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Electrocoagulation as an Efficient Technique to Remove Dyestuffs from Aqueous Mediums

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A B S T R A C T

In recent years, attention has been directed towards the application of electrocoagulation (EC) for treatment of wastewater contains toxic and non-biodegradable organic pollutants. It has flexibility in the design, operation and the cost. In our study, EC was adopted to treat a lab-synthetic wastewater polluted by methylene blue dye in a batch cell of iron screen anode and two iron cathodes, inner rod and outer cylindrical sheet. The effects of current density, electrolyte concentration and the initial concentration of pollutant on process performance and energy consumption were investigated for a definite process time. The results showed that process efficiency has a direct proportional with applied current density and electrolyte concentration in contrast with the initial dye concentration. Furthermore, for 293 mg/L initial dye concentration and 16 g/L NaCl, removal efficiency reached 93% for current density of 2.67A/m^2 compared with 71% at 0.77A/m^2 during 15 min process time while 99.5% removal was achieved after 25 min for 2.67A/m^2 current density when other parameters remain constant. The optimum operating conditions is calculated from economic point of view based on the requested characteristics of treated water.

1. Introduction

Many of the water systems that keep ecosystems thriving and feed a growing human population have become stressed. Some of rivers, lakes and aquifers are drying up or becoming too polluted to use that caused more than half the world's wetlands have disappeared. Agriculture consumes more water than any other source, 70% of the accessible freshwater, and wastes much of that, about 60%, through inefficiencies. UNICEF report in 2021shows that climate and environmental hazards have devastating impacts on the wellbeing of children globally where a greater than one-third of children over the world are currently highly exposed to water scarcity. In Egypt, the issue of water scarcity is taken very seriously as a major challenge now and for the next years.

Many of industrial activities (textile, leather, pulp and paper, printing, photographs, cosmetics, pharmaceutical, food) deliver huge amount of effluents contain dyes. As an example, textile industry utilizes about 10,000 dyes and pigments and about 20–50% of reactive dyes used in textile fabrics can be released into waterways [1]. Several techniques have been used for color removal from aqueous solutions, includes biological treatments, chemical oxidation, electrochemical, photo-degradation, adsorption, membrane separation, chemical coagulation and the electrolytic treatments [2].

At the same time as global demand for high-quality drinking water rises, environmental laws governing wastewater discharge become stricter. As a result, more effective treatment technologies for water and wastewater have to be developed, or the existing treatment methods must be improved. This, together with the environmental benefits, has sparked a surge in global interest in electrochemical processes, particularly electrocoagulation as a research topic. Publication in the environmental sector has showed a growing interest in the application of EC during the last few decades [3].

Electrocoagulation (EC) is a class related to the conventional coagulation process, which has been used as a method (early as 2000 BC) for water treatment and clarification [4] and nowadays, it is still extensively used [5]. It is dual technology that combines the advantages of conventional coagulation and flotation in water and wastewater treatment. Coagulation is a conventional physicochemical treatment that uses phase separation to decontaminate wastewaters before they are discharged into the environment [6]. The mechanism of the procedure is based on the production and aggregation of a colloidal solution, which is then further coagulated with the help of coagulating agents. Metallic and organic contaminants are precipitated with coagula from the aqueous phase and then removed from the treated water [7, 8]. The addition of coagulating agents (M^+, such as Al^{3+} or/and Fe^{3+} salts) helps the formation of pollutant aggregates [9], then separated physically from water by precipitation or flotation [10]. Thus by means of EC technology, which is cost and safety effective, we can help to reduce concentrations of organic pollutants to an extent as minimum as possible and comply the environmental Egyptian law for its limits acceptable of zero COD in case of hazard organic pollutants.

The EC technique uses a current to generate metal ions (M^+) by dissolve metals (i.e. Fe, Al, Mg and Zn) as sacrificial anodes immersed in the polluted aqueous mediums. The electro-disintegration promotes an increase on the metal ions in solution or their complex intermediates with hydroxide ion depending on the pH conditions and the sacrificial anode used. These species act as coagulants or destabilization agents, enhances precipitation or adsorption of contaminants from wastewaters [11, 12]. The EC process involves four different phases, controlling steps:

1. Anodic reaction to produce the metal ions parallel with catholic reaction and H_2 gas evolution.
2. Breaking emulsions and destabilization of pollutants.
3. Formation of aggregates and flocs,
4. Separation of the coagulated species, flocs, by sedimentation or flotation using H_2 bubbles generated.

Our study aims to investigate application of EC technique to remove Methylene Blue dye from aqueous solutions. We constructed a new modified electrocoagulation cell that consists of a single iron screen anode with two different cathodes; inner cathode was iron rod while the outer cathode was a cylindrical

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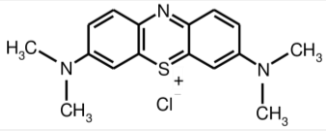
sheet. Both color removal efficiency and energy consumption were calculated under the different operating conditions, current density, electrolyte concentration and the pollutant initial concentration.

2. Materials and Methods

The commercial dye used in this study was Methylene Blue (MB) purchased from Al-Nasr Company, Cairo-Egypt. Table 1 shows the chemical structure and other characteristics of the dye. Dye solutions were prepared by dissolving dyes in distilled water. A stock solution (1g/L) was prepared where other initial dye concentrations are obtained by dilution with double distilled water (DDW).

Table 1: Characteristics of Methylene Blue dye.

Property	Value
Scientific name	**Basic Blue 9**
Chemical formula	**$C_{16}H_{18}N_3OS.3H_2O$**
Molecular weight	**373.5 (g/g-mol)**
Molecular structure	
Maximum wavelength (nm)	**665**
Molecular diffusivity (at 25 °C)	**$4.7X10^{-6}$ (cm^2/s)**
Solubility in water	**Soluble in water**
Color Index (C.I.)	**52030**
Color	**Dark green to blue powder**
Molecular volume	**390.2 (cm^3/g-mol)**

A batch experimental cell is constructed as shown in Figure 1. The cell is a glass cylindrical tube of 8.0cm inner diameter and 12.5cm height fitted with a cylindrical steel screen anode (4.0cm diameter, 5.0cm height, mesh number 32) and two different cathodes, inner iron rod (2.0cm diameter and 5.0cm height) and outer cylindrical iron sheet (6.0cm diameter and 5.0cm height) enclosing the anode. This arrangement gives 1.0cm inner gap between the three electrodes. The electrodes were connected to a DC power source with galvanostatic operating options to manage the current density, which is an important parameter in the electrocoagulation process. The conductivity of dye solutions was adjusted at different values by addition of NaCl salt (AR grad). All the runs were performed isothermally at room temperature. After each experiment, samples were allowed to settle for enough time, then analyzed from their absorbance characteristics using UV–Visible Spectrophotometer (8700 series Unican UV/V). All experiments were repeated twice in order to eliminate the error where, average values were used for the calculations. After treatment process, the dye removal efficiency R% (process performance) is calculated using this formula:

$$R\% = \left(\frac{C_0 - C}{C_0}\right)100$$

Where; C_o and C are the dye concentrations before and after treatment step respectively (mg/L).

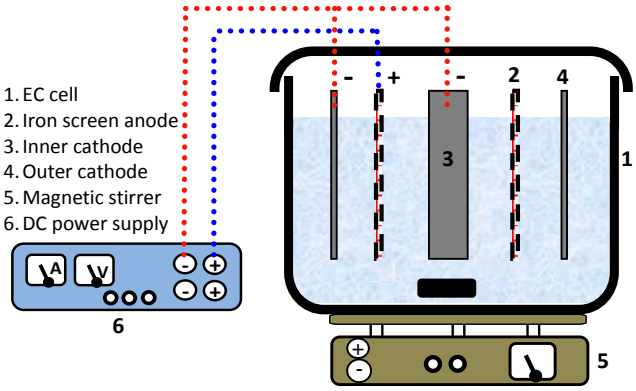


Figure 1: Schematic diagram of experimental setup.

3. Process Mechanism

EC process occurs when direct electric current passes through electrolyte, producing chemical reactions at electrodes. In our case iron electrodes are used, upon oxidation in an electrolytic system, it produces iron hydroxide, $Fe(OH)_n$ where n = 2 or 3 according two different mechanisms as follow [13], Figure 2.

The first mechanism:

Cathodic reaction:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \quad (1)$$

Anodic reactions:

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

$$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2 \quad (3)$$

The overall reaction:

$$Fe + 2H_2O \rightarrow Fe(OH)_2 + H_2 \quad (4)$$

The second mechanism:

Cathodic reaction:

$$8H^+ + 8e^- \rightarrow 4H_2 \quad (5)$$

Anodic reactions:

$$4Fe \rightarrow 4Fe^{2+} + 8e^- \quad (6)$$

$$4Fe^{2+} + 10H_2O + O_2 \rightarrow 4Fe(OH)_3 + 8H^+ \quad (7)$$

The overall reaction:

$$4Fe + 10H_2O + O_2 \rightarrow 4Fe(OH)_3 + 4H_2 \quad (8)$$

The insoluble metal hydroxides of iron that has a large surface area acts for trapping dye molecules by electrostatic attraction or/and surface complexation. Finally, these produced flocs are easily removed from aqueous medium by settling or flotation processes.

$$Dye + Fe(OH)_n \rightarrow [Sludge] \quad (9)$$

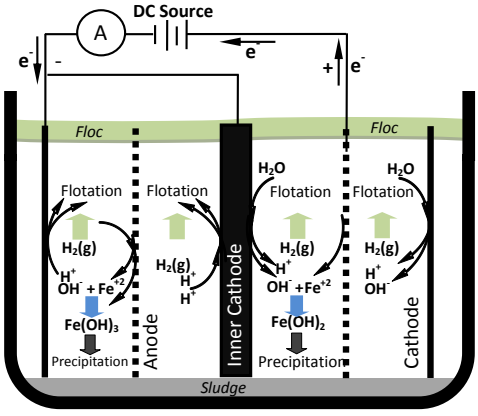


Figure 2: Representative diagram for anodic and cathodic reactions (first & second mechanism).

4. Results and Discussion

The performance of all electrochemical processes are influenced by several operating variables, reaction time, current density (CD), electrolyte conductivity, solution pH, and the pollutants concentrations. Time is one of the important operating parameters, as it effects on energy consumption, as well as the anode consumption, thus must be determined accurately. Preliminary experiments were done to determine the process time; since all next experiments will allow to run for this time. Figure 3 depicts change of dye concentration as a function of reaction time for, C_0 = 200 mg/L, CD = 0.77 A/m^2 at to two different electrolyte conductivities indicated by NaCl concentrations.

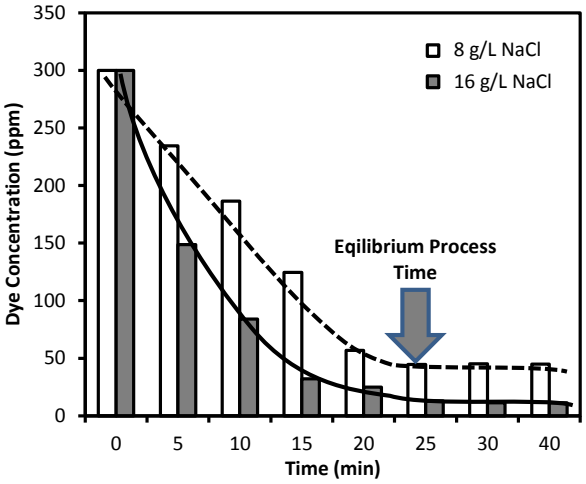


Figure 3: Dye concentration versus process time at 8 and 16 g/L NaCl concentrations.

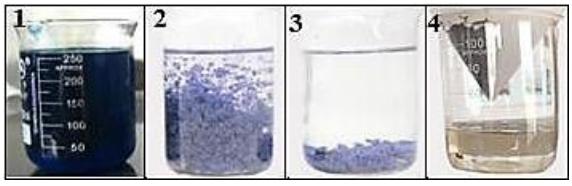


Figure 4: Image of wastewater during the sequence phases of EC. (1) at the beginning, (2) after 10 min., (3) after 20 min., (4) after 25 min.

It is clear that dye concentration decreases sharply in first ten minutes, then gradually decreases and reached to equilibrium (process time) during 25 minutes. Appearance of wastewater at different phases of the process is presented in Figure 4.

4.1. Effect of Current Density

Cell current (CD) density is the most important process variable for controlling reaction rate, that determine generation rate of coagulant as well as rate and size of H_2 bubble production. In order to evaluate the effect of CD on the removal efficiency, a series of experiments were carried out for different current densities, 0.77, 1.47, 2.03 and 2.67 A/m^2 at constant conductivity, 16g/L NaCl and initial dye concentration of 293 mg/L, Figure 5. It was found that increasing current density causes a corresponding increase in oxidizing rate of iron electrode, produces more ions of iron hydroxides, $Fe(OH)_n$, that accelerate trapping of dye molecules from water. Results depicted that an increase in current density from 0.77 to 2.67 A/cm^2 enhances the color removal from 72.8 to 96.3% after 15 min reaction time in contrast, after full process time (25 min) it reaches 99.5 and 86.5% for the highest and lowest CD respectively.

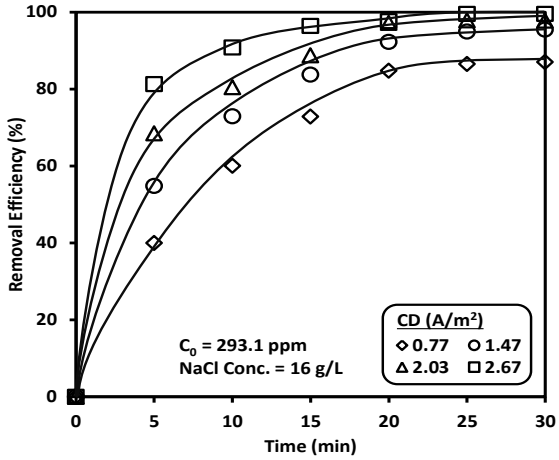


Figure 5: Removal efficiency versus process time at different CD.

From an economic point of view, electrical energy consumption was also calculated as kW.h/g-dye removed for the different applied CD, Figure 6.

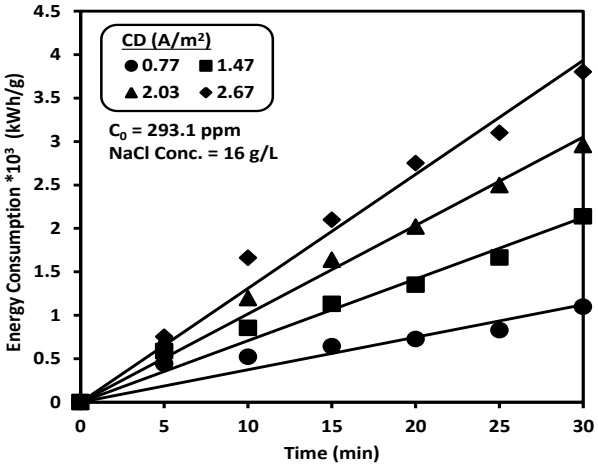


Figure 6: Energy consumption versus process time at different CD.

The energy consumption was calculated using the commonly equation of:

$$E = \frac{I * V * t_{EC}}{(C_0 - C) * Q} \qquad (10)$$

Where, E is the electrical energy in kWh/g-dye removed, V the cell voltage (Volt), I the current (Ampere), t_{EC} is the time of EC process (h) and Q the treated dye solution volume (L). The results showed that an increase in CD increases the energy consumptions, thus, to achieve an optimized current density, both color removal percent and energy consumption should be taken into account. Other factors may affect the optimum CD such as water-pH and temperature as well as quantity or flow rate [14].

4.2. Effect of the Conductivity

Sodium chloride salt is usually employed to adjust the conductivity of wastewater to be treated. In order to evaluate the conductance effect on EC performance, a series of experiments were done at different NaCl concentrations (8:20 g/L) where other parameters remain constant (C_0= 293.0 mg/L, CD = 1.47 A/m^2), Figure 7. It is evident from results that removal efficiency increases as the NaCl concentration increases. It was found that, when sodium chloride concentration increases from 8 to 20g/L, the percentage removal increases from 35.8 to 91.2 during 10min reaction

time. This result can be explained by; the solution conductivity affects both current efficiency and cell voltage. In addition, in real wastewater, it has other advantages, i.e. chloride ions could significantly reduce the adverse effects of other anions, such as bicarbonate and sulfate. The existence of the carbonate ion would lead to the precipitation of Ca^{2+} ion that forms an insulating layer on the surface of the cathode [14]. Enhancement of process efficiency with increasing NaCl concentration at constant CD will improves the energy consumption also.

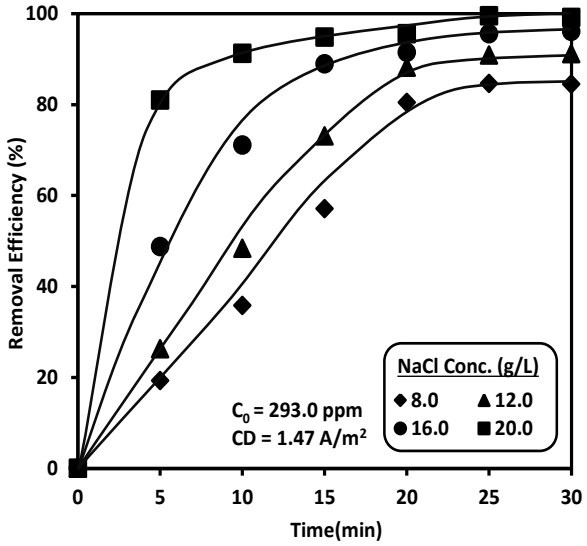


Figure 7: Removal efficiency versus process time at different NaCl concentrations.

4.3. Effect of Initial Concentration

In order to study the effect initial concentration of the dye solutions viz., 122.5, 198.5 and 293.0 mg/L were taken, while the CD and NaCl remained fixed at 1.47A/m^2 and 16g/L respectively. Figure 8 shows R% versus electrolysis time at different initial concentration. The results showed that removal rate has decreased upon increasing initial concentration. This is ascribed to the fact that at a constant current density, the same amount of metal ions passes through the dye solution consequently, the formed amount of complex metal hydroxides was insufficient to coagulate the greater number of dye molecules at higher concentrations. Also, the results indicated that residual dye is not detectible after 15 min process time for 122.5 mg/L initial dye concentration that means it completely removed. As expected, energy consumption as kW.h/g-dye removed has an inverse proportional with concentration, due to the chance to coagulate a greater number of dye molecules at higher concentrations for the same metal hydroxide in contrast at lower concentrations.

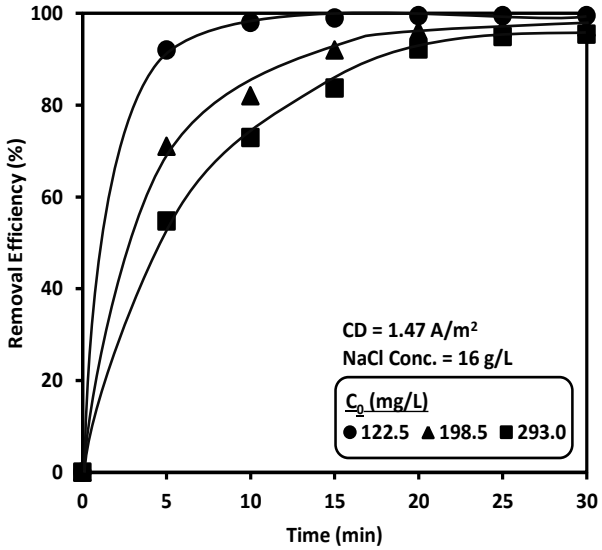


Figure 8: Removal efficiency versus process time at different initial dye concentrations.

4.4. Complete Outline of EC Process

The EC technology is applied to remove contaminants including dissolved ionic species such as dyestuffs; suspended colloidal materials such as emulsified oil from water or wastewater. This section concerned with a suggestion for a complete wastewater treatment train (prototype pilot plant) as presented in Figure 9. After receiving wastewater, it firstly passes through a physical separation unit (coarse filtration unit) to isolate any coarse and floated materials, and then send to influent storage tank. This tank collects contaminant streams to provide surge capacity during continuous operation and sometimes pH adjusted in range of optimum value for the EC process. The EC system induces coagulation of contaminants by means of DC electrolytic process where floccules are formed, residence time 20:30 min. The treated water with its floccules is allowed to settle in the clarifier. Treated water exits the clarifier as the overflow while settled floces is removed in the underflow Treated water is then passed through two filters in sequence, sand and carbon filter, to remove any residuals suspended as well as colure and odor. The treated water is discharged from filters for disposal or reuse. Contaminants are concentrated in floes are dewatered and discharged to drums for ultimate disposal or reclamation while filtrate is recycled through the wastewater reservoir.

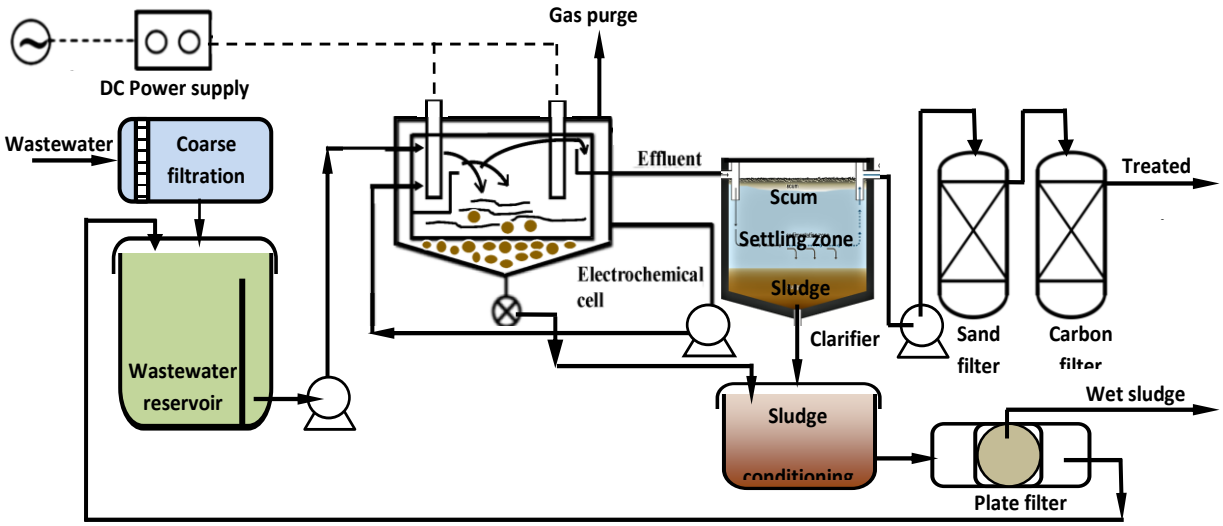


Figure 9: Schematic diagram of pilot prototype electrocoagulation treatment process.

5. CONCLUSION

Electrocoagulation was used to treat a colored wastewater containing methylene blue dye. A batch reactor was design to investigate effect of operating variables, applied current density, medium conductivity and dye initial concentration, on the process performance. Results indicated the optimal electrolysis time for removal of color resulting from methylene blue dye was 25 min where highly percentage was remove in the first ten min. It was found that applying high current density could achieve approximately complete color removal but consumed more energy. For 2.67 A/m^2 applied CD, 16g/L NaCl and 293 mg/L initial dye concentration, the removal efficiency for MB dye was 96.3% and 99.5% after 15 and 25 min reaction time respectively. The dye solution decolorized more efficiently when its conductance increased by adding proper amount of NaCl (8, 12, 16 & 20 mg/L) which normally caused reduction in power consumption, in contrast with effect of initial dye concentrations. To conclude, EC offers a lot of potential for decolorizing water and wastewater, and it appears to be a viable and cost-effective option in this field, while more research is needed, particularly with larger-scale and continuous systems.

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