory direct current (DC) power supply (TEKTRONIX PS280, Oregon, USA). All of the experiments were performed at room temperature (20 °C) (Katal and Pahlavanzadeh 2011). The duration of electrolysis was up to 15 min. A multimeter (M1750, Elenco, IL, USA), non-conductive material cell (acrylic), and Cayman cables were used.

Sampling and Electrocoagulation Experiments

The samples were collected before their arrival at the wastewater treatment plant for recycled paper production located in northern Mexico. The samples were stored at 4 °C before use. The initial characterization is presented in Table 1.

To evaluate the electrocoagulation efficiency, the experiment was conducted with the electrocoagulation prototype. The water volume was 2 L and the tests were done at room temperature. The electrode gaps were 5 mm, 10 mm, and 20 mm, the current densities were 7 A/m^2, 8 A/m^2, 9 A/m^2, 10 A/m^2, and 11 A/m^2, and the reaction times were 5 min, 8 min, 12 min, and 15 min. A current density of approximately 10 A/m^2 was recommended

by Pouet *et al.* (1991). After the different reaction times, the samples were allowed to settle for 60 min.

Table 1. Physicochemical Characterization of the Samples

Parameter	Value
pH	7.95 ± 0.13
Conductivity (µS/cm)	2022 ± 14
Chlorides (mg/L)	77 ± 5.5
Hardness as $CaCO_3$ (mg/L)	451 ± 15
Alkalinity (mg/L)	790 ± 20
Sulfates (mg/L)	232 ± 15
Turbidity (NTU)	26.7 ± 2.9
TSS (mg/L)	247 ± 12
COD (mg/L)	4441.8 ± 10

TSS – total suspended solids; COD – chemical oxygen demand

Methods

Analytical methods

In the initial and final characterizations, the following parameters were measured. The electrical conductivity (EC) and pH were determined *in situ* with an HQ40d Portable device (HACH, Loveland, CO, USA). The total suspended solids (TSS) were analyzed gravimetrically. The chemical oxygen demand (COD) was measured using the closed reflux (colorimetric) method described in section 5220-D of APHA (2005), and the turbidity was determined *via* a spectrophotometric method that analyzed the water and wastewater (Rice *et al.* 2012). The total hardness as $CaCO_3$, alkalinity, and chloride content were measured *via* the volumetric method (Adams 2017). Using a photometer (HI83225, HANNA Instruments Inc., Woonsocket, RI, USA), the sulfate levels were determined.

Statistical analysis

During the tests, three factors were studied: the electrode gap, current density, and reaction time. A general full factorial design with different levels was used. The data analysis was performed using Statistica 7 software (StatSoft Inc., Oklahoma City, OK, USA). For modelling and optimization of the parameters, 60 runs were considered, and each experimental condition was evaluated in duplicate.

RESULTS AND DISCUSSION

Parameter Removal

For the electrode gaps of 5 mm, 10 mm, and 20 mm, the maximum turbidity removals were 92.7%, 89.5%, and 91.8%, respectively (Fig. A1 in the Appendix). At 20 mm of electrode gap and a reaction time of 5 min it was possible to reach 90% removal. In the study by Behrooz *et al.* (2011) it took twice as long to achieve similar results. Camcioglu *et al.* (2017) studied wastewater from wood fibers, where the maximum turbidity removal of more than 99% was achieved with twice electrolysis time.

For the electrode gaps of 5 mm, 10 mm, and 20 mm, the maximum removals of TSS were 66.7% (Fig. A2a), 91.3% (Fig. A2b), and 89.3% (Fig. A2b), respectively. These results reveal that this study was better, due to the fact that less reaction time is required. For example, Camcioglu *et al.* (2017), using an EC-Fe system, predicted 70% of the TSS

would be removed after 25 min to 35 min of electrolysis. Jaafarzadeh *et al.* (2016) revealed that a TSS removal efficiency of 100% was obtained, and optimized the process *via* a statistical model. Uğurlu *et al.* (2008) used Fe electrodes and achieved a TSS removal efficiency of 77%.

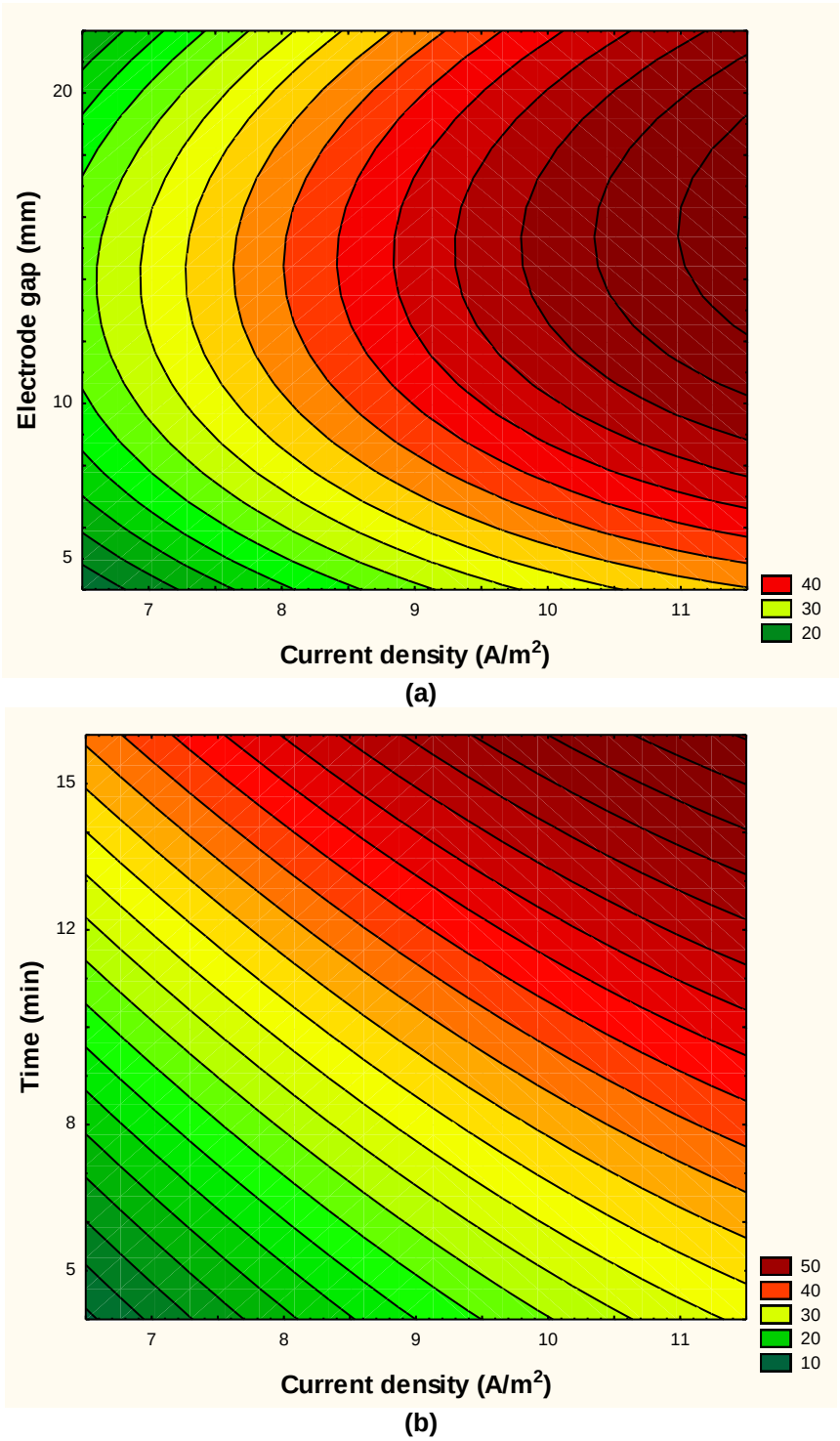

Fig. 2. 3D contour plot of the COD removals: (a) current density *vs.* electrode gap and (b) time

For chloride removal using a 5-mm electrode gap (Fig. A3a), it was possible to reach 70.4% removal. The maximum chloride removal efficiency with a 10-mm electrode gap (Fig. A3b) was 66%, and a 20-mm electrode gap achieved a maximum removal efficiency of 65.21%.

The maximum sulfate removal efficiency was 66.7% for a 5-mm electrode gap (Fig. A4a), 52.6% for a 10-mm gap (Fig. A4b), and 52.2% for a 20-mm gap (Fig. A4b).

In Fig. A5, the COD removal was 45%, 61%, and 64% with the 5-mm, 10-mm, and 20-mm gaps, respectively. In this study, using an Al-Fe electrode configuration (initial COD = 444.2 mg/L at 11 A/m^2), better removals were achieved than those reported by Zodi *et al.* (2011) (initial COD = 285 mg/L at 100 A/m^2). Camcioglu *et al.* (2017) used an Al electrode system, which showed an increase of 31.7% in the COD removal efficiencies. Jaafarzadeh *et al.* (2016) revealed that the highest and lowest COD removal rates were 60.1% and 38.0%, respectively. Khansorthong and Hunsom (2009) used electrocoagulation in the continuous mode with a current density of 20.7 A/m^2 for 45 min and obtained a removal efficiency of 91.2% for the COD and 89.8% for the TSS. Uğurlu *et al.* (2008) used Al electrodes with a current density of 77.13 mA and an electrolysis time of 2 min and obtained a COD removal efficiency of 75%.

The results indicated that an electrode gap of 10 mm to 20 mm (Fig. 2a) achieved the highest COD removal efficiencies. Furthermore, exceeding 20 mm decreased the removal efficiency. A gap less than 10 mm prevented the movement of the electrolyte in the system, which affected the removal efficiency (Mahesh *et al.* 2006). Figures 2a and 2b show that the highest COD removal levels were found with a current density greater than 10 A/m^2, a run time greater than 15 min (Fig. 2b), and an electrode gap of 15 mm (Fig. 2a). This was because the current density affects the number of ions produced from the electrodes (Al-Shannag *et al.* 2012). However, with these times it was not possible to achieve a COD removal of 100%.

Statistical Analysis

Table 2 shows the correlation of the physicochemical parameters studied. It was observed that the COD removal efficiency was related to most of the parameters. There was a high correlation between the COD removal efficiency and the TSS removal efficiency (91%), hardness decrease (61%), and sulfate removal efficiency (53%).

Table 2. Correlation of the Parameters

Variable	Conductivity	Chlorides	Hardness as $CaCO_3$	Alkalinity	Sulfates	Turbidity	TSS	COD
Conductivity	1.00	0.23*	0.72*	0.15	0.56*	0.24*	0.77*	0.84*
Chlorides		1.00	0.39*	-0.02	0.50*	0.20*	-0.03	0.09
Hardness as $CaCO_3$			1.00	0.11	0.66*	0.15	0.51*	0.61*
Alkalinity				1.00	-0.02	0.26*	0.33*	0.31*
Sulfates					1.00	0.30*	0.41*	0.53*
Turbidity						1.00	0.20*	0.26*
TSS							1.00	0.91*
COD								1.00
* Correlations are significant at $p < 0.05$; N = 119								

Model Estimation

The increase in the COD removal efficiency reflected a decrease in the other parameters. Therefore, COD is the appropriate variable to understand the behavior of the system. The model that explained the COD removal behavior in the *electrocoagulation* systems had an adjusted coefficient of determination (R^2) of 0.92 (Table 3).

Table 3. Test of the SS Whole Model *vs.* SS Residual (COD Removal)

Dependent	Variable	COD Removal
Multiple	*R*	0.962809
Multiple	R^2	0.927001
Adjusted	R^2	0.923091
SS	Model	14215.81
df	Model	6
MS	Model	2369.301
SS	Residual	1119.453
df	Residual	112
MS	Residual	9.995112
F		237.046
p		0

The univariate significance test (Table 4) showed that the current density, electrode gap distance, and time had a significant difference with a *p* less than 0.05. Therefore, they were important factors in the COD removal efficiency under these experimental conditions.

The model was validated, and it was observed that it did not violate any of the assumptions. The residuals behaved normally (Fig. A6); therefore, the interpretation and inferences of the model were reliable (Razali and Wah 2011). The plot of the dispersion of residuals against the predicted amount presented a cloud of points without a pattern (random behavior) (Fig. A7). The residues were not self-correlated, so they were independent (Table A1).

Table 4. Univariate Tests of Significance for the COD (Sigma-restricted Parameterization Effective Hypothesis Decomposition)

Effect	SS	Degrees of Freedom	MS	*F*	*p*
Intercept	225.422	1	225.422	22.5532	0.000006
Current Density	94.471	1	94.471	9.4517	0.002650
Current Density^2	32.022	1	32.022	3.2038	0.076172
Electrode Gap	1051.359	1	1051.359	105.1873	0.000000
Electrode Gap^2	791.550	1	791.550	79.1937	0.000000
Time	145.240	1	145.240	14.5311	0.000226
Time^2	0.237	1	0.237	0.0237	0.878020
Error	1119.453	112	9.995		

The behavior of the electrocoagulation process for COD removal *via* estimation of the model parameters is explained in Table 5.

Table 5. Parameter Estimates (Sigma-restricted Parameterization)

Effect	COD Param.	COD Std. Err.	COD *t*	COD *p*	-95.00% Cnf. Lmt.	+95.00% Cnf. Lmt.
Intercept	-67.4511	14.20314	-4.74903	0.000006	-95.5928	-39.3094
Current Density	9.6361	3.13434	3.07437	0.002650	3.4258	15.8464
Current Density^2	-0.3107	0.17358	-1.78990	0.076172	-0.6546	0.0332
Electrode Gap	3.3236	0.32406	10.25609	0.000000	2.6815	3.9657
Electrode Gap^2	-0.1111	0.01248	-8.89908	0.000000	-0.1358	-0.0863
Time	2.1216	0.55657	3.81197	0.000226	1.0189	3.2244

The equation that explains the COD removal behavior of the system is shown as Eq. 6,

$$COD\ removal = -67.4511 + 9.6361CD - 0.3107CD^2 + 3.3236EG - 0.1111EG^2 + 2.1216T + 0.0042T^2 \quad (6)$$

where CD is the current density (A/m^2), EG is the electrode gap (mm), and T is the time (min).

Using Eq. 6, it was possible to estimate the optimum time to remove 100% of the COD (Fig. 2) at an optimum electrode gap of 15 mm. The optimum time was similar when using 10-mm and 20-mm electrode gaps. The optimum time values for a 15-mm electrode gap and different current densities were 32.8 min to 39.5 min (Fig. 3). These times were lower than those reported by Sharma *et al.* (2014) (120 min at 15 A/m^2) and Asaithambi (2016) (120 min). Perng and Wang (2016) reported lower times (16 min), but the density they reported exceeded the density of this work by more than eight times.

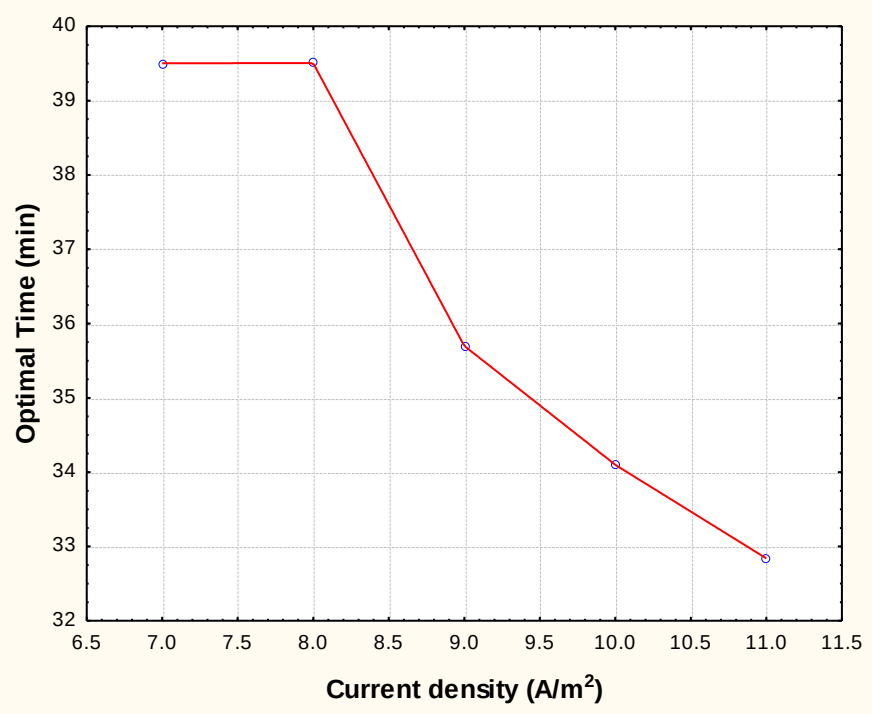

Fig. 1. Optimum time in relation to the current density

CONCLUSIONS

1. The prototype electrocoagulation system demonstrated a significant level of efficacy. The model generated made it possible to estimate the optimum time to remove 100% of the COD. Furthermore, there was a high correlation between the COD removal efficiency and the TSS removal efficiency (91%).
2. The optimum operating conditions were a current density of 7 A/m^2 to 11 A/m^2 and a 15-mm electrode gap.
3. After using electrocoagulation in the treatment of wastewater, these can be used in boilers once they go through a softening process, making it necessary to perform analyzes that demonstrate that they can be reused by reducing the hardness completely.

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APPENDIX

The Appendix containing 8 pages with 7 figures and 1 table.

Parameters removal

Figure A1a shows that with a separation of 5 mm it was possible to remove 84.2 to 92.7% of the turbidity. When the separation was 10 mm, the maximum turbidity removal was 76.5 to 89.5% (Figure A1b). The maximum turbidity removal with a separation of 20 mm was 66.7 to 91.8% (Figure A1c).

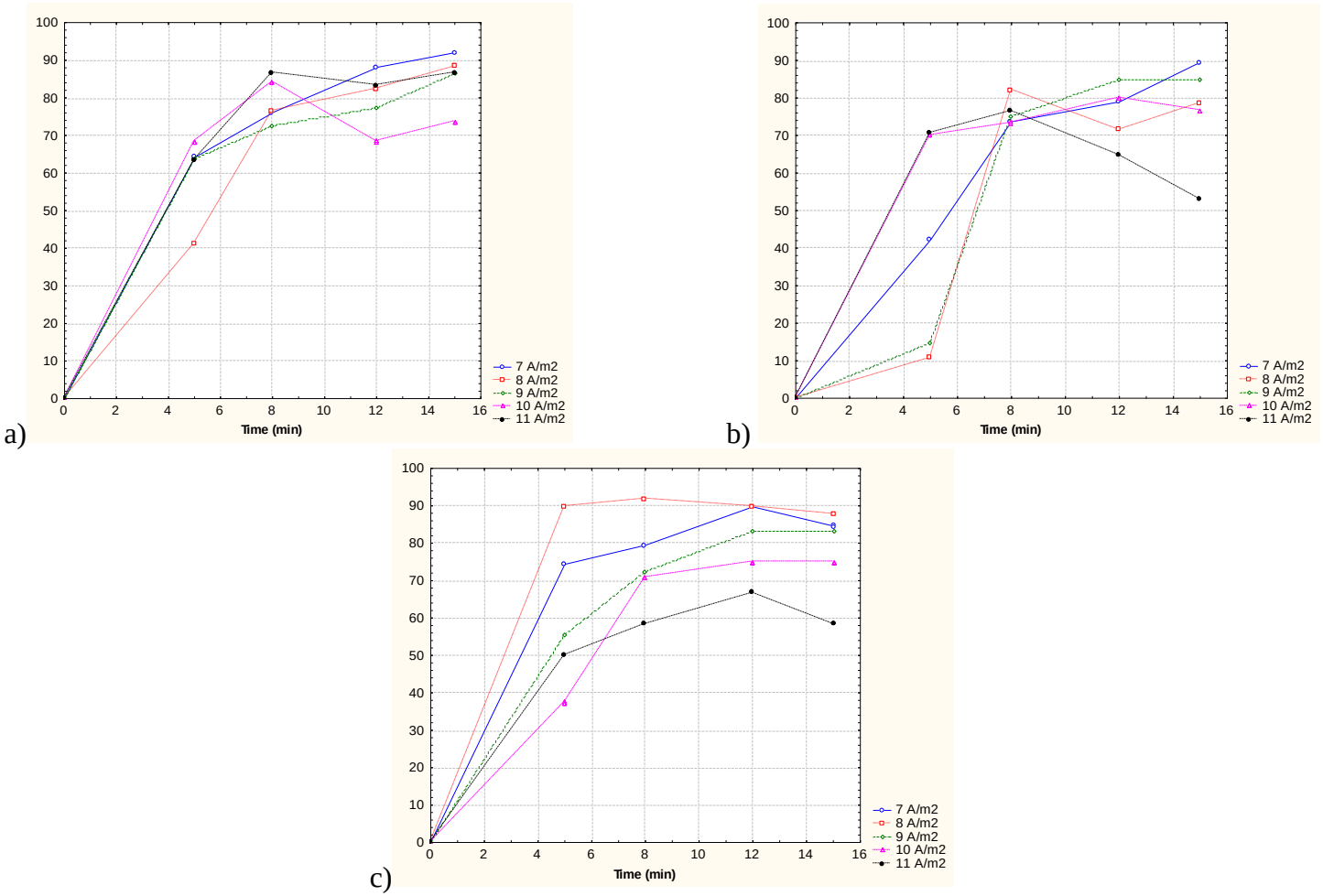

Fig. A1. Turbidity removal respect to time at a) 5 mm b) 10 mm and c) 20 mm of electrode gap

In the removal of the TSS, a linear behavior was observed that has a positive linear relationship with respect to time. With the electrode gap of 5, 10, and 20 mm the maximum removals were reached on a range of 42.8 to 66.7, 53.3 to 91.3, and 67.8 to 89.3, respectively.

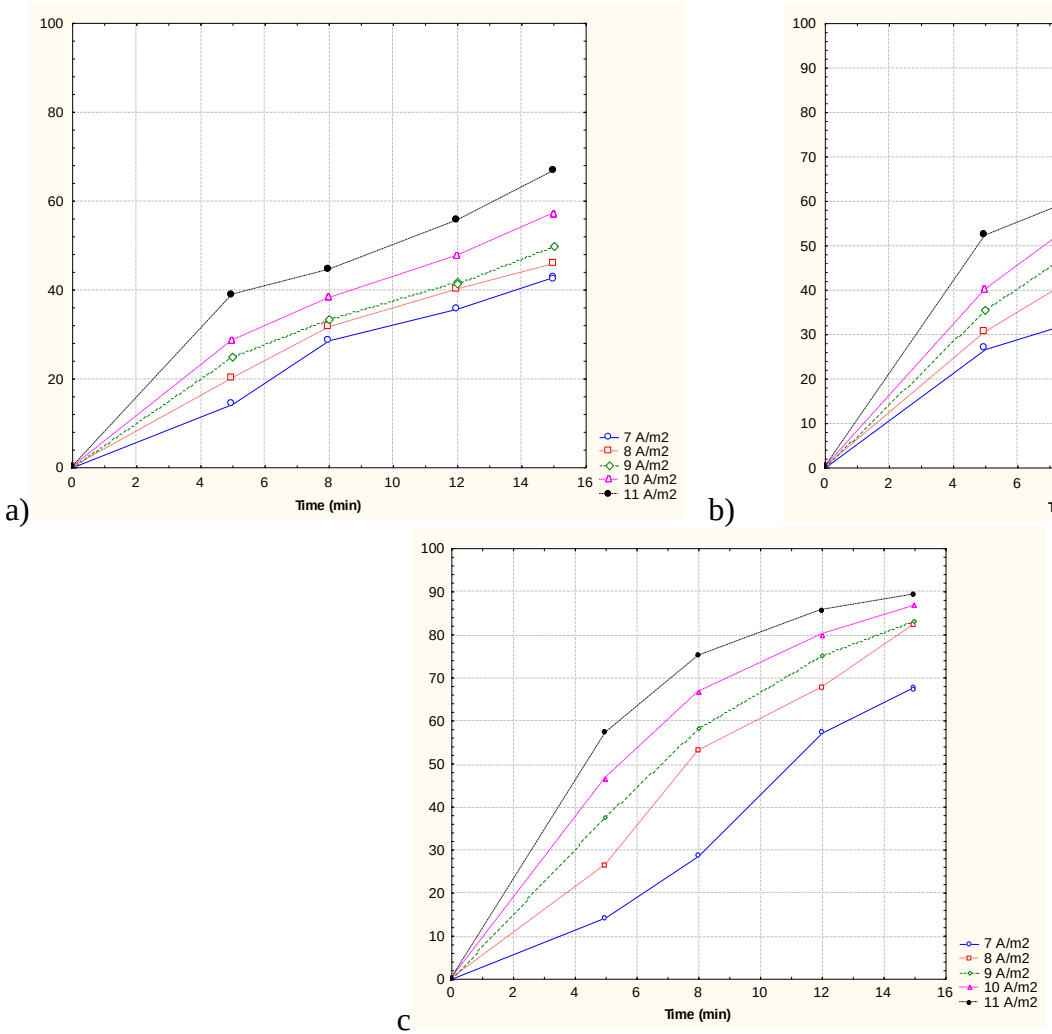

Fig. A2. Decreasing of TSS respect to time at a) 5 mm b) 10 mm and c) 20 mm of electrode gap

For the removal of chlorides using a 5 mm electrode gap (Fig. A3a) it was possible to reach 53.1 to 70.4%. The maximum chlorides removal range with a 10 mm electrode gap (Fig. A3b) was 40 to 66%. And for the electrode gap of 20 was 27.0 to 65.2%.

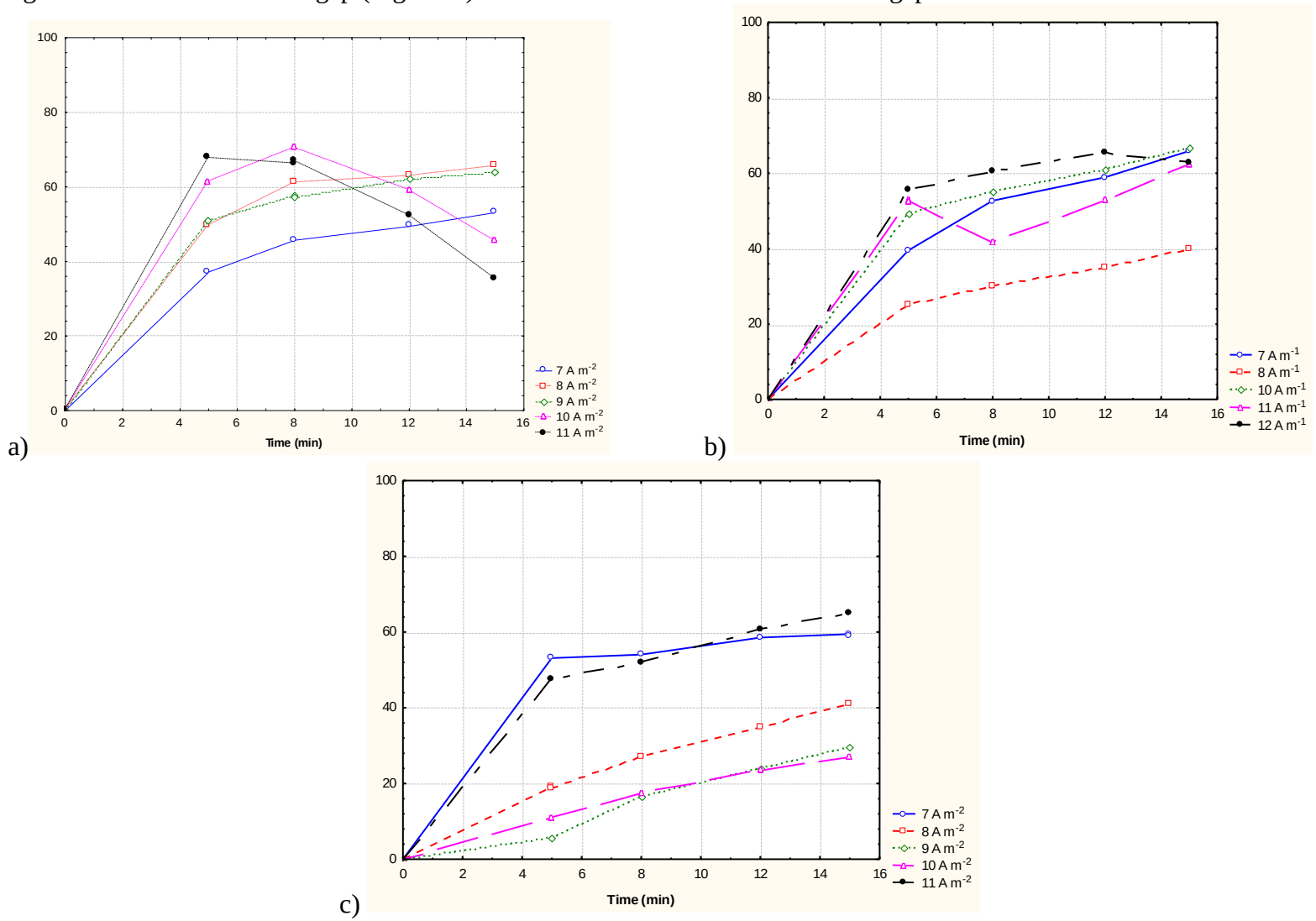

Fig. A3. Chlorides removal respect to time at a) 5 mm b) 10 mm and c) 20 mm of electrode gap

Sulfate removal was 42.9 to 66.7% for a 5 mm electrode gap (Fig. A4a), 46.6 to 52.6% for 10 mm (Fig. A4b) and 27.3 to 52.2% for 20 mm (Fig. A4b).

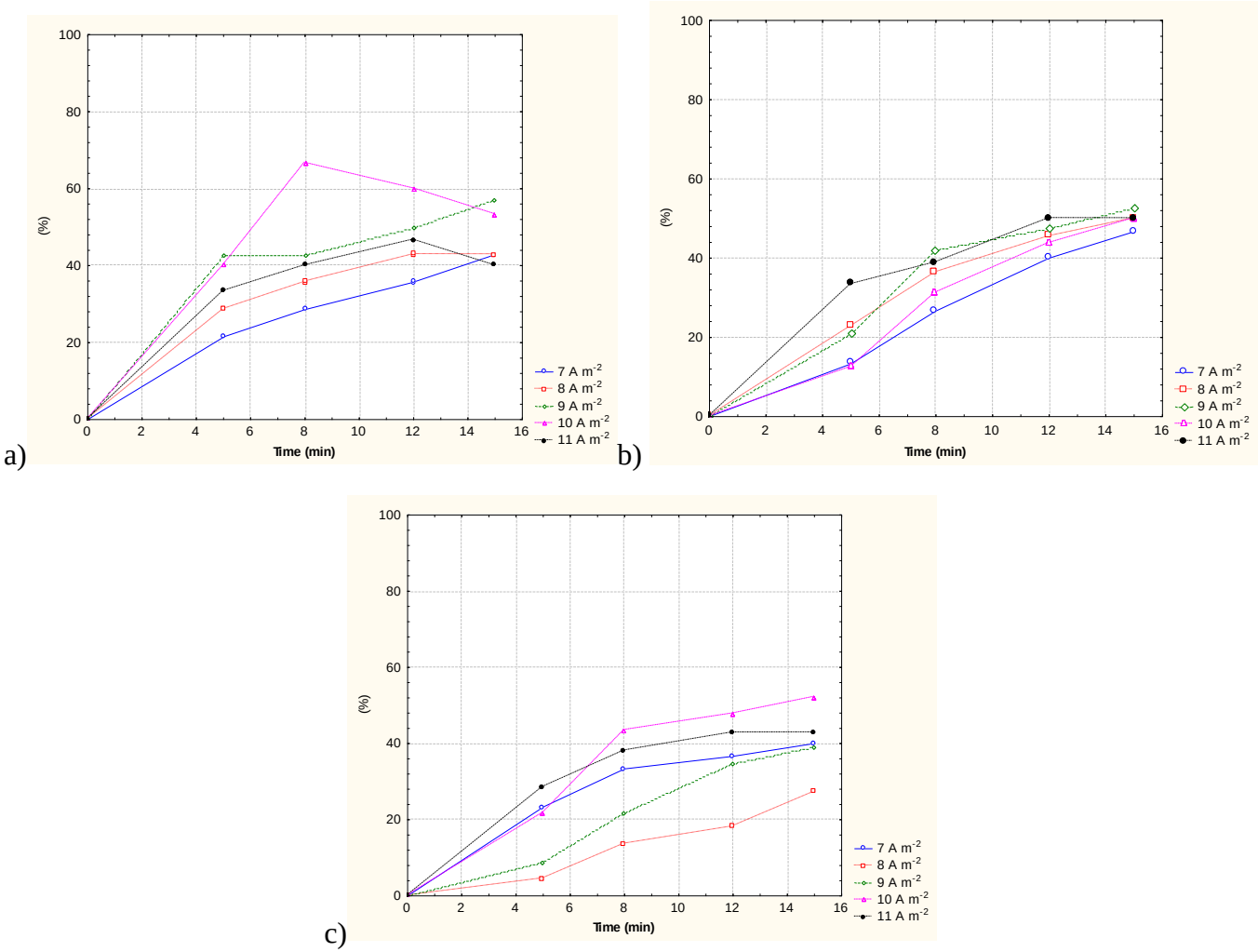

Fig. A4. Sulfates removal respect to time at a) 5 mm b) 10 mm and c) 20 mm of electrode gap

In the Fig. A5, the COD removal was observed at 5, 10 and 20 mm, reaching 36.5 to 45.4, 41.5 to 61 and 34.1 to 64%, respectively.

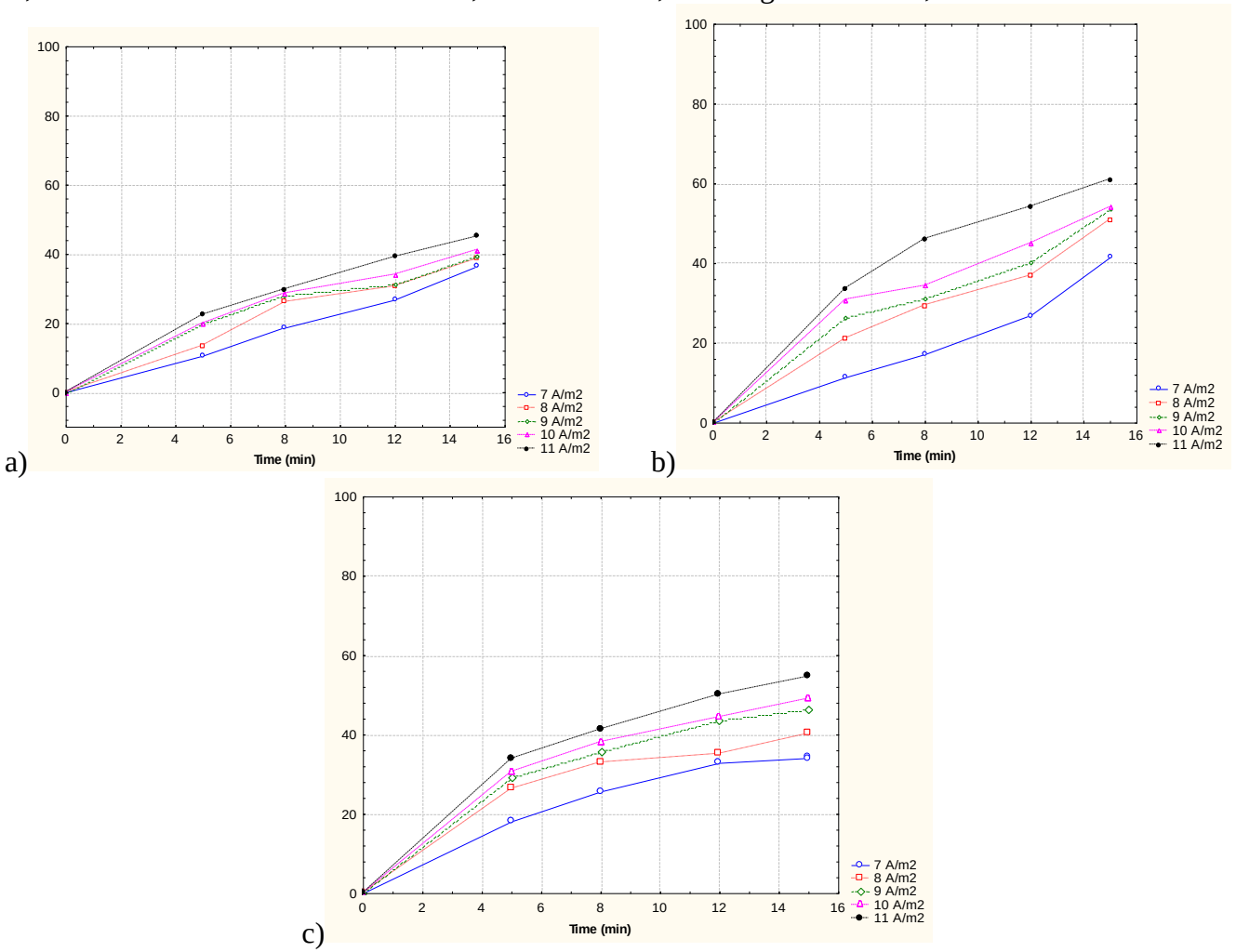

Fig. A5. COD removal efficiency with respect to time at a) 5 mm b) 10 mm and c) 20 mm of electrode gap

2 Model validation

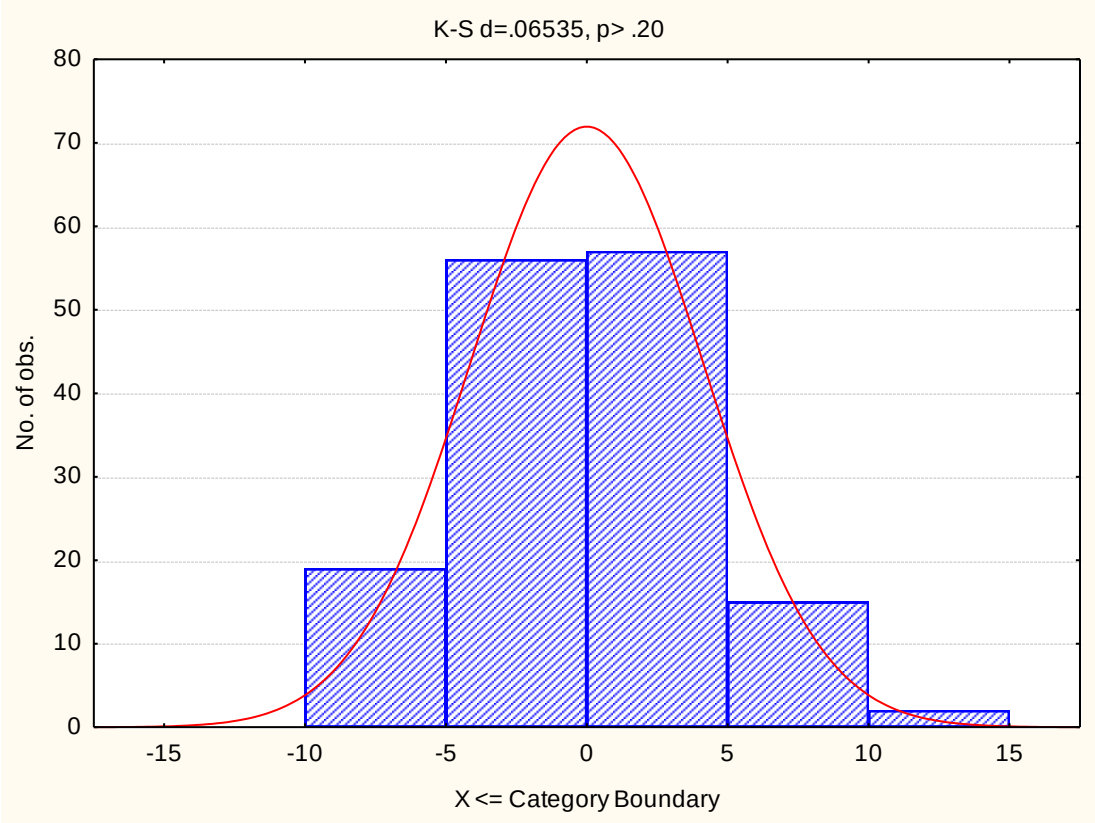

Fig. A6. Kolmogorov-Smirnov test, normality of residuals with a level of significance of 0.05

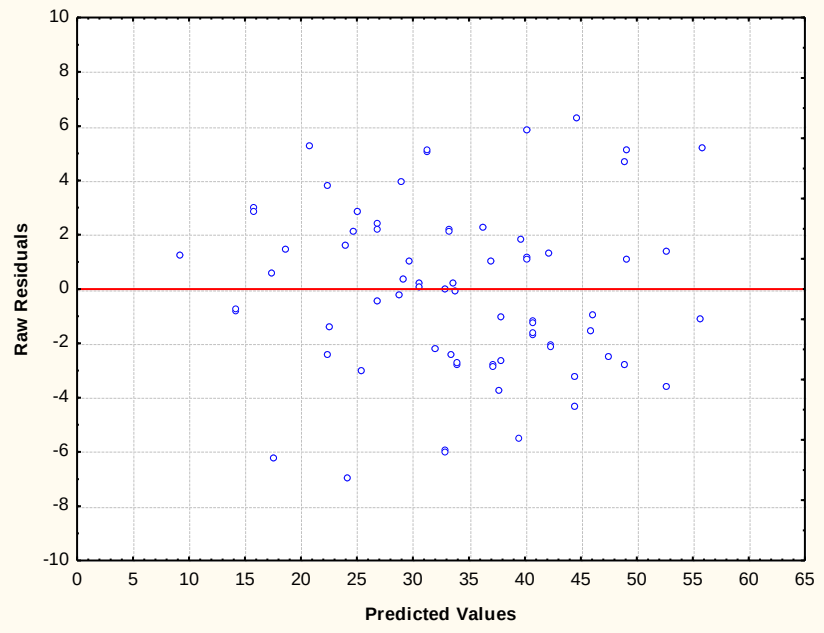

Fig. A7. Predicted vs. Residual Values. Dependent variable: COD removal

Table A1. Correlations*

Variable	Predicted	Residuals
Predicted	1.00	0.00
Residuals	0.00	1.00

* Marked correlations are significant at p < .05000. N=119 (Casewise deletion of missing data)