Investigating Changes in the Physical and Chemical Effects of Tartarazine Dye Remove on Industrial Wastewater by Electrocoagulation Method

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ABSTRACT: *This study has been accomplished to investigate the efficiency of electrocoagulation (EC) in aqua and determine operative condition and the effect of elements (Iron, Aluminum as an electrode), as well as, different connections in the amount of removing the tartrazine color of aqua using DC sources in an electrochemical cell, including anode and cathode the effect of each of varieties like the density of electricity circulation, time of the electrolyze distance between two electrodes, the density of electrolytic and pH which can have the most effect on color destroy. For solution 500 ppm NaCl +50 ppm tartrazine, COD (the amount O_2 in chemical activity) almost equals 40 ppm in pH about 6, the density of electricity flow was about 125 Am^{-2} and distance between two electrodes was 2 cm in 8 min from electrolyzing almost 100% color and 90% COD were canceled in next stage. We compared the efficiency of EC cells, including monopole electrodes in parallel connection and series with EC cells including polarize electrode along with one simple cell of EC. The result of this experiment demonstrated that EC cells including monopole electrode with series connection with sacrifices electrode of aluminum and cathode, anode iron had more efficiency and when using EC simple cells, including an ironic anode and cathodic aluminum. We could see better results rather than EC simple cells including ironic anode cathode.*

KEYWORDS: *Electro coagulant; Tartrazine color; Impure color; Combination wastewater; Industrial wastewater.*

INTRODUCTION

According to the previous studies, only an iron victim electrode has been used for the color removal process, and the changes in acidity or temperature were undesirable [1]. In another study, it was decided to remove cation using electrocoagulation, and only the victimized aluminum electrode was utilized [2]. In the study, acidity changes varied between 2 to 11 and the electrodes are the victims of iron and aluminum, but they have not focused on how electrodes are connected [3, 4]. However in the present paper, the pH changes were also higher, and the connection

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1021-9986/2021/1/123-132 10/$/6.00

mode and the shelf life of the sediment are also considered. Table 1 shows the type of connections and the type of victim's electrodes. Moreover, the time and flow rates were also studied. Another important point is the distance between the electrodes, which is assumed to be variable. In general, the present research attempts to investigate all the weaknesses and problems that have not been addressed by the previous scholars. This research helps to provide a complete way relative to the previous methods, because the methods used in previous papers depend on the conditions and facilities with constraints, as a result some factors were also taken into account by the present paper, which are not considered by the previous studies.

Color materials have been always considered as one of the added particles which are used in textile, food and drug industries. This is why a part of the waste of these industries are infected by these chemicals. Thus, one of major problems for most waste industrial products, as well as, cities is to remove these color materials having diazo group in their structure and solvability of their cancellation is problematic. So far, researchers have studies and proposed different methods for treating colored wastewater. The chemical coagulation process is not suitable for treatment of wastewater containing soluble colors [5]. The cost of adsorption by using high active carbon is high. The oxidation process using ozone and hypochlorite is an effective way to stain the wastewater, but the operating and equipment expenses are relatively high in this case. In addition, the presence of excess chlorine in the solution creates contamination. We have reported the catalytic ozonation of *para*-nitrophenol using TiO_2 in water [6].

In 2004, by Selcuk, the dyeing and decontamination of textile wastewater was studied by processes of ozonation and chemical coagulation by the cluster of aluminum sulfate and ferro sulfate. In this study, when using 1000 mg L^{-1} sulfate or 500 mg/L aluminum sulfate, the removal of color, COD and toxic waste were 60% -50%, 60% and 80% -70%, respectively. It has been reported that, due to the formation of large quantities of chemical sludge in the chemical coagulation process and removal of more than 98% of the color, was found to be more suitable for ointment in the dyeing and reduction of toxicity of the effluent, due to contact with the ozone for 20 min [7].

EXPERIMENTAL SECTION

Operating conditions

All experiments were performed at a discontinuous reactor at a temperature of about 295 K and a solution of 250 mL with iron and aluminum electrodes, and due to the low conductivity of the test specimen (57.9 μS / cm at 20 °C) to increase the conductivity, aqueous solution of tartrazine was used as electrolyte carrier. In order to control the flow and apply the voltage the rectifier device was utilized. At the end of each experiment, the contents of the reactor were transferred to a 250 ml cylinder and held at a standstill for 20 minutes to allow the flocks to settle. The produced clusters were initially smaller and placed at the top of the column, but after some time some of these clusters continuously produced bulky clusters and began to settle, which during sedimentation, a large part of the colored molecules absorbed the clots, were removed and excluded. After 20 minutes, the solution above the cylinder was straightened by a sheet of paper. In order to evaluate the efficiency of the electrical coagulation method, the color concentration was measured by measuring the absorbance of the solution before and after electrolysis by the UV/Vis spectrophotometer and using the calibration graph the remaining color concentration was measured.

Tartrazine solution concentration measurement method

To do this, first, a two-radial spectrophotometer was designed to determine the maximum absorption wavelength (λ_{max}) of the tartrazine solution spectrum in the wavelength range of 190-800 nm. Its absorption spectrum shows two severe absorption during one wave at 428 nm and another at 254 nm. Therefore, these same wavelengths are chosen to measure the concentration of tartrazine in the visible and ultraviolet regions [8]. After determining the maximum absorption wavelength in each of the visible and ultraviolet regions, the calibration charts for each of these λ_{max} are required to determine the concentration of the unknown samples [9]. Therefore, seven solutions with different concentrations of tartrazine and their absorption at 428 nm and 254 nm were measured by a UV/Vis spectrophotometer, and the calibration charts were based on available and curve data for each wavelength of 428 nm and 254 nm separate drawing [10].

COD measurement method

To measure the amount of soluble COD, a special kits containing potassium dichromate, sulfuric acid, and silver sulfate have been used. In this way, the special pipette added exactly 2 ml of the solution to the kite solution, and after tight closing it, the kit was shaken and then placed in a thermo reactor for 2 hours at a temperature of 148 °C. After two hours, the kit was removed from the thermocouple and as the temperature of the kit reached the laboratory temperature, the COD sample was read using the Phot Lab Spectral [11]. The solutions of COD include 40 ppm of tartrazine and 400 ppm of salt before electrolysis and after electrolysis by an electrical coagulation cell including an anode and an iron cathode, an electrochemical coagulation cell including iron anode and aluminum cathode, an electrical coagulation cell including iron anode and cathode and aluminum sacrifice electrodes. In a serial connection and an electrical coagulation cell, including anode and cathode, and aluminum bipolar sacrificial electrodes, a parallel connection is provided [12].

How to present the results: In all experiments, the following equation is used to measure the removal percentage [13]:

$$CR\% = \frac{C_0 - C}{C_0} \times 100$$

In this equation, C_0 denotes the initial concentration of colored solution and C refers to the concentration of solution at the end of each test [14]

Procedure details

20 mL test specimens and 10 mL of potassium dichromate solution (high concentration) in distillation balloons. Were used for analysis. Then, 30 mL of sulfuric acid containing silver carefully were added to the solution. Afterwards, the solution is refluxed. Finally, the solution is cooled and the volume of the solution in the balloon was about 150 mL. We added 3 drops of ferrin detector solution with ferammonium sulfate solution. Let's do the titration. The detector changes to the reddish-brown color at the final point of the reaction from greenish-blue to reddish-brown. At the same time, control sample containing 20 mL distilled water was measured.

$$COD\,(mg/L) = \frac{(a - b) \times f \times 200}{V}$$

a: Freamonium sulfate volume used for experimental specimen

b: Freamonium sulfate consumption for control sample

f: Fraction of titration of ferammonium sulfate solution

v: Sample sample size

RESULTS AND DISCUSSION

Electric coagulation process is a simple and effective method for water treatment and many other types. It is also possible to eliminate algae and microorganisms in this process [15]. Therefore, the electrical coagulation process is an ideal choice to stain color solutions.

Effect of kind and electrode binding method on removal of color and reduction of COD

According to the results presented in Table 1, in the case of using iron anode and an electrical coagulation cell including single polar electrodes in the series connection, the highest removal percentage of tartrazine will be obtained. The iron anode preference for the aluminum anode in this experiment can be attributed to the following reasons: (a) The process of removal by an aluminum anode is essentially accomplished using electric coagulation process, but in the case of iron anode, the total of two processes of electrical coagulation and electrical oxidation are involved. (b) The absorption capacity of pollutants on aluminum hydroxide clots is much lower than that of Ferric hydroxide. Table 2 shows the amount of tartrazine elimination. It shows the amount of color removal by the time of absorption of light by a spectrophotometer at a suitable wavelength.

Effect of the time remaining on the precipitates removing the paint

Electrolysis results under different concentrations of sodium sulfate are shown in Fig. 1(A). Moreover, the effect of electrolysis time on removal efficiency related to 254 nm wavelength and iron cathode are the effect of the sludge contact time on the efficiency of tartrazine. As it can be seen, with increase in sludge contact time, the removal efficiency increases to the optimum point and then decreases, since the contact with the solution due to its incomplete absorption capacity Repeated color, due to admixture with the solution, can increase the efficiency of removal. However, when the electrolyzed sample contacts time not only does not increase the efficiency of the electrolyte, but by stirring the solution, the absorbed colors are re-introduced into the solution, and consequently the removal efficiency is gradually reduced. The maximum

Table 1: The effect of kind and the method of connecting the victim's electrodes to removal efficiency ($C_{0[Dye]}$ = 50 ppm, C_{NaCl} = 500 ppm, $t_{elec.}$ = 5 min, I = 0.43 A, d = 1 cm, retention time = 20 min).

CR% [254 nm]	CR% [428 nm]	Number of Electrodes	Type of electrodes	Connection shape of the electrodes
54.69	100	4	Single polar electrodes Victim electrodes: iron Anodes: iron and cathode: acetyl 304	Serial connection
88.63	100	4	Single polar electrodes Victim electrodes: iron Anodes: aluminum and cathode: acetyl 304	Serial connection
66.07	69.95	4	Single polar electrodes Victim electrodes: aluminum Anodes: aluminum and cathode: acetyl 304	Serial connection
50.50	98.27	4	Single polar electrodes Victim electrodes: iron Anodes: iron and cathode: acetyl 304	Parallel connection
57.48	100	4	Single polar electrodes Victim electrodes: iron Anodes: aluminum and cathode: acetyl 304	Parallel connection
86	99.57	4	Single polar electrodes Victim electrodes: aluminum Anodes: aluminum and cathode: acetyl 304	Parallel connection

Table 2: Effect of electrolysis time on removal efficiency related to 254 nm wavelength and iron cathode (C0[Dye]=50 ppm,CNaCl= 500 ppm,[i] =120 A m-2, d=1.5 cm).

CR%	CDye (ppm)	absorption	Electrolysis time (min)
57.73	30.27	0.854	5
59.97	29.40	0.758	6
69.95	26.02	0.653	15
71.02	25.52	0.602	30
73.88	24.45	0.489	45
76.25	23.13	0.402	60
82.69	20.62	0.389	75
89.79	8.86	0.311	120

color removal was obtained at 15 min of sludge contact. In electrolysis, oxidation and resuscitation reactions occur. The result is slime or sediment. Which indicates the removal. By measuring the ambient color and the amount of sludge, electrolysis can be observed. The equilibrium relationship is always between the sludge and the solution. At that time, it plays an important role.

Effect of soluble pH on the removal efficiency of the paint

Fig. 1 (B) shows the variations in the efficiency of tartrazine elimination by changing the pH It can be deduced that low removal efficiency occurs at low pH, because at high concentrations of proton, the hydroxides produced by the reduction of water in the cathode are quickly neutralized and the iron oxidation is low due to the oxidation of these ions. The main reason for the removal of soluble color is reduced, and on the other hand, the soluble protons [H^+] in the cathode instead of the water molecules are decreased and the amount of hydroxide ion is reduced to the same amount, the result of the removal efficiency is reduced. The reason for reducing the removal efficiency at higher pH can be explained by the fact that at higher pH, due to the concentration of the solution from the hydroxide ions, some of these ions are oxidized and, as a result, exhaust from the working environment, and the oxidation of iron is low due to the oxidation of these ions. Due to reduced iron oxide concentration, the removal efficiency is low. Because the initial pH of the tartrazin solution to the concentration of 40 ppm is about 5.78 and at this pH, the removal efficiency is equal to 99.32%. Therefore, in order to remove the color of the solution of tartrazine, it is not necessary to adjust pH to carry out the electrical coagulation process.

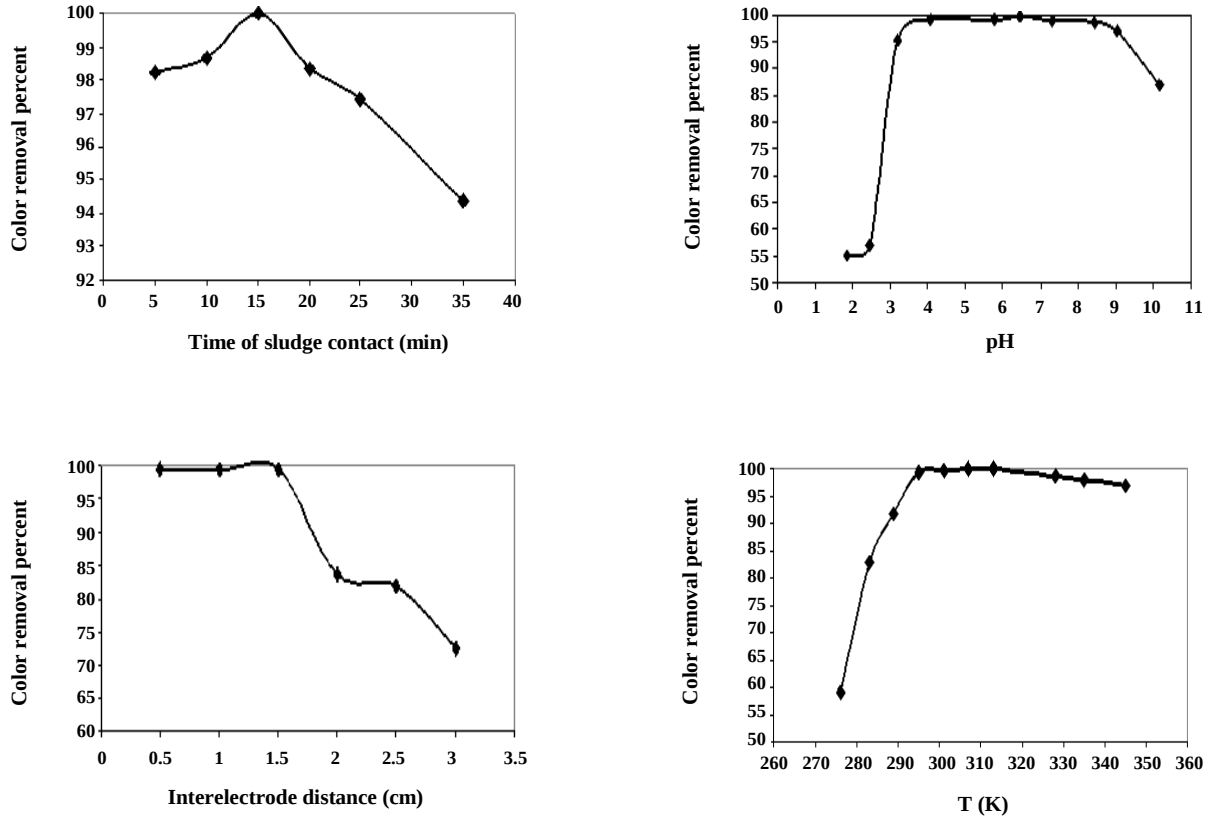

Fig. 1: Variation curves of variables. a) Effect of the sludge contact time on the removal of paint, b) Effect of initial pH of removal solution, c) Effect of electromotive distance on removal efficiency, d) Effect of water temperature removal.

The effect of the gap between the bivalent on the color removal efficiency

Fig. 1(C) shows the variation in the efficiency of removal of tartrazine by increasing the distance between the two electrodes. It should be noted that with increasing distance between the two electrodes, the removal efficiency decreases, which is due to the decrease in the pollutant density with the polymer hydroxide and as a result of the reduction of the electrostatic gravity, while with the decrease of the distance between the two electrodes, the interactions and reactions are increased and the removal efficiency is increased due to increasing local concentration. Meanwhile, by decreasing the distance between two electrodes, due to the decrease in the electrical resistance of the solution, the voltage decreases to achieve the specific current density. In this study, because of the same removal efficiency at intervals of 0.5, 1.0 and 1.5 cm, the ease of operation is considered as an optimum distance of 1.5 cm.

Effect of temperature on color removal efficiency

Fig. 1(D) shows the variation in the efficiency of tartrazine removal as a result of temperature increase. It can be inferred that an increase in temperature up to K 314 increases the removal efficiency, and then reduces the removal efficiency. With increasing temperature from K 278 to K 314, the removal efficiency 41.88% of the sample is due to the increase in the temperature due to increased particle crash, the speed of the electrolysis process and also the spread between the pollutant particles and the hydroxyl polymers. But after K 314, with increasing temperature, the removal efficiency decreases because excessive temperature rise prevents better accumulation of particles, and, along with the adsorbed particles on the sediments, due to increased particle collisions, they release, and at higher temperatures, the sediment increases. Fig. 2 through 5 show the connections used in the tests.

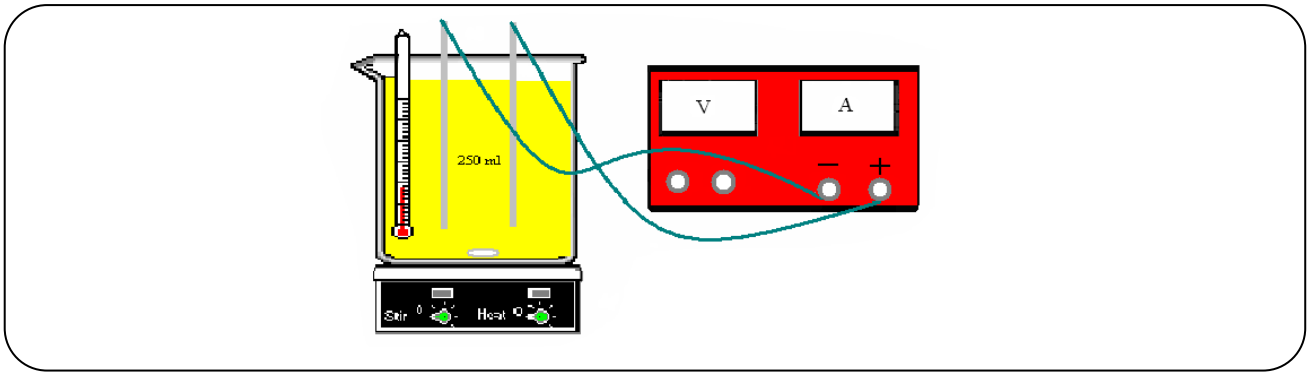

Fig. 2: Electrocoagulation apparatus.

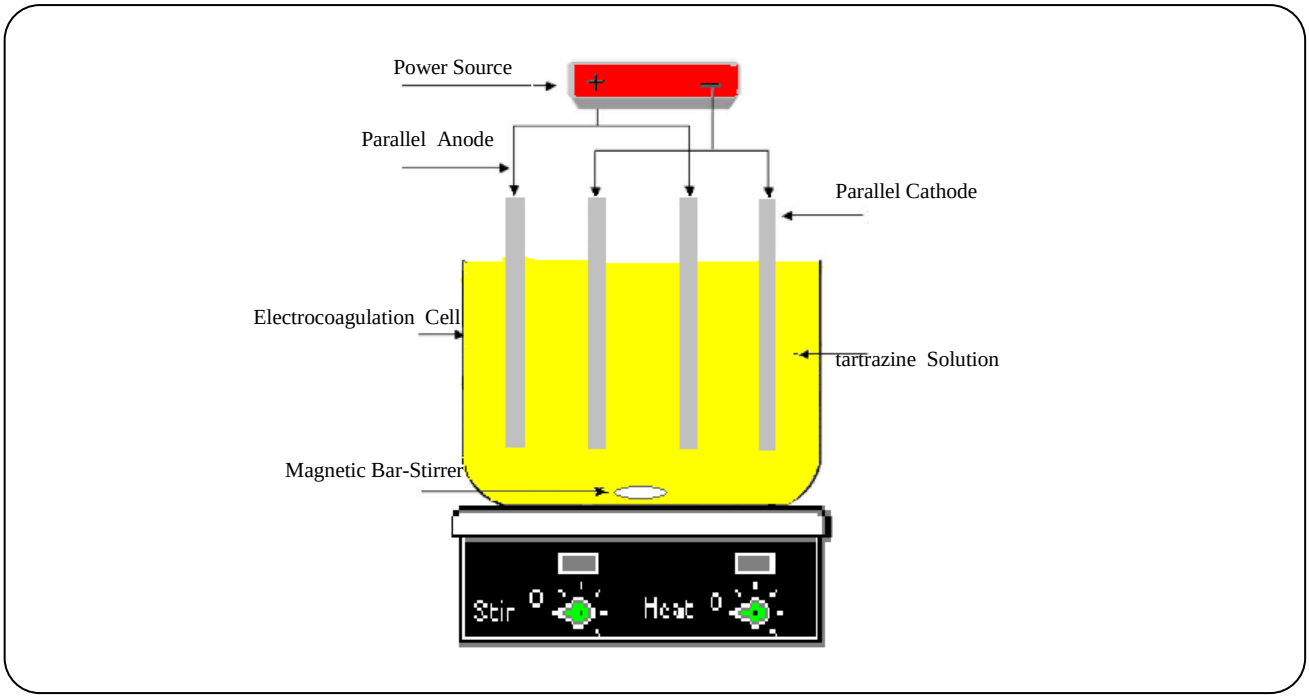

Fig. 3: Bench-scale EC reactor with monopolar electrodes in parallel connections.

Discussion

For *Kiran et al.* (2018), Bioremediation should be regarded as the man process for removal of recalcitrants, as it is deemed that it is common to observe the microorganisms with capability of vitiating effluents in the ecosystem. They declare that there exist two types of assimilation, namely assimilation of carbon and assimilation of other elements. During assimilation of carbon, the percentage of C components is converted into cells including biodegradation, while during assimilation of other elements, the molecules, such as N, P and S and energy are adopted for producing microorganisms [16]. In another research, *Kiran et al.* (2017) reviewed the most recent trends used in textile effluent treatments. They state that for realization of the proper color and design for textiles, using dyes, auxiliaries, energy and water is a must, as a result various methods are proposed for exploring the best condition and parameters for textile effluent treatment and various techniques are available to remove these pollutants, which are eco-friendly, rapid and cost-effective [17]. Some of these methods are as follows: Coagulation and Flocculation, membrane processes, Ultra Filtration, Micellar-enhance Ultrafiltration (MEUF), Nano-Filtration, Photo Catalytic Degradation of Dyes, Oxidation and Photocatalysis with Hydrogen Peroxide, Ozonation, Degradation of Dyes Using Sodium

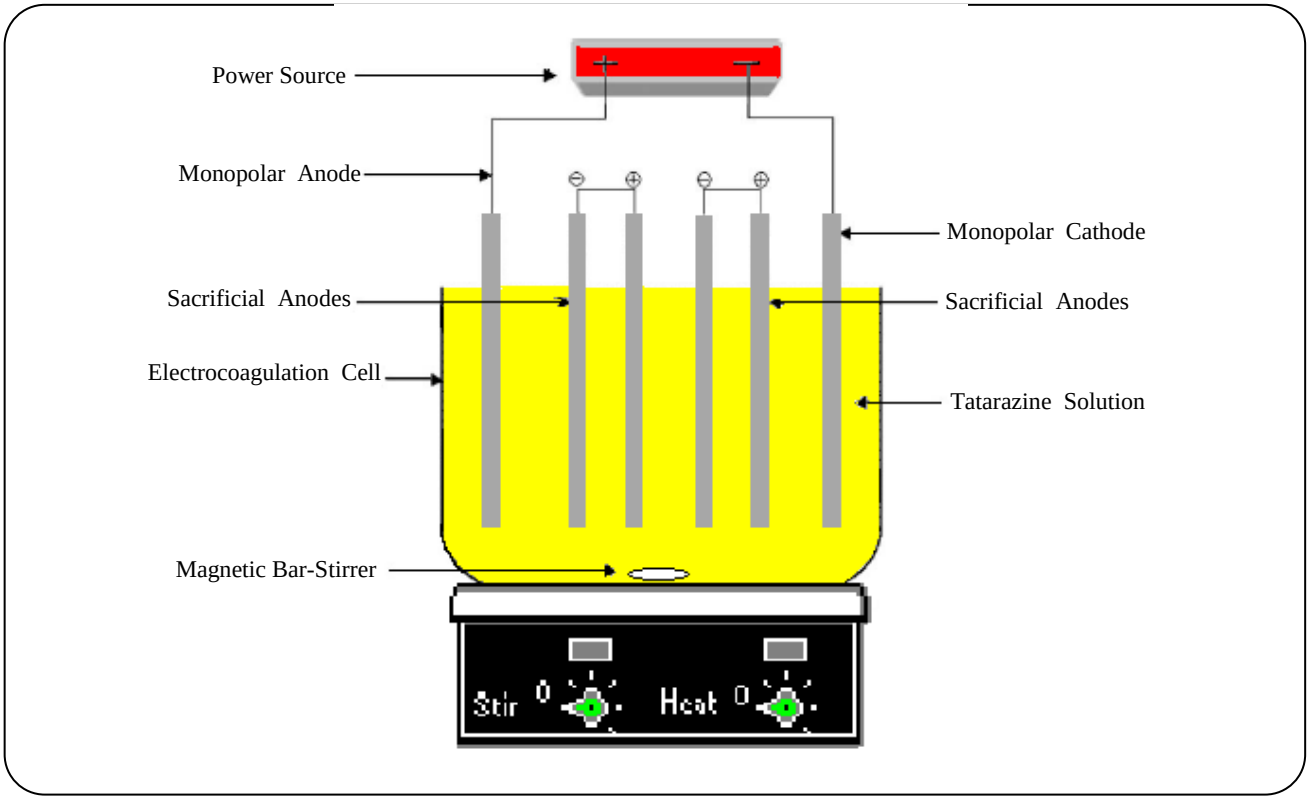

Fig. 4: Bench-scale EC reactor with monopolar electrodes in series connections..

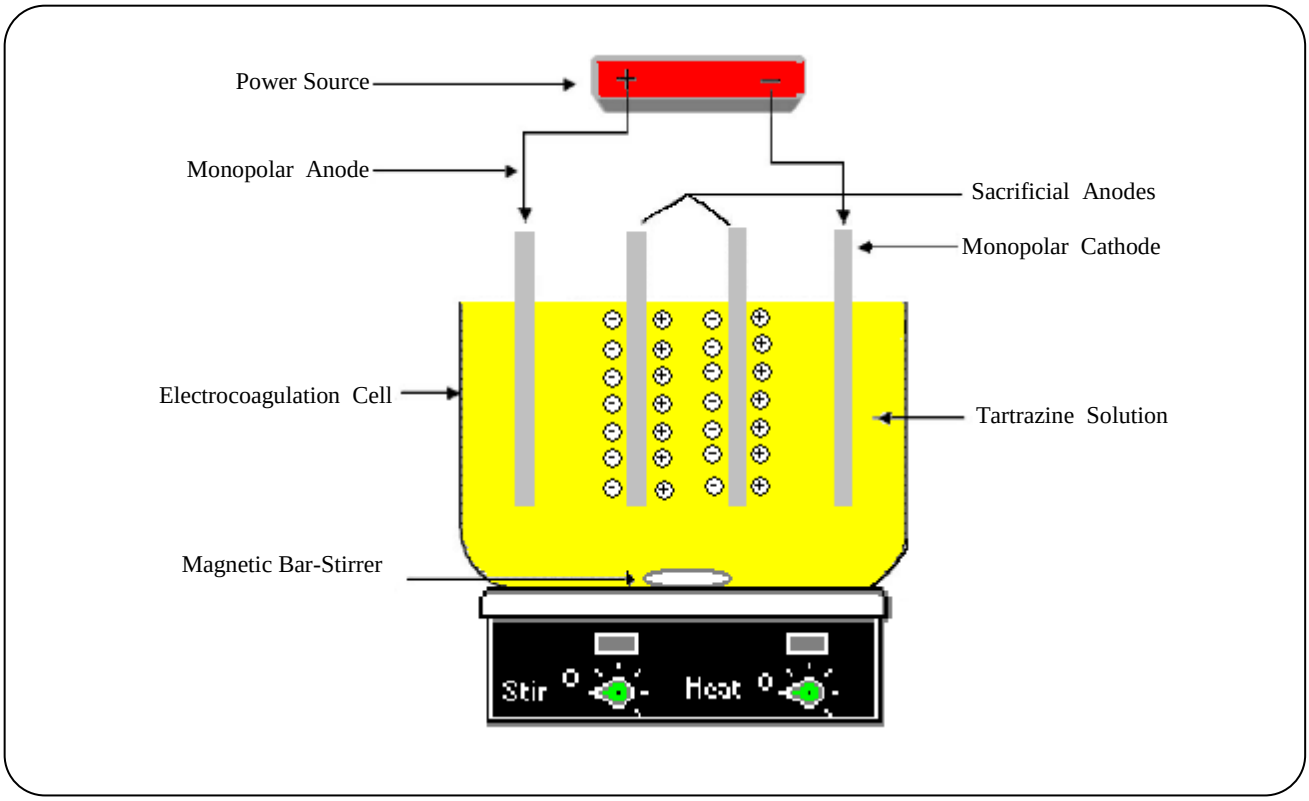

Fig. 3: Bench-scale EC reactor with monopolar electrodes in parallel connections.

Hypochlorite (NaOCl), Electro-chemical Method. *Saeed et al.* (2016) studied the degradation of direct black dye which was catalyzed by nickel hydroxide within aqueous solution. For this purpose, they utilized nickel sulfatehexahydrate, sodium hydroxide and sodium hypochlorite to synthesize the nickel hydroxide catalyst. They observed degradation reaction due to the Hinshelwood mechanism, and the reactant reacted in the form of absorption at the nickel hydroxide catalyst surface [18]. *Saeed et al.* (2015) used 5% Pt supported on alumina as the catalyst induring decolorization of the Rhodamine B dye. They also studied the effects on various parameters, such as temperature, reaction duration, agitation rate, catalyst dose and initial concentration of Rhodamine b dye on decolorization process. The results demonstrated that fresh and used catalyst exhibits the same catalytic performance and the decolorization reaction has Langmuir-Hinshelwood mechanism, in which adsorption of Rhodamine B at surface of catalyst followed Freundlich adsorption isotherm [19]. *Saeed et al.* (2017a) believes that the azo dyes are mulish pollutants with high capability in damaging the environment. For catalyzing the degradation treatment of methyl orange, they used Pt/Al_2O_3 [5]. The findings ascertained that an increase in the temperature improves the degradation efficiency and any increase in the initial concentration mitigates the degradation. According to the resulted derived from kinetics analyses, the degradation reaction follows Langmuir-Hinshelwood mechanism. Activation energy, entropy, enthalpy of adsorption and Gibbs free energy were computed to be equal to 29 kJ/mol, 94.9 J/K.mol,-18.2 kJ/mol, and -10.6 kJ/mol [20]. *Saeed et al.* (2017b) prepared ZnO NPs via reaction of zinc acetate dehydrate and sodium hydroxide in distilled water. They found that the oxygen absorbs at the surface, while the methyl orange reacts during fluid stage. The findings showed that the active radicals are capable of mineralizing the dye through water and carbon dioxide [21]. In addition, *Saeed et al.* (2016a) conducted a study on oxidative degradation of Methyl Orange which was catalyzed by nickel hydroxide within aqueous medium. In the first step, the reactants absorbed at the catalyst surface, which was followed by interaction between the observed reactants during the second phase [22]. *Saeed et al.* (2016b) studied the degradation of direct black dye which was catalyzed using nickel hydroxide. The catalytic efficiency of the nickel hydroxide was assessed regarding the discrete parameters, such as temperature effect, concentration of dye, stirring speed, reaction length, etc. [23]. It was observed that the degradation reaction follows the Langmuir-Hinshelwood mechanism. *Muneer et al* (2015) has investigated the removal of reactive orange P3R dye using oxidative pathway method. They point out that the dye concentration, gamma ray absorbed dose and concentration of hydrogen peroxide have been enhanced for attaining the maximum degradation of dye [24]. *Zhian et al.* (2018) have proposed a Novel Designed Ligands of 4,4,6',6'-Tetramethyl-3'-aryl-3',5',6',7'- tetrahydrospiro[cyclohexane -1,2'-indole]-4',6(1'H)-diones for metal cations removal from water and wastewater. They synthesized novel oxime and azo dye based compounds from spiro dihydrofurans. According to the obtained results, the UV-visible spectral data showed that 4a selectively absorbed Ni^{2+} at room temperature and natural pH condition [25]. In another study, *Shokri* (2018) used electrocoagulation process for the removal of Acid Orange 5 in synthetic wastewater and they revealed that the optimum conditions was realized at initial pH of 7, current density at 2 mA/cm^2, 60 mg/L of Acid orange 5 and time of reaction at 60 min. Under optimum circumstances, the removal efficiency of Acid Orange 5 and Chemical Oxygen Demand (COD) was equal to 99.3 and 85.5%, respectively [26].

CONCLUSIONS

The electrical coagulation method is a suitable method for purifying the diazo aqueous materials in industrial effluents such that in solutions 500 ml of color solution containing 50 ppm of tartrazine and 500 ppm of salt. The electrical current of about 120 mA was measured by electrochemical coagulation using an electrochemical cell including iron anode and aluminum cathode at pH 5.78, about 40 ppm. The distance between the electrodes is 1.5 cm in 6 minutes 30, at a temperature of 295 K (22 °C), 90% of the COD, content of the solution is reduced. By changing the carrier electrolyte, we can find other results, which can be described as the best electrolyte, given that sodium chloride can easily be tested. The above method can be considered for a variety of other colors, and even with the change of solvent, the changes and favorable conditions are determined. Because after the coagulation process of the particles that are formed from the tartrazine-induced sacrificial electrodes, they are insoluble

in sediment, the resulting deposits are detachable. If oxides of particles are likely to oxidize, they produce aluminum oxide and iron oxide, which are also insoluble oxides. And can be separated by filtering. In order to determine the amount of precipitated light. The electrodes can then be weighed after testing. And by weight loss, we can get the amount of milled columned color.

Although this research focuses on the competitive response between elements, the physical effects such as temperature and electrode distances that play an important role in electrolysis are also studied. So that, it is possible to obtain the best suitable method for removing water-soluble dyes. The important thing in this process is to control each physical and chemical variables, each of which can have positive or negative impact on the reaction result. Based on the experience and coordination, each of the above-mentioned variable can be situated in the best possible conditions. Of course, according to Table 1, we set the conditions for doing the research in as follow: (C0 [Dye] = 50 ppm, CNaCl = 500 ppm, telec. = 5 min, I = 0.43 A, d = 1 cm, retention time = 20 min) It should be noted that changing any variable, new conditions can be achieved. But according to experience, we have introduced the best conditions.

Acknowledgment

We gratefully acknowledge financial support from the Research Council of Islamic Azad university Mahabad and Urmia university.

Received : Feb. 23, 2019 ; Accepted :Sep. 16, 2019

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