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A comparative study of hydrogen peroxide oxidation and electrocoagulation using aluminum, iron, and zinc electrodes for urban sludge disintegration

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ABSTRACT

Municipal and industrial wastewater treatment plants produce large amounts of sludge containing high levels of organic, chemical, and microbial pollutants. Unless stabilized completely and discharged safely, they may become potential pollution sources threatening soil and water bodies. This study investigated H₂O₂ oxidation and electrocoagulation as pretreatments to improve stabilization of an urban sludge. The H₂O₂ oxidation was optimized with respect to H₂O₂ dosage and initial sludge pH-H₂O₂. Batch electrocoagulation experiments were conducted using aluminum, iron, and zinc electrodes to investigate the effect of treatment period, current density, and pH. The effectiveness was compared in terms of solubilization of sludge, disintegration degree, and reduction of total solids. Sludge settling velocity after disintegration by both H₂O₂ oxidation and electrocoagulation were measured with respect to the operating conditions. The obtained results indicated that the high rate of sludge disintegration (63.3%) was obtained with aluminum electrodes, which has lower operating costs than iron and zinc electrodes. The H₂O₂ oxidation reached a maximum disintegration degree of 50%. Additionally, with aluminum and iron electrodes, sludge settleability was enhanced with both H₂O₂ oxidation and electrocoagulation.

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Urban sludge; disintegration; H₂O₂; electrocoagulation; sludge settleability; energy consumption

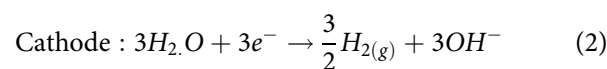
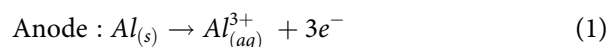
Introduction

Wastewater sludge consists of high rates of organic, chemical, and microbial pollutants that may threaten the health of soil and water bodies, if not stabilized appropriately and discharged safely. In Algeria, about 550 tons of dry sludge are produced daily,^[1] with an increased production rate expected, because of more stringent criteria for wastewater plant effluent treatment and the construction of additional wastewater treatment plants to service Algeria's growing population. Both drivers are representative of global trends of international concern. Worldwide, disposal costs for dewatered sludge can be half of a plant's total management costs (including operational energy, personnel, and ordinary maintenance expenditures).^[2] Arguably sludge minimization is a more sustainable approach than sludge recycling or re-use. Therefore, there is growing interest in methods to reduce sludge volume and mass and, therefore, reduce environmental pollution.

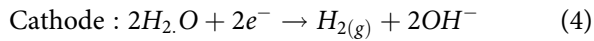
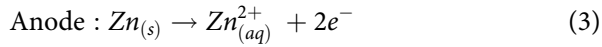
Over the past decades, several innovative treatment processes have attracted significant attention for their potential suitability to alter the structure and composition of sludge biomass and, hence, enhance sludge stabilization and production. Pretreatment disintegration

methods in which biological cells are broken down or lose cohesion can be achieved by sonication,^[3] acid/alkaline treatment,^[4] chemical oxidation,^[5–7] and thermal treatment,^[8] and electrochemical processes such as electrokinetics,^[9] electro-oxidation,^[10] electrolysis^[11] and advanced oxidation process (AOP) are gaining widespread adoption or sludge digestion and disintegration,^[9–12] as summarized by both Ushani et al. (2022)^[13] and Qian et al. (2022).^[14] Among the electrochemical processes, electrocoagulation (EC) processes appear the most promising in removing pollutants from waste water.^[15,16] EC employs in-situ generation of coagulants that are provided from the sacrificial metallic anode dissolution, while hydrogen gas is released simultaneously at the cathode. Electrode material can be aluminum,^[16] graphite,^[17] zinc,^[18] or iron.^[15] Selection depends on cost, oxidation potential, and targeted pollutant.

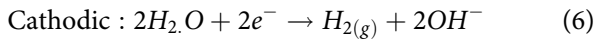
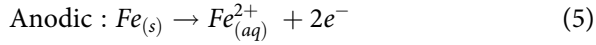
(i) Reactions with aluminum anodes:



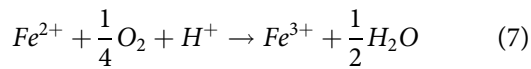
(ii) Reactions with zinc anode:



(iii) When iron is used as anode, the reactions are as follows:



Generally, the ferrous form exists in water, in the presence of oxygen and a pH below 6.5 and immediately undergoes oxidation reactions^[19]:



Under adequate pH conditions,^[20] the resulting Al^{3+} and Zn^{2+} and Fe^{3+} ions are immediately hydrolyzed to yield the corresponding polyhydroxides or hydroxides and effectively destabilize the pollutants. The eradication mechanism may be through charge neutralization, sweep coagulation, or adsorption. Destabilized pollutants, can then be separated from the treated solution either by flotation^[21] or settling.^[22]

Recent EC studies on wastewater sludge monitored with aluminum,^[23] iron^[24] under various current density, treatment time, and pH condition reported encouraging results, but the influence of electrode material on organic matter solubilization and the mechanism involved through EC was not considered. Furthermore, while wastewater EC treatment with aluminum and iron are have been studied significantly, comparatively, few studies included zinc, despite its reported ability to remove wastewater pollutants at high rates (e.g., urea up to 66%), improve chemical oxygen demand (COD) [50%],^[18] and enhance phenolic content (84.2%).^[25] Yet, zinc electrodes for sludge disintegration by electrocoagulation has not been reported alone, nor in direct comparison to aluminum or iron.

Among chemical methods for sludge disintegration, chemical oxidation is common^[5,6,7]. Hydrogen peroxide is an environmentally friendly oxide that can oxidate organic pollutants efficiently and economically.^[24] Glaze et al. (1990),^[26] showed the effectiveness of H_2O_2 for disinfection, as it destroys pathogenic organisms in wastewater without noxious or polluting byproducts^[27] However, safety handling of H_2O_2 requires use of low concentrations, adequate ventilation, and temperature control. No previous studies on sludge disintegration have consider H_2O_2 oxidation and electrocoagulation in a side-by-side study. The work

presented herein aims to address this gap and to compare electrode materials (aluminum, iron and zinc) in electrocoagulation. The comparison considers soluble SCOD reduction, disintegration degree (DD), total solid, (TS) and sludge settleability (SVI). Operating costs of both hydrogen peroxide oxidation and electrocoagulation.

Sludge sampling and characterization

For this study, waste sludge was collected from a municipal urban wastewater treatment plant located in Annaba, Algeria. The sludge sample was a mixture of a primary sludge and a biologic sludge stored at 4°C. Using standard analytical methods,^[28] the sludge was found to contain 30.84 g/L of total solid (TS), an average 3063 mg/L total chemical oxygen demand (TCOD), and 510 mg/l of soluble chemical oxygen demand (SCOD), as well as 1.13 g.L⁻¹ of volatile matter. The sludge's natural pH was 7.46, conductivity 1.34 mS.cm⁻¹, and sludge volume index 120 mL/g.

Analytical methods

The sludge samples were centrifuged at 150 rpm for 10 min. Then the supernatant was filtrated with 0.45 µm filters. Next the COD of the filtrate was analyzed and characterized by its SCOD. Sludge solids obtained by centrifugation and filtration treatment were used for TS testing. Sludge disintegration efficiency was evaluated by disintegration degree DD_{COD} (%), directly informs the part of the particulate substances solubilized by treatment, as per Muller (2000).^[29]

$$DD_{COD}(\%) = \frac{SCOD - SCOD_0}{TCOD - SCOD_0} * 100 \quad (8)$$

where, $SCOD_0$ and $SCOD$ are soluble COD concentrations in the raw sludge and disintegrated sludge, respectively. Solubilization indicates the transfer of COD from the particulate fraction of sludge to the soluble fraction. $TCOD$ is the soluble COD concentration in the sludge after alkali hydrolysis. This was determined for the sludge samples by mixing 0.5 mol/l NaOH for 24 h followed by filtering using a 0.45 µm round filter to separate the soluble part. The filtrate's COD was measured as the $TCOD$.^[30] The COD and $TCOD$ analyses were determined by titration, after a total digestion with H_2SO_4 and potassium dichromate in a heated MARCONI® DRY BLOCK MA 4004 digester block at 150°C for 2 h. Absorbance was read on a spectrophotometer GBC® model UV/VIS 911A at 600 nm.^[28] Before COD measurement, the unreacted (residual) H_2O_2 in the pre-treated sludge was quenched

using MnO_2 to prevent the positive interference of H_2O_2 on the COD analysis.^[31] Three replicates of each experimental condition were performed with the sample analysis in triplicate to ensure the reproducibility of results. The average of the values was reported.

Hydrogen peroxide oxidation

Hydrogen peroxide oxidation was investigated through a series of bench-scale experiments were conducted with a sample volume of 100 mL. A mechanical stirrer was used to ensure sufficient mixing. The mixing speed was maintained at 200 rpm, which dispersed the reagents and prevented the sludge from settling. An amount of 10 ml of Hydrogen peroxide (35%) was used at 4 different concentrations (0.4, 0.8, 1.2 and 1.6 M) as an additive to the sludge. For the experiments on the effect of pH on sludge reduction, the initial pH of the sludge was modified to be 3.0, 5.0, 9, 10.5, and 12, by adding 1N sodium hydroxide (NaOH) and 1N sulfuric acid (H_2SO_4). The experiments were conducted at an ambient temperature of 25°C. At the end of the 2 hours of reaction, samples were collected, settled and filtered. The solid residue was then dried overnight at 105°C to obtain the total solid concentration measurement (TS).

Electrocoagulation

For comparison, electrocoagulation of the wastewater sludge (WWS) effluent was conducted in a 100 mL beaker in a batch mode as described in the authors' previous study.^[16] Preliminary experimental tests showed that adding 0.25 g of KCl added to the sludge was reasonable to adjust sludge conductivity. The pH was adjusted to the desired value (3, 5, real pH = 7.46, 10.5 and 12) using H_2SO_4 and NaOH solutions. For testing, a pair of electrodes (aluminum, iron, or zinc) with the dimensions 12 cm × 3 cm × 1 cm and an effective surface area of 13.63 cm² were separated by 1.5 cm and dipped in the wastewater. The current was provided by a direct current power supply (GW GPR3030D).

The effluent under treatment was homogenized by gentle magnetic stirring at 300 rpm, which allowed the separation of gasses formed from the solution, thus avoiding the formation of foam, which can affect the batch processing. After testing, the electrodes were sponge washed and then cleaned using hydrochloric acid solution (15%) after each experiment.

Settling tests

By definition, settling is the separation by gravity of two immiscible phases, of which at least one is liquid. Sludge settleability can be expressed by means of the sludge volume index (SVI).^[32] SVI is often recommended for characterization of sludge

formation and is defined by considering a particular point of the settling curve, namely $t = 30$ min as follows:

$$\text{SVI} \left[\frac{\text{mL}}{\text{g}} \right] = \frac{V_{30}}{\text{TS}} \quad (9)$$

where V_{30} (ml/l) is the volume of settled sludge after 30 min of settling, and TS is the total amount of solid waste remaining (g/l) after the disintegration treatment period. Sludge with good settling properties has an SVI of 100 mL/g. In the presence of bulking, this can reach 500 mL/g.^[32]

Estimation of operating cost

Operating costs is a critical parameter in treatment method selection, as it determines the viability of that method.

For the H_2O_2 oxidation treatment, the total operating cost was calculated at the optimum conditions included the costs of the reagents (H_2O_2 and NaOH) and the electricity consumed by the device (magnetic stirrer of 640 W) during the 2 hours. In the Algerian market in March 2023, the electrical energy price was \$0.038 kWh for the first 125 kWh and \$0.03473/kWh beyond 125 kWh.

For lab-scale electrocoagulation units, the cost of energy and electrode material are the major operating expenses, which can be calculated using the following equation^[33]:

$$\begin{aligned} \text{Operating cost} &= \text{Energy}_{\text{consumption}} \\ &+ \text{Electrode}_{\text{consumption}} \\ &+ \text{Chemical}_{\text{consumption}} \end{aligned} \quad (10)$$

The electrode material price was \$1.5/kg (aluminum), \$1.27/kg (iron), and \$3.12/kg (zinc). Specific electrical energy consumption SEEC is calculated using Eq 11.

$$\text{SEEC} \left[\frac{\text{KWh}}{\text{m}^3} \right] = \frac{IUt}{V} \quad (11)$$

where I is the current intensity (A), U is the cell voltage (V), t is the operating time (H), and V is the liquid volume (m³), and the amount of aluminum, iron and zinc in solution is calculated using Eq12.

$$m = \frac{ItM}{zF} \quad (12)$$

where m is the released coagulants from the anode (g), M is the atomic weight of the electrodes ($M = 26.98$ g/mol for Al, 55.84 g/mol for Fe and 65.38 g/mol for Zn), z is the number of electrons transferred in the anodic dissolution, F is the Faraday constant (96486 C.mol⁻¹), and t is the electrolysis time in seconds.

Statistical analysis

Multivariate analysis of variance MANOVA test was conducted to examine the significance of the effects of electrode type on DD, SCOD and TS. This was followed by a Bonferroni post-hoc analysis for multiple comparisons. The MANCOVA test was then applied as part of a multivariate analysis of the covariance to determine significance of the effects of electrode type on DD, SCOD and TS after controlling for time treatment, current density and pH. A p -value $< .05$ was considered statistically significant. This analysis was performed using SPSS Statistics software version 26 SPSS (IBM, New York, USA).

Results and discussion

Sludge disintegration by H_2O_2 oxidation

Effects of hydrogen peroxide oxidation

To examine the influence of H_2O_2 dosages on sludge disintegration, the runs were conducted by varying dosages of 0.6 to 2 M at an initial pH of 7. Figure 1a illustrates how the disintegration efficiencies improved gradually from 18.57% to 32.86%, as the H_2O_2 dosage was increased from 0.6 to 1.4 M. Notably, beyond an H_2O_2 dosage of 1.4 M organic matter solubilization was not improved. These results confirmed that H_2O_2 oxidation could solubilize the particulate component of sludge into the soluble form. Devi et al. (2016),^[34] attributed the cell-killing power of H_2O_2 to the oxidation of intracellular constituents. Song and Hiraoka. (1992),^[35] related the enhancement of sludge biodegradability to the intermediates produced from ramified aliphatic chain in hydrogen peroxide oxidation, among

others short-chain carboxylic acid, which are easily degraded.

Previous studies by Feki et al. (2015) and Gogate and Pandit. (2004),^[36,37] showed sludge disintegration efficiency decreases with increasing levels of H_2O_2 .^[36,37] In the latter study 1.4 M was also found to be an upper bound. This value is within the range of similar works.^[5,35] Kenge et al. (2009)^[38] recommended a H_2O_2 /sludge ratio of 0.8.

The effects of different H_2O_2 dosages on sludge solid content after treatment are shown in Fig. 1b. For dosages of H_2O_2 smaller or equal to 1.4 M, there were improvements in the TS reduction. SCOD production was also enhanced in these conditions, thereby showing a reasonable connection between the destruction of TS and the increase in SCOD (Fig. 1a). The increase in H_2O_2 up to 1.4 M inhibited this improvement and organic compounds did not solubilize and, thus, remained in the sludge.

Effect of pH

Different pH levels were tested in the presence of 1.4 M of H_2O_2 (the previously established optimum dose for disintegration of wastewater sludge). Figure 2a indicates that acidification did not have a favorable effect on the disintegration efficiency, and the DD value did not exceed 21.8%. However, when alkaline values of pH were used, the DD of the sludge increased to 32.86%, 41.73% and 50.00% for pH values of 7.00, 10.5 and 12.00, respectively. This is explained by H_2O_2 being stable under acidic conditions but active under alkaline conditions where it decomposes easily.^[39] This outcome is consistent with previously

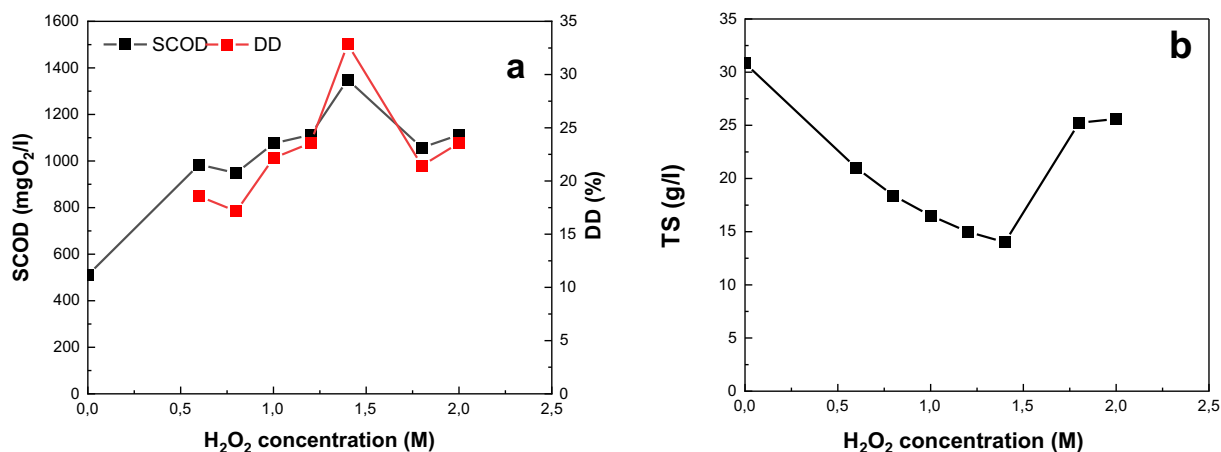


Figure 1. The change in a) SCOD and DD b) TS at different H_2O_2 concentration. Operating conditions: $[H_2O_2] = 0.4, 0.8, 1.2$ and 1.6 M, treatment time = 2 h, $pH_{\text{natural}} = 7.46$

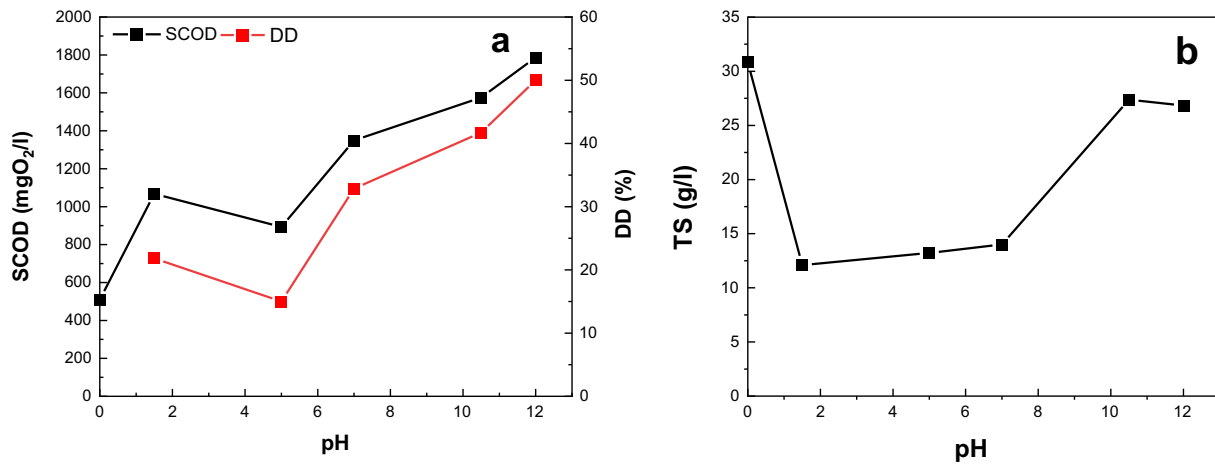


Figure 2. pH dependent change of a) COD and DD b) TS. Operating conditions: pH = 3, 5, pH_{natural} = 7.46, 9, 10.5 and 12, [H₂O₂] = 1.4 M, treatment time = 2 H

published results by Kim et al. (2009)^[5] and Ksibi (2006)^[40] who showed that an alkaline medium enhanced solubilization of wastewater by using H₂O₂. This is because the alkaline decomposed the lipids, hydrocarbon, and proteins into smaller soluble substances such as aliphatic acids, polysaccharides, and amino acids.^[41,42] Chen et al. (2007)^[43] compared the effectiveness of both acidic and alkali treatment of wastewater sludge and found that alkali conditions were more effective than acidic one. They also observed that the soluble proteins and carbohydrates were the most important components of the SCOD and that their concentrations were also higher at alkaline pH conditions.

As seen in Figure (2a, 2b), alkaline conditions led to a higher production of SCOD, while acidic one promoted TS diminution due to the negatively charged colloidal matter (including the bacterial cells in the sludge)^[44] reacting with the H⁺ brought to the medium which disturbs their stability and leads to their destruction and, consequently, TS decreases. Previous studies reported that a low pH was found to be favorable for the release of heavy metals, phosphorus, and ammonia and decreases in TS.^[45,46]

However, at pH equal to or above 7, the protons OH⁻ dominate, which creates high electrostatic repulsion. The repulsion causes desorption of some parts of the extracellular polymers which leads to the augmentation of TS. Similar results were obtained by Li et al. (2008),^[47] when 0.05 M of Ca(OH)₂ was used for sludge disintegration, where the weight of the sludge increased by 24.3%. Monje et al. (2021)^[48] also reported that in high alkaline conditions, particulates COD and precipitates of inorganic compounds are retained in the solid content of the sludge thereby increasing the TS amount.

Sludge disintegration by electrocoagulation

Effect of treatment period and electrodes material on SCOD, DD and TS

Figure 3 illustrates the effects of treatment period and electrode material selection on SCOD, DD and TS. Under a constant current density of 14.67 mA/cm² and a pH of 7.48, SCOD was analyzed at 5–10–15–20–25–30–45–60–90–120–150 min periods, and the accompanying DD values were calculated to determine the optimum reaction time. SCOD increase is a key indicator of the pretreatment performance,^[49] as shown in Fig. 3a. When the treatment period increased, the solubilization efficiencies were enhanced up to 60 min (Fig. 3a); according to Asaithambi et al. (2016),^[45] lengthening the reaction time resulted in higher removal efficiency due to the generation of hydroxyl ions. SCOD and DD values reached 1075.86 mg/L and 28.44% for aluminum and 837.64 mg/L and 14.69% for iron electrodes. In comparison to the EC treatment with aluminum and iron, the zinc electrodes provided the worst treatment outcomes. At 14.67 mA applied current for 60 min, the highest SCOD and DD levels observed were only 689.98 mg/L and 3.39%, respectively.

The optimum treatment time was established as 60 min. At longer treatment times, the SCOD concentration remained stable. A similar observation was reported by Erkan and Engin (2020)^[50] and Erden. (2019).^[51] These results may be attributable to the additional deposition of cations and sludge particles on the surface of electrode material which form an oxide layer, which would inhibit the dissolution of metal and, thus, reduce electrode dissolution with additional treatment time.^[52,53] However, Mechelhoff et al. (2013)^[54] showed that such phenomena could be avoided under certain

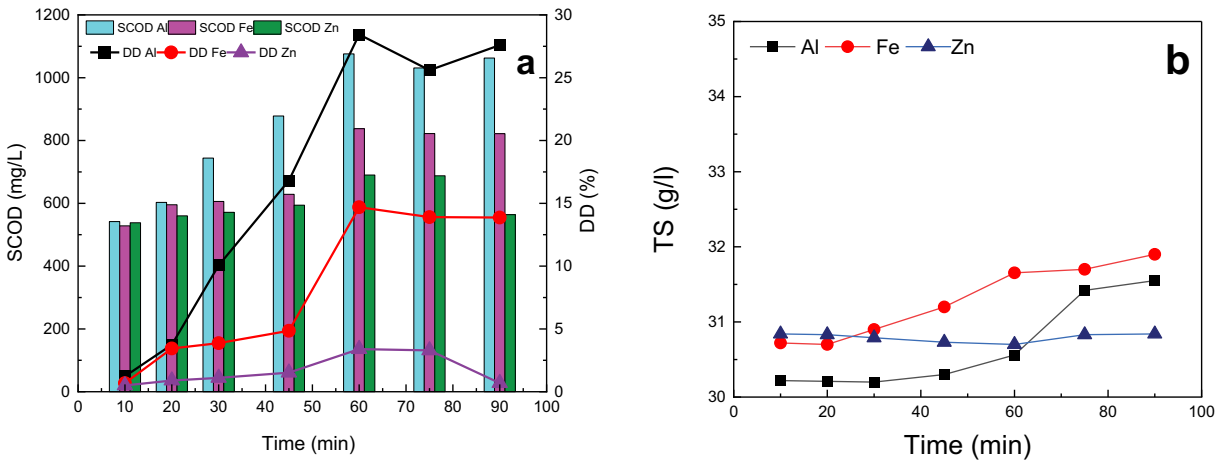


Figure 3. Effect of treatment period and electrodes material on: a) SCOD and DD b) TS. Operating conditions: treatment time = 90 min, current density = 14.67 mA/cm², pH_{natural} = 7.46.

working conditions such as the appropriate KCl concentration, surface electrode selection, and water composition.

Figure 3b illustrates the aluminum electrode's production of more total flocs after 30 min compared to the iron and zinc electrodes, thereby yielding more TS effluent. The use of the zinc electrodes led to a slight reduction of TS.

Effect of current density and electrodes material on SCOD, DD, TS and SEEC

Applied current density is the major operating parameter governing the coagulant production rate, oxygen evolution, heat generation, and bubble production rate. Hence, the current density controls the EC process performance and operating costs.^[55] The effects of current density and electrode material on SCOD, DD, and TS are seen in Fig. 4. The results indicate that the

current density has a positive influence on the solubilization of sludge. After 60 min, the SCOD concentration significantly increased the treatment efficiency (SCOD and DD). Higher currents during EC process increased the total ions produced as Al³⁺, Fe²⁺ or Zn²⁺. Therefore, the metal hydroxide formation rate grew with the increase in the metal ion quantities, which dissolved into the solution and led to the pollutant adsorption on the surface of the hydroxides, oxyhydroxides, and metal oxides.^[56] Destabilization of pollutants can also be attributed to the phenomena of the compression of the double layer, neutralization of the charge, and lastly, floc creation.^[56] Furthermore, the increase in current density resulted in more hydrogen bubble production, which leads to the elimination of the pollutant through the flotation phenomenon. These trends were consistent with previously reported results, which suggest a direct

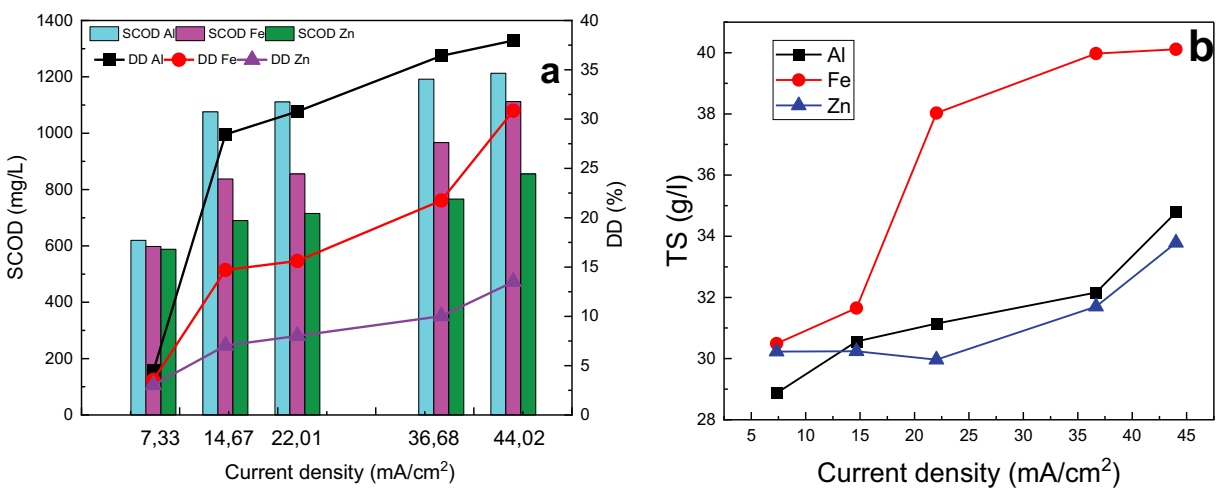


Figure 4. Effect of current density and electrodes material on: a) SCOD and DD, b) TS. Operating conditions: treatment time = 60 min, current density (mA/cm²) = 7.33, 14.67, 22.01, 36.68, 44.02, pH_{natural} = 7.46

relationship between applied current density and sludge solubilization.^[57] Zeng et al (2019),^[57] showed that the increase in the SCOD concentration is attributed to the intensive disruption effects of electrocoagulation, resulting in the liberation of intracellular and/or extracellular compounds from the sludge to the solution.

For a current density of 44 mA/cm², the COD and DD were 1212.92 mg/L and 37.96%, respectively with aluminum electrodes, 1112.33 mg/l and 30.84% with iron electrodes, and 855.33 mg/L and 13.5% with zinc electrodes (Fig. 4a). In most studies to date, aluminum electrodes were reported as more efficient than iron ones in term of harvesting efficiency due to the greater charge of Al (3+) is than Fe (2+).^[20,58] In addition, the initial sludge pH (6.48) encourages precipitation of the aluminum hydroxides more than iron. For the zinc, minimum values of SCOD and DD were obtained even at the highest applied currents compared to those of Al and Fe. According to Pourbaix (1996),^[58] the precipitation of Zn(OH)₂ would only be significant at pH level equal or greater than 8.6. Thus, the amount of coagulants in the water is insufficient to destabilize the sludge compounds.

Figure 4b illustrates the evolution of TS as a function of the applied current density for the three types of electrodes. Under these operating conditions (natural pH = 7.48), electrode material selection was more notable on the SCOD and DD than on the TS reduction. TS content grew by increasing current density.

The proposed mechanism for the electrocoagulation sludge treatment is shown in Fig. 5. This includes (i): cationic ions generated from metallic anode (Mⁿ⁺) combined with negative surface of colloidal matter of sludge,^[44] composed principally of fatty acids, extracellular polysaccharides, compounds with nitrogen and bacteria cells,^[59] which leads to its destabilization and dissolution into dissolved proteins, lipids, carbohydrates, humic substances^[60,61]; and (ii): Mⁿ⁺ hydrolyzed under adequate conditions of pH to form an amorphous M(OH)_n with large surface area, suspended organic matter of sludge was adsorbed into M(OH)_n (iii): The destabilized particles aggregate to form flocs that can be separated from the treated solution either by flotation or settling.

Figure 6 shows the consumption of energy needed for treating wastewater sludge versus current density with different electrode materials. As expected, the calculated specific energy consumption per cubic meter of wastewater treated (SEEC) increased with greater applied current densities. Energy consumption varied with the square of the current density. This trend was confirmed experimentally and agrees qualitatively with previously reported data.^[62] The outcomes indicate that the consumption of energy for the aluminum was higher than those with the iron and zinc electrodes. For $j = 44$ mA/cm², the SEEC was 79.00 kWh/m³, which corresponds to 37.496% of the DD with aluminum, and 66.75 kWh/

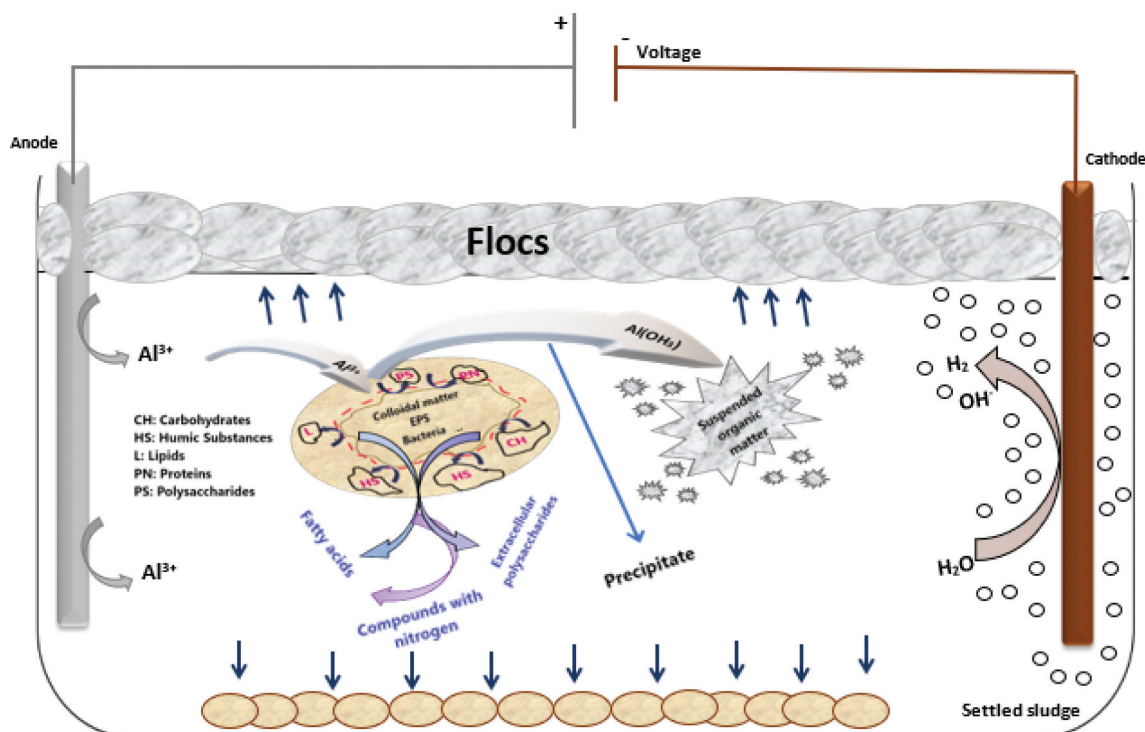


Figure 5. Proposed electrocoagulation mechanisms of the sludge disinteg.

m^3 , which corresponds to 30.84% of DD with iron, and 52.5 kWh/m^3 which corresponds 13.5% with zinc. Thus, use of Aluminum electrodes can be considered better for DD efficiency than those with iron or zinc.

Effect of pH and electrodes material on SCOD, DD, TS and SEEC

The initial pH condition is an important operating factor that influences the performance of the electrochemical process.^[63,64,65] While keeping the natural pH of the solution is preferable to avoid the addition of chemicals, the efficiency of the kinetic rate can be very sensitive to the initial pH level, despite the buffer effect during the EC treatment, as seen in Fig. 7 which considers an initial pH range from 1.5 to 12 using Al, Fe and

Zn. With aluminum electrodes, the highest SCOD (1500.03 mg/l) and DD (63.3%) was achieved in acidic medium (pH = 5) [Fig. 7a]. At pH levels beyond 5, it dramatically dropped. With iron electrodes, an initial pH of 7.48 produced a SCOD of 1212.35 mg/l and DD of 37.92%. With a pH of 10, these dropped to 875.28 mg/l and 16.67%, and further declined to 784.69 mg/l and 12.03%, respectively with a pH of 12. With zinc electrodes, the best achievable was with a pH of 10, which resulted in a SCOD of 993.52 mg/l and an affiliated DD of 23.64%. In conclusion, in acidic medium, higher SCOD and DD efficiencies were obtained with aluminum, while in neutral and weakly alkaline medium, the efficiency of iron was enhanced. However, efficiency of the process was achieved at a pH higher than 8. These

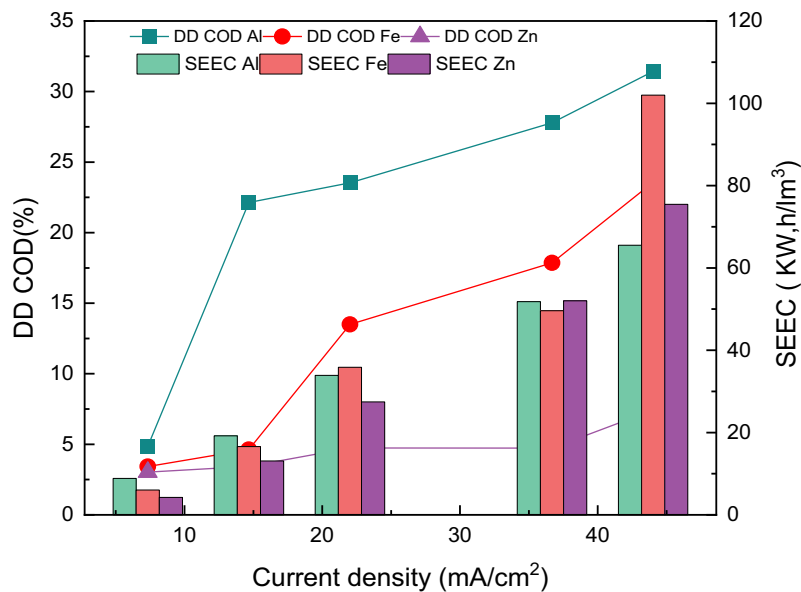


Figure 6. Profile of DD and SEEC under the effect of current density and electrodes material. Operating conditions: treatment time = 60 min, current density (mA/cm^2) = 7.33, 14.67, 22.01, 36.68, 44.02, $\text{pH}_{\text{natural}} = 7.46$

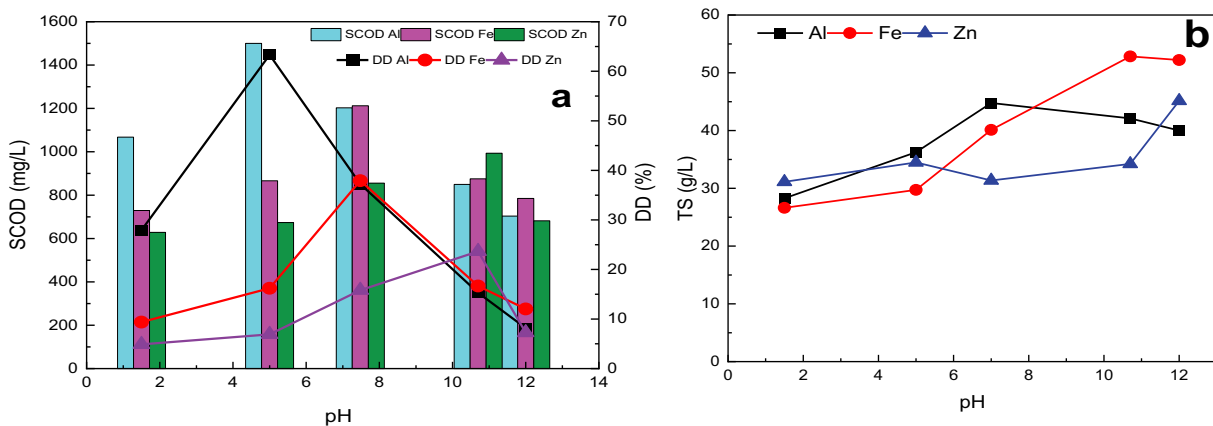


Figure 7. Profile of a) SCOD and DD b) TS under the effect of pH and electrodes material. Operating conditions: treatment time = 60 min, current density = 44.02 mA/cm^2 , $\text{pH} = 1.5, 5$, $\text{pH}_{\text{natural}} = 7.46, 10.5, 12$

results are attributable to the fact that these pH ranges enhanced the creation of freshly amorphous $\text{Al}(\text{OH})_3$ (s), $\text{Fe}(\text{OH})_3$ and $\text{Zn}(\text{OH})_2$ “sweep flocs” that have large surface areas, which is beneficial for rapid adsorption of soluble organic compounds.^[35–58] Furthermore, acidic conditions favored the formation of positively charged metal cations.

Figure 7b depicts the effects of the initial pH on TS reduction. The reduction in TS is not significant for the three tested electrodes. Acidic conditions (pH = 1.5) reduced TS to 28.27 g/L and 26.61 g/L for Al and Fe electrodes respectively for the reason that Al^{3+} and Fe^{2+} species in this pH range reacted with the negatively charged surface of colloidal matter of sludge and release the destabilized organic compounds into solution this is why TS slightly decrease in acidic medium. However, the amount of TS reached its maximum in neutral to weakly acidic pH. This is the result of the solid precipitates of Al, Fe and Zn being formed when sludge pH rose. Higher pH values promoted the formation of monomeric soluble anions like $\text{Al}(\text{OH})_4^-$, $\text{Fe}(\text{OH})_4^-$ and $\text{Zn}(\text{OH})_4^{2-}$. These anionic species do not react with the negatively charged surface of sludge particulate. Therefore, the TS content does not decrease after electrocoagulation treatment.

Figure 8 shows the evolution of DD (%) and SEEC as a function of the initial pH for three electrode materials. As expected, with all materials energy consumption increases steadily with higher pH levels, and the low values of SEEC were found when the pH was acidic. Concentrated acid was added to the wastewater sludge

to obtain pH values that ensured the presence of excess H^+ protons in the solution and also of metallic cationic species. This increased the wastewater sludge conductivity, which decreased the potential of the system under the constant current density of 44 mA/cm^2 . This decrease potential reduced energy consumption as per Equation 10.

For Al electrodes, the lowest energy consumption occurred at pH 1.5, but the best removal efficiency was obtained at pH 5 (DD = 52.29%) for an energy of SEEC = 46.5 KWh/m^3 . For Fe electrodes, the lowest energy consumption occurred at a pH of 1.5, but the best removal efficiency was obtained at a pH of 7 (DD = 37.92%) for 63.00 KWh/m^3 of energy consumed. With Zn electrodes, at pH levels of 1.5 to 5, the minimum electrical energy consumed coincided with slow disintegration efficiency. The maximum of DD rate occurred at a pH of 10 (DD = 23.64%) with an SEEC = 56.75 KWh/m^3 .

Effect of disintegration processes on sludge settleability

The effect of H_2O_2 oxidation parameters on sludge settleability are shown in Fig. 9. The SVI decreased from 99.04 ml/g to 57.85 ml/g when the H_2O_2 increased from 0.4 M to 1.4 M (Fig. 9a), this is consistent with the previously obtained results (Fig. 1a) when the addition of 1.4 M of H_2O_2 resulted in the greatest increase of the SCOD. Saby et al. (2002)^[7] related the poor sludge settling to the very fine particulates of the sludge and the amount of nonbiodegradable residual COD. The SVI values decreased more when alkaline conditions

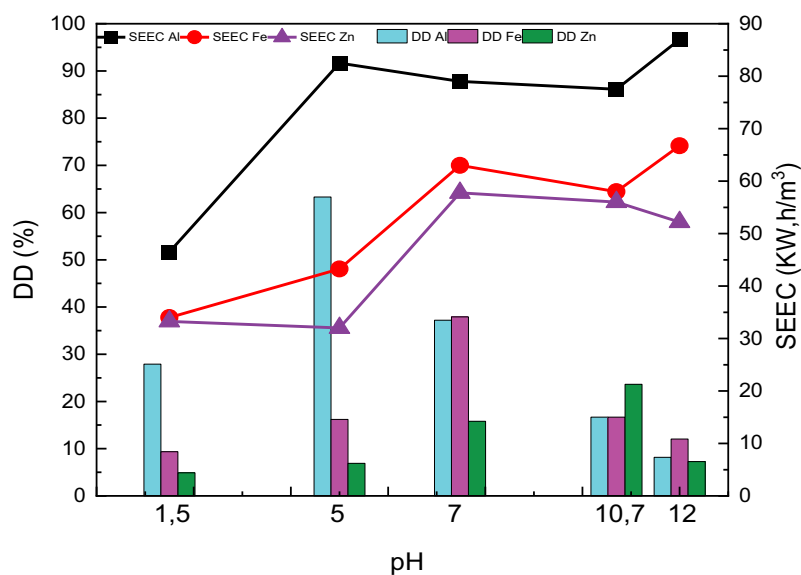


Figure 8. Profile of DD and SEEC under the effect of pH and electrodes material. Operating conditions: treatment time = 60 min, current density = 44.02 mA/cm^2 , pH = 1.5, 5, pH_{natural} = 7.46, 10.5, 12

were used as a pretreatment of hydrogen peroxide oxidation (Fig. 9b), as justified by the large amount of TS (Fig. 1b)

Figure 10 depicts the effect of electrocoagulation parameters on sludge settleability. As the EC treatment was extended from 10 min to 90 min, SVI values increased, especially when comparing the Al electrodes to the Fe ones (Fig. 10a). Those with Zn were large unaffected (Fig. 10a). Much longer disintegration times could provide more solubilization of COD, which may lead to better sludge settling. The present results may be to the low applied EC current being unable to generate sufficient coagulant species capable of destabilization and aggregation of organic matter. Similar results were obtained by Zodi.S et al. (2009)^[65] when SVI values increased with Al electrodes, as the EC period increased from 40 to 120 min. In that work, no explanation was given.

At various applied current densities (7 mA/cm²- 44 mA/cm²), the SVI with Fe electrodes (Fig. 10b) decreased more than those with Al going from 62 mL/g to 35-mL/g, as the current density increased. Sludge settling was expected be favored with Fe due to the formation of heavy flocs of iron hydroxides.^[63] With Al electrodes, the SVI decreased from 84 mL/g to 39 mL/g. At high applied currents, the formation of large flocs were observed, which made the reading of the settled sludge height after 30 min difficult and may have impacted the SVI calculation results. For Zn electrodes, there was no clear influence of the current on SVI values, which only varied from 93 mL/g to 95.2 mL/g. According to Song et al. (2008),^[66] the rate of bubble-generation increases and the bubble size decreases with greater current density: hydrogen bubbles can adsorb more easily onto the particles, and the settling velocity decreases accordingly.

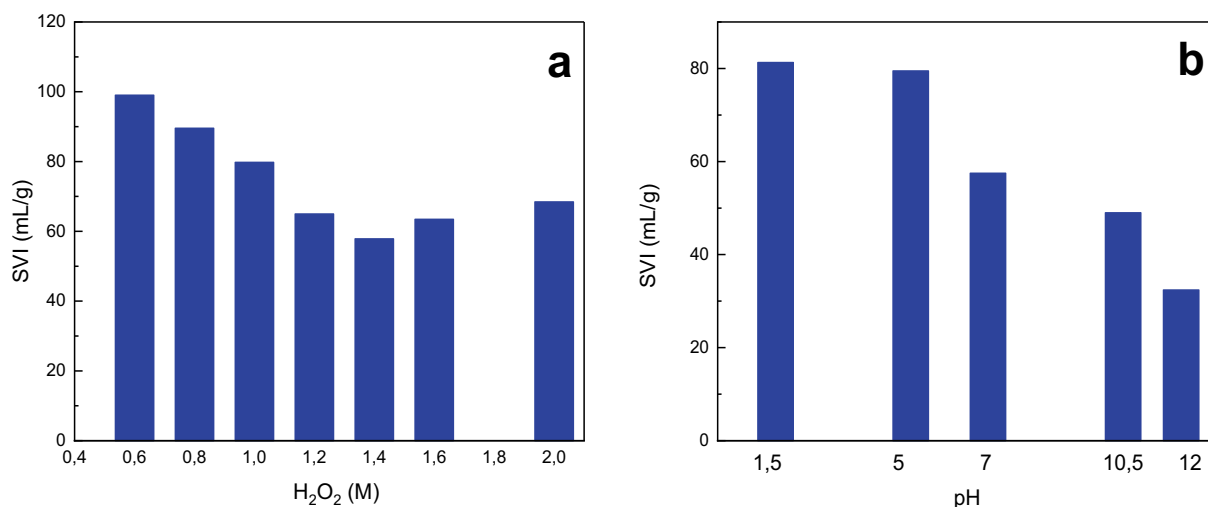


Figure 9. Change in SVI at different a) H₂O₂ concentration b) sludge pH.

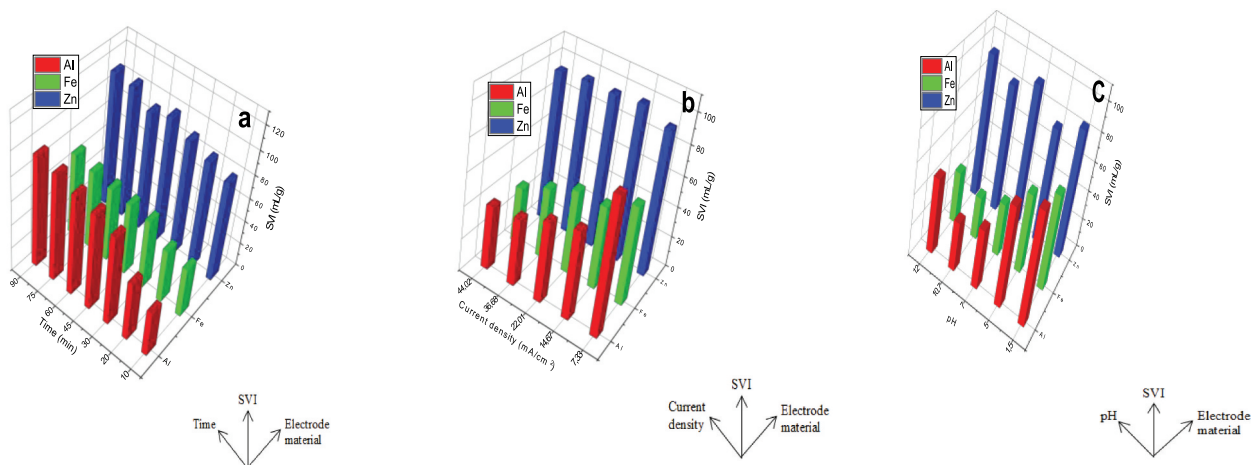


Figure 10. Impact of a) treatment period and b) current density and c) pH on SVI.

Higher SVI values were obtained in acidic conditions (pH = 5) with both Al and Fe electrodes, at 75 mL/g and 64 mL/g, respectively (Fig. 10c). The SVI then decreased gradually at higher pH values. At an alkaline pH, SVI levels of only 49 mL/g and 34 mL/g were obtained with Al and Fe electrodes, respectively. Higher SVI values were obtained with Zn in the range of 85 mL/g to 112 mL/g. The aluminum electrodes were more effective for sludge settleability than Zinc, but the iron electrodes were best. Previously, Zodi et al. (2009)^[65] showed that more rapid settling was observed with Fe than with Al in textile wastewater treatment effluent. They attributed this to the large concentration of aluminum hydroxide being produced, which resulted in a significant gel formation whose settling was hindered by high local viscosity and a low difference in density.

Interpretation of statistical analysis

A MANOVA analysis demonstrated electrode material selection had a statistically significant effect ($p < .001$) on the three responses DD ($p = .007$), SCOD ($p = .023$) and TS ($p = .050$) and that this effect occurred regardless of treatment time and pH control, but not current density. Electrode type changes were not statistically significant for SCOD ($p = .057$) and TS ($p = .054$) but were statistically significant effect ($p = .002$ and $p = .001$ for SCOD and TS, respectively) when controlling for current density.

Discussion of the results of optimum operational conditions and comparison with literature

Based on the optimized experimental results reported above, the cost analyses for H₂O₂ oxidation and electrocoagulation were calculated, as presented in Table 1. Measured energy consumption and calculated operation costs were in the range of several

previously reported, pretreatment technologies.^[24,67,68]

At \$71.17/m³, the H₂O₂ oxidation process was not economical compared to EC and reached a maximum of 50% of sludge disintegration. Using aluminum a better DD value was obtained (63.3%) at barely more than a third of the cost.

Compared to published experimental (Table 2), H₂O₂ is a potential solution to the stabilization and minimizing of sludge problem. Specifically, Wassate et al. (2014)^[71] and Feki et al. (2015)^[36] both showed that H₂O₂ alone is insufficient for the removal of specific compounds from a wastewater sludge but could be improved through combination with other processes like thermo-alkaline hydrolysis and electro-oxidation. Usually, chemical pretreatment methods are criticized because of their potential corrosive impacts on the equipment. Sludge pretreated with acid or alkali requires re-neutralization, which leads to high operational and maintenance costs.

Table 2, also demonstrates that the disintegration efficiency is high with electrocoagulation compared to other techniques, and that the processes can be enhanced by employing specific metal electrodes. Electrocoagulation has the benefit that it does not necessitate the addition of chemical catalysts or use of acid and base products for adjustment of the pH of the medium both of which increase costs.

Table 2 also demonstrates that wastewater sludge disintegration by electrochemical processes can be enhanced by employing different metal electrodes and specific oxidants. The disintegration efficiency is high with electrocoagulation compared to other techniques. The choice of electrode material significantly influenced the process performance. Under optimum conditions, specific electrical energy consumption was in the range of several pretreatment technologies.^[11,72] For a thorough comparison of pretreatment effectiveness, a comprehensive cost analysis

Table 1. Operating cost calculation for H₂O₂ oxidation and EC process.

	Operating conditions	Cost calculation	Sludge disintegration
H ₂ O ₂ Oxidation	H ₂ O ₂ = 1.4 M T = 2 hours pH = 12	Reagent price=\$ 52.17/m ³ E _{consumption} price =\$19/m ³ Cost=\$71.17/m ³	DD = 50%
Electrocoagulation			
Electrode	Al	Fe	Zn
Operating conditions	T = 60 min, I = 44.02 mA/cm ² , pH = 5	T = 60 min, I = 44.02 mA/cm ² , pH = 7	T = 60 min, I = 44.02 mA/cm ² , pH = 10.5
Cost calculation	m _{consumption} = 2 Kg/m ³ E _{consumption} = 82.5KWh/m ³ Reagent price=\$20 Cost= \$26.13/m ³	m _{consumption} = 6 Kg/m ³ E _{consumption} = 43.25 KWh/m ³ reagent price=\$0/m ³ Cost=\$9.26	m _{consumption} = 7 Kg/m ³ E _{consumption} = 56 KWh/m ³ Reagent price=\$10/m ³ Cost=\$24.96
Sludge disintegration	DD = 63.03%	DD = 37.92%	DD = 23.64%

Table 2. Select experimental results of chemical oxidation and electrochemical wastewater sludge disintegration.

Type of pretreatment	Sludge characteristics	Optimum conditions	Removal yield (COD/DD)	Reference
Electrochemical	TSS = 12.8 g/L SCOD = 29 mg/L	Ti/RuO ₂ anode and carbon cathode, V = 14.5 V, t = 0.14 h	R(SCOD) = 30%	[9]
Electrocoagulation	SCOD = 179.2 mg/L	Fe electrode, pH 7, t = 30 min J = 150 A/m ²	DD = 84.23%	[24]
Electrocoagulation.	SCOD = 510.58 mg/l TS = 30.84 g/l	J = 44.02 mA/cm ² , t = 30 min pH = 5 : Al electrodes pH = 8: Fe electrodes pH = 10 : Zn electrodes	Al (DD = 63.03%), Fe (DD = 37.92%), Zn (DD = 23.64%)	This study
Fenton	SCOD = 88.76 mg/l TSS = 34.60 g/L	pH = 5.0, H ₂ O ₂ /Fe ²⁺ :24:1, T = 70°C, t = 90 min.	SCOD = 3 714.64 mg/l R(SCOD) = 41.85%	[69]
H ₂ O ₂ oxidation	SCOD = 190 mg/l TS = 34 g/l	[H ₂ O ₂] = 1.8 M, t = 2 h pH = 3.0	R(SCOD) = 19.2%	[36]
H ₂ O ₂ oxidation	SCOD = 510.58 mg/l TS = 30.84 g/l	[H ₂ O ₂] = 1.4 M, pH = 12, t = 2 h	DD = 50%	This study
KMnO ₄ oxidation	TS = 9580 mg/L SCOD = 93.18 mg/L	[KMnO ₄] = 500 mg/L	R(SCOD) = 89.6% DD = 32.3%	[6]
Potassium Ferrate disintegration	TSS = 13.05 g/l SCOD = 211 mg/l	[K ₂ FeO ₄] = 1.58 g/SS	R(SCOD) = 35.87%	[70]
Sludge chlorination	SCOD = 37 mg/l	[Cl ₂] = 133 mg/L	R(SCOD) = 40%	[7]

and an evaluation of the composition of soluble organics released after pretreatment are needed. In this study, only the effect of H₂O₂ oxidation and electrocoagulation on sludge disintegration at a laboratory scale for a single sludge type was investigated. Future work should expand the types of sludge and include the influences of EC pre-treatment on sludge biodegradability and methane production. Additionally, based on the work by Van et al. (2014)^[73] and Zeng et al. (2019)^[57] who piloted studies showing the potential to convert sludge wastes into methane, hydrogen gas and volatile fatty acids, this should be explored in an anaerobic electrocoagulation reactor with the sludge investigated herein.

Conclusions

This study compared electrocoagulation (EC) using aluminum, iron, and zinc electrodes for sludge disintegration an H₂O₂ oxidation process. Effects of the H₂O₂ dosage and initial sludge pH-H₂O₂ levels, as well as the EC operating conditions (i.e. treatment period, current density and initial sludge pH) on sewage sludge disintegration were evaluated based on sludge solubilization, disintegration degree, total solids, and sludge volume index, and operational cost.

The experimental data and analysis led to the following conclusions:

- Under moderate energy consumption, sewage sludge was more effectively treated with an EC process of only 60 min compared to an H₂O₂ oxidation of 120 min.
- Aluminum electrodes were more effective than iron and zinc ones at solubilizing organic matter

in the sludge. With aluminum electrodes a total of 63.03% DD was achieved at optimum conditions (pH_{real} = 5, j = 44.02 mA/cm² and 1 h) compared to 37.92% with iron (optimum pH_{real} = 7.48, j = 44.02 mA/cm² and 1 h) and 23.64% with zinc conditions (optimum pH = 10, j = 44.02 mA/cm² and 1 h).

- H₂O₂ oxidation under optimum conditions (pH 12.00, 1.4 M H₂O₂ and 2 H) increased solubilization to 50.00%.
- TS reduction was more effective with H₂O₂ oxidation under acidic conditions (TS = 14 g/l) when organics were partially decomposed by H₂O₂ oxidant and neutralized by H⁺ protons. EC with aluminum achieved TS = 36.25 g/l (with aluminum), 40.11 g/l (with iron), and 34.23 g/l (with zinc); probably due to electrode dissolution and metal hydroxide formation.
- Sludge settleability was enhanced with H₂O₂ oxidation. The SVI reached 32.4 mL/g under optimum conditions (1.4 M H₂O₂, pH 12.00 and 2 H). With EC, sludge settleability was most enhanced with Fe (SVI 39 mL/g), Al (35 mL/g) and Zn 95.2 mL/g under optimum conditions for each. Formation of flocs and metal hydroxides occurred, which obfuscates the relationship between the solubilization of COD, TS and SVI and needs to be further explored in future investigations.

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Disclosure statement

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Statement of novelty

I declare that the theme is new. The disintegration of wastewater sludge by H₂O₂ was addressed alone. The application of electrocoagulation for the disintegration of sewage sludge is new. There are only two publications in this field, the difference with our work is to apply different electrode materials (Al, Fe and Zn) with the follow-up of several parameters such as: the degree of disintegration, COD, dry matter, sludge settling but also the calculation of the energy consumed for electrocoagulation method. Finally, a comparative table with the other disintegration techniques was given.

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