Indian Journal of Chemical Technology
Vol. 32, May 2025, pp. 387-394
DOI: 10.56042/ijct.v32i3.9929

Removal of golden yellow dye from aqueous solution using layered double hydroxide: Optimization using response surface methodology

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Received 27 March 2024; accepted 19 March 2025

The magnesium-aluminium layered double hydroxide (MAN-LDH) is used as an adsorbent to remove golden yellow dye from aqueous solutions. The response surface methodology (RSM) based on a four-level-four variables Central Composite Rotatable Design (CCRD) has been employed to evaluate the interactive effects of the various optimization parameters. The parameters are contact time (1-3 h), solution pH (4 – 10), adsorbent dosage (0.02 – 0.04 g) and dye concentration (75-200 mg/L). Simultaneously by increasing the contact time, the initial concentration and the amount of dosage, the quantity of golden yellow dye removal is increased. The optimum conditions derived via RSM for the removal of golden yellow dye are a reaction time of 2.98 h, a dye concentration of 185.74 mg/L, a solution pH of 5.24 and an adsorbent dosage of 0.03 g. The experimental percentage removal is 89.58% under optimum conditions, which compared well with the maximum predicted value of 87.83%.

Keywords: Adsorption, Golden yellow dye, Layered double hydroxide, Optimization, Response surface methodology

Introduction

Dyes have long been used in dyeing, paper and pulp, textiles, plastics, leather, cosmetics and food industries[1]. Colour stuff discharged from these industries poses certain hazards and environmental problems. These coloured compounds are not only aesthetically displeasing but also inhibiting sunlight penetration into the stream and affecting aquatic ecosystem[2]. Dyes are a class of anthropogenic organic substances that pose serious threat to the environment[3] and usually have complex aromatic molecular structures which make them more stable and difficult to biodegrade[4]. Furthermore, many dyes are toxic to some microorganisms and may cause direct destruction or inhibition of their catalytic capabilities[5]. Therefore, the removal of dyes from wastewater and water are becoming important in order to protect the environment and also a public health. There are several methods that can be used to remove dyes from aqueous solution such as coagulation and flocculation[6], oxidation and ozonation[7] and adsorption[8]. Recently, adsorption has becoming a widely used technique due its simplicity, potential to regeneration and sludge-free operation. Moreover, it can be an attractive technique if the adsorbent that used can be made cheaply, able to regenerate and readily used.

Layered double hydroxides (LDH) also known as hydrotalcite-like compounds or anionic clays, have received much attention in the past decades due to their wide spread applicability. LDH have positively charged layers of metal hydroxides and the anions and water molecules are located between the layers. The positive charges that are produced from the isomorphous substitution of divalent cations and trivalent cations are counter balanced by anions located between the layers[9]. LDHs have a general formula of $[M^{2+}_{1-x} M^{3+}_{x} (OH)_2] [A^{n-}_{x/n.} m H_2O]$, where M^{2+} and M^{3+} are divalent and trivalent metal cations, respectively, A is the anion, and x is ratio $M^{3+}/(M^{2+} + M^{3+})$[(Ref.10)]. The anions between the layers can be polymers, organic dyes, surfactants and organic acids. Many substances in effluents wastewater containing pollutants carry positive and negative charges. The LDH anion exchange ability, large surface area and regeneration ability ensures that this adsorbent can be effectively utilized in wastewater purification.

Conventional and classical methods that normally being used to study optimization process required large number of experiments in order to determine the optimum level and the interactions between the parameters studied is almost impossible to study. The limitations posed by using a conventional analytical method can be eliminated by optimizing all the

affecting parameters collectively by using a statistical experiment design such as Response surface methodology (RSM). RSM is an effective statistical technique, which provides an investigative approach towards optimization. In addition, it is a collection of mathematical and statistical techniques used in significance of several affecting factors in an optimum manner, even in the presence of complex interactions[11].

Recent studies of LDH as an adsorbent to remove Alizarine Red-S dye with several optimization parameters has been reported by Ahmed *et al.,*[12] however, did not include RSM as a statistical tool in their experiment. In the previous year, Ghanbari and Ghafuri,[13] has reported excellent properties of LDH as an efficient and recyclable adsorbent for removing methylene blue (MB) dye from water. Despite the adsorption capacity of dye being reported as high, the optimization parameters were conducted conventionally and did not comprise RSM. Considering the above drawbacks, this present research aims to investigate for the first attempt the behaviour of LDH on the removal of golden yellow dye from the aqueous solution via RSM based on a four-level-four variables Central Composite Rotatable Design (CCRD) at different contact time, solution pH, adsorbent dosage, and dye concentration. The main reason for implementing RSM is to determine the optimum operational conditions for the process or to determine a region that satisfies the operating specifications[14]. RSM comprises of a five level-three-factor Central Composite Rotatable Design (CCRD), which is used in this study to evaluate the interactive effects and to obtain the optimum conditions for golden yellow dye removal from aqueous solutions using layered double hydroxides as an adsorbent.

Experimental Section

Materials

Magnesium sulphate hydrate [$Mg(SO_4)$. $7H_2O$], aluminium sulphate hydrate [$Al_2(SO_4)_3$. $16H_2O$] and sodium carbonate [Na_2CO_3] were obtained from Sigma (St Louis, MO, USA). All chemicals used were of analytical grade. Golden yellow dye was obtained from textile laboratory, Faculty of Applied Sciences, Universiti Teknologi MARA, Malaysia. All other chemicals used were of analytical grade.

Synthesis and characterization of layered double hydroxide

The co-precipitation method was adopted to synthesize Mg-Al-SO_4 in this work following Reichle's procedure[15]. During the preparation, an aqueous solution of magnesium sulphate hydrate [$Mg(SO_4)$. $7H_2O$] was added to aluminium sulphate hydrate [Al_2 $(SO_4)_3$. $16H_2O$] to give Mg^{2+} / Al^{3+} ratio, R=4. Aqueous solution of NaOH and Na_2CO_3 (2.0 M) was then added to the mixture dropwise, with vigorous stirring at room temperature and the pH was adjusted to 10.0 ± 0.2. The precipitate formed was aged at 70ºC in an oil bath shaker for 24 h, cooled, centrifuged, washed several times with deionized water and dried in an oven for 48 h. The resulting layered double hydroxide was ground into powder and kept in sample bottles for further use and characterization. X-ray diffraction (XRD) pattern of the sample powder was recorded on a Siemen diffractometer D500 with Ni filtered, Cu Kα radiation at 40 kV and 20 mA. The sample was mounted on a glass slide and scans at 2°-65° 2θ/min at 0.003° steps. The basal spacing was determined via the powder technique.

Experimental Design

A four-level-four factor CCRD design was employed in this study, leading to a set of 15 experiments. The variables and their levels selected for the study of the removal of golden yellow dye using layered double hydroxide were contact time (1-3 h), adsorbent dosage (0.02 – 0.04), pH of dye solution (4-10) and initial golden yellow dye solution concentration (75–200 mg/L), based on the preliminary experiments using conventional optimization method. The variables and their respective levels are presented in Table 1, which represents the actual experiments (in triplicate) carried out for developing the model. The data obtained were fitted to a second-order polynomial equation:

$$Y = \beta_0 + \sum_{i=1}^{4} \beta_i x_i + \sum_{i=1}^{4} \beta_{ii} x^2 + \sum_{i=1}^{3} \sum_{j=1+1}^{4} \beta_{ij} x_{ij}$$

where, Y is percentage of golden yellow removed; β_0, β_i, β_{ii}, β_{ij} are constant coefficients and x_i are the uncoded independent variables. Subsequent regression analyses, analyses of variance (ANOVA) and response surfaces were performed using the Design Expert Software (Version 6.0.4) from Stat-Ease Inc. (Mineapolis, MN, USA). Optimal reaction parameters for maximum removal were generated using the software's numerical optimization function.

Table 1 — Central composite rotatable quadratic polynomial model, experimental data, actual and predicted values for five-level-four-factor response surface analysis

Run	A	B	C	D	Actual (% removal)	Predicted(% removal)
1	2.00	7.00	0.03	12.50	98.66	99.42
2	3.00	10.00	0.02	200.00	97.82	97.00
3	2.00	7.00	0.03	137.50	96.75	97.63
4	4.00	7.00	0.03	137.50	99.49	83.26
5	1.00	4.00	0.02	200.00	90.04	80.39
6	3.00	10.00	0.04	200.00	98.79	99.63
7	2.00	7.00	0.05	137.50	98.45	99.53
8	1.00	4.00	0.04	75.00	99.76	89.08
9	2.00	13.00	0.03	137.50	63.1	79.55
10	2.00	7.00	0.03	137.50	98.45	97.63
11	3.00	10.00	0.02	75.00	95.95	97.57
12	3.00	4.00	0.04	200.00	99.97	99.95
13	1.00	10.00	0.04	75.00	97.86	85.38
14	1.00	10.00	0.04	200.00	98.24	83.21
15	2.00	7.00	0.03	262.50	96.96	99.65
16	3.00	4.00	0.02	75.00	99.83	99.35
17	1.00	10.00	0.02	75.00	99.32	83.84
18	2.00	7.00	0.03	137.50	98.12	97.63
19	1.00	4.00	0.04	200.00	97.45	85.89
20	3.00	4.00	0.04	75.00	100	99.85
21	2.00	7.00	0.01	137.50	97.42	99.79
22	1.00	10.00	0.02	200.00	88.42	77.30
23	1.00	4.00	0.02	75.00	98.75	87.97
24	3.00	4.00	0.02	200.00	99.21	99.75
25	2.00	1.00	0.03	137.50	83	88.00
26	3.00	10.00	0.04	75.00	97.68	95.82
27	0.00	7.00	0.03	137.50	13.79	51.46
28	2.00	7.00	0.03	137.50	98.49	97.63
29	2.00	7.00	0.03	137.50	95.84	97.63
30	2.00	7.00	0.03	137.50	98.11	97.63

A = time (h); B = pH; C = dosage (mg); D = concentration (mg/L)

Batch adsorption and analysis

Stock solutions of golden yellow dye were prepared using distilled water and diluted to a predetermined set of working concentrations. Golden yellow dye concentrations were measured using a Lambda 20 UV-visible spectrophotometer. 0.25 g of synthesized layered double hydroxide was placed in 25 mL of 50-100 mg/L golden yellow dye solution. 25 mL of golden yellow dye solution placed in a 250 mL conical flask was contacted with 0.25 g of layered double hydroxide and agitated using a water batch shaker operating at 120 rpm. Experiments were performed according to the Central Composite Rotatable Design (CCRD) presented in Table 1. The response was expressed as follows.

$$\% \text{ of dye removal} = [(C_o - C_t) / C_o] \times 100.$$

Results and Discussion

Characterization of layered double hydroxides

The X-ray diffraction (XRD) patterns of original layered double hydroxides at different ratios are shown in Fig. 1. Based on the patterns as shown in Fig. 1, original layered double hydroxides represent a strong sharp and symmetrical peak assigned to (003) and (006) planes, respectively as agreed by George and Saravanakumar[16]. The pattern also demonstrated general features of layered double hydroxides with good crystallinity, which was supported by the d-spacing and recorded at 7.9Å[17]. The interlayer spacing of the sample corresponding to the (006) plane was recorded at 4.5Å. The same result on the XRD patterns of layered double hydroxides was also reported previously[18] which describes the sharp peaks signifying high crystallinity related to layered double hydroxides. Layered double hydroxide with the synthesized ratio of 4 was chosen as an adsorbent for the removal of golden yellow dye.

Model development

The coefficients of the empirical model and their statistical analyses, evaluated using the Design Expert Software, are presented in Table 2. The predicted

values were obtained from the model-fitting technique using the Design Expert Software version 6.0.4 and were seen to be sufficiently correlated with the observed values. Fitting the data to various models (linear, two factorial, quadratic and cubic) and their subsequent ANOVA showed that the removal of golden yellow dye from aqueous solution using layered double hydroxide was suitably described by quadratic model. From the Design Expert Software, the quadratic polynomial is given as follows:

$$\text{Removal (\%)} = 97.63 + 7.95A - 2.11B + 0.94C - 0.94D - 7.57A^2 - 3.46B^2 + 2.76C^2 + 2.73D^2 - 0.41AB - 0.82AC + 1.49AD + 0.11BC + 0.26BD + 1.09CD$$

Where A is contact time, B is solution pH, C is the amount of dosage and D is the concentration.

The computed model *F*-value of 18.9 was higher than the tabular value of $F(0.01, 18, 5) = 9.61$, implying the model is significant at 1% confidence level. The model also showed a very low value of pure error of 5.86, which indicates good reproducibility of the data obtained. The high coefficient of determination ($R^2 = 0.9398$) showed that the quadratic polynomial was highly significant and sufficient to represent the actual relationship between the removal (%) and the significant variables.

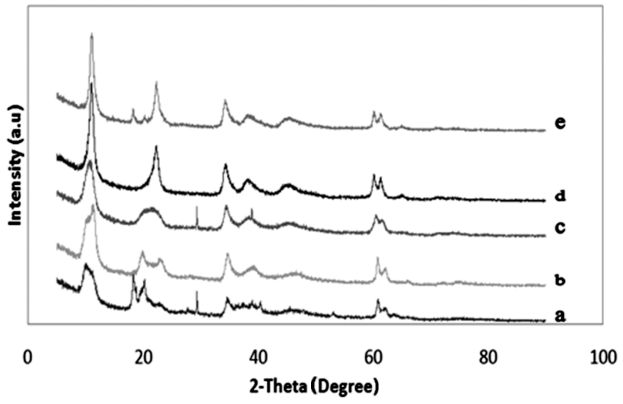

Fig. 1 — XRD of LDH-MAN at different Mg/Al ratio; (a) MAN 1, (b) MAN 2, (c) MAN 3, (d) MAN 4 and (e) MAN 4 after adsorption of golden yellow dye

Response Surface Plots

The quadratic polynomial equation was then used to facilitate the plotting of response surfaces. Two parameters were plotted at any one time on the x_1 and x_2 axes, respectively, with the other remaining parameters set at their center points values (coded level:0). Fig. 2 shows the profile of contact time *versus* pH of dye solution for removal of golden yellow dye from aqueous solution using layered double hydroxide. A response surface plot for interaction between contact time and pH of dye solution was generated with the parameters of the amount of dosage and concentrations fixed at center points of (0.03 g) and (150 mg/L), respectively, under any given conditions. From Fig. 2, at any given pH of dye solutions from 4 to 10, an increase in contact time led to a higher percentage of removal. The effect of

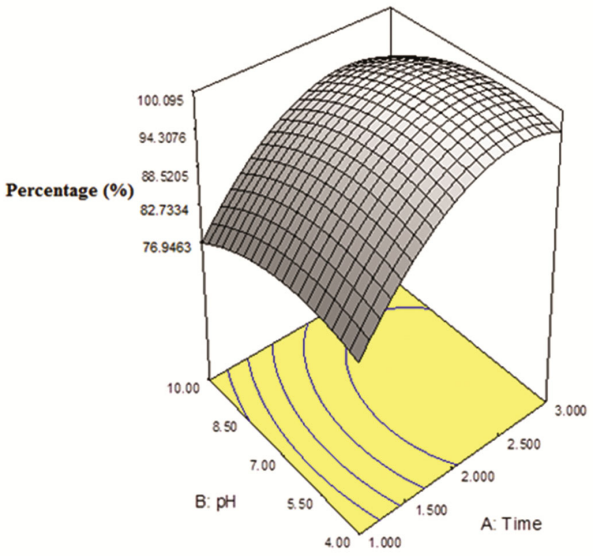

Fig. 2 — Response surface plot showing the effect of contact time and pH and their mutual effect on the removal of golden yellow dye using layered double hydroxide

Table 2 — ANOVA for removal of golden yellow dye from aqueous solution using layered double hydroxide

Source	Degree of freedom	Sum of squares	Mean square	*F*-value	*P*
Model	14	4334.67	309.62	18.90	< 0.0010
Residual	15	3695.32	246.35		
Lack of fit	10	3689.46	368.95	1.25	Not significant
Pure error	5	5.86	1.17		
Total	29	8029.99			
Time (A)	1	1517.02	1517.02	6.16	0.0254
pH (B)	1	107.23	107.23	0.44	< 0.0010
Dosage (C)	1	21.04	21.04	0.085	< 0.0010
Concentration (D)	1	21.30	21.30	0.086	< 0.0010

contact time has been demonstrated by Ramadhani & Kurniawati[19] on theadsorption process of MB dyes. According to the author, contact time is the essential duration required for optimal interaction between the biosorbent and adsorbate. As contact time increases, the biosorption rate rises until equilibrium is reached. However, beyond this point, prolonged contact does not enhance adsorption but may cause desorption, reducing adsorption capacity.

An increase in pH up to 10 resulted in a slight decrease in the percentage removal of golden yellow dye. Removal of the dye from aqueous solution using layered double hydroxide as an adsorbent with moderate pH and maximum contact time favoured maximal percentage removal. A similar trend profile of contact time versus pH of dye solutions has indeed been reported[18].

Response surfaces predicted for the interaction of contact time and amount of adsorbent dosage are illustrated in Fig. 3 with the concentration of golden dye solution set at center point value 150 mg/L. As shown in Fig. 3, at any given amount of layered double hydroxide dosage from 0.02 to 0.04 g, an increase in contact time led to a higher percentage removal of golden yellow dye. Removal with the highest dosage and highest reaction time (3 h) favoured maximal percentage removal. A similar trend profile in contact time versus amount of dosage has indeed been reported[20]. Also, Sibhat *et al.*,[21] investigatethe effect of adsorbent dosage on the adsorption of MB dye. Their finding revealed that the percentage removal efficiency MB was increased with an increase in the adsorbent dose. This was attributed to the direct correlation between surface area and the number of active sites available for adsorption. Similarly, Assila *et al.*,[22] investigated the effect of particle size on adsorption using natural materials from Morocco. Their findings revealed that smaller particle diameters (40 µm) enhanced adsorption efficiency compared to larger particles (125 µm), highlighting the importance of surface area in adsorption performance.

Fig. 4 depicts the response surface plot showing the effect of contact time and concentration of dye solution at a fixed pH and adsorbent dosage. A shown here, the percentage removal increased with increasing contact time and concentrations. Similar trend of profile for the effect of contact time and concentration was reported for dyes removal using a novel adsorbent[11], in which the highest removal efficiency was recorded at the highest contact time between the dye and adsorbent. As shown in Fig. 4, the adsorption was fast at the early stages and approached equilibrium after 3 h. The same trend of adsorption was reported in the removal of humic acid using anionic clay hydrotalcite[23] in which the percentage removal increased with the increase in the concentration of humic acid. Maximal percentage removal favoured high initial concentration of golden yellow dye and a longer contact time between the dye and adsorbent.

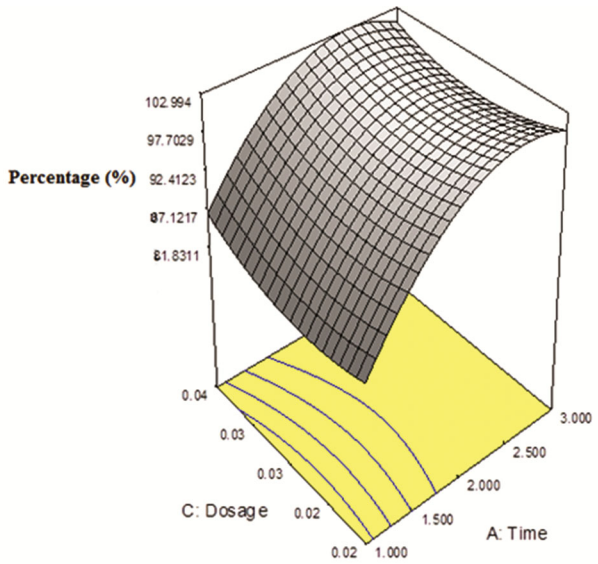

Fig. 3 —Response surface plot showing the effect of contact time and amount of dosage and their mutual effect on the removal of golden yellow dye using layered double hydroxide

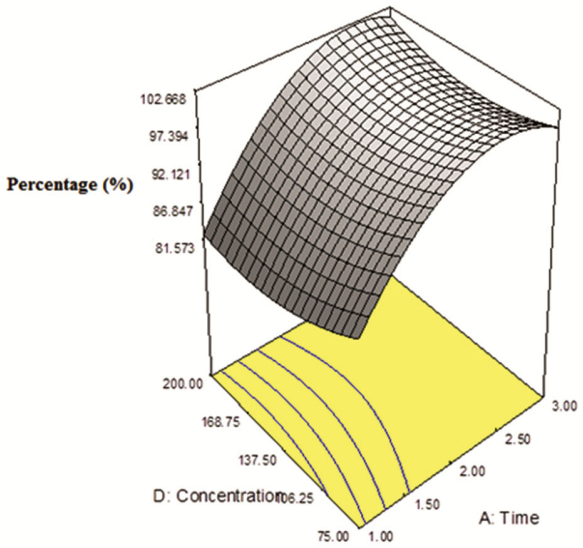

Fig. 4 — Response surface plot showing the effect of contact time and concentration of dye and their mutual effect on the removal of golden yellow dye using layered double hydroxide

The response surface plot presented in Fig. 5 shows the interaction of pH and amount of adsorbent dosage at a fixed contact time and initial concentration for removal of golden yellow dye from aqueous solution using layered double hydroxide. It is observed that an increase in dosage led to higher percentage removal but further increased of pH will steadily decreased the percentage removal of golden yellow dye. Hence, the weak acid is the ideal condition for golden yellow dye removal using layered double hydroxide. Fig. 6 shows the profile of pH *versus* concentration of dye solution at a fixed contact time and adsorbent dosage for removal of golden yellow dye. As expected, the percentage removal of golden yellow dye increase with an increase of adsorbent dosage and steadily decreased in percentage with an increase of solution pH. The golden yellow dye is acidic in nature and its percentage removal is better at a lower pH, which is in good agreement with other studies reported by other authors who also performed optimization studies[24].

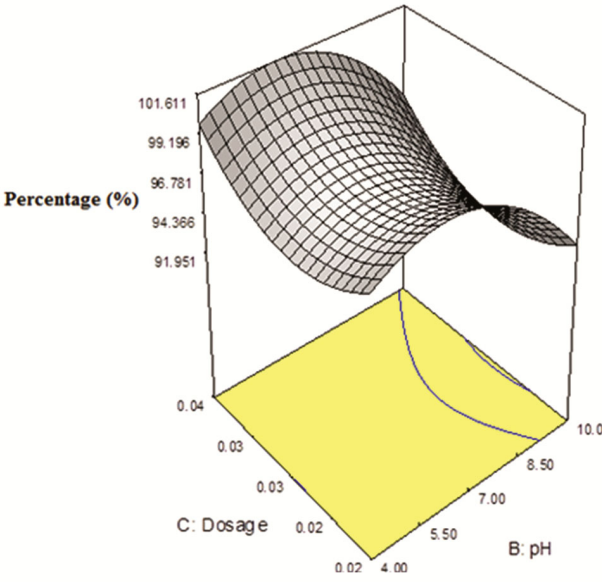

Fig. 5 — Response surface plot showing the effect of pH and amount of dosage and their mutual effect on the removal of golden yellow dye using layered double hydroxide

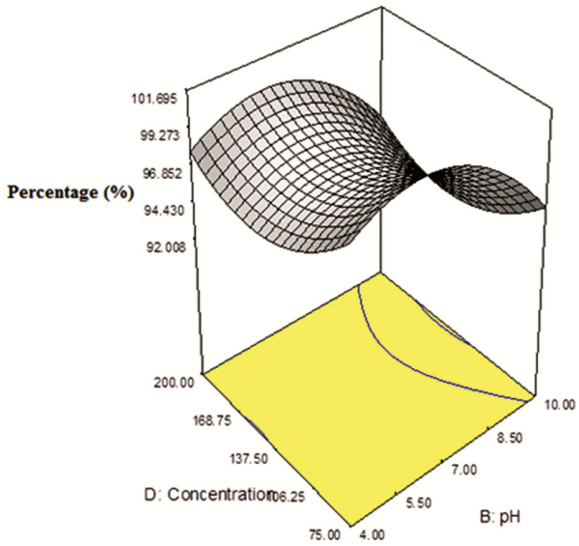

Fig. 6 —Response surface plot showing the effect of pH and concentration of dye and their mutual effect on the removal of golden yellow dye using layered double hydroxide

Fig. 7 presents the profile of golden yellow dye concentration versus the amount of layered double hydroxide dosage at a fixed contact time and pH of golden yellow dye solution. As shown in Fig. 7, at low dosages of layered double hydroxide and low concentrations of golden yellow dye, the percentage removal was lower, and this could be attributed to a more limited affective adsorption area and the decreased probability of a dye molecule to find an adsorption site. A higher percentage removal was achieved with high amounts of layered double hydroxide and high initial concentrations of the golden yellow dye solution.

Optimum Conditions

Within the experimental range studied, optimum conditions for removal of golden yellow dye using layered double hydroxide were predicted using the optimization function of the Design Expert. These are

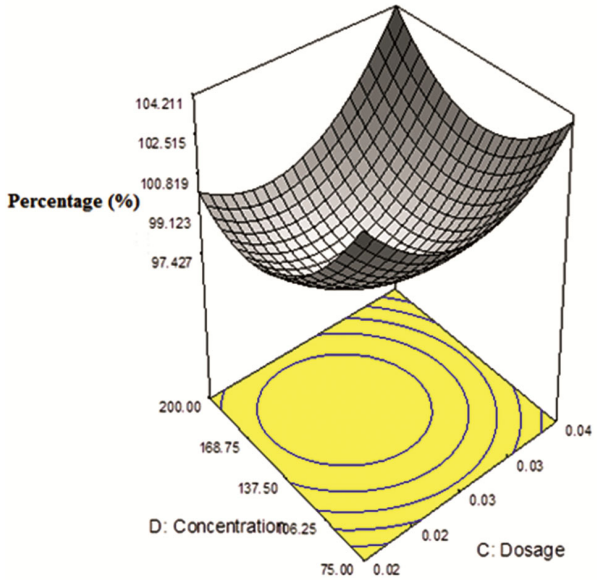

Fig. 7 — Response surface plot showing the effect concentration of dye and amount of dosage and their mutual effect on the removal of golden yellow dye using layered double hydroxide

Table 3 — Optimum conditions derived by RSM

Optimal conditions				Actual removal	Predicted removal
A	B	C	D	(%)	(%)
2.98	5.24	0.03	185.74	89.58	87.83

presented in Table 3 along with their predicted and actual values. The analysis indicated that maximum percentage removal of golden yellow dye was at a contact time of 2.98 h, pH of 5.24, dosage of 0.03 g and 185.74 of golden yellow dye concentration. The response behaviour of contact time and pH of golden yellow dye (Fig. 1) were followed while holding the other reactions parameters constant at suggested point. Therefore, the conditions (contact time 2.98 h, pH of 5.24, dosage of 0.03 and dye concentration of 185.74) were recommended as optimal conditions for removal of golden yellow dye from aqueous solution using layered double hydroxide as an adsorbent. Comparison of predicted and experimental values revealed good correspondence between them, implying that empirical models derived from the RSM could be used adequately describe the relationship between the factors and response in removal of golden yellow dye from aqueous solution

Conclusion

The study demonstrated the effectiveness of magnesium-aluminium layered double hydroxide (MAN-LDH) as an adsorbent for the removal of golden yellow dye from aqueous solutions. The optimum conditions for maximum dye removal (89.58%) were identified as a reaction time of 2.98 h, a dye concentration of 185.74 mg/L, a solution pH of 5.24, and an adsorbent dosage of 0.03 g, closely aligning with the predicted removal efficiency of 87.83%. These findings highlight the potential of MAN-LDH as an efficient adsorbent for dye wastewater treatment, contributing to sustainable water purification strategies.

Acknowledgments

This work was supported by the Ministry of Higher education Malaysia under Fundamental Research Grant Scheme.

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