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A Comparative Study of Electro and Chemical Coagulation for Efficient Removal of Lignin and Some Other Pollutants from Industrial Wastewater

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Abstract

The industrial factories especially pulp and paper as well as textile effluents are characterized by high amount of lignin. In addition, food industrial effluents contain fats, oil and grease, nitrogen phosphors, potassium and other pollutants. The producing wastewater containing lignin and several other contaminants becomes making many problems in the environment. The present work involves application of an electrochemical treatment (EC) to remove lignin and other contaminants from the industrial wastewater. Electrochemical coagulation cell was established and used for this purpose. The study was performed to check the removal efficiency of lignin and some other contaminants such as biological oxygen demand (BOD), chemical oxygen demand (COD), turbidity, total dissolved solids (TDS) and total suspended solids (TSS) from wastewater. The percentage removal of BOD, COD and TSS from wastewater by electrochemical coagulation are compared with those obtained using chemical coagulation with ferric chloride and both. The removal efficiency of these parameters when using electrocoagulation was 53%, 71% and 98% respectively. While the chemical coagulation by using ferric chloride was 42, 32 and 35 % respectively. The obtained results in this work reveal that electrochemical coagulation procedure is more effective than chemical coagulation when used in the treatment of industrial wastewater. In addition, the using of both technique in the same time give an excellent water clarity.

Keywords: Electrochemical cell; industrial wastewater; chemical coagulation; electrochemical coagulation.

1. Introduction

Sometimes the traditional methods of water treatment may become complex, expensive, sophisticated; and may require specialized technical knowledge [1]. Moreover, some of these methods may not be economically viable for some industries [2]. The treatment of raw and industrial wastewater by using electrocoagulation flotation method is more effective than the classic chemical coagulation for many reasons. Traditionally, chemical coagulation includes the use of ferric chloride (FeCl3), alum (aluminum sulfate), or ferrous sulfate (FeSO4). Traditionally, chemical coagulation method may be very expensive depending on the volume of treated water [3-5]. On the other hand, the electrocoagulation process works on the principle of oxidative or reductive chemistry and it needs relatively simple equipment at ambient temperature and pressure. Moreover, electrocoagulation is environmentally compatible, small area demanded, small volume of sludge produced, and short treatment time required [6, 7]. The sludge formed during electrocoagulation process can be used for extracting different valuable

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elements or as fertilizers [8-11]. Also, the effluent formed from the electrocoagulation can be used for irrigation, drinking and industrial purposes.

A limited study on the use of electrocoagulation were performed on industrial effluents [12, 13], pharmaceutical wastewater [14], and textile dye wastewater [15]. However, until today, electrocoagulation was not used in the large scales.

Some industries are categorized as heavily consuming water such as food processing, dairy products, paper mill, steam power plants, metal finishing and petroleum industry and this water needs some treatment before discharging [16-18]. Therefore, large amounts of industrial wastewater were discharged to the surrounding environment (public sewer system, underground water and surface water). These wastewaters must be subjected to pretreatment prior to discharge in an environmentally safe way. The treatment cost should be as low as possible to be economically valued. However, most of the conventional treatment techniques are highly cost. Therefore, there is a need to apply some modified techniques to reduce the treatment cost, to reuse or recycle the treated wastewater and to minimize the sludge produced from this treatment.

Water containing lignin is turned in importance from an environmental stand point, where most paper and food industries discharge this kind of wastewater directly into the surface water. It has to meet and subjugated to environmental protection agency (EPA) regulations on the entrained and dissolved lignin and other chemicals included in the wastewater [19].

Lignin is quite complex, with different structures and repeating units depending on the plant species, growing location, and growth duration [20, 21]. It is widely known that lignin is an amorphous phenolic polymer consisting of phenyl propane type fragments: sinapyl alcohol, p-coumaryl alcohol and coniferyl alcohol as shown in Figure (1) [20, 22].

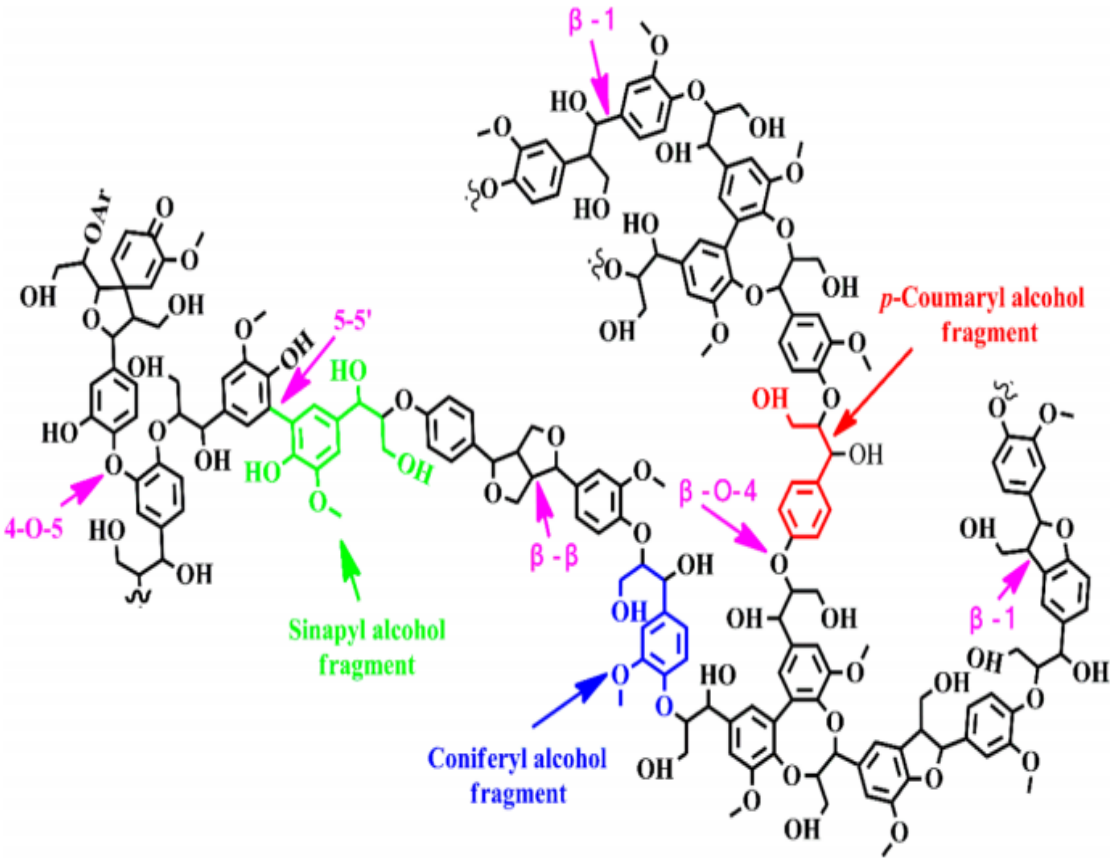

Fig.1. Structure of lignin.

Usually, lignin produced from hardwood is consists of equal amounts of coniferyl and sinapyl units and that obtained from softwood contains a large section of 92−95 coniferyl units. Lignin obtained from grass is typically composed of all three units [23]. The inter unit linkage, β-O-4 between the second C atom (β) of the side chain in one unit and the O atom connected to the fourth C atom of the benzene ring in the next unit represents the most common type among linkages lignin macromolecule units (~40−60%) [23].

On other hand, presence of lignin, biological oxygen demand (BOD), chemical oxygen demand (COD) and some heavy metals in water makes many troubles to the worker in the fields of wastewater treatment and they treat them as impurities. These impurities put the aquatic life in danger and leads to mutation and release of carcinogenic materials in several cases [24-28].

The specific objective of this study is to investigate the using of electrocoagulation treatment as an alternative technique for removing lignin, (BOD) and (COD) from aqueous solutions as a preliminary step for further large-scale application in industrial wastewater like paper and food industries in Egypt.

2. Experimental

2.1. Isolation of lignin

Lignin isolation process is accomplished in this work using an alkali method [29]. 20 grams of grinded Apricot kernel is inserted in the neck of 500 ml flask, then 120 mL demineralized water containing (1% H_2O_2) was added; and pH 9 was adjusted using 2N NaOH solution. The mixture was then isolated; heated and kept at 100˚C using a heating oil bath. Thereafter, the obtained solution (liquor) was filtered out and separated from the solids using Whatman filter paper. The filtrate containing concentrated (lignin) was then analyzed using UV-Vis spectrophotometer. The product was stocked for the preparation of aqueous solutions containing several concentrations of lignin to be representative samples of wastewater containing lignin.

2.2. Instrumentations and methods of analysis

During the electrochemical treatment of wastewater an electric DC power source (GW lab GPR-3030) was used as the source of direct electric current applied. Two aluminum sheets were used as the working electrodes (anode and cathode) of the electrolytic cell and connected to the positive and negative terminals of DC Power respectively as illustrated in Figure (2).

The spectrophotometric analysis of raw water before and after the electrochemical treatment was carried out to discern the removal efficiency according to equation (1) as follows:

$$Efficiency\ \% = ((C_0-C_e)/C_0) \times 100 \qquad (1)$$

Where C_0 and C_e are the concentration of contaminants before and after treatment (mg/l), respectively.

-The concentration of lignin was measured by using spectronic 2000 spectrophotometer.

-Measuring BOD by using Dissolved Oxygen Meter with BOD Probe YSI Incorporated, USA Model 5100-230V S/N 15A 103,525 and BOD Incubator, Thermo scientific, USA Model 3730-146E S/N 300027894.

-Measuring COD by using Digital Dispenser 10 ml, Dispensette RS, Brand, Germany, Heater, Electro-thermal, UK S/N: 10953529 and Automatic Burette, DIGITRAT, JENCONS, UK.

-Both TDS and TSS were determined using oven, Fisher Scientific, USA Model 738 F S/N 126 and Analytical Balance: ADAM AFA Germany.

-Electric conductivity was measured using Conductivity Meter, JENWAY, UK Model 4520 S/N 56,923.

-Turbidity was measured using Turbidimeter, HF Scientific, Inc. USA Model Micro 1000, S/N 201503151.

2.3. Electrocoagulation cell in the constructed plant:

Figure (2) shows the schematic diagram of our constructed treatment plant, which contain wastewater basin, clarification and solid waste removal unit, electrochemical cell, filtration unit and clarified water basin.

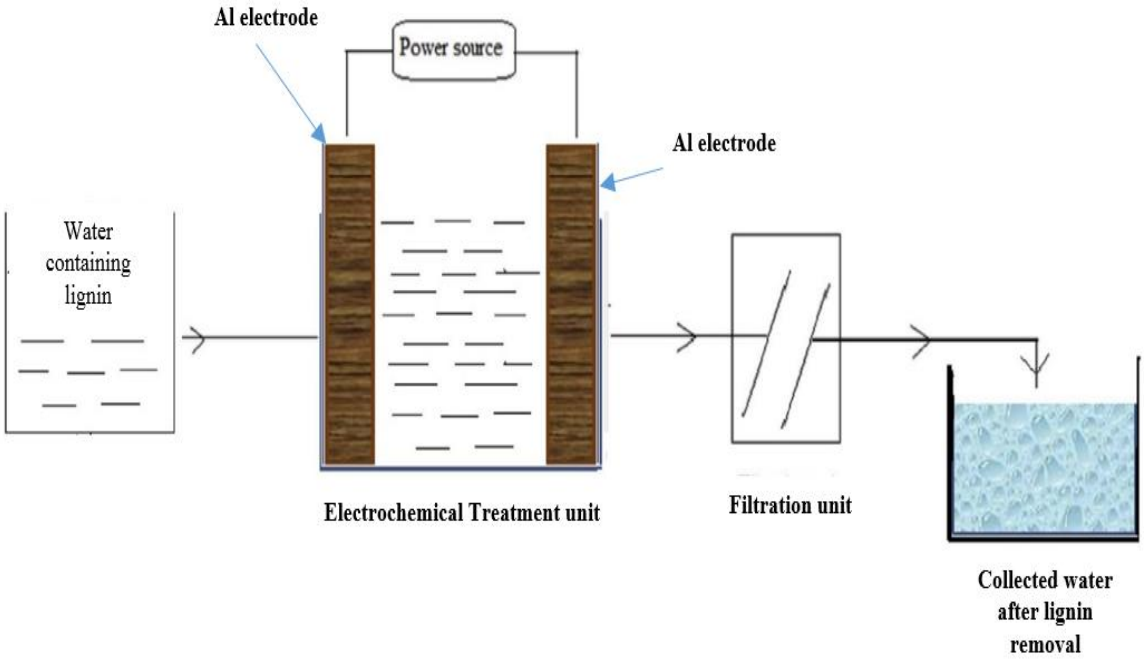

Fig. 2. Schematic diagram of treatment plant.

3. Results and discussion

3.1. Electrocoagulation mechanism

The Anode in the electrochemical cell produces the aluminum ions (Al^{3+}). During electrocoagulation these aluminum ions work as coagulant in situ by electrolytic oxidation. Charged ionic species are removed from wastewater by permitting it to react with either (i) ions having opposite charge, or (ii) flocs of metallic hydroxides produced within the effluent [30].

Electro-flotation, electro-oxidation and electro-coagulation occur jointly, and particles aggregate by combination of any of the above mechanisms or at least one of them. Flocculation can reinforce by continuous blending. Some of the reaction mechanisms that occur at the Aluminum (Al-Al) electrode (anode or cathode) are illustrated in equations (2-4) as follows:

$$Al_{(s)} \rightarrow Al^{3+}{}_{(aq)} + 3e^- \quad \text{(at anode)} \qquad (2)$$

$$3H_2O_{(l)} + 3e^- \rightarrow 3/2\ H_{2\,(g)} + 3OH^-{}_{(aq)} \quad \text{(at cathode)} \qquad (3)$$

Overall reaction:

$$Al^{3+}{}_{(aq)} + 3OH^-{}_{(aq)} \rightarrow Al(OH)_{3(s)}) \qquad (4)$$ [31-33].

The aluminum hydroxide flocs have large surface area, are adsorbed, trapped or polymerized colloidal particles, and can extracted from the aqueous solution. Aluminum hydroxide is also an important adsorbent of organic and inorganic ions, molecules and colloidal particles [34].

3.2. Lignin removal from water using electrocoagulation

The water containing lignin must be treated by using several processes such as chemical coagulation or electrochemical treatment, etc.…, before discharging it. This treatment is called water remediation. As shown in Fig.1, the hydroxyl groups in lignin macromolecules are crucial because they play

an important role in the chemical and physical properties of lignin such as reactivity, hydrophilicity, and functionality [35, 36], which responsible for metal ion absorptivity. Thus, the removing of lignin from water is usually accompanied by removing metal ion from that water.

3.2.1. FTIR of isolated lignin

The fine powder of isolated lignin from Apricot kernel was investigated using FTIR technique. The investigation was conducted to identify and characterize the product by comparing it with pure lignin. Thus, the FTIR spectra of the produced isolated lignin and pure one was studied to show its characteristics.

It was concluded from Figure (3) that all the peaks detected for the isolated lignin are similar to those of pure lignin and related to C-OH, CH, CH_2, CH_3 and C=O groups. The FTIR spectra of pure lignin showed a peak at 3413.44 cm^{-1} which assigned to OH stretching vibration of hydroxyl group of the lignin; and a symmetric stretch for CH_3 of methoxyl group appeared at 2840 cm^{-1}. The absorbance band detected at 2927.39 cm^{-1} appeared from C-H stretching in methyl and methylene group. A peak at 1713.83 cm^{-1} assigned to carbonyl stretching–unconjugated ketone and carboxyl groups. The peaks observed at 1508.81cm^{-1}, 1458.39 cm^{-1} and 1426.41 cm^{-1} are corresponding to the aromatic skeletal vibrations and there is β-O-4 ether bond band at 1119.78 cm^{-1}. A small peak at 1035 cm^{-1} may be due to aromatic CH in plane deformation, guaiacyl type and C-O stretching for primary alcohol.

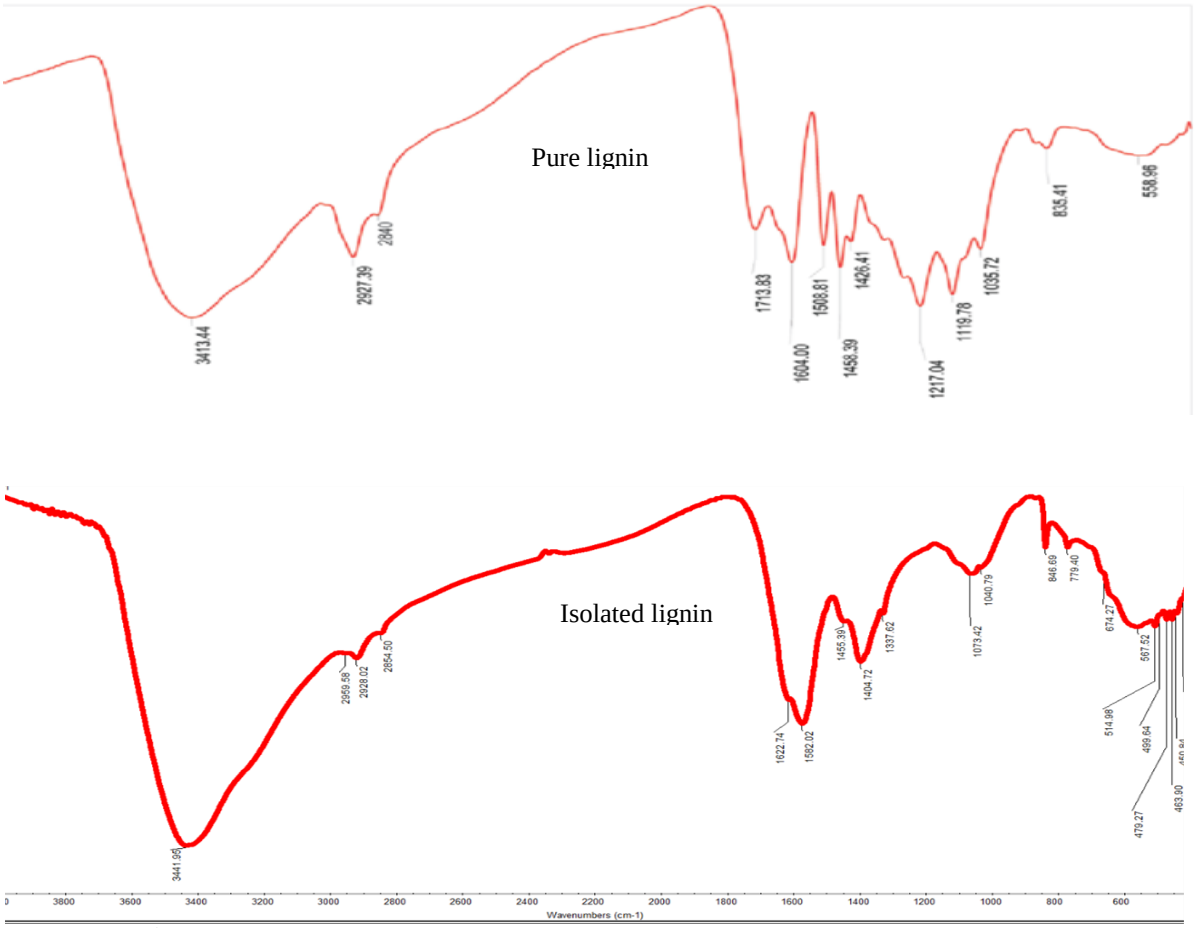

Fig. 3. FTIR of pure and isolated lignin.

3.2.2 Determination of lignin in water:

Lignin concentration of liquor was analyzed by UV-Vis spectrophotometer with a resolution of 1 nm. Firstly, the wavelength of maximum absorption was determined using 100 ppm standard solution of lignin by changing the wavelength and measuring the corresponding absorbance as demonstrated in Figure (4).

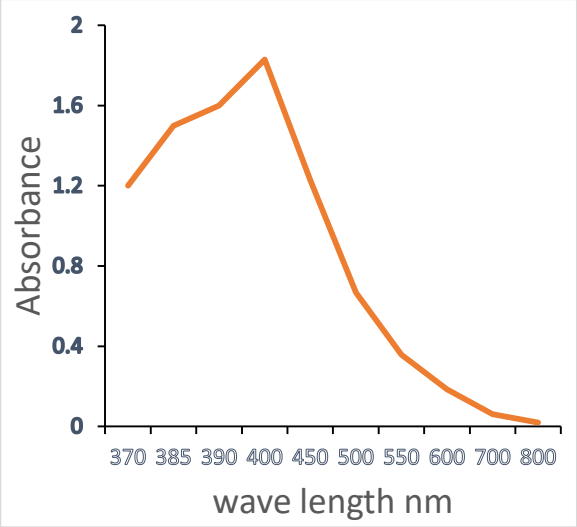

Fig.4. The variation of wavelength against absorption.

The maximum absorbance was detected at λ_{max} = 420 nm. It follows that, two series of different concentrations of lignin solution were prepared to formulate calibration curves for low and high standard concentrations of lignin solutions and illustrated in Figs (5) and (6) respectively.

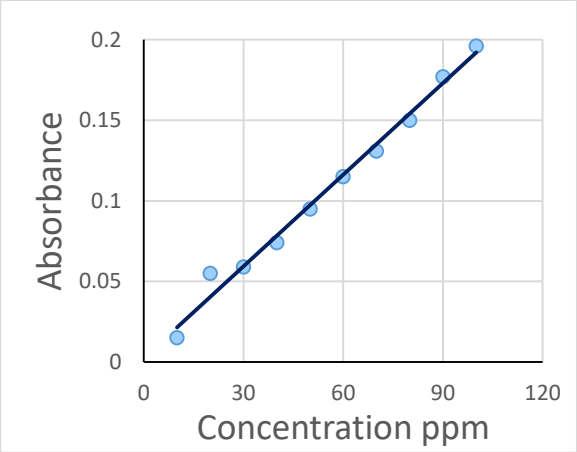

Fig.5. Calibration curve of low standard concentration lignin.

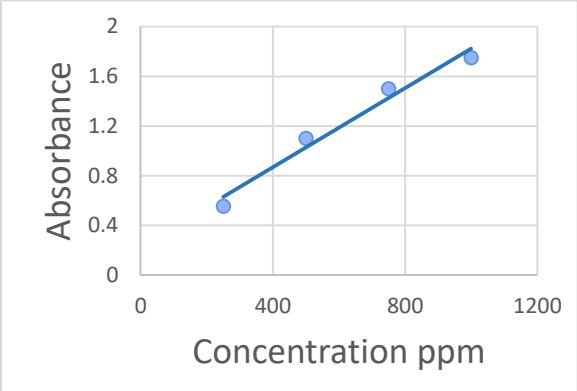

Fig. 6. Calibration curve of high standard concentration lignin.

3.2.3 Effect of applied potential:

A sample of industrial wastewater is prepared, and all specifications measured and listed in table (1).

The applied potential was changed at different time and pH values to study its impact on the removal of lignin. For each change, the concentration of lignin was determined spectrophotometrically and the removal percentage of lignin was calculated and the results were tabulated in tables (2-5). Firstly, concerning the potential, it is found that the removal percentage is increased by increasing the applied potential and the time taken to remove the lignin from the water is decreased.

Table 1
The specifications of wastewater sample

No.	Parameters	Result
1.	Lignin (ppm)	100
2.	pH	7.2
3.	Turbidity (NTU)	26.4
4.	Color	Deep brown
5.	BOD (5 day, 20°C) (ppm)	530
6.	COD Dichromate (ppm)	1857
7.	Total hardness as $CaCO_3$ (ppm)	250
8.	Calcium hardness as $CaCO_3$ (ppm)	167
9.	Magnesium hardness as $CaCO_3$ (ppm)	83
10.	Total alkalinity $CaCO_3$ (ppm)	149
11.	Total Dissolved solids (ppm)	1585
12.	Total Suspended solids (ppm)	36
13.	Oil and Grease (ppm)	11.55
14.	Organic matter as ($KMnO_4$) ppm	29

Secondly, with respect to effect of pH, as the pH of the solution increases at constant potential and time, in addition to increasing of removal percentage. The highest removal percentage value of lignin was found at pH 6 and 8 after 25 min. at applying potentials 10 and 12 volts, respectively. At these conditions the amount of aluminum ions released is suitable for the formation of aluminum hydroxide flocs of large surface area by which the colloidal lignin particles are adsorbed, trapped or polymerized leading to the maximum removal percentage of lignin from the wastewater sample.

Table 2
Variation of removal percentage of lignin with the operation conditions of electrochemical cell at (PH=2)

Applied potential (volts)	% Removal Time (min)				
	5	10	15	20	25
2v	20.1	39.3	45.3	49.4	51.3
4v	31.3	44.4	49.2	55.3	59.2
6 v	39.5	49.5	54.5	59.6	62.6
8v	47.3	53.3	59.1	64.2	65.7
10 v	51.4	58.7	62.3	66.6	68.4
12 v	53.4	59.7	64.3	67.2	69.5

Table 3
Variation of removal percentage of lignin with the operation conditions of electrochemical cell at (PH=4)

Applied potential (volts)	% Removal Time (min)				
	5	10	15	20	25
2v	25.1	44.5	52.3	56.3	59.3
4v	39.3	49.6	54.7	59.5	62.4
6 v	47.4	53.7	59.5	64.6	65.5
8v	51.3	58.3	62.9	66.7	68.2
10 v	56.4	64.4	68.3	72.1	77.7
12 v	57.4	66.5	69.6	74.3	79.2

Table 4
Variation of removal percentage of lignin with the operation conditions of electrochemical cell at (PH=6)

Applied potential (volts)	% Removal Time (min)				
	5	10	15	20	25
2v	49.5	59.3	64.5	79.3	82.3
4v	57.7	63.5	69.6	74.4	75.4
6 v	61.3	68.6	72.3	76.5	78.5
8v	66.4	74,7	78.3	82.6	87.2
10 v	74.6	82.3	87.5	92.6	97.2
12 v	76.7	84.2	89.7	94.7	97.7

Table 5
Variation of removal percentage of lignin with the operation conditions of electrochemical cell at (PH=8)

Applied potential (volts)	% Removal Time (min)				
	5	10	15	20	25
2v	57.4	63.4	69.4	74.7	75.5
4v	61.5	68.5	72.5	76.8	78.4
6 v	66.7	74.7	78.5	82.9	87.7
8v	74.5	82.3	87.5	92.3	98.0
10 v	76.8	84.4	91.5	95.7	98.0
12 v	77.2	85.6	92.5	96.7	98.0

3.3. Effect of applied potential and applied current density on the properties of cell electrodes

The relations between the weight loss of the cell aluminum electrodes and some applied parameters such as potential, current density and time are studied, and the obtained results were listed in Table (6). It has been found that the percentage of weight loss increases by increasing the applied potential. Also, the weight loss increases with increasing the applied current density increases at constant potential and time. The highest percentage of weight loss for (Al-Al) electrode was 7.8% at 12V and 3.67 A after 25min.

Table 6
variation of weight loss of ammonium electrode with the change of cell parameters

Applied potential (Volts)	Time (min)	Current (Amp.)	Weight % of (Al) electrode	%Weight loss
2	5	0.14	99.78	0.22
2	10	0.158	99.56	0.44
2	15	0.162	99.13	0.87
2	20	0.178	98.69	1.31
2	25	0.183	98.47	1.53
4	5	0.57	99.34	0.66
4	10	0.65	98.69	1.31
4	15	0.66	98.04	1.96
4	20	0.68	97.6	2.4
4	25	0.7	97.1	2.8
8	5	1.59	98.91	1.09
8	10	1.68	97.82	2.18
8	15	1.86	96.95	3.05
8	20	1.73	96.3	3.7
8	25	1.71	96.0	4.0
12	5	3.02	97.17	2.83
12	10	3.54	94.56	5.44
12	15	3.57	93.7	6.3
12	20	3.6	93.0	7.0
12	25	3.67	92.2	7.8

3.4. Cost estimation

The energy consumption (E) of the applied electrochemical cell could be calculated using the following equation [37].

Energy consumption (E) =
¼ Volt (V) x Current (A) x Time (sec) (5)
Or

$$E = 12 \times 1.5 \times (25 \times 60)$$
$$= 6750\ jl^{-1}$$
$$= 6750 / 1000 \times 3600$$
$$= 0.001875\ KW\ l^{-1}$$

These results indicate that the Al electrode achieve lowest consumption of electric current. In addition, it achieved the more efficient treatment as lignin isolation and water clarity.

3.5. Comparison between chemical and electrochemical coagulation methods on the removal percentage of (BOD), (COD) and (TSS) from wastewater

Concerning the effect of ferric chloride and polyelectrolyte as conventional method of chemical coagulation on the biochemical oxygen demand (BOD), chemical oxygen demand (COD) and total suspended solid (TSS) were studied and illustrated in Figs. (7-9). The lowest doses for maximum removal of BOD, COD and TSS were chosen; so, the dose of polyelectrolyte was kept constant at (0.6) mg/l and different doses of ferric chloride verses the measured BOD, COD and TSS were taken. Figure (7) showed that the dose of (30) and (0.6) mg/l of ferric chloride and polyelectrolyte respectively recoded. The maximum (BOD) removal reaching (42%) then remained constant by increasing ferric chloride dose.

The same trend was observed at (25) and (0.6) mg/l ferric chloride and polyelectrolyte respectively for each of COD and TSS, which gave percentage reduction reached to (32%) and (35%) respectively as can be seen from Figures (8 and 9).

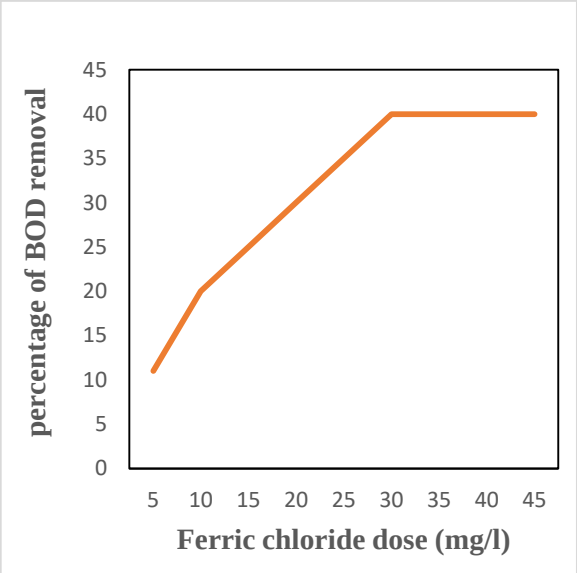

Fig.7. BOD removal from wastewater at different ferric chloride doses and (0.6) mg/l polyelectrolyte.

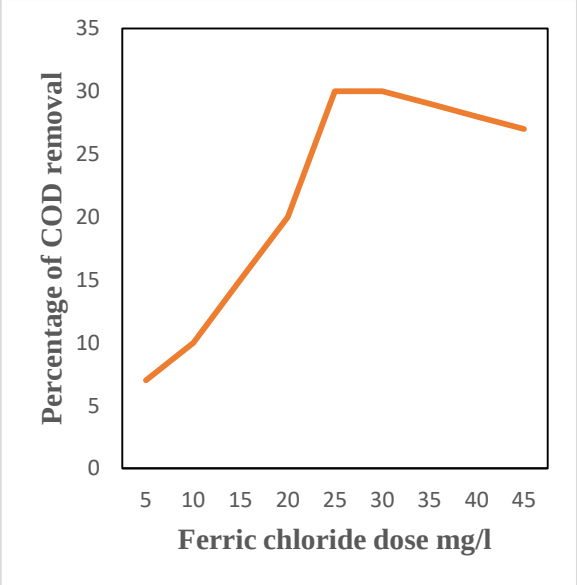

Fig.8. COD removal from wastewater at different ferric chloride doses and (0.6) mg/l polyelectrolyte.

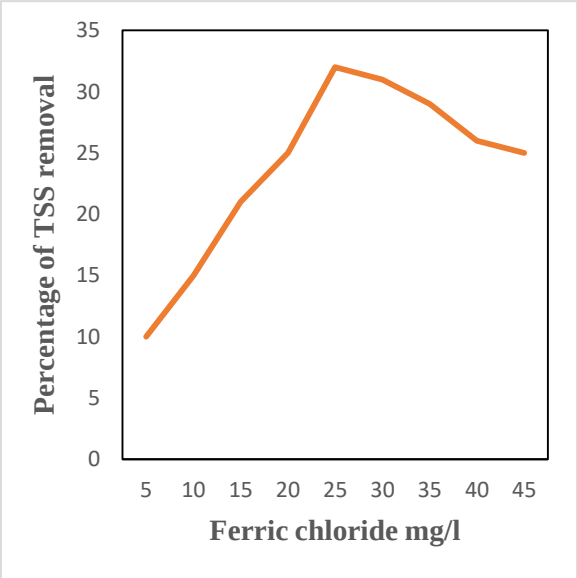

Fig.9. TSS removal from wastewater at different ferric chloride doses and (0.6) mg/l polyelectrolyte.

In addition to collective table (7) was made to explain the maximum percentage removal of the above-mentioned parameters (BOD, COD and TSS) where they are the most parameters used to show the efficiency of clarification technique.

Table 7
Removal efficiency for (BOD, COD and TSS) of wastewater by using Chemical and electrochemical coagulation

Method of clarification	Removal efficiency %		
	BOD	COD	TSS
Electro chemical coagulation	53	71	98
Chemical coagulation by ferric chloride	42	32	35

3.6. Effect of electro, chemical and both on the specifications of treated water

Some of previous work are listed in literature concerning removal of lignin and other contaminants by using chemical and electro coagulation [38-42] but there is a shortage in the comparison between these techniques. Therefore, the effect of electro and chemical coagulation and combination of both on the specification of treated water compared and studied. Table (8) showed that the parameter studied for treated water by using electrocoagulation technique in this work was enhanced and gives better results than chemical coagulation. In addition, they have given an excellent water clarity when used together in the clarification of the examined industrial wastewater.

Table 8
Effect of electro and chemical coagulation and the combination of both on the specification of treated water

Parameters	Industrial wastewater	Treated water using electro coagulation	Treated water using chemical coagulation	Treated water using both techniques
Lignin (ppm)	100	2	11	Nil
pH	7.2	7.1	6.7	7
Turbidity (NTU)	26.4	3.2	8.7	0.9
Color	Deep brown	Clear	Light brown	Very clear
BOD (5 day, 20°C) (ppm)	530	249	307	42.4
COD Dichromate (ppm)	1857	525.75	1262.76	241.41
Total hardness as $CaCO_3$ (ppm)	250	145	180	137
Calcium hardness as $CaCO_3$ (ppm)	167	90	110	83
Magnesium hardness as $CaCO_3$ (ppm)	83	55	57	54
Total alkalinity $CaCO_3$ (ppm)	149	140	117	135
Total Dissolved solids (ppm)	1585	850	136	790
Total Suspended solids (ppm)	36	0.72	23.4	0.36
Oil and Grease (ppm)	11.55	2.9	5.2	2.1
Organic matter as ($KMnO_4$) ppm	29	4.3	6.7	3.8

4. Conclusions

The suggested electrochemical cell in this study was applied to remove lignin and some contaminants from industrial wastewater. The percentage removal efficiency of lignin polymer from waste water and some parameters such as (BOD, COD and TSS) was compared between chemical coagulation by using ferric chloride and electrochemical coagulation, the results obtained revealed that electrochemical coagulation is more effective than chemical coagulation for that parameters. Nevertheless, combination between both electrochemical and chemical coagulation gives an excellent water clarity when used together.

5. Conflicts of interest

"There are no conflicts to declare".

6. Acknowledgments

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