



## Research Article

# Factorial experimental design for removal of Indigo Carmine and Brilliant Yellow dyes from solutions by coagulation

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## ABSTRACT

Textile and food industries produce huge amounts of wastewaters containing dye residues. When these wastewaters are discharged to receiving surface waters like as lakes and rivers, aesthetically unpleasant situations form. Therefore, these wastewaters should be treated. Wastewater treatment is sometimes an expensive operation and cheap methods should be developed. The removal of Indigo Carmine (I.C., Acid dye) and Brilliant Yellow (B.Y., Azo dye) from synthetically prepared solutions was studied by coagulation using iron chloride salt in a batch reactor at room temperature. As an experimental approach, two leveled factorial design with three factors was applied as a function of pH (4–12), iron chloride amount (0.1–0.4 g/500 mL) and dye concentration (100–200 mg/L). Low pHs supported to removal of these two dyes. The results showed that 100% I.C. dye removal and 90.5% B.Y. dye removal were achieved. The all parameters were statistically insignificant for both the dyes. Indigo Carmine and Brilliant Yellow dyes were removed from solutions successfully. The applied treatment method was evaluated as promising due to low sludge production, low cost, low coagulation duration and high performance. A time span of 5 minutes was found as enough for removals of both of the dyes. After treatment of I.C. and B.Y. dyes by coagulation, the coagulated dyes were determined as un reusable due to iron complex by these dyes. Flocculation was found to be ineffective. A continuous flow reactor was successfully adopted for these dyes.

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## INTRODUCTION

Textile and food industries use many different types of dyes to color their products [1]. Approximately 1.6 million tons of textile dyes are produced yearly throughout the World and 10–15% of this volume is scrapped as wastewater [2]. The wastewaters of textile industries contain dense color, heavy metals, high COD and BOI, suspended solid materials and salt at high concentration [3]. Some textile dyes are toxic for humans and animals [4]. The receiving mediums like as lakes and rivers are badly affected by these dye wastewaters as dyes aesthetically ruin water view, inhibit the ox-

xygen and the sunlight penetration into receiving water, and thus dyes prevent the photosynthesis activity in the water body [5]. Therefore, food coloring and textile industries wastewaters should be treated by applying one of the following methods like as adsorption, coagulation, electrocoagulation, biosorption, membrane separation and advanced oxidation. An adsorption method can be applied effectively, if the adsorbent material is regenerable and has a high adsorption capacity. Adsorption is a remarkable method due to its simple and low cost application and high efficiency [2]. In electrocoagulation treatment of dyes, iron or aluminum plates are used. An electrocoagulation method works

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by electrical current transfer from a cathode plate to an anode plate which is dissolved to produce coagulating cations like as iron and aluminum for removals of dye molecules [6, 7]. Dye molecules are coagulated by dissolved cations and also converted to  $H_2O$  and  $CO_2$  in an electrocoagulation reactor by electrical deterioration of organic dye [7]. Electrocoagulation produces high iron or aluminum sludge at high current densities and long operation durations. Similar to adsorption, biosorption of dyes is suitable in the usage of the regenerable biosorbent with high capacity. Also, surface functional groups of biosorbent play an important role for dye adsorption and adsorbent regeneration [8]. Further, the surface area of the biosorbent as well as the other key point for dye adsorption [9]. Membrane separation is an appropriate process, if the pore diameter of the used membrane is smaller than molecule size of dye [10]. An electrooxidation process works by electrical current transportation from a cathode plate to an anode plate and has a high dye shredding performance to  $H_2O$  and  $CO_2$  [7]. The difference between electrooxidation and electrocoagulation is the absence of coagulating cation and is that the plates in electrooxidation are indissoluble. The plates in electrooxidation can be graphite, boron-doped diamond, and rubeidum-coated titanium or others. A coagulation process forms from rapidly dispersion of coagulant and the flocculation [11]. In dye removal by coagulation using iron or aluminum chloride salts, the cations aggregate dye molecules and enable to filtration, sedimentation or centrifugation of these flocks. A coagulation process for dye removal is the more advantageous and cheap technology because aluminum or iron salts are cheap and vast in marked. Also, there would not be any important sludge amount. In addition to this, coagulation-flocculation of dyes by these harmless cations is generally a rapid process. Therefore, the reactor volume for dye removal by coagulation and flocculation process would be small. But, dye recovery after the coagulation process may be impossible due to iron or aluminum complexion with treated dye. Brilliant Yellow dye is an anionic azo dye that can be used in the electrochemical synthesis of polypyrrol films and to dye wool, nylon, and silk fibers [12]. Indigo Carmine dye is a colorant mainly used in the pharmacy, textile, leather, and food industries [13].

Optimization is an useful approach for treatment of wastewaters by applying one of the methods like as taguchi, response surface, artificial neural network and factorial design [14]. In this study, the factorial design method was applied. Factorial optimization design is sometimes used to be initial stage for central composite design. If the experimental matrix limits are not certain for factorial design, it is necessary to carry out several preliminary experiments to determine the experimental matrix. When this situation is required in factorial optimization method, the most suitable way of determination of experimental matrix is the classical single parameter experiments. However, in other optimization experiments (Central composite design and Taguchi), the optimum condition can be determined in a wider cage and in these statistical methods, experimental numbers are too than the factorial design [14]. Thus, the factorial design is

advantageous. This applied coagulation process using iron chloride is a cheap method for dyes (Indigo Carmine and Brilliant Yellow) removal from the textile and food industries wastewaters. In a study, Disperse Blue 106, Disperse Yellow 54, Reactive Blue 49, Reactive Yellow 84 dyes removals were studied using ferric chloride and optimum pHs were determined as 6, 5, 6, 6, respectively [15]. Optimum  $FeCl_3 \cdot 6H_2O$  dosages for these dyes were reported between 1 and 2 mM [15]. In another study, Acid Black 1, Acid Violet 5, Reactive Black 5 dyes removals were reported to be increase at 35–50 mg/L chitosan concentrations in coagulation treatment [16]. Shi et al. [17] studied the removal of Direct Black 19, Direct Red 28, Direct Blue 86 dyes by different aluminum species and removals of dyes increased by increasing amounts of aluminum species which were  $AlCl_3$ ,  $PACl$  and  $Al_{13}$ .

In this study, Brilliant Yellow (B.Y., Azo dye) and Indigo Carmine (I.C., Acid dye) removals by iron chloride coagulation in a batch reactor applying  $2^3$  factorial design were investigated at room temperature. The applied experimental parameters were pHs (4 and 12), concentrations (100 and 200 mg/L) and iron chloride dosages (0.1 and 0.4 g/500 mL). The advantages and selection reasons of the applied coagulation method from other methods can be summarized as follows. The coagulation method is more effective than adsorption, electrocoagulation, and biosorption because the produced sludge is low. The coagulation method is superior to electrocoagulation due to zero electrical requirements. Membrane separation has an electrical cost for pressure pump operation and membrane fouling is the drawback. Electrooxidation is a very effective technology for dye removal from wastewaters but the residual persistent dye by-products are still a very important problem for treated water discharge. Therefore, iron coagulation of Brilliant Yellow and Indigo Carmine dyes is very advantageous due to the low cost of operation, the easily handling of process and the low operation duration. In addition to batch reactor, a continuous reactor was applied to these two dyes. The I.C. is used in the food and textile industries, and B.Y. is used in the textile industry, therefore, their removals were selected.

## MATERIALS AND METHODS

### pH Effect Experiments

Experiments for pH effect were carried out in a temperature controlled incubator shaker. For this purpose, 50 mL solutions with 100 mg/L concentrations were adjusted to 4, 6, 8, 10 and 12 pHs and 0.01 g  $FeCl_3$  was added to each 100 mL polyethylene bottles with 50 mL dye solution. The other parameters were 20 °C, 5 minutes agitation time and 100 rpm agitation speed. These experiments were carried out for both Indigo Carmine and Brilliant Yellow dyes. After coagulation, 10 mL dye solutions were filtered with Whatman filter paper and measured by using UV-vis spectrophotometer at 610 nm for Indigo Carmine dye and 400 nm for Brilliant Yellow dye. The reason of selection 100 mg/L concentration was the concentration range of 100 and 200 mg/L in factorial design.

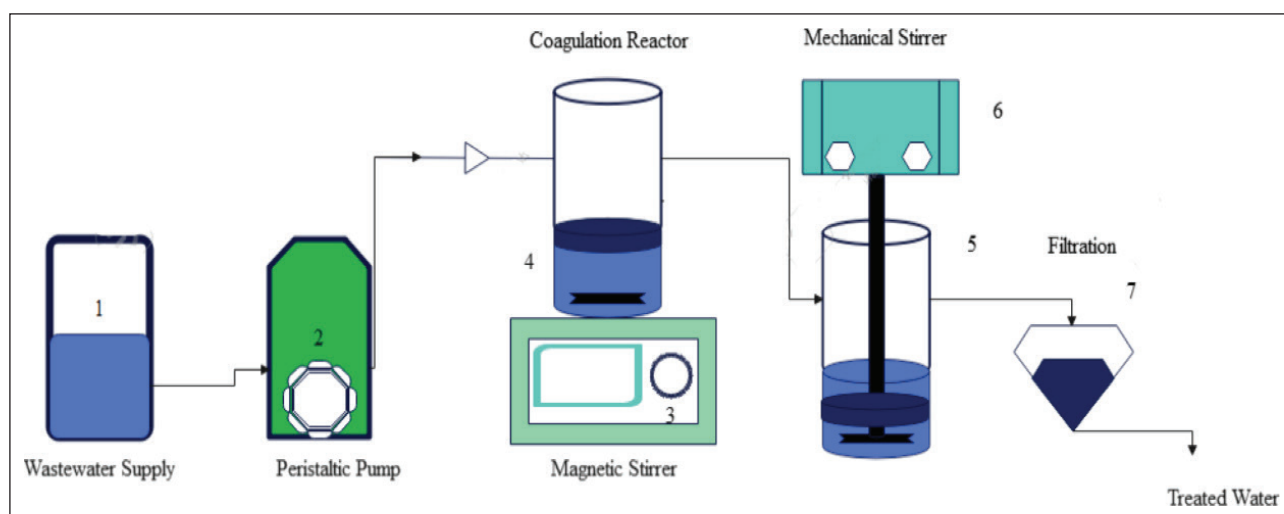


Figure 1. Total Experimental setup.

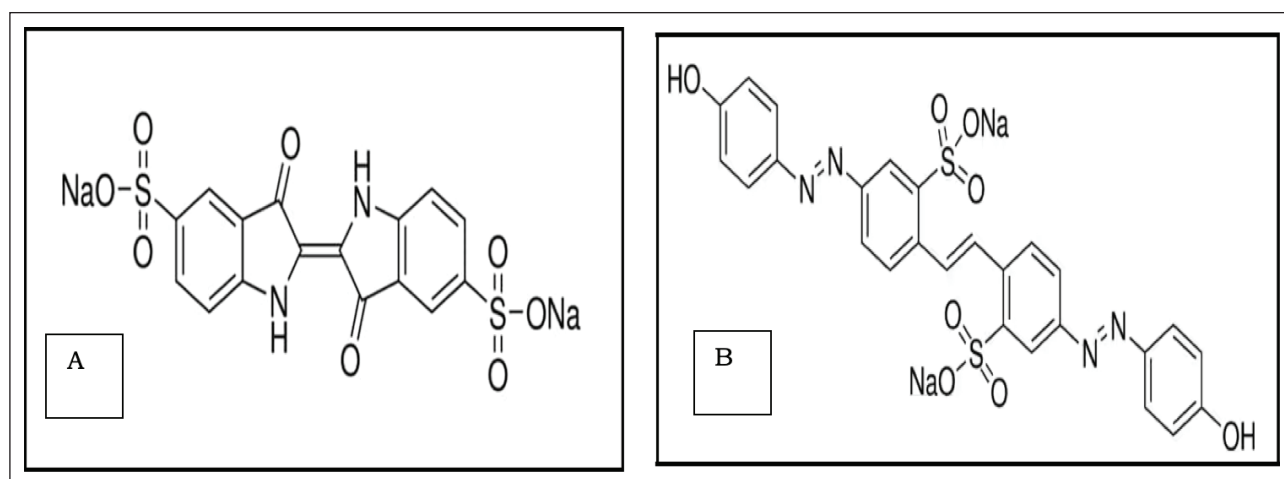


Figure 2. (a) Chemical structure of Indigo Carmine dye. (b) Chemical structure of Brilliant Yellow dye.

**Factorial Experimental Design Experiments for Coagulation of Dyes**

In this study, Indigo Carmine and Brilliant Yellow dyes removals by coagulation using iron chloride were studied. The experimental setup is given in Figure 1. The batch coagulation reactor had 1.3 L volume and was filled with 500 mL dye solution. In this study, only coagulation was applied and flocculation was omitted. The coagulation reactor was stirred at 1,000 rpm stirring rate by a magnetic stirrer. In batch experiments, 2<sup>3</sup> factorial design was applied for pH, concentration and iron chloride dosage. Parameters were iron chloride dosage (0.1–0.4 g/500 mL), concentration (100–200 mg/L), and pH (4–12). Firstly, dye concentrations of working solutions with 500 mL volume were prepared from 2,000 mg/L stock dye solution (I.C. and B.Y.). pHs of solutions were adjusted and iron chloride salt solution at predetermined volume was added from 10 g FeCl<sub>3</sub>/250 mL stock solution to the working solutions. The reactor content was stirred during 5 minutes and a volume of 10 mL of treated solution was pipetted by automatic pipette and filtered by Whatman filter paper. The filtered solution was diluted and its absorbance value was measured by a spectrophotometer (UV-visible) at 610 nm

for I.C. and at 400 nm for B.Y. The diluted solutions had absorbance values within calibration curve. The experimental error due to dilution was around 3% in removal percentages. The experiments were carried out according to optimization experimental matrix parameters determined by Minitab 16.0 software. The dye chemical structures are given in Figure 2. The iron chloride salt had 98% purity (Merck product). The solution temperatures were 20 °C throughout the experiments. The pure water was used in preparation of dye solutions. Calibration curves of dyes (I.C. and B.Y.) were prepared within 0–10 mg/L concentrations. B.Y. dye had 70% purity and this ratio was taken into consideration in dye concentration calculation. I.C. dye had analytical grade.

**Flocculation Experiments**

Solutions with 200 mg/L concentrations for B.Y. and I.C. dyes were treated with 0.4 g FeCl<sub>3</sub> in a batch reactor. Coagulation experiments were carried out in a batch reactor and then flocculation experiments were carried out in a Jar test device. The other parameters were kept as constant at 20 °C, 1,000 rpm, 500 mL and natural pHs for coagulation. The flocculation experiments were carried out at 20

**Table 1.** The intervals of the factors used in optimization

Dye type	Parameter	Abbreviation	Low level (-1)	High level (1)
Indigo Carmine	pH	pH	4	12
	Coagulant Dosage (g/500 mL)	CD	0.1	0.4
	Concentration (mg/L)	C	100	200
Brilliant Yellow	pH	pH	4	12
	Coagulant Dosage (g/500 mL)	CD	0.1	0.4
	Concentration (mg/L)	C	100	200

**Table 2.** Experimental matrix for dye removal

Trial	pH	(CD, g/500 mL)	(C, mg/L)	I.C. (%)	I.C. model (%)	B.Y. (%)	B.Y. model (%)
1	-1	-1	-1	79.69	86.86	82.14	83.52
2	-1	-1	1	77.73	70.36	90.50	87.12
3	-1	1	-1	100.00	92.62	88.10	84.72
4	-1	1	1	95.31	102.02	79.17	80.56
5	1	-1	-1	47.66	40.48	58.33	56.94
6	1	-1	1	6.64	14.02	55.36	58.74
7	1	1	-1	12.50	19.88	34.52	37.90
8	1	1	1	26.95	19.78	33.33	31.94

CD: Coagulant dosage, C: Concentration.

°C, 60 rpm, natural pHs. After treatment of dye solutions with iron chloride, 500 mL solutions were flocculated and filtered with Whatman filter paper. Because the gathered flocks should be dewatered by filtration.

Dye removal percentages of I.C. and B.Y. were calculated using following equation.

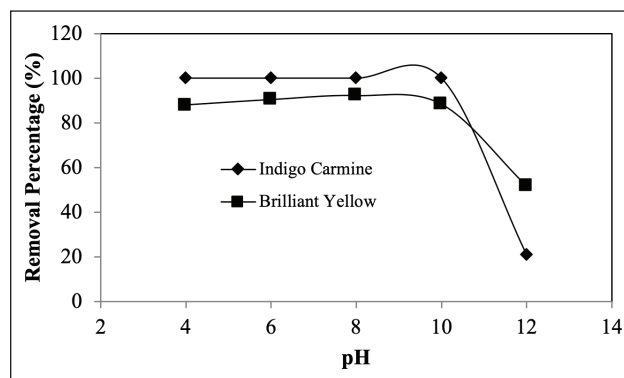
$$\eta = \left( \frac{C_0 - C_t}{C_0} \right) \times 100 \quad (\text{Eq. 1})$$

Here,  $\eta$  is removal percentage (%),  $C_0$  is initial dye concentration (mg/L),  $C_t$  is treated water dye concentration (mg/L).

## RESULTS AND DISCUSSION

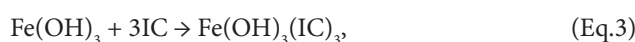
### pH Effect

The effect of pH was studied at range of 4–12 for Indigo Carmine and Brilliant Yellow dyes removal by coagulation using iron chloride salt. Indigo Carmine and Brilliant yellow dyes removal as a function of pH were carried out at 100 rpm agitation speed, 20 °C, 50 mL solution volume, 100 mg/L dye concentration, 5 minutes coagulation time, 0.01 g/50 mL iron chloride dosage. The removal was carried out in an incubator shaker. The results are given in Figure 3. The results showed that I.C. and B.Y. dyes removals were constant at 100% and 90 % removal yields from pH 4 to pH 10, respectively, but dyes removals decreased at pH 12. This result can be related with competitive adsorption of anionic dyes and hydroxyl ions for iron cation and iron hydroxyl surface at pH 12. Similar results were reported for anionic dyes adsorption onto coffee waste [18]. In another



**Figure 3.** pH effect on removal of Indigo Carmine and Brilliant Yellow dyes from water (100 rpm, 20 °C, 50 mL, 100 mg/L dye, 5 minutes, 0.01 g FeCl<sub>3</sub>/50 mL).

study, the anionic dyes removals by spent mushroom waste were decreased at high pHs [19]. The removal mechanism of I.C and B.Y. dyes by iron cation and iron hydroxide can be summarized as follows.

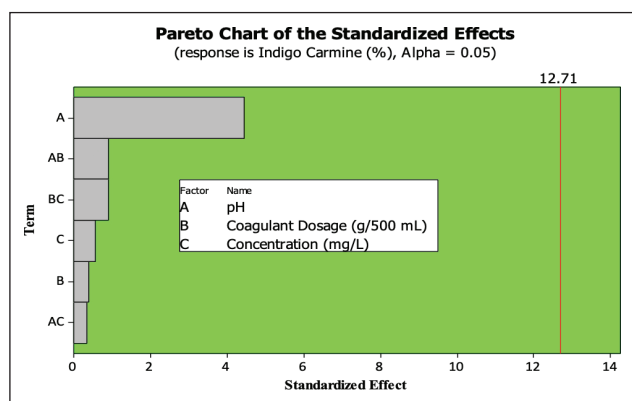


### Factorial Optimization Design of Experiments

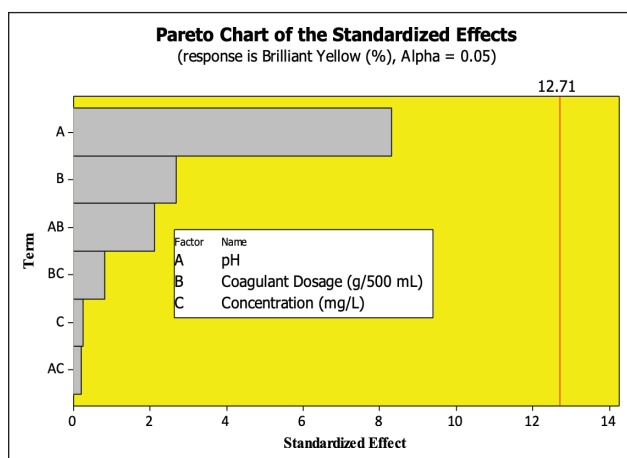
Science and industry sometimes use optimization methods to upgrade their outputs. Optimization of experiments generally reduces the experiment number. There

**Table 3.** Analysis of ANOVA

Term	Indigo Carmine			Brilliant Yellow		
	Coefficient	T-value	p	Coefficient	T-value	p
Constant	55.81	7.67	0.083	65.18	27.34	0.023
pH	-32.37	-4.45	0.141	-19.80	-8.30	0.076
Coagulant Dosage (CD)	2.88	0.40	0.760	-6.40	-2.69	0.227
Concentration (C)	-4.15	-0.57	0.670	-0.59	-0.25	0.845
pH-CD	-6.59	-0.91	0.531	-5.06	-2.12	0.280
pH-C	-2.49	-0.34	0.790	-0.45	-0.19	0.882
CD-C	6.59	0.91	0.531	-1.94	-0.81	0.565



**Figure 4.** Pareto chart of analysis (Indigo Carmine dye).



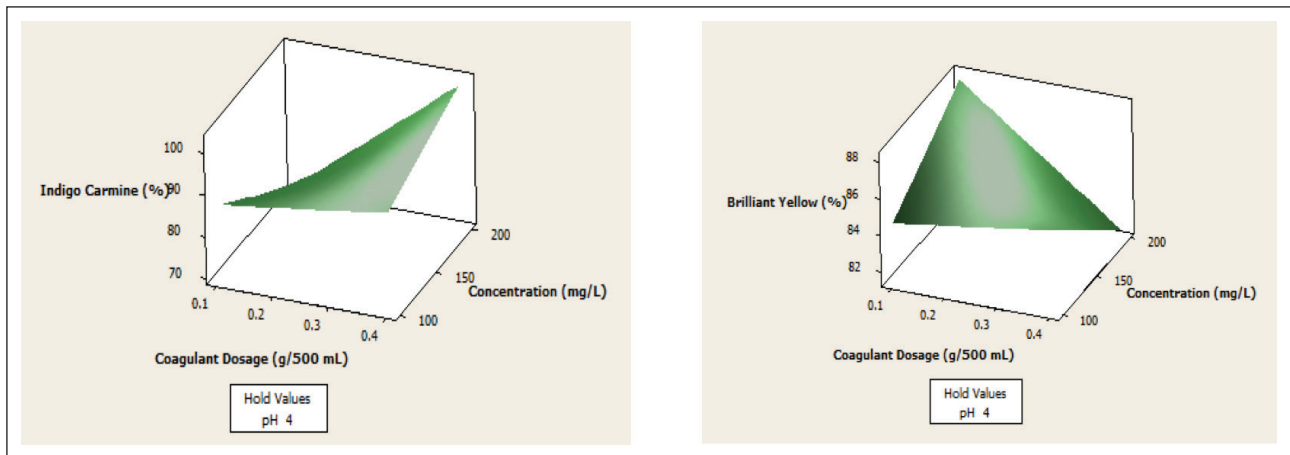
**Figure 5.** Pareto chart of analysis (Brilliant Yellow dye).

are several optimization methods like as response surface, taguchi, full factorial design, artificial neural network and traditional single parameter optimization. The central composite design tool of response surface method requires more experimental runs containing the matrix conditions of factorial design. If the limits of parameters matrix conditions are unknown, the analysis of central composite design would become more complicated and effort requiring and costly. Traditional single parameter optimization analyzes data separately, and therefore it is not advantageous. Therefore, the factorial design is advantageous than single parameter experiments and central composite design. A factorial experimental design was applied to Indigo Carmine and Brilliant Yellow dyes removals. For analysis of main and interaction effects of parameters, the experimental matrix of the factorial design should be determined before. For the present study, the effect of parameters such as pH, coagulant dosage and concentration were optimized by 2<sup>3</sup> factorial design using Minitab 16.0 software. In this study, the response for the analysis and model development was selected as removal percentage. The p values (confidence constants) were used as control parameter to check the reliability of the developed statistical model. When the p value is small from confidence level, the corresponding parameter or interaction were determined as statistically important [20, 21]. The confidence level was selected as 95% for the present study. An example of the general regression model equation for the factorial design can be given as follows.

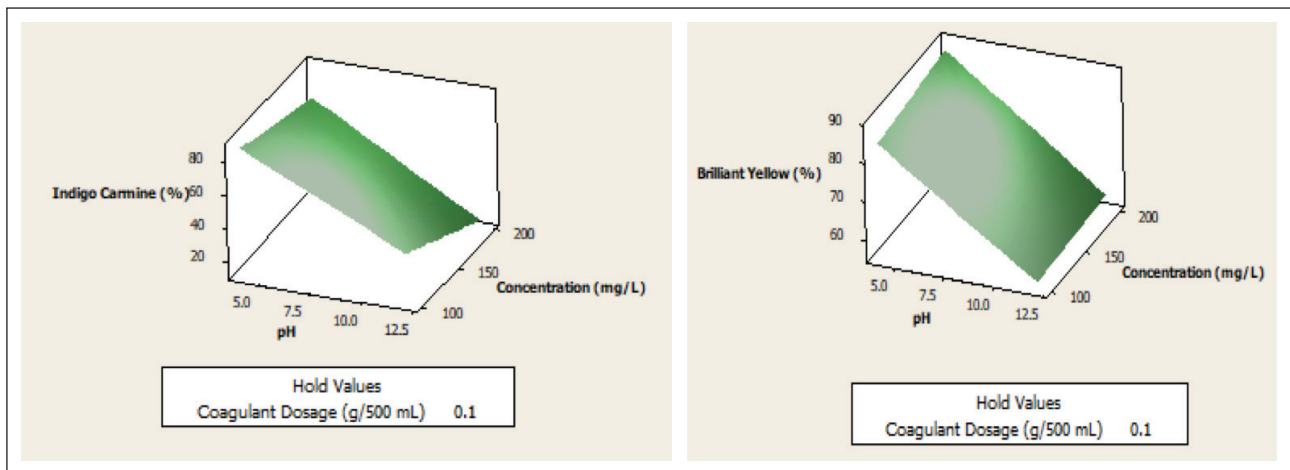
$$\text{Removal (\%)} = b + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_1X_2 + b_5X_1X_3 + b_7X_2X_3 + b_8X_1X_2X_3 \quad (\text{Eq.6})$$

Here; b, b<sub>1</sub>, b<sub>2</sub>, b<sub>3</sub>, b<sub>4</sub>, b<sub>5</sub>, b<sub>7</sub>, b<sub>8</sub> are model constants and X<sub>1</sub>, X<sub>2</sub>, X<sub>3</sub> are coded factors representing pH, coagulant dosage, concentration. X<sub>1</sub>X<sub>2</sub>, X<sub>1</sub>X<sub>3</sub>, X<sub>2</sub>X<sub>3</sub>, X<sub>1</sub>X<sub>2</sub>X<sub>3</sub> are their interactions.

The low and high levels of parameters are given in Table 1. The low and high levels of coagulant dosage (iron chloride) were 0.1 and 0.4 g/500 mL, the low and high levels of pH were 4 and 12 and the low and high levels of concentration were 100 and 200 mg/L. The factorial matrix of experimental design is given in Table 2. The ANOVA analysis (student-t test and confidence levels, p) was performed and was given in Table 3. The confidence limit value (p) for the main and the interaction effects of the parameters was selected as 95%. The maximum dye removal was obtained as 100% in I.C. for pH (4), 0.4 g/500 mL coagulant dosage, 100 mg/L dye concentration. The maximum dye removal was obtained as 90% in B.Y. for pH (4), 0.1 g/500 mL coagulant dosage, 200 mg/L dye concentration. All the parameters were found as statistically insignificant (p values above 0.05). Pareto charts of removals are given in Figures 4 and 5 showing parameters effect above significance. As can be seen in Figures 4 and 5, the all parameters were determined as above confidence values because the parameters lines below the limit value of 12.71. This nu-



**Figure 6.** Concentration-coagulant dosage effect on Indigo carmine and Brilliant yellow removal.



**Figure 7.** pH-concentration effect on Indigo carmine and Brilliant yellow removal.

merical value is determined by the Minitab 16.0 software. Experimental matrix is given in Table 2. As can be seen in Table 2, I.C. dye removal is changed from 6.64 to 100% and B.Y. dye removal is changed from 33.33 to 90.50%. For Indigo Carmine dye, correlation coefficients were calculated as  $R\text{-Sq}=95.66\%$ ,  $R\text{-Sq}(\text{predicted})=0.00\%$ ,  $R\text{-Sq}(\text{Adjusted})=69.62\%$ . For Brilliant Yellow dye, correlation coefficients were calculated as  $R\text{-Sq}=98.79\%$ ,  $R\text{-Sq}(\text{predicted})=22.51\%$ ,  $R\text{-Sq}(\text{Adjusted})=91.52\%$ . According to model estimation values of removal percentages, the dye removal percentages were well recalculated and given in Table 2. The Anova analysis of data was done according to coded factors. The  $R\text{-Sq}$  value for Indigo Carmine dye was calculated as 95.66% and the  $R\text{-Sq}$  value for Brilliant Yellow dye was calculated as 98.79%. These results show that the developed regression models estimated the data at enough degree and these conclusions were corrected with recalculated data given in Table 2. The statistically insignificant parameter effects are thought that the factorial design assumes the linear lines in two values of one factor and there is a slope. For instance, pH shows very different slopes for all parameter interactions like as pH-concentration, pH-dosage and these different slopes can cause to statistically insignificant ( $p$ ) values. Also, these results were

related with selection of confidence limit value in Minitab 16.0 software. In this study, the confidence value was selected as 95% and it can be selected to be less. In a study, the confidence value was reported as 90% [14] and in another study, the confidence value was reported as 88% [22]. In Table 3, while the negative coefficients indicate the decreasing effect on data calculation by regression model, the positive coefficients indicate the increasing effect on data calculation by regression model.

The regression models were developed as follows.

$$\text{I.C. removal (\%)} = 55.81 - 32.37 \cdot \text{pH} + 2.88 \cdot \text{CD} - 4.15 \cdot \text{C} - 6.59 \cdot \text{pH} \cdot \text{CD} - 2.49 \cdot \text{pH} \cdot \text{C} + 6.59 \cdot \text{CD} \cdot \text{C} \quad (\text{Eq. 7})$$

$$\text{B.Y. removal (\%)} = 65.18 - 19.80 \cdot \text{pH} - 6.40 \cdot \text{CD} - 0.59 \cdot \text{C} - 5.06 \cdot \text{pH} \cdot \text{CD} - 0.45 \cdot \text{pH} \cdot \text{C} - 1.94 \cdot \text{CD} \cdot \text{C} \quad (\text{Eq. 8})$$

#### Concentration-Coagulant Dosage Effect on Indigo Carmine and Brilliant Yellow Removal

The results are given in Figure 6. For Indigo Carmine dye removal, high concentration and high coagulant dosage increased the removal and maximum removal was obtained as 100%. For Brilliant Yellow dye removal, maximum removal was obtained at high concentration and low coagulant dosage and maximum removal was obtained as

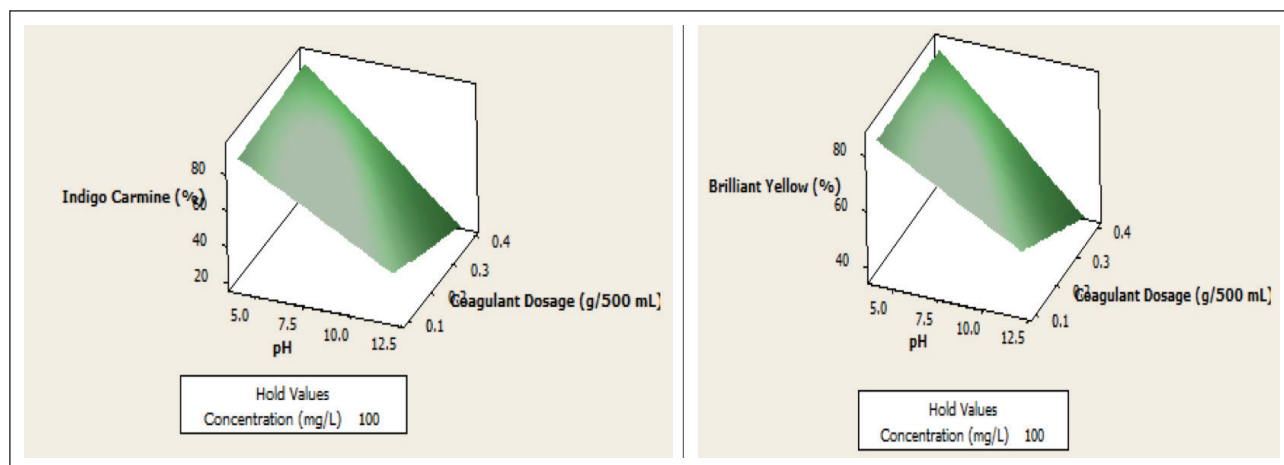


Figure 8. pH-coagulant dosage effect on Indigo Carmine and Brilliant Yellow removal.

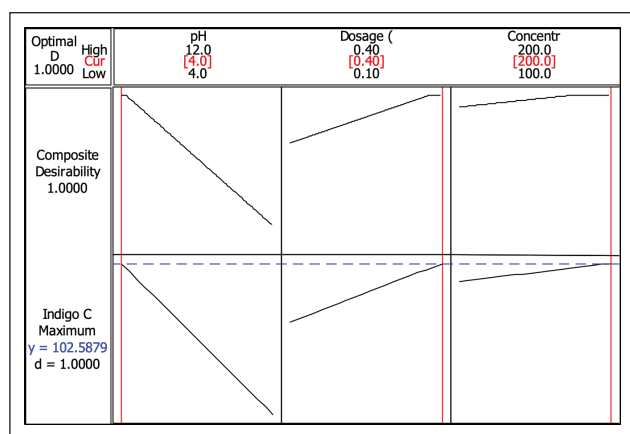


Figure 9. Optimization graphs of Indigo Carmine dye.

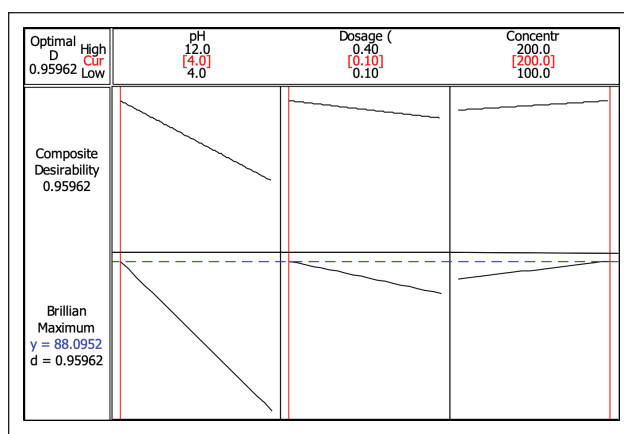


Figure 10. Optimization graphs of Brilliant Yellow dye.

87%. In Indigo Carmine removal, the reason of maximum removal at high concentration and high coagulation dosage was increasing active site number for increasing dosage and driving force of high concentration [9]. The maximum removal reason at low coagulant dosage and high concentration was probably the equilibrium between dye and coagulant for Brilliant Yellow [23]. In different studies, iron chloride dosages were applied as 100 mg/L and 5.6 mg/L for dye removals [24, 25]. Dye concentration was selected as 50 mg/L in a study [25].

### pH-Concentration Effect on Indigo Carmine and Brilliant Yellow Removal

The results are given in Figure 7. The Indigo Carmine dye removal increased at low concentration and low pH. The brilliant yellow dye removal increased at high concentration and low pH. At low pHs, the Indigo Carmine dye adsorption was realized at more positive surface of iron hydroxide and iron cations [9]. Also, low concentration of Indigo Carmine dye increased the removal percentage due to high surface of coagulant for decreasing concentration [9]. Low pHs for Brilliant Yellow dye solution increased the positive surface charge of iron hydroxide and iron cations for more removal [9]. High concentration of Brilliant Yellow dye increased the removal due to high driving force [23].

### pH-Coagulant Dosage Effect on Indigo Carmine and Brilliant Yellow Removal

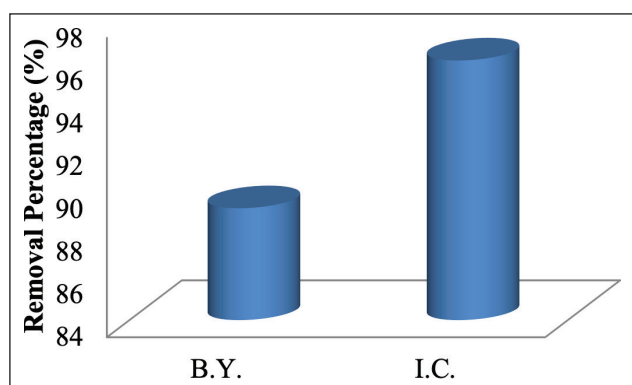
The results are given in Figure 8. Indigo Carmine dye removal increased at low pHs and constant coagulant dosages. Brilliant Yellow dye removal increased at low pHs and constant coagulant dosage. While low pHs increase the positive surface charge of coagulants for adsorption of both of the dyes, constant coagulant effect was related with solid-to-solution ratio [9, 22].

### Optimization of Indigo Carmine Dye and Brilliant Yellow Dye Removal by Coagulation

Indigo Carmine and Brilliant Yellow dyes removals by iron coagulation were optimized and given in Figures 9 and 10. Maximum removals of I.C. and B.Y. dyes were selected from experimental matrix as 100% and 90.4%, respectively. The removal of I.C. dye at 100% extent was obtained at conditions of pH (4), dosage (0.4 g/500 mL) and concentration (200 mg/L). The removal of B.Y. dye at 88.1% extent was obtained at conditions of pH (4), dosage (0.1 g/500 mL) and concentration (200 mg/L). Optimization graphs of dyes are given in Figures 9 and 10.

### Mechanism of Removal of Indigo Carmine and Brilliant Yellow Dyes

Both the dyes have dissociable sodium cation releasing to solution after dissolution by water. Sulfite groups of both of the



**Figure 11.** Brilliant Yellow and Indigo Carmine removal after 3.5 hydraulic retention time (The exit dye was collected completely in a baker and the color value was measured as was).

dyes molecules produce negative molecule tips that can form flocks by iron (III) cations. Iron cations combined with the three dye molecules for both of the dyes and long chains for coagulated flocks formed. This formed dye-iron (III) bonds were thought as to be partial ionic and partial covalent bonds between oxygen atoms of sulfite groups and iron (III) ions. These bonding types are depended on bond energies [26].

#### Continuous Reactor Application for Brilliant Yellow and Indigo Carmine Dye Removal

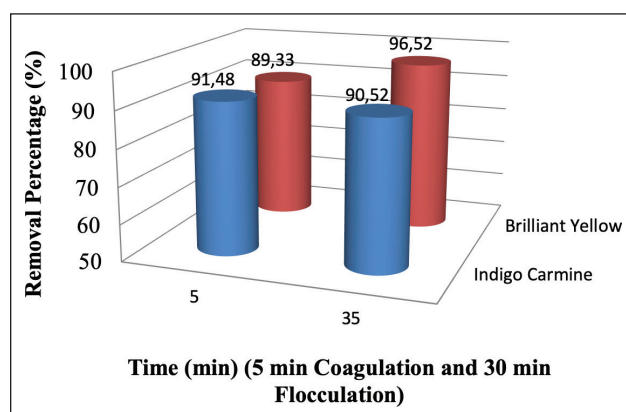
Brilliant Yellow and Indigo Carmine dyes removals were studied in a continuous reactor with 100 mL volume. In the experiments, natural pH, 1,000 rpm stirring speed, 20 °C, per 5 minutes 0.04 g FeCl<sub>3</sub> dosage, 400 mL dye reservoir volume and 10.084 mL/min volumetric flow rate were applied. The results are given in Figure 11. As can be seen in Figure 11, I.C. dye removal was 96% and B.Y. dye removal was 89%. These results were accordance with the results of factorial design but, B.Y. dye removal was found as high. These results showed that the batch coagulation reactor could be operated as successfully by converting to the continuous reactor. The results belong to total collected 400 mL treated water (3.5 hydraulic retention time) for B.Y. and I.C. dyes, separately.

#### Flocculation Removal of Indigo Carmine and Brilliant Yellow Dyes

After coagulation of Indigo Carmine and Brilliant Yellow dyes, flocculation of these dyes by iron cations and iron hydroxides were realized at 20 °C, 60 rpm stirring speed, 500 mL volume and 0.4 g FeCl<sub>3</sub>/500 mL. After flocculation operation, I.C. dye removal remained as the same with coagulation, but B.Y. dye removal increased at about 7%. B.Y. dye adsorption on Whatman filter paper was measured below 5%. Therefore, flocculation for these two dyes was ineffective. I.C. and B.Y. dyes removals were 90.52 and 96.52, respectively. Results are given in Figure 12.

#### CONCLUSION

Indigo Carmine (I.C.) and Brilliant Yellow (B.Y.) dyes removals were investigated using 2<sup>3</sup> factorial design. Optimum conditions for I.C. were determined as 100 mg/L



**Figure 12.** Coagulation and flocculation effect for removal of dyes.

concentration, 0.4 g/500 mL coagulant dosage and pH (4). Optimum conditions for B.Y. were determined as 200 mg/L concentration, 0.1 g/500 mL coagulant dosage and pH (4). Maximum removal for I.C. and B.Y. were obtained as 100% and 90.5%, respectively. The removal of both of dyes by iron coagulation was found as successful. The all parameters were insignificant in respect to statistic for both the dyes.

- For Indigo Carmine dye removal, high concentration and high coagulant dosage increased the removal and maximum removal was obtained as 100%. For Brilliant Yellow dye removal, maximum removal was obtained at high concentration and low coagulant dosage and maximum removal was obtained as 87%.
- The Indigo Carmine dye removal increased at low concentration and low pH. The Brilliant Yellow dye removal increased at high concentration and low pH.
- Indigo Carmine dye removal increased at low pHs and constant coagulant dosage. Brilliant Yellow dye removal increased at low pHs and constant coagulant dosage.
- Optimization graph was designed by selection maximum removal values of 100 and 90.4% for I.C. and B.Y. dyes, respectively and desirability values were obtained as 1 and 0.95962, respectively.
- After treatment of 400 mL Brilliant Yellow and Indigo Carmine dyes in a 100 mL continuous reactor, removal percentages for dyes were calculated as 89 and 96%, respectively.
- Flocculation was determined as ineffective.

#### DATA AVAILABILITY STATEMENT

The author confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

#### CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.



## USE OF AI FOR WRITING ASSISTANCE

Not declared.

## ETHICS

There are no ethical issues with the publication of this manuscript.

## REFERENCES

- [1] X. R. Xu, H.B. Li, W.H. Wang, and J.D. Gu, “Degradation of dyes in aqueous solutions by the Fenton process,” *Chemosphere*, Vol. 57(7), pp. 595–600, 2004. [CrossRef]
- [2] B. Erdem, S. B. Avşar, S. Erdem, and N. Tekin, “Adsorption of light green and brilliant yellow anionic dyes using amino functionalized magnetic silica (Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@NH<sub>2</sub>) nanocomposite,” *Journal of Dispersion Science and Technology*, Vol. 40 (9), pp.1227–1235, 2019. [CrossRef]
- [3] N. Viktorová, A. Szarka, and S. Hrouzková, “Recent developments and emerging trends in paint industry wastewater treatment methods,” *Applied Science*, Vol. 12, Article 10678, 2022. [CrossRef]
- [4] B. A. Fil, M. Korkmaz, and C. Özmetin, “Application of nonlinear regression analysis for methyl violet (MV) dye adsorption from solutions onto illite clay,” *Journal of Dispersion Science and Technology*, Vol. 37, pp. 991–1001, 2016. [CrossRef]
- [5] V. Selvaraj, T.S. Karthika, C. Mansiya, and M. Alagar, “An over review on recently developed techniques, mechanisms and intermediate involved in the advanced azo dye degradation for industrial applications,” *Journal of Molecular Structure*, Vol. 1224, Article 129195, 2021.
- [6] T. Arslan, “Kompleks olarak bağlı ağır metal içerikli atıkların elektrokoagülasyon ile arıtımı” Available at: <https://tez.yok.gov.tr/UlusalTezMerkezi/tezSorguSonucYeni.jsp> Accessed on Nov 06, 2023.
- [7] D. R. Ryan, “Electrocoagulation-electrooxidation for mitigating trace organic compounds in model source waters” Available at: [https://epublications.marquette.edu/cgi/viewcontent.cgi?article=1532&context=theses\\_open](https://epublications.marquette.edu/cgi/viewcontent.cgi?article=1532&context=theses_open), Accessed on Nov 06, 2023.
- [8] Y. S. Ho, G. McKay, “Sorption of dye from aqueous solution by peat,” *Chem Eng J*, Vol. 70, pp. 115–124, 1998. [CrossRef]
- [9] M. Korkmaz, C. Özmetin, B.A. Fil, E. Özmetin, and Y. Yaşar, “Methyl violete dye adsorption onto clinoptilolite (natural zeolite): isotherm and kinetic study,” *Fresenius Environmental Bulletin*, Vol. 22(5a), pp. 1524–1533, 2013.
- [10] Y. Hu, and W. Wu “Application of membrane filtration to cold sterilization of drinks and establishment of aseptic workshop food and environmental,” *Virolgy*, Vol. 15, pp. 89–106, 2023. [CrossRef]
- [11] Davis, M. L. *Water and wastewater engineering design principles and practice*; The McGraw-Hill Companies, Inc., Newyork (USA) 2010.
- [12] D. Bingol, N. Tekin, and M. Alkan, “Brilliant Yellow dye adsorption onto sepiolite using a full factorial design,” *Applied Clay Science*, Vol. 50(3), pp. 315–321, 2010. [CrossRef]
- [13] M. B. Ahmad, U. Soomro, M. Muqet, and Z. Ahmed, “Adsorption of Indigo Carmine dye onto the surface-modified adsorbent prepared from municipal waste and simulation using deep neural network,” *Journal of Hazardous Materials*, Vol.408, Article 124433, 2021. [CrossRef]
- [14] M. Korkmaz, C. Özmetin, E. Özmetin, E. Çalgan, and Y. Süzen, “Boron removal by aluminum modified pumice and aluminum hydroxide from boron mine wastewater-Full factorial experimental design,” *Nevşehir Bilim ve Teknoloji Dergisi*, Vol. 10 (1), pp. 1–13, 2021.
- [15] T. H. Kima, C. Park, J. Yang, and S. Kima, “Comparison of disperse and reactive dye removals by chemical coagulation and fenton oxidation,” *Journal of Hazardous Materials*, Vol. 112, pp. 95–103, 2004. [CrossRef]
- [16] A. Szygula, E. Guibal, M. Ruiz, and A. M. Sastrec, “The removal of sulphonated azo-dyes by coagulation with chitosan,” *Colloids and Surfaces A: Physicochemical Engineering Aspects*, Vol. 330, pp. 219–226, 2008. [CrossRef]
- [17] B. Shi, G. Li, D. Wang, C. Feng, and H. Tang, “Removal of direct dyes by coagulation: The performance of preformed polymeric aluminum species,” *Journal of Hazardous Materials*, Vol. 143, pp. 567–574, 2007. [CrossRef]
- [18] S. Wong, N. A. Ghafar, N. Ngadi, F. A. Razmi, I. M. Inuwa, R. Mat, and N. A. S. Amin, “Effective removal of anionic textile dyes using adsorbent synthesized from coffee waste,” *Scientific Reports*, Vol. 10, Article 2928, 2020. [CrossRef]
- [19] A. Alhujaily, H. Yu, X. Zhang, and F. Ma, “Adsorptive removal of anionic dyes from aqueous solutions using spent mushroom waste,” *Applied Water Science*, Vol. 10, pp. 183–195, 2020. [CrossRef]
- [20] M. Korkmaz, B. A. Fil, C. Özmetin, and Y. Yaşar, “Full factorial design of experiments for boron removal from Colemanite mine wastewater using Purolite S 108 resin,” *Bulgarian Chemical Communications*, Vol. 46, pp. 594–601, 2014.
- [21] D. Kavak, “Removal of boron from aqueous solutions by batch adsorption on calcined alunite using experimental design,” *Journal of Hazardous Materials*, Vol. 163, pp. 308–314, 2009. [CrossRef]
- [22] M. Korkmaz, C. Özmetin, E. Özmetin, E. Çalgan and Ö. Ziyank, “Boron removal from colemanite mine wastewater by coagulation using zinc hydroxide–A factorial optimization study” *Celal Bayar University Journal of Science* Vol. 18(1), pp. 77–83, 2022. [CrossRef]
- [23] C. Özmetin, and M. Korkmaz, “ Full factorial design of experiments for boron removal by iron hydroxide from colemanite mine wastewater,” *Journal of BAUN Institute Science and Technology*, Vol. 21(1),

- pp. 244–253, 2019. [\[CrossRef\]](#)
- [24] F. Mcyottoa, Q. Wei, D.K. Macharia, M. Huang, C. Shen, C.W.K. Chow, “Effect of dye structure on color removal efficiency by coagulation,” *Chemical Engineering Journal*, Vol. 405 (1), Article 126674, 2021. [\[CrossRef\]](#)
- [25] Q. Wei, Y. Zhang, K. Zhang, J. I. Mwasiagi, X. Zhao, C. W. K. Chow and R. Tang, “Removal of direct dyes by coagulation: Adaptability and mechanism related to the molecular structure,” *Korean Journal of Chemical Engineering*, Vol. 39(7), pp. 1850–1862, 2022. [\[CrossRef\]](#)
- [26] C. E. Mortimer, “Modern Üniversite Kimyası,” Cilt 1, Çağlayan Kitap Evi, 1990.