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Investigation for the possibility of nitrogen removal from industrial effluent of bone glue industry using a batch electrocoagulation unit with monopolar horizontal electrodes

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ABSTRACT

The aim of the present work is to investigate the possibility of using the electrocoagulation technique (EC) for the treatment of wastewater generated from bone glue industry especially for the removal of nitrogen as pollutant. A batch electrochemical reactor with monopolar horizontally oriented aluminum electrodes was used. Variables studied were initial nitrogen concentration measured as total Kjeldahl nitrogen (TKN), applied current density, distance between the electrodes, sodium chloride concentration, electrode area ratio (cathode/anode) and time of electrolysis. The results show that up to 90% of TKN of a synthetic waste can be removed within 60 min. In addition, a real wastewater sample from a bone glue factory located in Alexandria, Egypt, was investigated under different current densities. The results show that 60–80% of TKN can be removed within 120 min depending on the operating conditions. At the same time, a reduction in BOD, COD, and TSS of about 68%, 61% and 85% respectively can be achieved within 120 min.

Keywords: Electrocoagulation; Nitrogen removal; Monopolar electrode; Wastewater treatment; Bone glue industry

1. Introduction

During the last few decades new wastewater treatment plants have been designed for tertiary treatment, especially for the removal of nutrients such as nitrogen and phosphorous. Nitrogen effluent from fertilizers, tanneries, sewage, bone glue industry, etc. is a pollutant of concern for a number of reasons. Nitrogen in the ammonia form is toxic to certain aquatic organisms. In the environment, ammonia is oxidized rapidly to nitrate, creating an oxygen demand and low dissolved oxygen in surface waters. Organic and inorganic forms of nitrogen may cause eutrophication (i.e., high productivity of algae) problems in nitrogen-limited freshwater lakes and coastal waters. Finally, high concentrations of nitrate can harm young children when ingested [1]. Nitrogen removal is increasingly required when onsite systems are on or near coastal waters or over sensitive, unconfined aquifers used for drinking water. Nitrogen removal systems are generally located at the end in the treatment train prior to disposal and may be followed by disinfection when the system must discharge into surface waters. Usually, the minimum quantity of total nitrogen that can be regularly met is about 10 mg/L [2]. Enhanced nitrogen removal systems can be categorized by their mode of removal. Physical and chemical systems such as ion exchange, volatilization, and membrane processes are capable of 80–90% removal. Ion exchange resins remove NH_4-N or NO_3-N.

Membrane processes employ a variety of membranes and pressures that all have a significant rejection rate. None of the above found industrial applications because of higher operation costs [3]. The vast majorities of practical nitrogen-removal systems employ nitrification and denitrification biological reactions. Most notable of these are recirculating sand filters (RSFs) with enhanced anoxic modifications, sequencing batch reactors (SBR), and an array of aerobic nitrification processes combined with an anoxic/anaerobic process to perform denitrification [3,4]. Nitrification is the biological oxidation of ammonium, with nitrate as the end product, while denitrification is the biological reduction of nitrate to molecular nitrogen. Nitrification denitrification technique can remove up to 90% of the total nitrogen content of wastewater, with extremely controlled conditions; otherwise the system will experience operational instability due to the formation of a floating layer composed of sludge flocks with attached nitrogen bubbles, which will eventually lead to loss of sludge due to unintentional discharge with the effluent. Of course, the presence of sludge in the effluent will lead to unacceptable high concentration of suspended solids and organic matter. Another negative aspect of the floating layer (sludge loss) is the reduction in sludge mass and sludge age. The remaining sludge mass may be insufficient to metabolize the influent organic matter, resulting in the reduction of secondary treatment efficiency. Further the reduction in sludge age may lead to washout of the nitrifiers and hence interruption of the nitrification process [5].

The objective of this research is the implementation of the electrochemical technique (electrocoagulation) for the removal of nitrogen from industrial waste water using a cell of horizontally oriented electrodes; previous studies on electrocoagulation [6] have shown that the performance of horizontal cells is superior to vertical cells. Electrocoagulation has proved to be a powerful tool for the treatment of wastewater contaminated with heavy metals [7–12], color [13], organic substances [13,14].clay minerals [15,16], as well as oil [17,18] and chemical oxygen demand [17–20]. Electrocoagulation safely eliminates the disadvantages of the classical treatment techniques to achieve a sustainable and economic treatment of polluted wastewater [21–23]. This technique does not require addition of supplementary chemicals, reduces the volume of produced sludge [22–24], and first economical studies indicate a financial advantage compared to the conventional methods [22].

Many investigations were carried out for the removal of nitrite and ammonia [25,26]. Lin and Wu [26] found that extended time is required for the removal of nitrite using the electrochemical techniques. Generally the results of electrocoagulation process for the treatment of nitrate, nitrite and ammonia indicate that using electrocoagulation removes the pollutants from aqueous solution Emamjomeh and Sivakumar [27] using batch electrocoagulation/electrofloatation reactor found that nitrate removal efficiency depends on electrolysis time and current values. However, further investigations need to be done when the removal mechanisms were not considered in the literature [28]. The above shows that scant investigations were carried out for the removal of nitrogen produced with wastewater especially that produced from bone glue industry. The aim of the present work is to implement electrocoagulation technique for the treatment of wastewater generated from bone glue industry especially for the removal of nitrogen in a batch monopolar electrode reactor.

2. Experimental setup and procedure

The experimental setup and electrical circuit used is shown in Fig. 1. The setup consists of 1000 ml cylindrical glass container of 11 cm diameter and 15 cm height, two monopolar aluminum electrodes (cathode and anode) placed horizontally, with the anode to the top of the cell parallel to the cathode at the bottom with electrode spacing ranging from 5 to 10 cm. Before each run aluminum electrodes were immersed in diluted carbon tetrachloride solution for a few minutes for removing greases or oxides, washed with distilled water, and finally connected to a power supply (DC, 5 V, 10 A) fitted with a voltage regulator. A multirange ammeter was connected in series with the cell and a DC voltmeter was connected in parallel with the cell to measure its voltage. In each run 1000 ml of synthetic wastewater were placed at the electrolytic cell with total Kjeldahl Nitrogen content (TKN) ranging from 168 to 813 ppm. The study was carried out using two different wastewater samples. The first is a synthetic waste prepared by dissolving a known weight of well dried bone glue (granular particles were used) in distilled water and the second is a real waste effluent

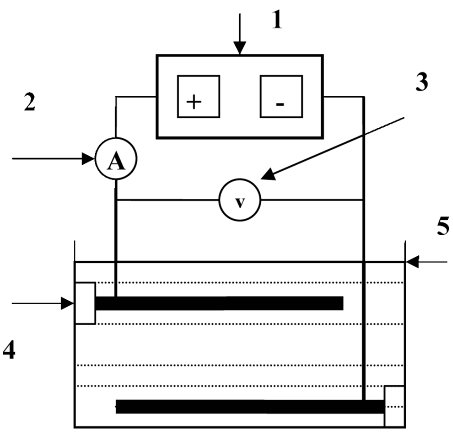

Fig. 1. Experimental setup and the electrical cell. 1 – DC power supply; 2 – ammeter; 3 –voltmeter; 4 – plastic support; 5 – wastewater level.

obtained from a bone glue factory located in Alexandria, Egypt. TKN for the fresh and treated wastewater was measured before and after each run [29]. Many variables were investigated for the effect on the removal efficiency of the TKN of wastewater such as, time of operation that ranged from 10 to 60 min, initial nitrogen concentration measured as (TKN), where 5 different concentrations ranging from 168 to 813 were examined, applied current density which ranged from 1 to 4 mA/cm^2, concentration of sodium chloride as electrolyte which ranged from 1 to 10 wt%, distance between electrodes from 3 to 9 cm was investigated and finally the area ratio (cathode/anode) in the range of 1–4 cm was investigated.

2.1. Description of the methods of analysis

The principle of the method used for organic nitrogen determination (total Kjeldahl nitrogen) is the conversion of the nitrogen of the nitrogenous substance into ammonia by boiling with concentrated sulphuric acid which is fixed by the excess of acid as ammonium sulphate. The latter is determined by adding an excess of caustic alkali to the solution after digestion with the acid, and distilling off the liberated ammonia into standard acid (boric acid was used). The acid (boric acid) is back titrated against standard sodium hydroxide in presence of methyl red indicator. Mercury (II) oxide and potassium sulphate was added to increase the speed of reaction. The described process is working well for nitrogen determination of proteins and also in amines and amides [29,30].

TSS, COD and BOD were determined in conformity with standard methods [29].

3. Results and discussion

3.1. Effect of applied current density

As shown in Fig. 2, the percentage of nitrogen removal increased by increasing the applied current density within the range of 1–4 mA/cm^2. These results can be explained by the fact that increasing current density will increase the dissolution rate of aluminium electrode according to Faraday's law with the formation Al^{3+} and hence the formation of Al(OH)$_3$ coagulant according to the following reactions:

Anode:

$$2Al \rightarrow 2Al^{3+} + 6e \quad (1)$$

Cathode:

$$6H_2O + 6e \rightarrow 3H_2 + 6OH^- \quad (2)$$

$$2Al^{3+} + 6OH^- \rightarrow 2Al(OH)_3 \quad (3)$$

The overall:

$$Al + 3H_2O \rightarrow Al(OH)_3 + 3/2\ H_2 \quad (4)$$

Higher rate of freshly formed amorphous Al(OH)$_3$ has a large surface area on which rapid adsorption of soluble nitrogen and trapping of colloidal particles take place with a consequent removal of nitrogen from wastewater. In addition, the cathodically evolved H$_2$ bubbles float Al(OH)$_3$ along with the adsorbed nitrogen compounds to the upper surface of the solution. Besides, the rising H$_2$ bubbles entrain solution in their wake [6]; the upward rising gas-liquid dispersion stirs the anode vicinity with a consequent decrease in the anode concentration polarization and the anode tendency to passivate. Fig. 3 shows the flow pattern of the rising gas–liquid dispersion.

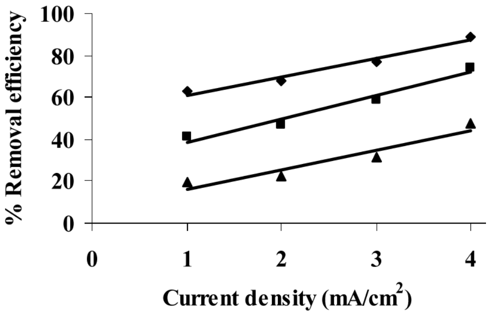

Fig. 2. % Removal efficiency vs. current density at different initial nitrogen concentrations. Distance between electrodes = 5 cm, Area ratio (cathode/anode) = 1, NaCl concentration = 3%, Operating time = 60 min, Initial nitrogen concentration (mg/L) ♦ 168, ■ 399, ▲ 813.

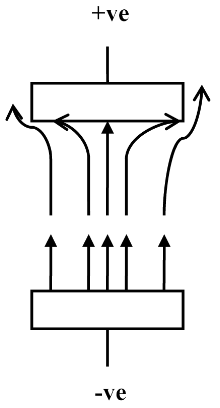

Fig. 3. Approximate flow pattern of the gas (H$_2$)–liquid dispersion in the cell.

3.2. Effect of initial nitrogen concentration

As shown in Fig. 4, the % nitrogen removal efficiency decreased by increasing initial nitrogen concentration in the range of 168–813 ppm. This can be ascribed to the fact that increasing nitrogen concentration would probably increase activation polarization via adsorption on the anode and cathode with a consequent decrease in the rate of aluminium dissolution at the anode and hydrogen evolution at the cathode. In addition, higher

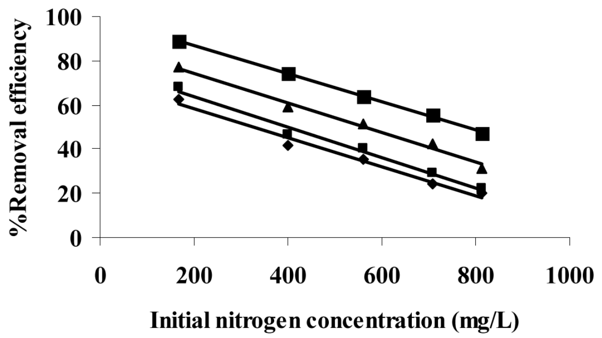

Fig. 4. % Removal efficiency vs. initial nitrogen concentration at different current densities. Distance between electrodes = 5 cm, Area ratio (cathode/anode) = 1, NaCl concentration = 3%, Operating time = 60 min, Current density (mA/cm^2) ♦ 1, ▪ 2, ▲ 3, ■ 4.

concentrations of nitrogen will block adsorption sites of $Al(OH)_3$ rapidly and decrease its ability to adsorb more nitrogen compound.

3.3. *Effect of NaCl concentration*

The effect of initial concentration of NaCl as electrolyte was investigated for different current densities as shown in Fig. 5, the results show that the % removal efficiency of nitrogen increases by increasing sodium chloride concentration in the range of 1–10% NaCl, this can be ascribed to the depassivation effect of chloride ion on aluminium electrode [31] which increases the rate of Al^{3+} and consequently $Al(OH)_3$ formation which increases the % of nitrogen removal efficiency up to 90%.

Increasing NaCl concentration above approximately 5% has no effect on increasing the % of nitrogen removal at different current densities, it seems that at 5% NaCl concentration complete depassivation of aluminium electrode takes place and further increase in NaCl concentration would have little effect on the removal efficiency of nitrogen compounds.

3.4. *Effect of distance between electrodes*

As shown in Fig. 6, the % removal efficiency of nitrogen increased by increasing the distance between the electrodes (electrode separation). The increase in the % removal efficiency with increasing interelectrode separation may be attributed to the decrease in concentration polarization at the Al anode as a result of the rapid transfer of dissolved Al^{3+} from the anode surface to the solution bulk by the combined diffusion and natural convection by virtue of increasing the volume of the solution located between the two electrodes. The higher the bulk volume the lower the bulk concentration of Al^{3+}, and the higher the driving force (concentration difference) for Al^{3+} diffusion from the anode surface to the bulk solution [31]. High concentration polarization tends to increase the anode potential and promote anode passivity which lowers the % removal efficiency owing to the decrease in the amount of dissolved Al^{3+}.

3.5. *Effect of area ratio (cathode/anode)*

As shown in Fig. 7, the % removal efficiency of nitrogen decreased by increasing the area ratio (cathode/anode). Increasing the cathode area lowers the current density on the cathode surface with a consequent production of a large number of small sized H_2 bubbles which are more effective in floatation than large sized particles [29] that would reduce the anode efficiency for Al^{3+} production.

3.6. *Effect of operating time*

The % removal efficiency of nitrogen was increased by increasing the operating time of the cell as shown in Fig. 8. It is clear that approximately about 60–90% of total

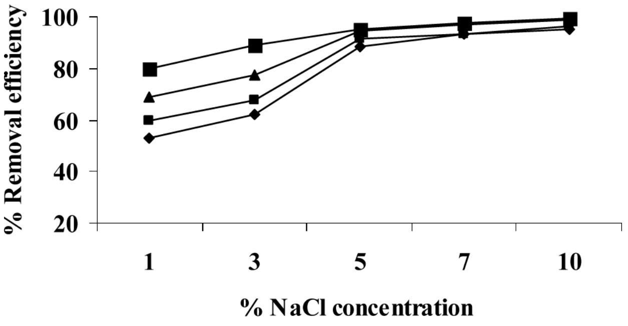

Fig. 5. % Removal efficiency vs. NaCl concentration at different current densities. Initial nitrogen concentration = 168 mg/L, Distance between electrodes = 5 cm, Area ratio (cathode/anode) = 1, Operating time = 60 min, Current density (mA/cm^2) ♦ 1, ▪ 2, ▲ 3, ■ 4.

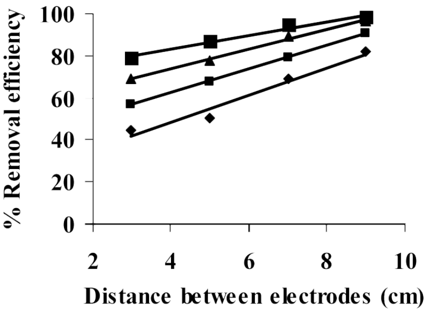

Fig. 6. % Removal efficiency vs. distance between electrodes at different current densities. Initial nitrogen concentration = 168 mg/L, Area ratio (cathode/anode) = 1, NaCl concentration = 3%, Operating time = 60 min, Current density (mA/cm^2) ♦ 1, ▪ 2, ▲ 3, ■ 4.

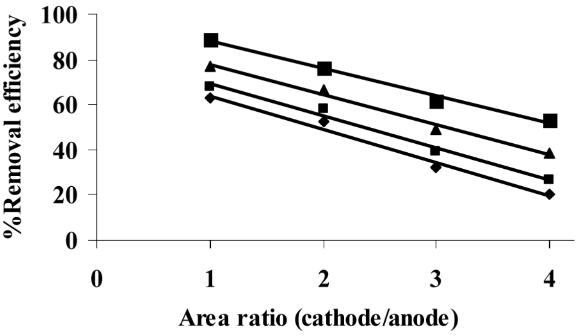

Fig. 7. % Removal efficiency vs. area ratio (cathode/anode) at different current densities. Initial nitrogen concentration = 168 mg/L, Distance between electrodes = 5 cm, NaCl concentration = 3%, Operating time = 60 min, Current density (mA/cm^2) ♦ 1, ▪ 2, ▲ 3, ■ 4.

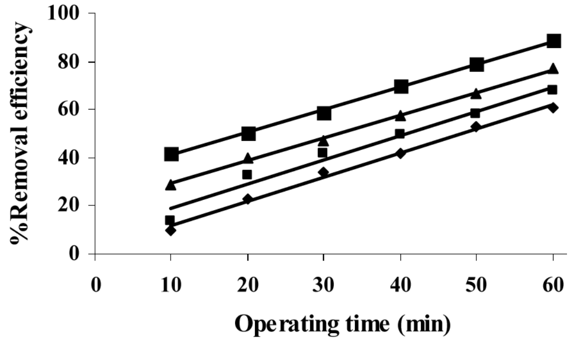

Fig. 8. % Removal efficiency vs. operating time at different current densities. Initial nitrogen concentration = 168 mg/L, Distance between electrodes = 5 cm, Area ratio (cathode/anode) = 1, NaCl concentration = 3%, Current density (mA/cm^2) ♦ 1, ▪ 2, ▲ 3, ■ 4.

nitrogen was removed within 60 min at current densities of 1–4 mA/cm^2, respectively. These results show that electrocoagulation of nitrogen is faster than the biological methods which require more duration for the same removal efficiencies; this would improve the capacity of the wastewater treatment plants [27,28,32].

Table 1
Characteristics of sample of industrial waste

pH	8–9.5
COD, mgL^{-1}	10,500
BOD, mgL^{-1}	6280
TSS (suspended matter), mgL^{-1}	2010
N_2, TKN	680
Oils and grease, mgL^{-1}	25

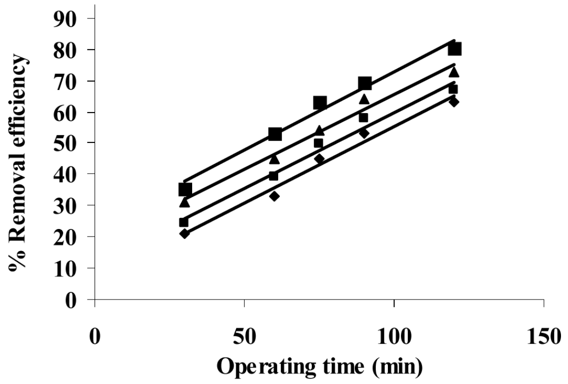

Fig. 9. % Removal efficiency vs. operating time at different current densities for real sample of bone glue wastewater plant. Distance between electrodes = 5 cm, Area ratio (cathode/anode) = 1, NaCl concentration = 3%, Initial N_2 concentration = 660 mg/L, Current density (mA/cm^2) ♦ 1, ▪ 2, ▲ 3, ■ 4.

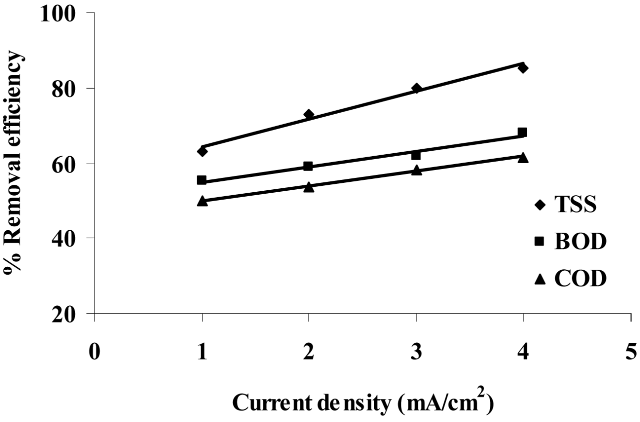

Fig. 10. % Removal efficiency of TSS, BOD and COD vs. current density for the real sample.

3.7. *Application of the present results to real industrial wastewater effluent from bone glue industry*

A representative real sample was collected from a plant producing glue. The analysis of the sample is shown in Table 1. Samples were treated under different conditions of current density ranging from 1 to 4 mA/cm^2 as shown in Fig. 9. The results show that about 60–80% of total nitrogen can be removed within 120 min depending on the applied current density. In addition for the true sample treatment a reduction in BOD, COD, and TSS up to 68%, 61% and 85% respectively can be achieved within the same time interval as shown in Fig. 10. The longer time required for the treatment of the real sample can be ascribed to the fact that part of the produced $Al(OH)_3$ is used for treating BOD, COD, suspended solids, etc. It has to be mentioned that removal of nitrogen is usually carried out at tertiary treatment steps, i.e. the effluent to the electrochemical unit will be of lower BOD and COD values, this would reduce $Al(OH)_3$ consumption and improve the unit performance. In addition to this, removal of the residual BOD and COD in the electrochemical unit will certainly improve the quality of treated water further more.

3.8. Sludge generation

The amount of sludge generated (dissolved electrode + adsorbed glue molecule) due to the electrocoagulation process was calculated under different conditions of current density ranging from 1 to 4 mA/cm^2. 500 mg of bone dried glue was dissolved in 1000 ml distilled water and the cell was connected for about 120 min. Longer time was used to ensure complete removal of the glue added. At the end of electrolysis time the formed sludge was collected on a filter cloth of known weight and then dried in an oven at 105°C for about 4 h to a constant weight. The difference in weight was considered as the sludge weight. The results show that the amount of sludge generated is proportional to the applied current density. Fig. 11 compares the amount of sludge generated with the calculated amount using Faraday's law. The results show that the amount of sludge generated is less than the calculated amount which can be attributed to the current inefficiency due to IR drop and cell resistance.

3.9. Electric power consumption

The electric power consumption of the process was calculated per m^3 of the waste solution using the equation that [32,33]:

$$P = EIt/1000V \quad (5)$$

where P is the specific power consumption (kWh/m^3), E is the cell voltage in volt (V), I is the current in ampere (A), t is the time of electrocoagulation in hour (h) and V is the solution volume (m^3). The time of operation was fixed at 60 min. It is clear from Fig. 12 that increasing the current density will increase both power consumption and percentage removal efficiency. The increase in power consumption can be ascribed to the increased polarization on the two electrodes by increasing the current density [34].

To throw some light on the role of current density in the economy of the process the ratio (% removal efficiency/power consumption) was plotted vs. current density as shown in Fig. 13, the results show that the ratio decreased from 1840 to 340 by increasing the current from 1 to 4 mA/m^2 which suggests that the economy of the process is favored by low current densities.

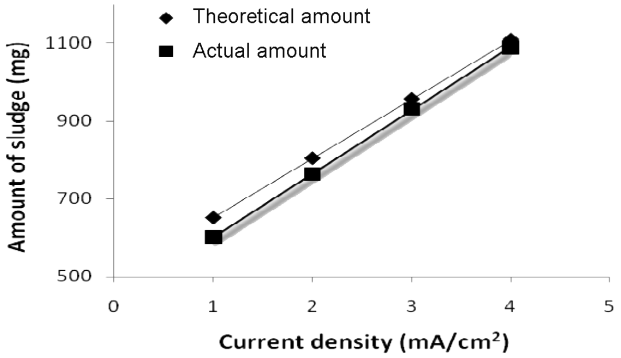

Fig. 11. Amount of sludge generated vs. current density for both theoretical and actual conditions..

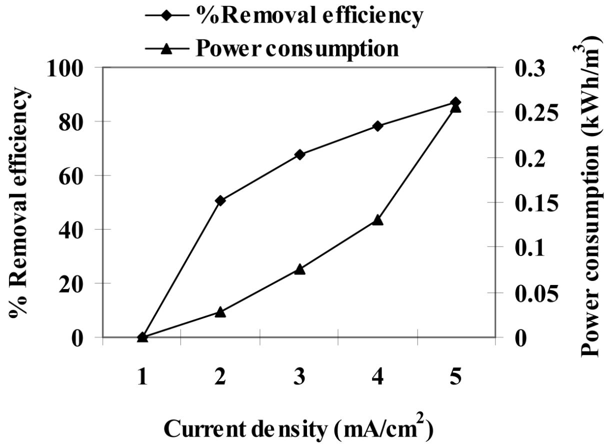

Fig. 12. Effect of current density on the power consumption and the% eemoval efficiency.

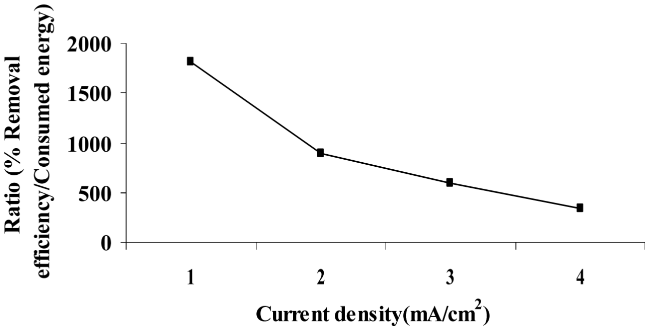

Fig. 13. The ratio (% removal efficiency/consumed energy) vs. current density

4. Conclusion

The present work proved that electrocoagulation technique using horizontally oriented aluminum electrodes can be used efficiently as a new technique for the removal of nitrogen from industrial wastewater especially for the treatment of industrial wastewater generated from bone glue industry. The results show that, the % removal efficiency of nitrogen has been increased by increasing the applied current density, distance between electrodes, sodium chloride concentration up to 5%, while it was decreased by increasing both initial nitrogen concentration and the area ratio (cathode/anode). The results show that about 60–95% of total nitrogen can be removed within 60 min in the case of using a synthesized solution of bone glue in distilled water, depending on the value of applied current density from 1 to 4 mA/cm^2 respectively and other operating conditions.

In addition treatment of a real sample collected from one of bone glue factories located in Alexandria, Egypt was carried out, under different current densities, the results show that about 60–80% of the total nitrogen can be removed within 120 min depending on the applied current density. In addition it was found that for the true sample treatment a reduction in BOD, COD, and TSS of about 68%, 61% and 85% respectively can be achieved within the same time interval.

The power consumption of the process was measured at different current densities. It was found that the power consumption increases by increasing the applied current. The results show that the economy of the process is favored by low current density.

Symbols and abbreviations

BOD — Biological oxygen demand, mg/l
COD — Chemical oxygen demand, mg/l
E — Cell voltage, V
I — Current applied, A
P — Specific power consumption, kWh/m^3
R — Electrical wire resistance
t — Time of electrolysis, h
TKN — Total Kjeldahl nitrogen, mg/l
TSS — Total suspended solids, mg/l
V — Solution volume, m^3

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