



## Removal of Reactive Green 19 dye from synthetic wastewater using electrocoagulation and aluminum electrodes

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### Original Article

#### Abstract

Textile dyeing is considered to be one of the major industrial sources of high rates of organic and aromatic compounds. Conversely, these compounds have become a significant environmental problem. Many methods have been investigated for color removal from dye-containing wastewater. In this research, the efficiency of the electrocoagulation (EC) process with aluminum electrodes in the removal of Reactive Green 19 (RG-19) dye from synthetic solutions was studied. The experiments were conducted in a batch reactor equipped with 4 aluminum electrodes with a volume of 2 l. Dye concentrations were measured ( $\lambda_{\max} = 630 \text{ nm}$ ). The effects of operating parameters, such as voltage, reaction time, initial dye concentration, energy consumption, pH, KCl concentration, and inter-electrode distance, on removal efficiency were investigated. The highest removal efficiency of RG-19 was found to be 33.49, 60.32, 72.43, 93.63, and 94.91 percent for initial voltage of 10, 20, 30, 40 and 50 v, respectively, in optimum conditions (pH = 11, KCL concentration = 0.005 M, and distance = 1 cm). The removal was effectively reduced to less than 99.88% when the initial dye concentration increased from 25 to 150 mg/l. In addition, by increasing KCl concentration and decreasing electrode distance, removal efficiency increased considerably. Based on the results, EC process by aluminum electrodes is an efficient and suitable method for reactive dye removal from wastewater.

**KEYWORDS:** Aluminum, Electrodes, Electrocoagulation, Textile Wastewater

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#### Introduction

Textile dyeing creates large volumes of effluent and is considered as one of the major industrial polluters. The large quantity of aqueous waste generated by textile industries has become a significant environmental problem.<sup>1,2</sup> The production of dyes-containing wastewater is common in industries like textile, paper, plastic, food, and mineral processing industries ;

furthermore, azo dyes constitute over 50% of all textile dyes used in the industry.<sup>3,4</sup> In developed countries, these types of wastewater are normally treated through traditional methods, but in developing countries, they are discharged into natural streams and/or any river. This causes serious problems as well as biological changes, consuming dissolved oxygen (DO) and demolishing aquatic life. Some dyes are carcinogenic, mutagenic, and toxic.<sup>5</sup> Therefore, it is necessary to treat dye effluents prior to their discharge into streams. Many methods have been investigated for color removal from dye-

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containing wastewater. In general, several techniques are used for the treatment of textile effluents including physiochemical methods of dye removal [such as filtration, chemical coagulation, activated carbon adsorption, ultrafiltration, ozonation, and electrocoagulation (EC)], chemical methods of dye removal (such as reduction, oxidation, ion exchange, and neutralization), and dye removal by means of biodegradation.<sup>6-9</sup> Some of these methods are effective, although they are quite expensive and have many disadvantages and limitations. In chemical oxidation or coagulation, added chemical substances lead to secondary pollution. Biological methods cannot be applied for most textile wastewater types, because most commercial dyes are toxic to the organisms used in the process.<sup>5,10,11</sup> In recent years, the successful EC treatment of various organic effluents has been reported by many authors.<sup>9-13</sup> The EC process provides a simple, reliable, and cost-effective method for the treatment of wastewater without any additional chemicals and secondary pollution. It also reduces the amount of disposed sludge and requires simple equipment and easy operation.<sup>9,14</sup> The dye is coagulated by aluminum hydrates or hydroxides produced from the sacrificial anode.<sup>15,16</sup> Reactive Orange 19 (RG-19) dye belongs to a class of organic compounds known as azo dyes, which are abundantly used in textile industries for dyeing.<sup>5</sup>

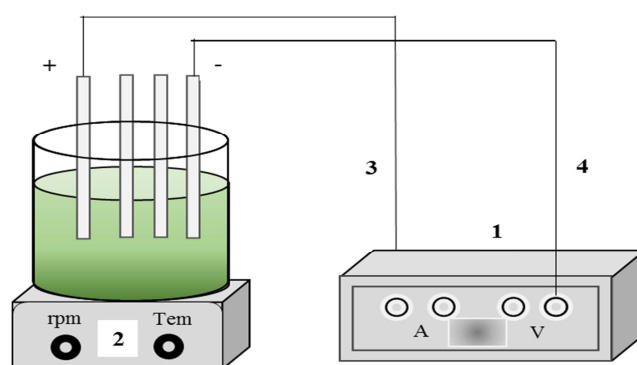
The present work was carried out to study the removal of RG-19 dye by EC with aluminum electrodes from synthetic wastewater. Wastewater parameters, such as dye adsorption, high salt concentration, voltage, initial concentration of dye, and pH, were also investigated to examine their effects on the dye.

## Materials and Methods

All chemicals used in this study were obtained from the Merck Company (Germany). The chemical structure of RG-19 is  $C_{40}H_{23}Cl_2N_{15}Na_6O_{19}S_6$  with a molecular weight of

1418.93 g/mol. The pH was adjusted using either 0.15 M NaOH or 0.1 M  $H_2SO_4$  as necessary.

Figure 1 illustrates the batch reactor apparatus used in the study. It consisted of 4 bipolar aluminum electrodes; 2 anodes and 2 cathodes of the same dimensions ( $10 \times 15$  cm) placed 1 cm apart. A digital DC power apparatus (ADAK PS808 model, Iran) was used in all experiments.



**Figure 1. An electrocoagulation cell apparatus: (1) digital DC power apparatus; (2) magnetic stirrer bar; (3) anode; (4) cathode (bipolar electrodes in parallel connection)**

RG-19 dye solutions were prepared by dissolving stock solution in distilled water and used without any further treatment. In each run, 1000 ml of the dye solution was added to the reactor. The pH of the solution was measured before and after each experiment. The electrodes were connected to a digital direct current (DC) power supply to measure the current and the potential between the two electrodes. All runs were performed at the constant temperature of  $22 \pm 2$  °C. In each run, 2 l of the wastewater solution was added to the electrolytic cell. At the end of EC, the solution was filtered and the filtrate was centrifuged at 4000 rpm for 5 minutes, and then, was analyzed. Before each run, the electrodes were washed by dipping for 5 minutes in a solution freshly prepared by HCl solution (35%) to remove impurities on the surface of the aluminum electrodes.<sup>17</sup> Finally, the electrodes were washed thoroughly with diluted

water to remove any solid residues on the surfaces, dried, and re-weighted. Moreover, in the EC method, as a treatment technology with aluminum electrodes, some factors such as initial pH, contact time, voltage, energy consumption rate, and initial concentration of dyes were studied. Table 1 shows the range used for these experiments.

The concentration of dye remnants were measured ( $\lambda_{\max} = 630 \text{ nm}$ ) using a spectrophotometer (Shimadzu, Tokyo, Japan; Model 1601). The efficiency of dye (RG-19) removal, % Removal, after EC treatment was calculated as follows:

$$RE(\%) = \frac{(C_0 - C)}{C_0} \times 100 \quad (1)$$

Where  $C_0$  is the initial concentration (mg/l) and  $C$  is the final dye concentration (mg/l). Electric energy consumption was also calculated using the commonly used equation<sup>18</sup>:

$$E = \frac{UIt_{EC}}{V}$$

Where  $E$  is electric energy (kWh/m<sup>3</sup>),  $U$  the cell voltage in volt (V),  $I$  the current in ampere (A),  $V$  is the volume of solution (m<sup>3</sup>), and  $t_{EC}$  is the duration of EC process (minute).

## Results and Discussion

### Effect of initial pH and voltage

The pH of the solution is one of the most important parameters in EC process and dye removal from aqueous solutions. Using the EC process at its optimum pH results in maximum pollutant removal.<sup>19</sup> To achieve an optimum voltage and pH, dye removal percentage should be evaluated. Figures 2-6 and the results show that an increase in voltage from 10 to 50 v caused an increase in dye removal efficiency at

different pH values; maximum color removal was achieved at a pH of 11. The highest removal percentages were found to be 33.49, 60.32, 72.43, 93.63, and 94.91% for initial voltages of 10, 20, 30, 40, and 50 v, respectively, at a pH of 11.

The mechanism of the electrochemical process in aqueous systems is complex. With an increase in voltage, the amount of aluminum ion produced increased. Therefore, there was an increase in flock production, and hence, an improvement in dye removal efficiency.<sup>20</sup> In addition, the results showed that the EC process with aluminum electrodes had greater effect on the pH of the samples and caused an increase in final solution pH. As in the study performed by Sengil and Ozacar on the decolorization of C.I. Reactive Black 5 in aqueous solution in 2009, the results showed that by increasing pH, the dye removal efficiency increased.<sup>21</sup> In another research by Basiri Parsa et al. on the removal of acid brown 14 from aqueous media, the findings proved that, in time, the final pH solution enhanced.<sup>22</sup> The investigation by Daneshvar et al. illustrated that with increasing the primary voltage in EC process, the removal efficiency increased.<sup>23</sup>

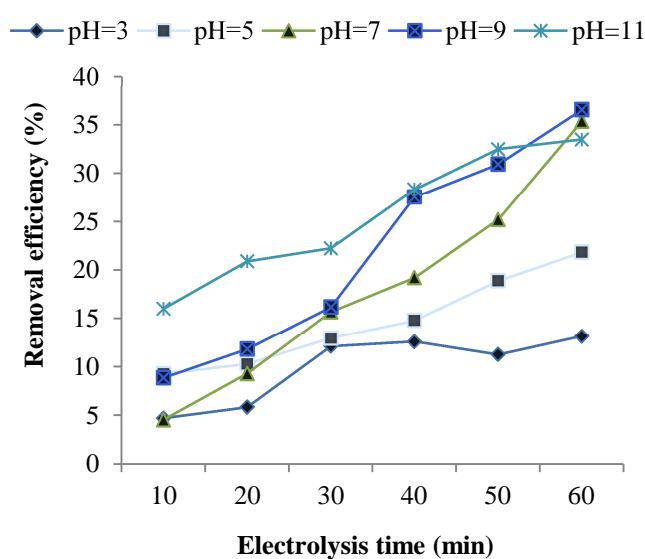
### Effect of initial concentration of dye

The effect of initial concentration of dye on the removal efficiency and energy consumption ( $E$ ) has been presented in table 2. The results showed that with increasing initial concentration of dye, both energy consumption and dye removal increased.

By using aluminum as sacrificial anode, the removal efficiency of RG-19 dye reduced dramatically to less than 99.88%, when the initial dye concentration increased from 25 to 150 mg/l. This is because of the decrease in aluminum ions in high concentration.

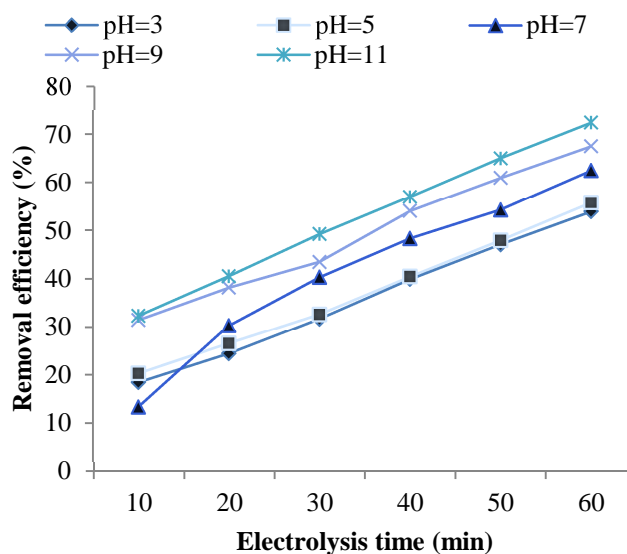
**Table 1. The ranges of experimental parameters**

pH	Voltage (v)	Dye concentration (mg/l)	Time of reaction (min)	KCl concentration (M)	electrode distance (cm)
3,5,7,9,11	10,20,30,40,50	25,50,75,100,150	10,20,30,40,50,60	0.0025,0.005,0.0075,0.01	1,2,3



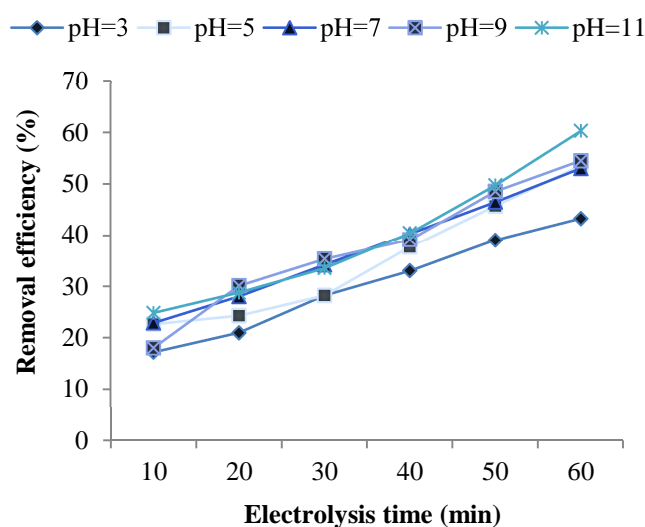
**Figure 2. Effect of initial pH on removal efficiency of Reactive Green 19 (RG-19) dye**

(Voltage = 10 v,  $C_0 = 50$  mg/l,  $d = 1$  cm, KCl = 0.005 M,  $T = 22 \pm 2$  °C)



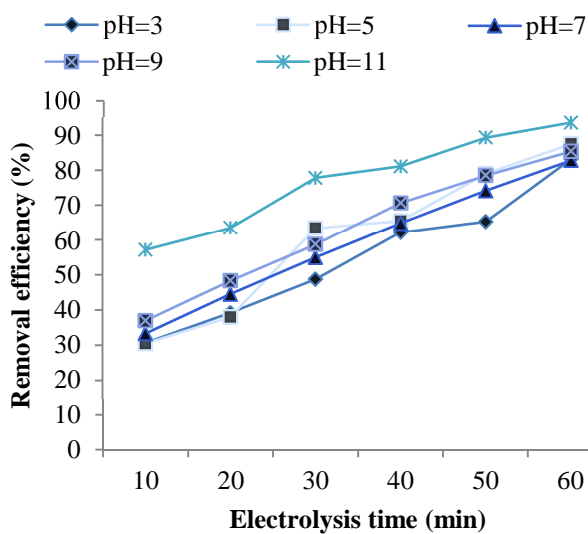
**Figure 4. Effect of initial pH on removal efficiency of Reactive Green 19 (RG-19) dye**

(Voltage = 30 v,  $C_0 = 50$  mg/l,  $d = 1$  cm, KCl = 0.005 M,  $T = 22 \pm 2$  °C)



**Figure 3. Effect of initial pH on removal efficiency of Reactive Green 19 (RG-19) dye**

(Voltage = 20 v,  $C_0 = 50$  mg/l,  $d = 1$  cm, KCl = 0.005 M,  $T = 22 \pm 2$  °C)



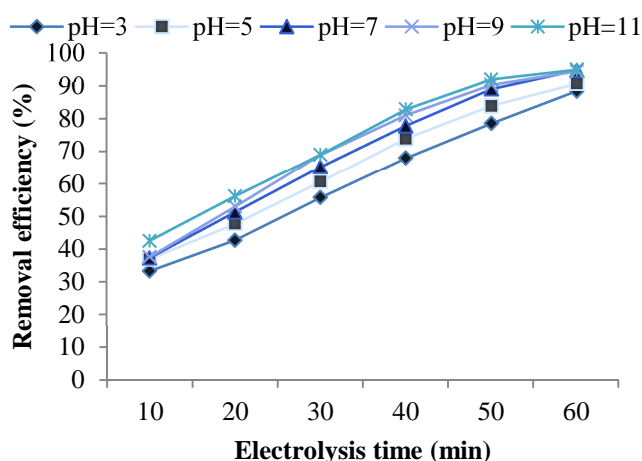
**Figure 5. Effect of initial pH on removal efficiency of Reactive Green 19 (RG-19) dye**

(Voltage = 40 v,  $C_0 = 50$  mg/l,  $d = 1$  cm, KCl = 0.005 M,  $T = 22 \pm 2$  °C)

At concentrations of higher than 50 mg/l (99.27%) the no reduction was observed in color removal rate and it was relatively constant. However, increasing initial dye concentration caused a steady increase in decolorization capacity.<sup>22</sup>

Moreover, up to the concentration of 50 mg/l, the energy consumption was constant, but above this amount, it increased. When initial concentration of the dye was raised from 25 to 150 mg/l at the electrolysis time of 60 minutes, the efficiency decreased from 99.88 to 99.56%.

On the contrary, the energy consumption increased from 10.25 to 13.18 kWh/(kg m<sup>3</sup>). Furthermore, the study by Daneshvar et al. has also proven the fact that an increase in initial concentration of dye can enhance energy consumption and increase dye removal from solution.<sup>23</sup>



**Figure 6. Effect of initial pH on removal efficiency of Reactive Green 19 (RG-19) dye**

(Voltage = 50 v, C<sub>0</sub> = 50 mg/l, d = 1 cm, KCl = 0.005 M, T = 22 ± 2 °C)

#### Effect of KCL concentration

The effect of KCl concentration on removal efficiency and electrode consumption has been shown in figures 7 and 8. With an increase in the concentration of KCl in solution, the conductivity of the solution and the current density also increased. The cell voltage decreased with increasing wastewater conductivity at constant current density.<sup>24</sup> According to the results, high color removal

with low energy consumption occurred at around 0.01 M of KCl, but electrode consumption ratio increased at this concentration. The results of the study by Sengil and Ozacar showed that high color removal percentage with low cell voltages and low energy consumption could be obtained in dye solutions containing around 3 g<sup>-1</sup> of NaCl.<sup>21</sup>

#### Effect of electrode distance

Figures 9 and 10 show the percentage of color removal against distance between the electrodes. The effect of inter-electrode distance on removal efficiency was examined at 1-3 cm. the efficiency decreased slightly, when the inter-electrode distance increased. The distance of 1 cm was selected as the optimum point. Moreover, low energy was consumed at this distance. The reason for this observation is thought to be the fact that when inter-electrode distance increases, the produced ions move more slowly in the solution. Hence, the voltage and energy consumption increase.<sup>24</sup>

### Conclusion

EC is one of the most effective techniques for removing color and organic pollutants from wastewater. In this research, the removal efficiency of EC process by means of aluminum electrodes for decolorization of RG-19 in batch system was studied. The results showed that the highest dye removal efficiency was 99.88%, at the optimized conditions of pH of 11, initial dye concentration of 25 mg/l, voltage of 50 v, electrolysis time of 60 minutes, and electrode

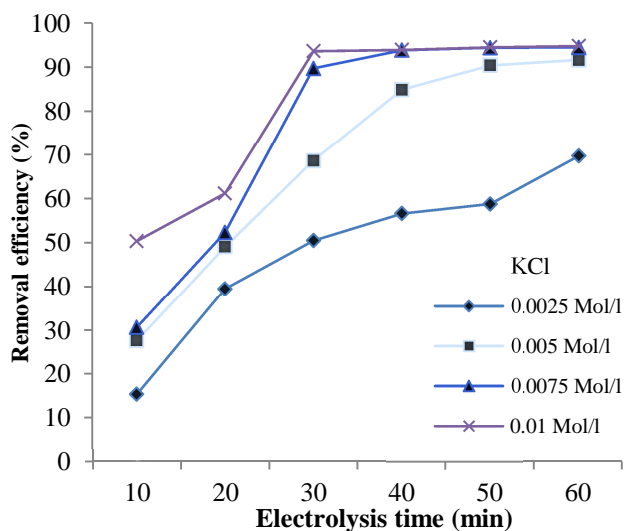
**Table 2. Effect of initial dye concentration on removal efficiency and energy consumption**

Time	Concentration of dye									
	25 mg/l		50 mg/l		75 mg/l		100 mg/l		150 mg/l	
	RE (%)	E (Kw.h/m <sup>3</sup> )	RE (%)	E (Kw.h/m <sup>3</sup> )	RE (%)	E (Kw.h/m <sup>3</sup> )	RE (%)	E (Kw.h/m <sup>3</sup> )	RE (%)	E (Kw.h/m <sup>3</sup> )
10	24.21	1.04	15.40	2.13	36.39	2.64	46.11	2.80	63.45	4.62
20	48.60	2.83	49.14	3.24	53.71	3.84	65.46	4.11	63.75	5.36
30	68.85	4.38	68.69	4.40	80.08	4.46	81.42	5.29	66.89	7.33
40	92.10	5.92	84.91	6.17	98.48	7.33	94.71	6.53	91.77	9.75
50	95.68	7.38	90.44	7.93	99.51	9.06	99.49	9.94	99.50	11.08
60	99.88	10.25	91.66	10.27	99.62	12.41	99.57	11.87	99.56	13.18

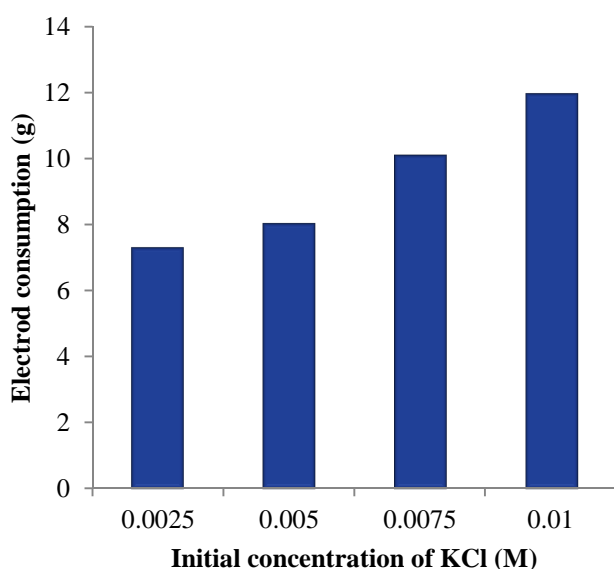
RE: Removal; E: Energy Consumption



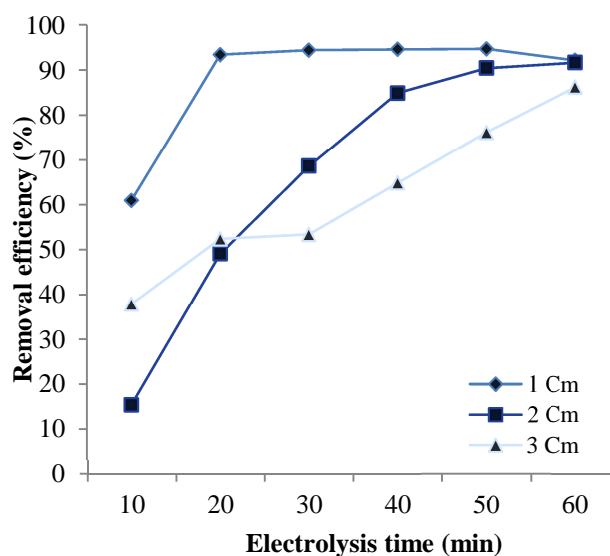
distance of approximately 1 cm. The removal efficiency increased with the increase in inter-electrode distance to 1 cm and KCl concentration to 0.01 M. It can be concluded that the EC process by aluminum electrodes is an efficient method for reactive dye removal from colored solutions.



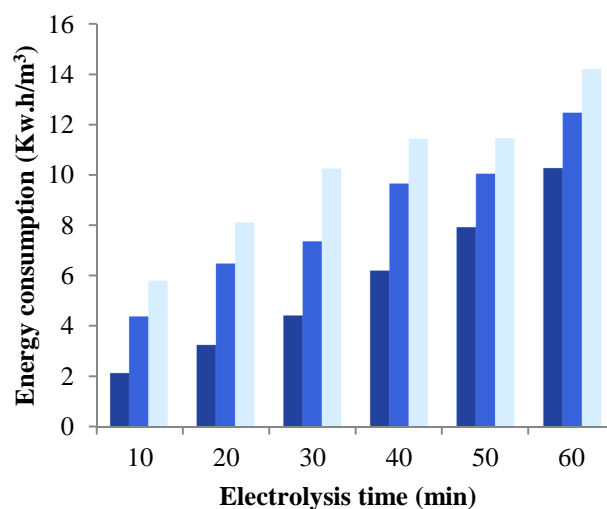
**Figure 7. Effect of KCl concentration on the removal efficiency of Reactive Green 19 (RG-19) dye** (Voltage = 50 v,  $C_0 = 50$  mg/l,  $d = 1$  cm, pH = 11,  $T = 22 \pm 2$  °C)



**Figure 8. Effect of KCl concentration on electrode consumption** (Voltage = 50 v,  $C_0 = 50$  mg/l,  $d = 1$  cm, pH = 11,  $T = 22 \pm 2$  °C)



**Figure 9. Effect of distance between electrodes on removal efficiency of Reactive Green 19 (RG-19) dye** (Voltage = 50 v,  $C_0 = 50$  mg/l, KCl = 0.01 M, pH = 11,  $T = 22 \pm 2$  °C)



**Figure 10. Effect of distance between electrodes on energy consumption** (Voltage = 50 v,  $C_0 = 50$  mg/l, KCl = 0.01 M, pH = 11,  $T = 22 \pm 2$  °C)

### Conflict of Interests

Authors have no conflict of interests.

### Acknowledgements

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