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The Efficiency of Separate and Combined Modified Electrolysis Processes and Electric Coagulation in Removing Yellow Gold Dye (Gold Yellow X-GL) from Aqueous Solutions

Reza Ali Fallahzadeh [1, 2], Davoud Ghadirian [1], Ahmad Fattahi [1], Pejman Amjadian [1], Samaneh Mozaffary [1], Nasrin Homayonibezi [3*]

[1]Genetic and Environmental Adventures Research Center, School of Abarkouh Paramedicine, Member of Student Research Committee, Shahid Sadoughi University of Medical Sciences, Yazd, Iran.
2 Environmental Science and Technology Research Center, Department of Environmental Health Engineering, School of Public Health, Shahid Sadoughi University of Medical Sciences, Yazd, Iran.
[3] Department of Environmental Health Engineering, School of Public Health, Bushehr University of Medical Sciences, Bushehr, Iran.

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***Corresponding Author:**
Nasrin Homayonibezi

Email:
n.homayoni73@gmail.com

Tel:
+989162523893

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ABSTRACT

Introduction: Disposal of the wastewater produced in the paper and paperboard industry without the refining process can add substances to the environment, which are harmful to humans, environment, and other organisms. With regard to process efficiency, the current methods used in wastewater treatment of this industry are economically unjustifiable. Therefore, in this research, the electrolysis / electrocoagulation method was investigated using a graphite / iron electrode for synthetic aqueous solutions containing golden yellow X-GL.

Materials and Methods: In this test, two Plexiglas reactors with a volume of 3 liters were used. The impact of the operation parameters such as voltage, initial dye concentration, and reaction time were investigated. Dye concentration in specimens was determined by visible spectrophotometry using DR-5000 at 438nm wavelength.

Results: In optimal conditions of voltage 12V, dye concentration 10 mg/l, and time 60 min, the removal efficiency rates of electrolysis reactors, electric coagulation, and combined reactor were 64.17%, 75.42%, and 84.19% respectively. By increasing the dye concentration and decreasing the voltage, the dye removal efficiency decreased.

Conclusion: The electric coagulation process using an iron electrode is a suitable method for removing the yellow color of colored aqueous solutions from the paper and paperboard industry.

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Introduction

In recent years, many industries has paid attention to water pollution[1]. Water shortage has caused these industrials to reuse the wastewater produced in their industries or other downstream industries applying modern methods[2-6]. The paper and paperboard industry is one of the water-based industries in which high volumes (approximately 20 to 250 cubic meters per ton of pulp) of aqueous solutions are produced for paper production. The

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produced aqueous solutions has a high concentration of lignin[7]. Other materials in the wastewater of the paper industry include fatty acids, tannins, resin acids, sulfur compounds, phenols, and their derivatives[8]. Among the pollutants in the textile industry, dyes play the most significant role. Typical methods such as adsorption, chemical oxidation, and use of chemical coagulants produce sludge, which is a threat to the environment[9]. So far, various methods have been developed to remove these dyes from the aqueous solutions, but they often have major problems. For example, the purification method with activated carbon is not economical and its recovery process as well as waste disposal are costly and difficult[10,11]. Moreover, photo-oxidation by UV/H_2O_2 and UV/TiO_2 requires additional chemical materials, which causes secondary contamination and additional costs[12]. In recent years, use of the electrocoagulation process has been developed by coagulant produced from electro-oxidation of an anode electrode[13]. Many studies dealt with the treatment of the paper industry wastewater using the electrocoagulation process[7, 14-16].

In recent years, electrochemical processes have been widely applied for the destruction of toxic organic pollutants [17]. Some of these methods have been successfully used to treat industrial wastewater[18]. However, the efficiency of the electrochemical process is low to remove some dyes and organic materials molecules with high solubility in water and low molecular weight[19]. During the electrolysis process, by electrical current between the electrodes, the anodic and cathodic reactions produce chlorine ions, hydroxyl, and hydrogen, which remove contaminants during the reaction.

Equations 1 to 4 show the mechanism of the reactions during the electrolysis process using graphite electrodes as the cathode and anode[20]:

Cathode reaction:

1) $Na^+ + e^- \rightarrow Na \qquad E^\circ red = -2.71\,V$

2) $2H_2O + 2e^- \rightarrow H_2 + 2OH^- E^\circ red = -0.83\,V$

Anodic reaction:

3) $2Cl^- \rightarrow Cl_2 + 2e^- E^\circ ox = -1.36\,V$

4) $2H_2O \rightarrow O_2 + 4H^+ + 4e^- E^\circ ox = -1.23\,V$

Due to the poor performance of the conventional wastewater treatment methods to remove this sample of dye and considering its non-degradation, it is necessary to introduce new methods for dye removing from wastewater and investigate its efficiency. Therefore, in this study, we attempted to treat aqueous solutions using separate and combined modified electrolysis processes and electrical coagulation by removing golden yellow X-GL dye.

Materials and Methods

In this study, different concentrations of 10, 50, and 100 mg/l of X-GL yellow golden aqueous solution were prepared by adding the yellow golden industrial X-GL (Gold Yellow X-GL, 98%, China) todistilled water. Then, the sample passed through a reactorand the results were analyzed. A pre-determined dose of NaCl (from Merck GmbH Darmastadt, Germany) was added to the solution as electrolyte.

Figure 1 represents the image of the reactor. The graphite and iron electrodes were used in the electrolysis reactor for electrocoagulation process. The voltage rates used in these reactors were 2V, 6V, and 12V. The input voltage to the reactor was controlled by a DC power supply (DAZHENG PS-305D, China).

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1. Power supply
2. Multi-meter
3. Graphite electrode
4. Output flow control valve
5. Flow path tube
6. Electrolysis reactor output flow tank
7. Electrocoagulation reactor

▲ Sampling point

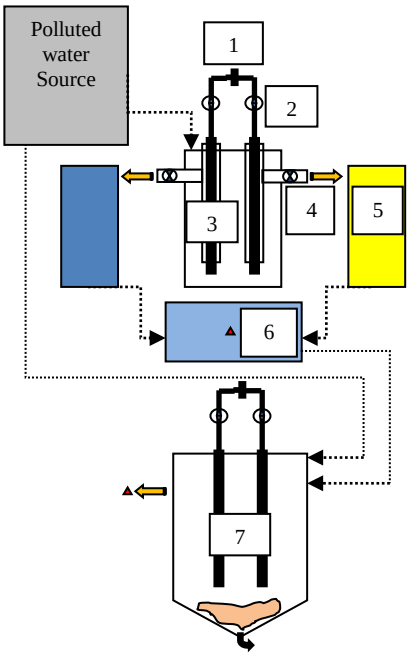

Figure 1: Scheme of the used electrolysis and electrocoagulation reactor

After preparing different concentrations of X-GL golden yellow dye, they entered into the electrochemical reactor. The reaction time in this reactor was set at 10, 30, 60, and 90 minutes using the lit/min input control. At the set times, the outputs of both poles were combined and the color was determined. Totally, 72 samples were taken and tested with two repetitions. In this step, the most efficient voltages and concentrations were determined.

In the case of electrocoagulation, various concentrations of 10, 50, and 100 mg/l of X-GL golden yellow dye entered into the electrocoagulation reactor and the samples were taken at 10, 30, 60, and 90 minutes using voltage of 2, 6, and 12 volts. Totally, 72 samples were taken and tested with two repetitions.

In order to test the electrolysis-electrocoagulation combined reactor, the optimal conditions were determined for removing the yellow X-GL gold dye in the electrolysis reactor. Later, the effluent was entered into the electrolytic-electrocoagulation reactor under the optimal conditions of operation; maximum removal efficiency (in terms of time and voltage). The pollutant removal test in the electrocoagulation reactor was tested only in terms of the voltage and time required for the reaction and the results of the color changes were investigated. At all stages, the amount of total dissolved solids (TDS) and pH was considered to be the same as normal aqueous solutions. According to the study, the TDS and pH values were 350 mg/l and 7.9, respectively.

Ethical issues

This study was approved by the Medical Ethics Committee of Shahid Sadoughi University of Medical Sciences and Health Services No: (IR.SSU.SPH.REC.1397.087)

Results

In the first step, the effect of dye removal on electric potential (volt) and time (minute) was investigated in the electrolysis reactor by synthetic aqueous solutions containing golden yellow dye in concentrations of 10, 50, and 100 mg/l (Figure 2). The results showed that at all concentrations, increasing the detention time and voltage of the aqueous solutions increased the removal efficiency.

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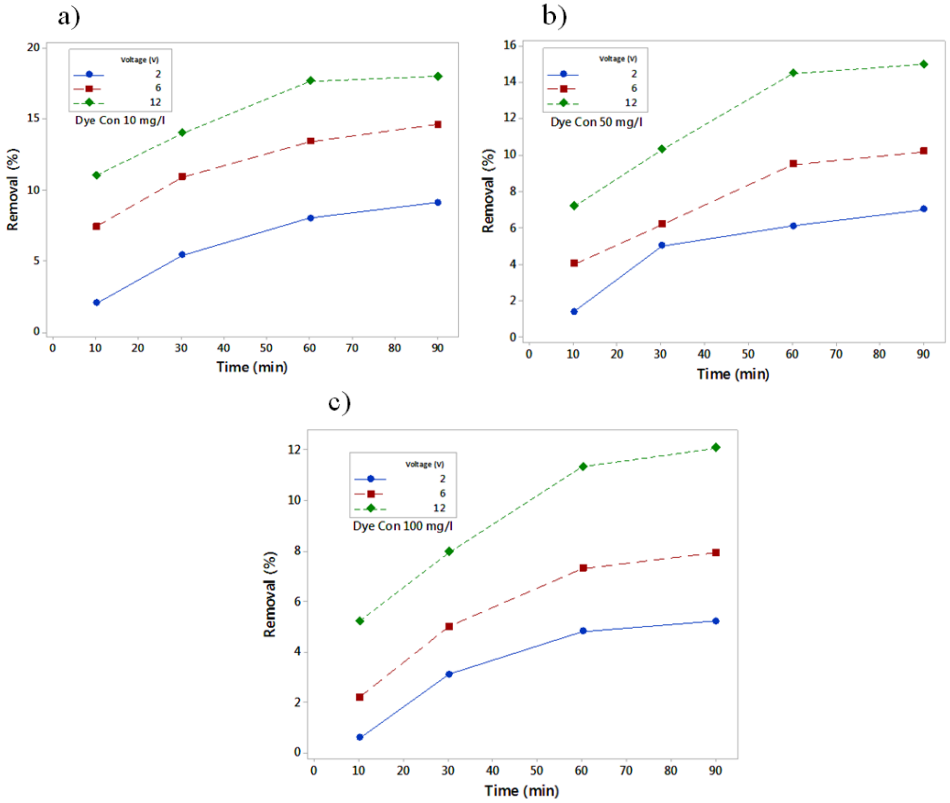
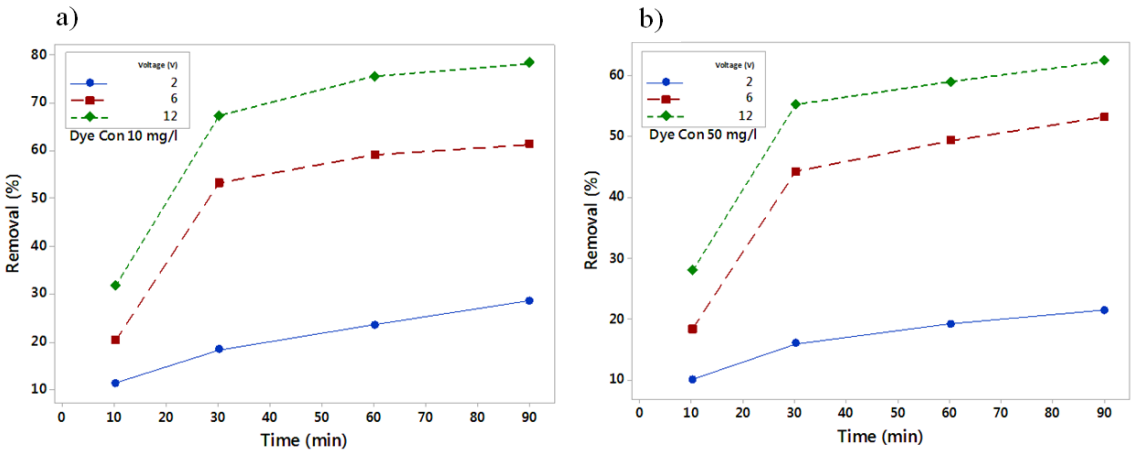


Figure 2: Efficiency of dye removal at voltage and different times at a concentration of a) 10 mg/l b)50 mg/l c)100 mg/l in electrolysis reactor

In the second stage, with the entry of synthetic aqueous solutions containing golden yellow dye at concentrations of 10, 50 and 100 mg/l, the efficiency of dye removal in the electrocoagulation reactor was investigated based on the electric potential based on volt and time in minutes (Figure3).The results showed that removal efficiency improved by increasing the detention time and voltage in all concentrations of pollutants.



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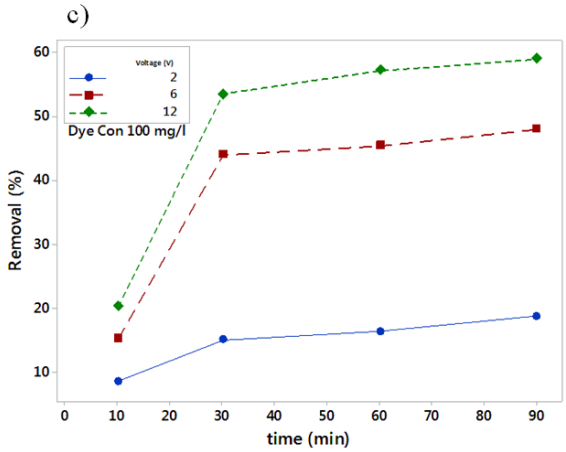

Figure 3: Efficiency of dye removal at electric potential and different times in a)10 mg/l b)50 mg/l c)100 mg/l in the electrocoagulation reactor

Figure 3 shows the removal efficiency of golden yellow dye at different times based on the dye concentration at optimum voltage of 12 volts in the electrocoagulation reactor. Based on the plot, the removal efficiency is reduced by increasing dye concentration.

In the third step, the removal efficiency for electrolysis and electrocoagulation reactors were studied separately and combined in the following optimum conditions: time 60min, dye concentration 10 mg/l, and voltage 12V. Figure 4 illustrates the results of the removal efficiency in these reactors. As it is represented, the efficiency of the electrocoagulation reactor is better than the electrolysis reactor in optimum conditions. Furthermore, the removal efficiency in the combined reactor is better than each individual reactor.

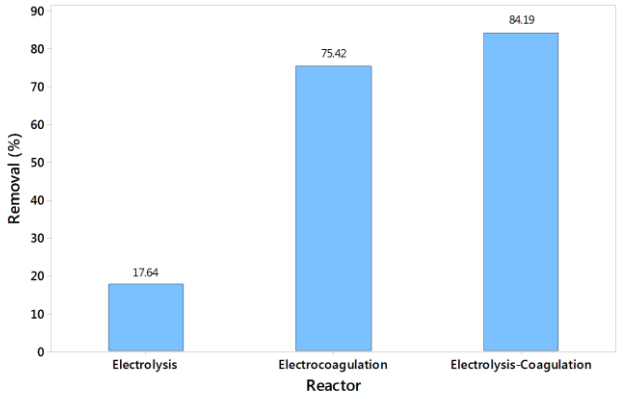

Figure 4: Comparison of removal efficiency in three studied reactors under optimum conditions (dye concentration; 10 mg /l, time; 60 min, voltage; 12)

Discussion

According to the results, the efficiency of removal has increased by increasing voltage. As a result, organic pollutants in the effluent can be destroyed due to electrochemical oxidation coming from the formation of a strong electric field and the production of free radicals such as H^+, O°, OH°, and H_2O_2 at higher voltage[21]. Increase of the cell voltage improves the anode potential, resulting in anode oxidation and increased reaction speed[22, 23].In a study on the removal of orange color by coagulation, Daneshvar et al. concluded that the removal of dye from the aqueous solutions was directly related to the voltage increase [24].

Studies show that the concentration of TDS present in the solution is important for the

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performance of the electrochemical processes, since it determines the electrolyte flow and affects the efficacy of removing pollutants. Increased flow of the electrolyte can improve the decomposition of organic matter as well as some side reactions, such as the production of H_2 in the cathode[21]. In this study, the concentration of TDS was fixed equal to urban water TDS, which was 375 mg/l. Generally, in most studies, NaCl is used to provide electrical conductivity in electrochemical processes[25, 26].In the case that the concentration of NaCl increases in the solution, the electrical conductivity and current densities are also increased; thus, the needed voltage and reaction time to provide the required current density are reduced. Therefore, the electrical energy consumed is reduced[25].

The main reactions that occur during the anode oxidation with organic compounds in the presence of NaCl are:

1.In the anodes:

Hypochlorite formation:

5) $Cl^- + 2OH^- \rightarrow OCl^- + H_2O + 2e^-$

Chlorate Formation:

6) $6ClO^- + 3H_2O \rightarrow ClO^-{}_3 + 4Cl^- + 6H^- + {}^3/_2\, O_2 + 6e^-$

Oxygen evolution:

7) $4OH^- \rightarrow 2H_2O + O_2 + 4e^-$

2.In the cathode:

Hydrogen evolution:

8) $H_2O + 2e^- \leftrightarrow H_2 + 2OH^-$

3.In the solution or near the surface of the anode, an indirect oxidation of organic compounds occurs.

9)

$$R \xrightarrow{active\ chlorine\ species} O_2 + H_2O + Chlorinated\ final\ product$$

In addition, the presence of chlorine ion in the electrolyte leads to indirect oxidation of the pollutants, because during the electrochemical processes a salt solution of products is produced such as CO_2, H_2, O_2, H_2O_2, $ClOH^{\circ}$, O°, OH°, O_3, ClO_2, and Cl_2. The $ClOH^{\circ}$, O°, and OH° radicals have a short lifespan. Due to their high oxidation potential, radicals can decompose other oxidants (CO_2, H_2,O_2, H_2O_2, O_3, ClO_2, Cl_2) and oxidize organic compounds.The primary (O_2 and Cl_2) and secondary (H_2O_2, O_3 and ClO_2) oxidants produced from radicular degradation have a long lifespan and are derived from the electrodes through the environment. They carry out the oxidation process and remove the dye[27]. Masudinejadet al. also investigated the electrolysis efficiency in removal of dye caused by phenolphthalein and phenol red. They observed that the concentration of salt had a direct correlation with the dye removal efficiency. This finding is consistent with the results of this study showing that increased electrolyte concentration improved the removal efficiency [28].

The results of this study indicated that with increasing initial dye concentration, the removal efficiency of the dye is decreased. Another study over the electrochemical process under constant conditions of the voltage and the contact time showed that increased initial dye concentration decreased the dye removal efficiency. This was due to the fact that in electrical conductivity and a constant voltage, a certain amount of oxidizing compounds is produced in the environment and this amount is capable of removing a certain amount of dye molecules[29]. Dalvand et al. also examined the removal of reactive red color by the electrocoagulation process and found that the removal efficiency was reduced by increasing the dye concentration, which is consistent with the results of the present study[30].

Conclusion

The maximum removal rates of golden yellow dyewere 17.64% and 18%by electrolysis process using graphite electrode at a voltage of 12V, for a dye concentration of 10 mg/l at the times of 60 and 90 minutes, respectively. The maximum removal efficiency rates of the electrocoagulation reactor were 67.12%, 75.42%, and 78.20% in conditions of 12 V at 30, 60, and 90 times for 10 mg/l, respectively. According to the results, the determined optimum conditions to investigate the electrolysis-electrocoagulation combined electrolysis reactor were voltage of 12V, time of 60 minutes, and dye concentration of 10 mg/l. The results of the combined

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reactor showed that the maximum achievable efficiency for dye removal was 81.19%. The results of the experiments showed that the removal efficiency of the dye was directly correlated with increased voltage and time and decreased dye concentration. In general, due to the amount of voltage and high speed of the reaction, the electrocoagulation method can remove the golden yellow dye with higher efficiency in the laboratory scale.

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Conflict of Interest

No conflict of interest has been stated by the authors.

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