

Review

Recent Applications of the Electrocoagulation Process on Agro-Based Industrial Wastewater: A Review

Rakhmania ^{1,†}, Hesam Kamyab ^{1,2,*,†} , Muhammad Ali Yuzir ¹, Norhayati Abdullah ¹, Le Minh Quan ¹ , Fatimah Azizah Riyadi ¹ and Riadh Marzouki ^{3,4} 

¹ Department of Chemical and Environmental Engineering (ChEE), Malaysia-Japan International Institute of Technology (MJIT), Universiti Teknologi Malaysia, Kuala Lumpur 54100, Malaysia; rakhmania99@gmail.com (R.); muhdaliyuzir@utm.my (M.A.Y.); norhayati@utm.my (N.A.); quanmle1505@gmail.com (L.M.Q.); fatimahriyadi@gmail.com (F.A.R.)

² Department of Electric Power Stations, Network and Supply Systems, South Ural State University (National Research University), 454080 Chelyabinsk, Russia

³ Chemistry Department, College of Science, King Khalid University, Abha 61413, Saudi Arabia; rmarzouki@kku.edu.sa

⁴ Chemistry Department, Faculty of Sciences of Sfax, University of Sfax, Sfax 3029, Tunisia

* Correspondence: hesam_kamyab@yahoo.com

† These authors contributed equally to this work.

Abstract: Agro-based final discharge is one of the major contributors to wastewater in the world. It creates high demand for efficient treatment. The electrocoagulation process can be used for agro-based wastewater treatment. The performance of the electrocoagulation process is based on several parameters, including the electrode materials, electrolysis time, current density, and electrolyte support. Agro-based industrial wastewater (AIW) treatment processes depend on the characteristics of the wastewater. The removal of organic content from various sources of AIW can reach up to more than 80%. Some studies show that the performance of the electrochemical process can be increased using a combination with other methods. Those other methods include biological and physical treatment. The results of previous research show that organic content and color can be degraded completely. The relationship between the energy consumption and operating cost was analyzed in order to show the efficiency of electrocoagulation treatment.

Keywords: wastewater treatment; color removal; oily wastewater; energy consumption



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1. Introduction

Many industrial activities nowadays have become a major source of pollutants due to their wastewater discharge [1,2]. One type of wastewater comes from the agro-industry [3]. Agro-based industrial wastewater (AIW) has a high number of organic compounds, inorganic salts and reactive dye [4]. These pollutant compounds cause several problems for the aquatic environment, such as eutrophication [5], pathogen contamination, oil and grease pollutants, etc. [6]. Therefore, it is necessary to treat the wastewater final discharge to reduce the toxicity for the environment [7,8].

There are several methods of treatment process according to the type and characteristics of the AIW, such as conventional treatment using the physicochemical method, and biological methods like microalgae treatment [9] for nitrogen [10] and phosphorus removal [11]. The selection of the method depends on the regulation of permissible wastewater released to the environment, the industrial stream capacity, the economical cost [12], the impact on the environment, and the possibility of toxic by-product formation [13]. Advanced technologies such as membrane filtration, ultrafiltration and reverse osmosis have also been used for AIW treatment [14]. The conventional treatment using physicochemical methods for AIW treatment includes coagulation–flocculation, adsorption, precipitation,

and flotation etc. They have several disadvantages, such as high chemical consumption [15], high sludge production and energy costs [16].

The conventional biological method includes aerobic and anaerobic processes. They possess a large number of microorganisms for the biodegradation of organic pollutants. However, this method is laborious, produces biological sludge, has low biodegradability on certain molecules (i.e., dyes), and is energy-intensive [17,18]. The sludge formation requires further treatment, which consumes about 50% of the operational cost of the wastewater treatment plant (WWTP) [19]. The sludge treatment including lime stabilization that commonly uses calcium hydroxide [20]. It can reduce the portion of solid organic matter to 30%, and can reduce 99% of fecal coliforms [21]. Wet air oxidation (WAO) has been used for sludge treatment [22]. It is suitable for the oily sludge type; it can remove 93.1% of the oil from the sludge, and can reduce the volume of oily sludge to 85.4% [23]. The thermal drying used for sludge treatment acquires 10–80% removal of alkylphenols, polycyclic aromatic compounds and mercaptobenzothiazole [24]. The sludge is then disposed of using several methods, such as composting, incineration, or open landfill, etc. [25]. The overall biological cost analysis at WWTP resulted in 0.1345 €/m³ of treated wastewater [26].

An application of microalgae on AIW treatment is used currently as a developed alternative approach. The microalgae method could remove the organic compound according to a previous study, but most of the performance achieves less than 50% removal [27,28]. A major challenge from the application of microalgae is the environmental stability of the culture when used outdoors [29]. The microbial fuel cell (MFC) system can be used for AIW treatment and the generation of bio-electricity at the same time [30]. In general, 1.06 kg of COD contained in wastewater could be converted to 4.41 kWh bio-electricity power [31]. Other than bio-electricity, this system also produces biogas during the process. The biogas usually contains H₂S, which is toxic and requires additional cost for removal [32].

Advanced-technology treatment has been applied for some types of AIW [33]. This system has been chosen due to its small space requirements and lack of chemicals required [34]. The main problem with membrane-related systems is fouling, which can reduce the membrane's efficiency performance [35]. In addition, this system also has disadvantages in terms of its high energy requirements due to high operation and maintenance costs, limited flow rates, the high expertise required for membrane selection, and not being suitable for low solution concentrations [36].

The electrocoagulation process is gaining attention for wastewater treatment due to its high efficiency to degrade pollutants, easy operation and maintenance, lack of chemical required, and relatively low energy consumption. In addition, this process can degrade organic pollutants completely into CO₂ and water [37]. Electrocoagulation has been applied and investigated in different stages of AIW wastewater treatment, including the pre-treatment, main treatment and polishing stages [13,38]. Studies conducted by some researchers show the use of the electrocoagulation process to treat different types of wastewaters, such as dairy wastewater [39], sugar industry wastewater [40], paper mill wastewater [31], and olive oil mill wastewater [41], etc. However, there is no report specifically reviewing the recent application of electrocoagulation treatment for AIW. Thus, this paper presents an exhaustive critical review of the recent application of the electrocoagulation process for the last nine years. The fundamental theories on electrocoagulation are described. The recent applications of the electrocoagulation process on various types of AIW are discussed, as well as the combination of electrocoagulation with other methods. Moreover, the relationship between specific energy consumption and the operating cost was analyzed.

2. Methods

This section analyzes all of the literature related to the AIW treatment by the electrocoagulation process identified during the period 2013–2021.

2.1. Data Source and Strategy

An exhaustive search of peer-reviewed literature was conducted in the Scopus database to select the scientific articles related to the electrocoagulation process on AIW treatment. The various types of AIW include olive oil mill wastewater, sugar industry wastewater, pulp and paper mill wastewater, palm oil mill effluent, coffee industry wastewater, vegetable oil refinery wastewater and nut processing wastewater. Because there are redundant papers for the electrocoagulation of AIW, the keywords are specified. The specified selected keywords were electrocoagulation, olive oil mill, sugar industry, pulp and paper mill, palm oil mill effluent, the coffee industry, vegetable oil and nuts. The keyword of wastewater was excluded in order to reduce the probability of redundant papers.

The literature was limited to the publications of scientific articles written in English from 2013 to May 2021, covering the period of the last nine years. The publication date, names of the authors, title and abstract of each paper found were listed to be further revised individually and manually. All of the publications with specified selected keywords were included. Review papers, conference papers, letters to the editor, books and short surveys were excluded.

2.2. Bibliometric Analysis

Figure 1 shows the annual distribution of a total of 71 publications that were found. Even though there were a number of publications from 2015 until 2021 (May), the highest number of publications in 2017 and 2019 indicate that the electrocoagulation process has become of great attention for AIW treatment. According to the selected publications, around 74% use a single treatment of electrocoagulation and 25% use electrocoagulation combined with other methods. It was also found that two scientific articles reported on biogas production from the electrocoagulation treatment of AIW.

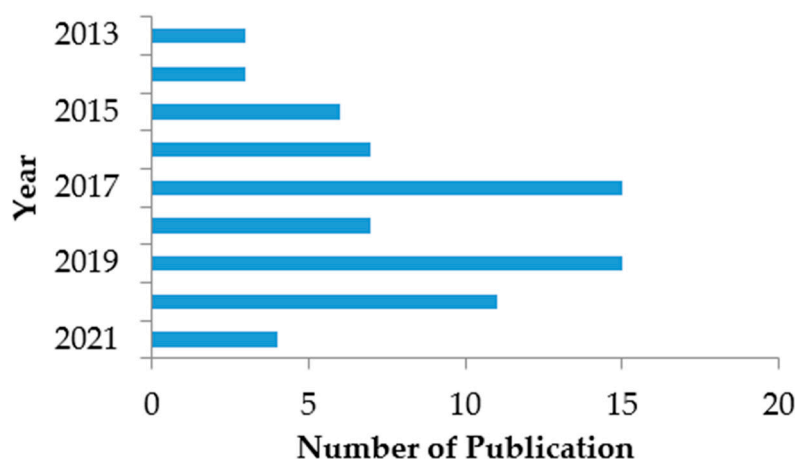


Figure 1. Number of publications by year.

Most of the publications use real AIW for the treatment by the electrocoagulation process, and only one scientific article reported on the electrocoagulation process using simulated wastewater. As the recent application of electrocoagulation is still on the laboratory scale, it was noticeable that there was one scientific article reporting the application of the electrocoagulation process at the pilot scale in 2019. This indicates an important challenge for future research on the scaling-up of the electrocoagulation process at the industrial level.

3. Fundamental

3.1. Coagulation Principal

Coagulation is a traditional physicochemical treatment using phase separation for the pollutants of wastewaters before they are released to the environment [42]. The addition

of inorganic or organic compounds to the coagulation process destabilizes the particles, affecting the electrical double layer (EDL). The impact to the environment from the coagulation process in the generation of sludge makes the concern more serious in terms of management, treatment and operational cost [43]. The sludge also contains residual metal ions from the coagulation process [44].

Chemicals known as coagulants lower the energy barrier between particles. As a result of the weak bond, the particles can agglomerate more easily [45]. Aluminium and iron metal salts are common coagulants used in wastewater treatment. Both metals can produce multivalent ions such as Al^{3+} , Fe^{2+} , and Fe^{3+} , as well as a variety of hydrolysis products. Because it oxidizes to Fe (III) during the coagulation process to increase efficiency, Fe (II) is a poor coagulant [46]. Simple aluminium, iron sulfates, and chlorides are the most commonly used salts [47]. The non-hydrolyzed metal coagulants include $\text{Al}_2(\text{SO}_4)_3$, FeSO_4 , AlCl_3 and FeCl_3 [48]. Meanwhile, there is another metal coagulant that comes with several advantages, which is named pre-hydrolyzed metal coagulant [49]. Examples of pre-hydrolyzed metal coagulants are polyaluminum chloride and sulfates [50]. The pre-hydrolyzed metal coagulants have advantages such as being more effective than non-hydrolyzed metal coagulants and less sensitive to changes in pH and temperature [51].

3.2. Electrocoagulation

Electrocoagulation by using metal hydroxide has a high ability to adsorb pollutants [52]. The electrode materials commonly use Al and Fe. When the electricity is supplied into the electrode, the electrode will generate ions (Fe^{2+} , Fe^{3+} , and Al^{3+}) and produce the coagulant. The electrocoagulation reaction is usually followed by an electroflotation reaction [53]. The aluminium anode generates cationic forms such as Al^{3+} and $\text{Al}(\text{OH})^{2+}$ in acidic conditions. However, in alkaline conditions, these species are transformed into $\text{Al}(\text{OH})_3$, dimer compounds such as $\text{Al}_2(\text{O})(\text{OH})_4$ and $\text{Al}_2(\text{OH})_2^{4+}$. They can also can be transformed into a more complex compound [54].

Figure 2 shows the schematic diagram of electrocoagulation. Several reactions happen during electrocoagulation for the production of hydroxides. For the electrode materials using aluminium, the oxidation takes place at the anode, as follows:

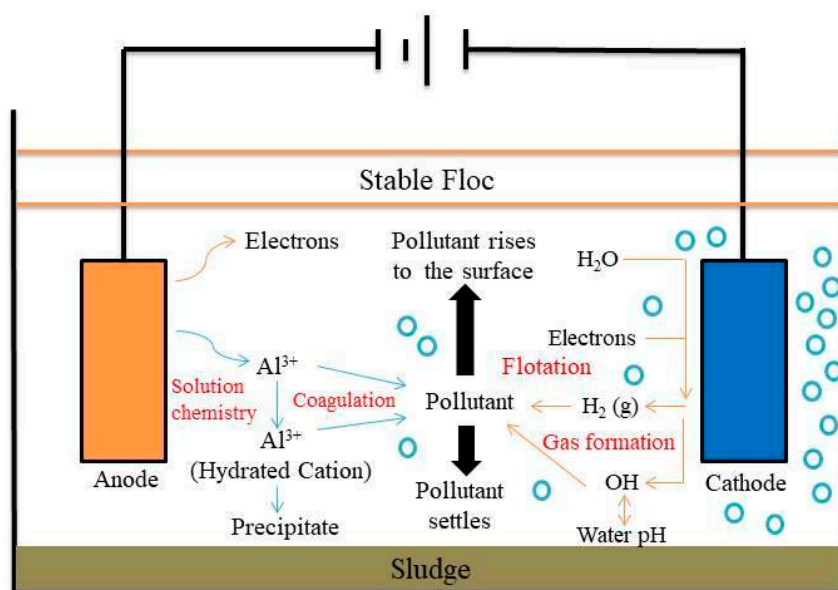
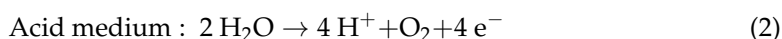
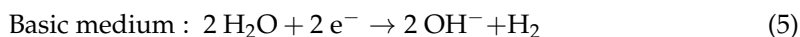
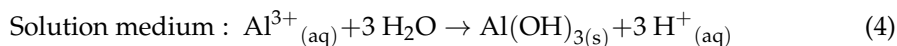
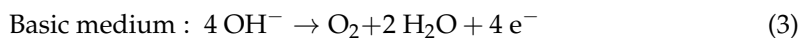


Figure 2. Schematic diagram of the electrocoagulation process.

In addition, a secondary reaction possibly occurs during the oxidation process. The formation of oxygen by the electrolysis of water is divided into two conditions: acidic and basic media.



The reaction that happened in the cathode is a reduction in water based on different conditions, as follows.

In Equation (1), the Al^{3+} produced on the electrode react to form several species, including:

- monomeric species such as $\text{Al}(\text{OH})^{2+}$, $\text{Al}(\text{OH})_2^+$, $\text{Al}_2(\text{OH})_2^{4+}$, and $\text{Al}(\text{OH})_4^-$;
- polymeric species such as $\text{Al}_6(\text{OH})_{15}^{3+}$, $\text{Al}_7(\text{OH})_{17}^{4+}$, $\text{Al}_8(\text{OH})_{20}^{4+}$, $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ and $\text{Al}_{13}(\text{OH})_{34}^{5+}$;
- amorphous species with very low solubility, such as $\text{Al}(\text{OH})_3$ and Al_2O_3 [55].

The formation of amorphous $\text{Al}(\text{OH})_3$, also called sweep flocs, needs surface areas which are useful for the adsorption of soluble pollutants and the trapping of the colloidal compounds [56]. The polymerization of flocs is as follows:

The flocs can be easily removed from an aqueous medium using the sedimentation method and H_2 flotation. However, aluminium hydroxide usually acts as an adsorbent or traps the pollutants. It would be a great composition for the removal of the organic compound from the solution [57].

4. Factors Affecting Electrocoagulation

There are several factors that affect anodic oxidation performance in wastewater treatment. There are electrode materials, electrode distance, electrode arrangements, pH, current density, reaction time, conductivity and temperature.

4.1. Electrode Materials

Electrode materials are one of the important factors affecting the electrocoagulation process performance [58]. Several types of anode materials have been used in the electrocoagulation process, such as Fe, Graphite, Al, stainless steel, and steel wool, etc. [59–61]. One of the anode materials available in the electrochemical industry is a lead-based anode. It is widely used due to its low cost in comparison to other materials. They are including lead–calcium (0.7%)–tin, lead–strontium (0.05%)–tin (0.6%), and lead–antimony (6%) [62]. The lead-based anode has significant drawbacks, including high energy consumption and low corrosion resistance [63]. Corrosion gives effects in terms of the electrode lifespan and the production of a poor quality of electrochemical reaction [64].

Three-phase-3D electrode electrocoagulation can potentially be used for agro-based industrial wastewater. The previous study reported the performance of chromium removal from wastewater using 3D NiO/NF electrodes. This achieved the 99.5% removal of chromium within 20 min at the applied potential of 0.97 V [65]. Another study reported on ground wastewater treatment using 3D Al electrode resulted in 98.6% arsenic removal [66].

4.2. Electrode Distance

The distance between the anode and the cathode has a crucial role due to the electrostatic field that forms in the gap [67]. The electrostatic field strength is substantial, as it attracts the ions generated from the plates during the EC process [68]. The distance between the electrodes varies from 1 cm to 11.5 cm. When the distance is too close, the transfer of solids and fluids is hampered. The accumulation of solid particles and bubbles between the anodes and cathodes results in increased electrical resistance. However, when

the distance is longer, it increases the travel time of the ions, thus leading to a decrease in the electrostatic attraction which subsequently reduces the formation of flocs [69].



4.3. Electrolysis Time

Faraday's law states that the amount of metal ions released from an anode is proportional to the electrolysis time and current intensity. The equation is expressed as follows:

$$m = \frac{i \times t \times M}{n \times F} \quad (7)$$

where m is mass of metal dissolution (g), i is current density (A), t is the electrolysis time, M is the molecular weight of the metal, n is the number of electrons involved in the oxidation reaction, and F is Faraday's constant ($96,500 \text{ Cmol}^{-1}$). When the electrolysis time is increased, then the metal ions released are high. Therefore, the performance of organic pollutant removal is also increased. However, at a certain point of electrolysis time, the rate of removal is slowed down [70]. This is due to the occurrence of electrode passivation, which occurs when the electrolysis process is operated for an extended period of time; it is recognized as being detrimental to the electrochemical process performance. The electrode material is also a factor in electrode passivation [71].

4.4. Current Density

One of the parameters is the current density, which is used in EC specifically for the kinetics of reduction of COD and decolourization. Because of the high dissolution of the electrodes, the electrolysis time decreases as the current density increases. This results in the further destabilization of the pollutant particles. Furthermore, as the current density increases, the rate of production of hydrogen bubbles increases, while their size decreases. All of these effects are advantageous for efficient pollutant removal via flotation [72].

4.5. Electrolyte Support

The role of electrolyte support is to increase conductivity [73]. Therefore, the voltage applied between the electrodes at a constant current density due to the decrease in the resistance of the polluted water can be reduced [74]. Some of the electrolyte supports that have been used for the electrochemical process include NaCl, Na_2SO_4 , and NaCO_3 , etc. Most of the studies use NaCl as the electrolyte support. This is because of the properties of NaCl which produce chloride ions. The chloride ions can reduce the significantly undesirable effects of other anions [75]. NaCl is also known as a contributor of strong oxidizing agents and hence the increase of color removal [76]. In comparison to sulfate and nitrate, chloride was the best supporting electrolyte for the electrochemical oxidation of refractory organic pollutants [77].

5. Type of Agro-Based Wastewater

5.1. Olive Oil Mill Wastewater

In general, olive oil mill wastewater (OOMW) consists of 80–83% water, 15–18% organic compounds, and 2% inorganic compounds [78]. OOMW has the characteristics of being dark in color, having an acidic pH (~5), and having highly toxic components including tannins, phenols and acid compounds making up about 37% of the total mass [79,80]. It has been reported that more than thirty types of phenolic compounds have been identified. Table 1 shows the characteristics of OOMW. The concentration ranges between 0.5 and 24 g/L. The COD content in OOMW is quite high, and can reach up to 175,000 mg/L. The BOD content is 25,000 mg/L, while the total solid content is 99,000 mg/L [81].

Table 1. Characteristics of olive oil mill wastewater [82,83].

Parameters	Units	OOMW
COD	mg/L	175,000
BOD	mg/L	25,000
pH	-	4
Oil and Grease	mg/L	13,000
NH ₃ -N	mg/L	37
TSS	mg/L	99,000
PO ₄ ³⁻	mg/L	57.5
NO ₃ ⁻	mg/L	395

The electrocoagulation process can be used for OOMW because it decomposes the organic compound and is commonly used for OOMW treatment [84]. Electrocoagulation has stability for colloids, suspension and emulsion, which is affected by electric charge. When an electric charge is supplied into a suitable electrode, the compound will be neutralized and the rest of it will aggregate together into a larger and separable compound [85].

A study showed that electrocoagulation can remove COD by up to 78% after 1 h of electrolysis time using Fe as the electrode. The energy consumption reaches 55 kWh m⁻³. However, using Al as the electrode, the removal of COD has a lower value than that using Fe. The removal efficiency is only 55% for 1 h of electrolysis time. It also acquires higher energy consumption until 62 kWh m⁻³ [86]. The reduction of the energy consumption using Al as an electrode can be achieved by diluting the sample 10 times. The study reported that the energy consumption was decreased to 1 kWh/L with 57% COD removal [87]. The kinetic model of the electrocoagulation process using the one-factor method increases the performance of COD removal to 99% with conditions of 60 min electrolysis time, with the current density at 12.5 mA/cm² and the addition of 400 mg/L NaCl [88]. The study conducted by Benekos [89] compared the performance of electrocoagulation on a laboratory scale and pilot scale. The results show on the laboratory scale that the COD and color removal (50% and 100%, respectively) is higher than that at the laboratory scale (42.5% and 85.3%, respectively).

The electrocoagulation process on OOMW is useful as a pre-treatment for biofuel production [85]. The methane production reaches 1902 kJ/L_{OOMW}. The treatment of COD removal also produces biogas, as reported by a previous study. The result shows that the COD removal of OOMW using the electrocoagulation process produces biogas around 0.741 g/L_{COD} [90].

5.2. Sugar Industry Wastewater

The sugar industry categorized as one of the largest agro-based industries. It uses about 1500–2000 dm³ water and produces about 1000 dm³ wastewater per ton of processing [91]. The wastewater mainly results from the process of floor washing, condensation, leakage, and spillage of solutions from the pipeline and valve [92]. The sugar wastewater industries (SIWW) have a high concentration of organic materials due to the presence of sugar and organic material in the cane or beet. SIWW contains organic compounds including COD ranging between 2300 and 8000 mg/dm⁻³, BOD of about 1700–6600 mg/dm⁻³, and TSS of about 5000 mg/dm⁻³ [93]. In addition, SIWW also possibly contains pesticides, herbicides and pathogens from the contaminated material and processes which are produced [91].

The electrocoagulation process becomes one of the choices to be considered for SIWW treatment when the conventional method can not reduce the pollution [94]. The previous study reported that COD removal on SIWW treatment can reach 86.36% for 8 h of electrolysis time. The 12 V voltage is applied to the system without any pre-treatment. Using iron as an electrode increases the COD removal of SIWW, which reaches up to 84% for 2 h electrolysis time. The color reduction is about 86% with the current density of 178 A/m². The lowest energy consumption utilized is 16.75 kWh/L. However, the performance of

COD removal decreases to 62%. This result does not suffice for the final discharge of wastewater; therefore, further advanced methods could possibly be applied to bring the wastewater to meet the final discharge [95].

Different cases happened with simulated sugar industrial effluent using the research surface method to study the effects of various parameters using the research surface method (RSM). The maximum COD removal of about 83.94% is obtained with the energy consumption of 6.64 kWh kg^{-1} . This system requires less energy. This study suggested that electrocoagulation potentially can be applied for the treatment of real sugar effluent [96].

5.3. Pulp and Paper Mill Wastewater

The pulp and paper industry produces highly polluted wastewater [97]. It forms black liquor as the main by-product, which contains about 50% lignin. The pulp and paper industry discards lignin to make a good quality of paper [98]. The release of lignin comes from the process of alkaline extraction at the bleaching stage. Lignin is a heterogeneous three-dimensional polymer that consists of oxy phenylpropanoid components. The contamination of phenolic compounds to the environment could damage the underground water and receiving water bodies [43].

The electrocoagulation process is one of the great alternatives to treat pulp and paper mill wastewater. Aluminium and iron are widely used as electrodes for the electrocoagulation process. A study showed that iron has greater efficiency for BOD and COD removal compared to aluminium. The performance of iron can reach up to the complete removal of COD and 90% removal of BOD within a 60 min reaction [99]. Other concerns for pulp and paper mill wastewater are lignin and phenol. By using iron as an electrode material, the electrochemical process can degrade lignin and phenol by up to 90% [93].

5.4. Palm Oil Mill Effluent

Palm oil mill effluent (POME)'s typical liquid waste results from the extraction of fresh fruit bunch (FFB) in the palm oil industry [100,101]. The characteristic of POME is a thick brownish liquid color due to the decomposition of lignocellulosic materials [102,103]. Table 2 shows the characteristics of POME. POME has acidity ranging from pH 4.0 to 5.0, chemical oxygen demand, biological oxygen demand, and total suspended solids ranging from 15,000 to 100,000 mg/L, 10,250 to 43,750 mg/L and 5000 to 54,000 mg/L, respectively [104].

Some studies have found the performance of the electrochemical process for POME treatment. Using Al as the electrode shows little removal of COD and BOD. The percentage of COD and BOD removal is 30% and 38%, respectively [105]. However, the addition of NaNO_3 could improve the performance of the electrochemical process using Al. The efficiency of COD removal increases by 64%. Using NaNO_3 as the electrolyte support is also beneficial for subsequent biological treatment [106].

Table 2. Characteristics of palm oil mill effluent [107–109].

Parameters	Units	POME
COD	mg/L	75,000
BOD	mg/L	18,200
pH	-	4.6
Oil and Grease	mg/L	2000
$\text{NH}_3\text{-N}$	mg/L	20
TSS	mg/L	50,000
PO_4^{3-}	mg/L	15
NO_3^-	mg/L	500

Another method uses iron for the electrochemical process. The performance of iron is quite great, with 89.2% removal of COD in 15 min of electrolysis time. For another good value, using iron as an electrode could remove the color by up to 90.4% [110]. In addition,

steel wool can be used as an electrode. It could remove COD by up to 74% and BOD by up to 70% with the improvement of the electrode arrangement [111].

5.5. Coffee Industry Wastewater

Coffee is one of the world's most well-known beverages, and it is the world's second-most-traded commodity after petroleum [112]. The wastewater produced from the coffee processing industry ranges from 40 to 45 L for every kg of coffee [113]. The process requires large amounts of water for every step; therefore, the wastewater produced is high [114]. Coffee fruits themselves are rich in caffeine, sugars, phenolic compounds, fatty acids, lignin, cellulose, pectic substances and other macromolecules [115]. Those compounds are not suitable to be released into the environment due to their toxicity [116].

Coffee processing wastewater (CPWW) is characteristically black in color. It is high in persistent compounds known as melanoidins, which are toxic, recalcitrant and non-biodegradable [117]. Releasing the untreated CPWW to the water body can promote eutrophication, reduce Secchi depth and prevent sunlight penetration. The concentration of oxygen in the water body will be depleted [118]. The electrocoagulation process can be used for the decolorization of CPWW treatment [119]. The addition of electrolyte support such as sodium chloride affects the removal efficiency due to the generation of active chlorine during the electrolysis process interacting with substances such as melanoidins [120]. Other than melanoidins, the mineralization of proteins and lipids are adsorbed on the surface of the precipitated hydroxides [121].

Previous studies reported that the use of a combination of iron and stainless steel as electrodes achieves 87% COD removal and 97.1% color removal [122]. When using iron alone as an electrode material, it removes 97% of COD and 89% of color [123]. The electrocoagulation process can also be used for caffeine recovery that reaches 96% recovery performance [124]. However, there is still no report for polygalacturonase characterization and production in the coffee industry using the electrocoagulation process.

5.6. Vegetable Oil Refinery Wastewater

Vegetable oil refinery wastewater (VORW) has a high amount of COD, phosphorus, sulphate, oil and grease [125]. The sources of vegetable oil manufacture are soybeans, groundnut, rapeseed, sunflower, safflower, cotton, sesame, coconut, mustard, rice bran, watermelon, and neem, etc. [126]. Another characteristic of VORW is that it has a low ratio of BOD/COD; therefore, it is not useful for the biodegradation method. Based on some studies, electrochemical technology could overcome that limitation [127].

A previous study reported that using iron as the electrode could remove 93.3% of COD after 60 min electrolysis time [128]. However, when aluminium is used as an electrode, the COD removal increases up to 98.9%, with 100% color removal in 90 min [129]. Another study using the Box-Behnken design for optimization attained 70.8% COD removal. Aluminium was used as an electrode, and the electrolysis time was 60 min [130]. On the other hand, another study reported that using the Box-Behnken design for the optimization of the electrocoagulation process of sunflower oil refinery could achieve 95% COD removal with a shorter time (18 min) [131].

5.7. Nuts Processing Wastewater

The nut processing industry includes many various types, including pistachio, almond, and cashew nut, etc. Pistachio is a kind of nut rich in organic nutrients. It contains 5.6% water, 19.6% protein, 53.2% fat, 19% carbohydrate, and 2.6% ash [132]. The pistachio industry produces approximately 1 Mm³ of wastewater and 50,000 tons of solid waste per year. Pistachio processing wastewater (PPW) has high COD, phenolic compounds and turbidity, which can cause harm to the aquatic ecosystem and terrestrial environment if no treatment of the PPW is performed [133].

Several processes are required in the almond processing industry, including cracking and blanching [134]. In the blanching step, the resulting product contains high levels of

organic compounds, suspended solids, turbidity, COD and color [135]. The effect of the blanching time on the moisture content is inversely proportional. This means that when the blanching time is longer, it results in a decreased moisture content. Meanwhile, the effect of the blanching time on the lipid content and color intensity is directly proportional. The longer blanching time will increase the lipid content and color intensity [136]. A previous study reported that the polyphenol content recovery in the almond industry using the blanching process acquired 0.53 g gallic acid equivalents (GAE)/kg [137], while the use of the microwave process recovered 0.42 g gallic acid equivalents (GAE)/kg [138]. Based on these studies, the polyphenol content recovery is higher using the blanching process. The use of blanching and microwave processes is recommended according to a previous study. It was reported that by conducting the blanching and microwave simultaneously, we can increase the polyphenol content recovery up to 11.9 g gallic acid equivalents (GAE)/kg [139].

The kinetic study showed that the sonicated almond acquires higher total phenolic compounds than non-sonicated almonds after 20 min extraction at room temperature (25 °C) [140]. Table 3 shows the performance of the electrocoagulation process on different types of nut processing wastewater. The electrocoagulation process has been shown to be a suitable method for the reduction of pollutants from cashew nut processing wastewater, as was reported in a previous work where COD reduced it by up to 80% [141]. According to a previous study, even though the COD removal for PPW is categorized as low, the phenolic compound removal shows greater efficiencies. In addition, using graphite as the electrode material on PPW could achieve the complete removal of phenolic compounds and the 99.79% removal of COD [142].

Table 3. Performance of the electrochemical process on nut processing wastewater.

Type of Nuts	Electrode Material	Electrode Type	COD removal	Phenolic removal	References
Pistachio processing industry wastewater	Al	Unipolar	60.1%	77.3%	[143]
Pistachio processing industry wastewater	Graphite	Unipolar	99.79%	100%	[144]
Pistachio processing industry wastewater	Al	Unipolar	57.4%	-	[134]
Pistachio processing industry wastewater	Al and stainless steel	Unipolar	60%	95%	[144]
Cashew nut processing industry wastewater	Fe, BDD (Boron Doped-Diamond) and stainless steel	Multipolar	80%	-	[145]

6. Integration of Electrochemical Treatment with Other Methods

Electrochemical treatment can be integrated with other methods. The processes include a combination of pre-treatment and main treatment. The purpose of this integration is to mitigate the limitations of other treatments. It could be an integration of the electrochemical method with physical, chemical, biological methods. The advantages of this integration are the effective removal of organic and inorganic pollutants [146], low operating costs [142], the increase of the performance of the electrodes and the reduction of the energy demand of the electrochemical method [147].

Table 4 shows the performance of the electrochemical method with other treatments. In the case of coffee industry wastewater, the electrochemical method is integrated with the coagulation method. The coagulation method was used as the pre-treatment. Several types of coagulants were used in the study, including FeSO_4 , FeCl_3 , $\text{Al}_2(\text{SO}_4)_3$, and AlCl_3 . Based on those coagulation processes, AlCl_3 shows the highest performance of TOC removal, at up to 28%. The treatment was continued with the electrochemical process.

A study on the treatment of pistachio processing wastewater showed improvement by combining electrochemical treatment with fungal treatment. The use of Al as an electrode showed the higher removal of COD and phenol compared to BDD, Fe and stainless steel. After the treatment with the electrochemical process, the treatment with the fungus was conducted. From the various types of fungal selection, *Penicillium glabrum* was chosen because it has a high capability to remove COD and phenol. The two combinations of this process could remove 90.1% of COD and 88.7% of phenol [148].

Table 4. Performance of the electrochemical process integrated with other methods on agro-based wastewater treatment.

Type of Agro-Based Wastewater	Electrode	Electrocoagulation with Other Methods	COD Removal	Color removal	References
Wood-based industry wastewater	Al	Adsorption and filtration	77%	-	[149]
Coffee wastewater	Al	Sequential batch reactor	84%	93%	[150]
Coffee pulp industry wastewater	Al	Anaerobic sequencing batch reactor	96%	-	[151]
Olive oil mill effluents	Fe	Electro-oxidation and electro-fenton	96%	-	[152]
Pulp and paper wastewater	Fe	UV-based sulfate radical	61%	-	[153]
Sugar industry wastewater	Fe	Thermal method	97.8%	99.7%	[154]
Palm oil mill effluent	Al and Fe	H ₂ O ₂ and coagulation	95.08%	-	[143]
Pulp and paper wastewater	Fe	H ₂ O ₂ , Co ₃ O ₄ /UV/ peroxymonosulfate and permanganate	95%	92%	[155]
Pulp and paper wastewater	Al	Sonication	90%	100%	[156]
Sugarcane industry wastewater	Al	Coagulation method	98%	99.5%	[157]
Olive oil mill effluents	Al	Coagulation	92%	-	[158]
Olive oil mill effluents	Fe	Photo-catalytic degradation	88%	100%	[155]
Coffee industry wastewater	Al	Electro-oxidation	74%	-	[159]
Coffee industry wastewater	Fe and stainless steel	PAC	80%	92%	[160]
Pistachio processing wastewater	Al and Fe	Anaerobic digestion	43.7%	-	[161]
Sugar industry wastewater	Fe	Thermal	97.8%	-	[124]
Soybean oil wastewater	Fe	H ₂ O ₂	94%	-	[162]
Palm oil mill effluent	Fe	Coagulation	68.84%	-	[163]
Olive oil mill wastewater	Al	Bio-augmentation	63.9	-	[164]
Sugar industry wastewater	Fe	Chemical method	82%	84%	[165]
Olive oil mill wastewater	Al	External loop airlift reactor	79.24%	-	[166]
Olive oil mill wastewater	Al	Catalytic ozonation and bio-degradation	98.4%	-	[167]
Sugar industry wastewater	Fe	Sequential batch reactor	87%	-	[168]
Pistachio processing industry wastewater	Fe, stainless steel, Al and BDD	Fungal treatment	90.1%	-	[169]
Palm oil mill effluent	Al	Adsorption method	44%	89%	[170]
Olive oil mill wastewater	Al	Impregnation method	78%	-	[150]
Olive oil mill wastewater	Al	Peroxone process	79.8%	-	[171]

7. Energy Consumption and Cost

Energy consumption is one of the main factors for cost evaluation in electrochemical treatment on a large scale. Less energy was consumed, thereby reducing the cost required

for the treatment [172]. The amount of energy consumed per unit mass of organic material removed is referred to as the specific energy consumption. It could be COD, ammoniacal nitrogen, or dye, etc. [147]. Specific energy consumption can be evaluated as follows:

$$SEC = \frac{U \times I \times t}{(COD_x - COD_y) \times V} \quad (8)$$

where SEC is the specific energy consumption (kWh/kg of $COD_{removed}$), U is the applied voltage, I is the current intensity, t is the retention time, COD_x is the chemical oxygen demand before treatment, COD_y is the chemical oxygen demand after treatment, and V is the volume of treated wastewater [173].

Some studies have shown that the energy consumption is based on the electrode and operating parameters. Using Al electrodes on pulp and paper wastewater, the specific energy consumption is 11.055 kWh/m³ and requires an operating cost of 1.56 USD/m³ [174]. A previous study on the electrochemical treatment of carwash wastewater reported that the total operating costs under optimum conditions using aluminium and iron as the electrode materials are US\$ 0.3/m³ and US\$ 0.6/m³, respectively. Their performance could achieve 88% COD removal and 90% oil and grease removal using iron, and 88% COD removal and 68% oil and grease removal using aluminium [175]. Using graphite as the electrode material on textile wastewater treatment results in 85% color removal; the initial COD concentration was 724.75 mg/L, and the operating costs reached US\$ 338.42/kg [176]. The increment happened for the specific energy consumption of pulp and paper mill wastewater treatment when using the electrocoagulation process within the membrane reactor. Its specific energy consumption is 67.5 kWh/m³, and it costs around 21.03 USD/m³ [177].

8. Conclusions

The treatment of agro-based industrial wastewater can be achieved using the electrocoagulation process combined with other methods, such as physical, chemical and biological methods. However, the application of the electrocoagulation process at the full scale still faces several challenges, as follows:

- i It requires the understanding of economic feasibility and the effective power consumption method.
- ii Different types of wastewaters require different types of electrode materials. They should fulfil the criteria to enhance the performance of the treatment. Future studies of the development of electrode materials and the design are required.
- iii Ultra-stable electrolyte support may be required to achieve complete degradation and prevent the formation of unwanted by-products.

The electrocoagulation process offers several advantages for the treatment of agro-based industrial wastewater. A little study shows the specific component removal, such as for oil and grease, phenolic compound, carboxylic acid, and palmitic acid, etc. In the future, these studies need to be further conducted in order to give a better understanding and broaden knowledge on the electrocoagulation process.

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