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The Impact of Sodium Chloride (NaCl) Concentrations on Electrocoagulation for Simultaneous Tartrazine Dye Removal and Hydrogen Production †

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Abstract: Liquid waste frequently contains a large variety of hazardous substances, including tartrazine-based dyes. These color compounds can present a significant hazard to both human health and the natural environment. Moreover, there is a growing demand for sustainable energy, and hydrogen is emerging as a promising alternative energy source that does not produce carbon emissions. To address the aforementioned concerns, it is necessary to conduct research aimed at eradicating tartrazine while concurrently generating hydrogen gas as a viable substitute for energy. This study aims to investigate the effect of different concentrations of NaCl electrolytes on the rate of simultaneous tartrazine elimination and hydrogen production using electrocoagulation. The electrocoagulation procedure was used with various concentrations of NaCl (0, 0.2, 0.4, 0.6, 0.8, and 1 g/L). UV–Vis spectrophotometers and gas chromatography were employed to evaluate the elimination of tartrazine and the rate of hydrogen production. The results show that the highest rate of tartrazine removal was 93%, which occurred at 0.8 and 1 g/L of NaCl at 240 min. It can be stated that higher electrolyte concentrations generally lead to an increase in tartrazine removal. The highest rate of hydrogen production was 217.44 mol H_2/m^2, which occurred at 1 g/L of NaCl at 240 min; thus, it can be concluded that higher electrolyte concentrations generally lead to an increase in hydrogen production.

Keywords: tartrazine; hydrogen production; NaCl; electrocoagulation

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1. Introduction

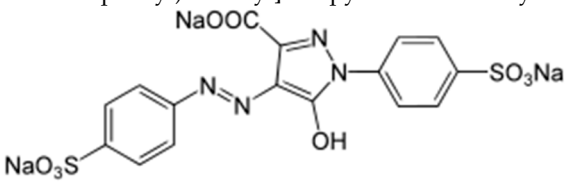
Tartrazine, a type of waste colorant, can present a significant risk to both human health and the environment [1]. Currently, there is a scarcity of studies on proper disposal methods for dye waste. The study conducted in [2] focused on the investigation of toxicity in single-waste materials. The efficient management of color waste is essential in order to minimize the detrimental effects of pollution on both human health and the environment. Furthermore, there is a growing demand for renewable energy, including hydrogen, which exhibits significant potential as a viable alternative energy source. To address this dilemma, it is crucial to adopt a cost-effective and eco-friendly method of producing hydrogen, hence decreasing our dependence on fossil fuels [3]. There are a lot of methods that have been evaluated for both dye removal and hydrogen production simultaneously, as stated in Refs. [4–7]. One of the promising methods is electrocoagulation, which has emerged as a viable method for mitigating the deleterious effects of environmental contamination. Electrocoagulation has the dual purpose of mitigating environmental removal [8–10] and generating renewable energy, such as hydrogen gas [11,12]

Electrocoagulation is a technique that employs an electric current to eliminate pollutants present in liquid media, whether they are suspended, emulsified, or dissolved [13]. The electrocoagulation reactor comprises electrolyte cells, anodes, and cathodes, with a conductive metal serving as the electrode [8]. The process involves the integration of electrochemistry, flotation, and coagulation techniques to generate hydrogen gas through the reduction of water on the cathode plate. Although electrocoagulation methods offer benefits such as the creation of H_2 gas, drawbacks may lie in their energy use, especially when handling high concentrations of liquid waste that require large electrical currents to produce sufficient coagulant for processing [14]. Several techniques can be employed to decrease electrical resistance values by enhancing the conductivity of a solution from a low electrolyte concentration to a high one [10]. We attempted to utilize a NaCl solution as a potent electrolyte to be subsequently incorporated into electrocoagulation system solutions. Prior investigations have explored the introduction of different potent electrolyte solutions into electrocoagulation systems, such as KCl, $NaSO_4$, and NaCl [7,15]. However, this study aims to introduce NaCl into the electrocoagulation process and manipulate its concentration to identify the most favorable conditions for effectively reducing tartrazine while simultaneously generating hydrogen. This study is unique in its focus on optimizing the electrocoagulation process through NaCl concentration variation.

2. Material and Methods

The chemical employed was an analytical compound procured from Sigma Aldrich. The experiment employed sodium chloride (NaCl) as the supporting electrolyte. The characteristics of tartrazine are presented in Table 1.

Table 1. The characteristics of tartrazine.

Name	Properties
IUPAC	Trisodium 5-hydroxy-1-(4-sulfonatophenyl)-4-[(E)-(4-sulfonatophenyl)diazenyl]-1H-pyrazole-3 carboxylate)
Chemical structure	
λ max (nm)	427
Molecular weight (g/mol)	534.36
Chemical class	Diazo

2.1. Electrocoagulation System

The process of electrocoagulation was carried out in an acrylic reactor with dimensions of 11 × 5 × 12 cm (length × width × height). Plat Al and SS-316 were used as the anode and the cathode, respectively. The anode and cathode had active surfaces of 4 cm × 8 cm × 1 mm, with a distance of 3 cm. A power supply (Zhaoxin RXN-605D, 60 V, 5 A, ZHAOXIN, Shenzhen, China) was used to control the current in the process. The schematic diagram is shown in Figure 1.

2.2. Analytic Procedure

Experiments were conducted on a small scale in a sealed reactor. The waste coloring model was created by dissolving tartrazine in distilled water at a temperature of 25 °C. The initial concentration of tartrazine was 10 ppm. The solution in the reactor had a capacity of 0.5 L. The concentration of the supporting electrolyte in the solution varied between 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L. The solution's pH was adjusted to an acidic level of 5. The rotational speed of the magnetic stirrer was set at 200 rpm.

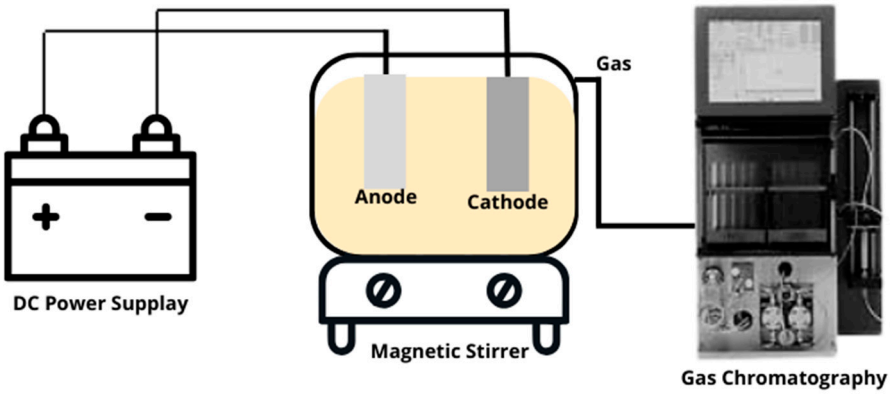


Figure 1. The schematic of the electrocoagulation process.

Each test set had a period of 240 min, and samples were systematically collected through wells in the reactor. In order to eliminate the silt, the sample underwent filtration using a 0.45 µm Millipore filter membrane. The experiment was conducted at a voltage of 5 volts.

The analysis of tartrazine's elimination was conducted using spectroscopy, specifically UV–Vis spectrometry (UV Mini 1240, Shimadzu, Kyoto, Japan). The absorption was determined using a maximum wavelength (λ) of 427 nm. The tartrazine concentration in the solution was determined using the Beer–Lambert law.

The quantity of H_2 gas produced in the reaction was measured using a Shimadzu GC-8A gas chromatography system, which included a molecular sieve (MS) hydrogen 5A column. Argon was used as a known gas carrier with retention time as a reference. The H_2 sample was collected every 30 min (0, 30, 60, 90, 120, 150, 180, 210, and 240 min) using a 1 mL gastight syringe.

3. Results and Discussion

3.1. The Effect of NaCl on the Current in Electrocoagulation

Figure 2a shows the effect of NaCl concentration on the current produced in the electrocoagulation process at 240 min, and Figure 2b shows the effect of NaCl concentration on the current produced at 60, 120, and 240 min. Based on Figure 2a, the average currents produced in the electrocoagulation at NaCl concentrations of 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L measured 0.022, 0.063, 0.107, 0.167, 0.186, and 0.194 Amperes. Generally, the current in the electrocoagulation system increased with the NaCl concentration [15]. The current tended to be stable from 0 to 240 min at each NaCl concentration. Based on Figure 2b, the currents under electrocoagulation at 60, 120, and 240 min were identical. The currents at NaCl concentrations of 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L at 60, 120, and 240 min measured 0.03, 0.02, 0.02; 0.063, 0.064, 0.062; 0.108, 0.107, 0.105; 0.166, 0.167, 0.166; 0.186, 0.187, 0.186; and 0.194, 0.194, 0.193 Ampere, respectively. This was because the addition of NaCl to the solution increased the amount of electrical current flowing in the electrocoagulation circuit, as has been reported in previous research [16,17]. The NaCl in the solution reduces the electrical resistance of the solution and changes the nature of the solution from a weak electrolyte to a strong electrolyte [17]. In other words, the addition of NaCl increases the electrical conductivity of the solution.

Based on previous work [17], the use of supporting electrolytes will trigger reactions that occur during the electrocoagulation process, as follows [4]:

Reaction at cathode (reduction):

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

$$2H^+ + 2e^- \rightarrow H_2 \quad (2)$$

Reaction at anode (oxidisation):

$$Al \rightarrow Al^{3+} + 3e^- \quad (3)$$

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (4)$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (5)$$

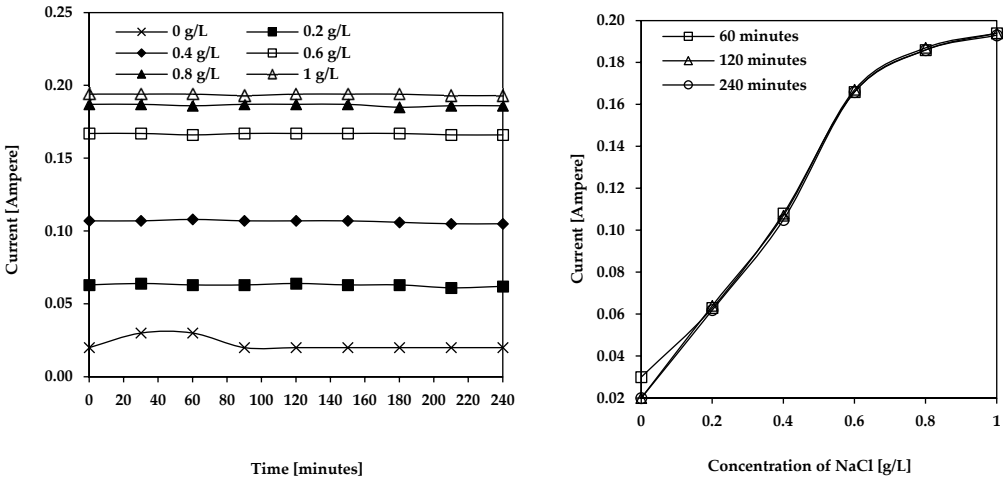
In bulk solution:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad (6)$$

$$Cl_2 + H_2O \rightarrow HOCl + HCl \quad (7)$$

$$HOCl \rightarrow H^+ + OCl^- \quad (8)$$

Equations (1) and (2) show an increase in conductivity due to the number of electrons on the cathode surface, which will later be used to reduce the hydrogen ions and water in the solution [6]. Meanwhile, the increasing conductivity also has an impact on the effectiveness of dissolving the electrode, in this case aluminum (Equation (3)), which then forms the coagulant $Al(OH)_3$ (Equation (6)) [5]. This increases the process capacity for the removal of tartrazine from the solution, as will be explained in Section 3.2.



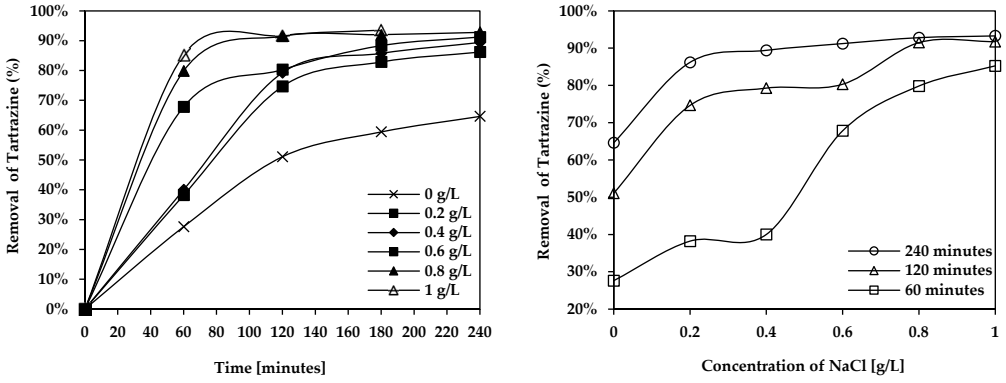
(**a**) (**b**)

Figure 2. (**a**) Current vs. time, and (**b**) current vs. NaCl concentration.

3.2. The Effect of NaCl on Tartrazine Dye Removal

Figure 3a shows the effect of NaCl concentration on the percent removal of tartrazine at 240 min, and Figure 3b shows the effect of NaCl concentration on the percent removal of tartrazine at 60, 120, and 240 min. Based on Figure 3a, the percent removal values of tartrazine at 240 min with NaCl concentrations of 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L are 65, 86, 89, 91, 93, and 93%, respectively. Generally, the tartrazine concentration decreases significantly with NaCl concentration [7]. This is caused by the change in the conductivity of the solution, as explained in the previous paragraph. Based on Figure 3b, the percentages of tartrazine removal at NaCl concentrations of 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L at 60, 120, and 240 min are 28, 51, 65%; 38, 75, 86%; 40, 79, 89%; 68, 80, 91%; 80, 92, 93%; and 85, 92, 93%, respectively.

Generally, a higher removal of tartrazine waste occurs with the increased conductivity of the solution [18]. This means that the electrode (aluminum) is dissolved effectively (Equation (3)), which then forms the coagulant $Al(OH)_3$ (Equation (6)) [17].



(**a**) (**b**)

Figure 3. (**a**) Percent removal of tartrazine vs. time, and (**b**) percent removal of tartrazine vs. NaCl concentration.

3.3. The Effect of NaCl on Hydrogen Production

Figure 4 shows the effect of NaCl concentration on the hydrogen produced in the electrocoagulation process at 240 min. Based on Figure 4, the hydrogen produced in the electrocoagulation process at NaCl concentrations of 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L measures 63.16, 99.06, 135.45, 177.80, 208.09, and 217.44 mol H_2/m^2, respectively. In general, the hydrogen produced in the electrocoagulation system increases with increasing NaCl concentration [7]. The hydrogen value tends to increase from 0 to 240 min at each concentration. Based on Figure 4, the hydrogen in the electrocoagulation system for NaCl concentrations of 0, 0.2, 0.4, 0.6, 0.8, and 1 g/L at 60, 120, and 240 min measures 8.64, 17.88, 63.16; 14.37, 56.74, 99.06; 45.00, 87.25, 135.45; 77.18, 129.74, 177.80; 97.04, 160.79, 208.09; and 135.16, 187.69, 217.44 mol H_2/m^2, respectively. According to the provided data, the longer durations of the electrocoagulation process and the higher concentrations of NaCl result in increased hydrogen production. The percent increases in hydrogen production from 0 g/L NaCl concentration to 1 g/L concentration at 60, 120, and 240 min are 1046.52%, 295.11%, and 124.07%, respectively.

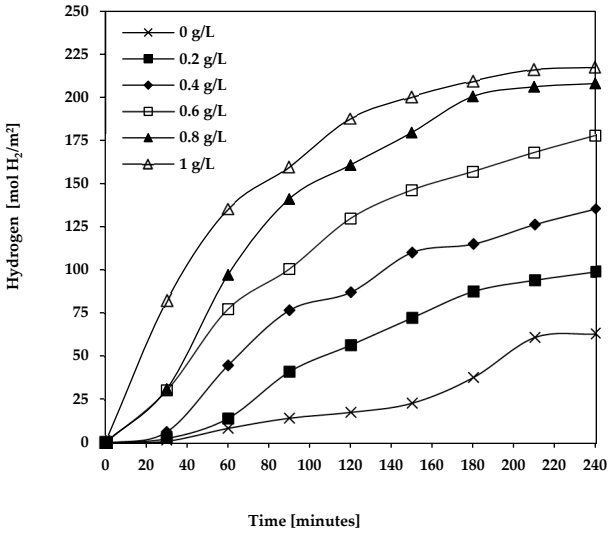


Figure 4. Hydrogen production vs. time at varied NaCl concentrations.

During the electrocoagulation process, the anode made of aluminum will undergo an oxidation reaction, where the metal ions formed will react with hydroxyl ions in the solution to form a coagulant. In this research, an anode in the form of an aluminum plate was used, so that in the electrocoagulation process, the aluminum would oxidize to form

Al^{3+} ions, which dissolve in the solution [19–21]. The dissolved Al^{3+} ions then bind to OH^- ions and form an $Al(OH)_3$ coagulant (Equation (6)). The $Al(OH)_3$ coagulant will adsorb pollutant compounds for the waste removal process, as stated in Equation (9). Meanwhile, electrons at the cathode will reduce the water to form $H_{2(g)}$ [13]. The reactions that occur at the anode and cathode in the electrocoagulation process can be written as stated in Equations (1)–(5) [22].

$$Al(OH)_{3(s)} + \text{Pollutants} \rightarrow Al(OH)_{3(s)} \cdot \text{Pollutants} \quad (9)$$

The hydrogen formed in the form of gas bubbles at the cathode will help bring coagulants that have absorbed pollutants to the surface of the solution. The bringing of coagulants to the surface of the solution by H_2 gas is called flotation. The flotation process causes flocs of coagulants and pollutants to concentrate on the surface of the solution so that they are easily separated from wastewater. The basic principle of the coagulation of pollutants in the electrocoagulation waste removal process is manipulating the electrostatic charge of the particles suspended in the water. Metal ions, as cationic species dissolved into the solution during the electrocoagulation reaction, in this case, Al^{3+} ions, will neutralize the negative charge of the dissolved pollutant, which will initiate coagulation. Pollutants that are neutralized by Al^{3+} will lose their ability to repel each other, so that coagulation between particles will occur, followed by the process of floc formation (flocculation). The flocculation process will help further clumping so that pollutants can be aggregated and precipitated at the bottom of the solution, or concentrated on the surface of the solution with the help of H_2 gas [23].

In the future, it is suggested that researchers should assess the removal of basic dyes from wastewater using parameters like adsorption capacity and other relevant metrics, as outlined in the kinetic adsorption models described in Ref. [24].

4. Conclusions

Research on the effect of different concentrations of NaCl electrolytes on the simultaneous removal of tartrazine and production of hydrogen using electrocoagulation has been conducted. The research shows that the highest rate of tartrazine removal was 93%, which occurred at 0.8 and 1 g/L of NaCl at 240 min. The highest rate of hydrogen production was 217.44 mol H_2/m^2, which occurred at 1 g/L of NaCl at 240 min. The percent increases in hydrogen production from 0 g/L NaCl concentration to 1 g/L concentration at 60, 120, and 240 min were 1046.52%, 295.11%, and 124.07%, respectively. It can be concluded that higher electrolyte concentrations generally lead to an increase in the proportion of tartrazine removal and hydrogen production. The significance of NaCl concentration in electrolytes in electrocoagulation for hydrogen production and dye waste removal underscores its potential for future applications.

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