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Primary treatment of dye wastewater using *aloe vera*-aided aluminium and magnesium hybrid coagulants

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Abstract

Hybrid coagulants, aluminium sulphate-*Aloe vera* (ALAV) and magnesium sulphate-*Aloe vera* (MGAV) were prepared for the primary treatment of methylene blue (MB) dye wastewater treatment through coagulation-flocculation process. The effects of the independent factors and their interaction on the dye removal (%) were determined using two independent factors, i.e. pH and dosage based on 2^2 full factorial design. All the independent factors and their interaction were significant in removing dye. The dye removal (%) for both ALAV and MGAV were then optimized through central composite design. ALAV was able to remove 50–55% of dye while MGAV was able to remove 60-70% of dye. Therefore, MGAV was proven to be a more effective hybrid coagulant in removing dye.

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Keywords: dye wastewater; hybrid coagulants; pollution; statistics; waste

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1. Introduction

Water pollution is a major problem to the environment and can negatively affect the sustainability of water resources. Textile industry is one of the industries that pose a high demand on water supply and produce large amounts of wastewater [1]. One of the best known textile industries in Malaysia is batik industry. Batik industry is usually operated in small scale by workshops and factories throughout the country without a regulated waste disposal system. The dye wastewater from the batik making process is usually being discharged directly into the waterways without any treatment, prompting both pollution and toxicological concerns [2]. One of the common dyestuffs, methylene blue ($C_{16}H_{18}N_3SCl$), is always being used in colouring cotton and wools. It forms a deep blue solution when dissolved in water. The degree of dye fixation on fabric is never complete during the dyeing process, resulting in coloured wastewater. Therefore, the removal of methylene blue from the dye wastewater is of utmost importance due to the serious environmental damage that can occur as a result of contact with it, particularly in the case of people [3].

Coagulation-flocculation is an important unit operation for the primary treatment of dye wastewater. Coagulation removes dissolved and colloidal substances in wastewater by overcoming the interparticle repulsive energy barrier by simply increasing the ionic strength and destabilizes colloids by neutralizing the forces that keep them apart. Flocculation occurs through the bridging between particles to form larger flocs for sedimentation to take place [4]. Hybrid coagulants have been proven to be effective in treating dye wastewater. Considering the increasing demand over the environmental friendly materials to be applied in coagulation-flocculation of wastewater, researchers have discovered the feasibility of several new natural polymeric materials to be composed in hybrid materials for coagulation-flocculation [5]. Inorganic-natural polymer hybrid coagulants using *Aloe vera* aided aluminium and magnesium salts, being relatively new combinations of hybrid material were of the interest of the present study.

In this study, two hybrid coagulants, aluminium sulphate-*Aloe vera* (ALAV) and magnesium sulphate-*Aloe vera* (MGAV) were prepared for the primary treatment of methylene blue (MB) dye wastewater treatment through coagulation-flocculation process. Two independent factors, i.e. pH and dosage, were taken into account of 2 [2] full factorial designs, to determine the effects of the independent factors and their interaction on the dye removal (%) [6]. Central composite design was then used to optimize the treatment of dye wastewater[4]. Regression models were presented to simulate the primary treatment of dye wastewater.

2. Materials and methods

2.1. Materials

Aluminium sulphate octadecahydrate (AL) (99%, R&M Chemicals), magnesium sulphate heptahydrate (MG) (99%, R&M Chemicals), Aloe vera extract (99%, Lotioncrafter LLC) and methylene blue (MB) (99%, R&M Chemicals) were used as is. Distilled water was used throughout the experiments.

2.2. Hybrid coagulants preparation and coagulation-flocculation procedures

Aqueous solutions of ALAV and MGAV (20g/L) were prepared through physical blending with the compositions of 90% AL : 10% AV and 90% MG : 10% AV, respectively. The aqueous solutions of ALAV and MGAV were allowed to age for 24 hours at room temperature prior to any application.

Coagulation-flocculation was carried out using a standard jar test. The pH of the 10 mg/L MB dye wastewater was adjusted using acid and alkaline. Desired dosage of hybrid coagulants was introduced into the dye wastewater according to the design of experiment prior to the 100 rpm mixing. Flocs formed were allowed to settle for 30 minutes. The supernatant was sampled and the absorbance was measured using a spectrophotometer (HACH, DR2800).

2.3. Experimental design

A 2 [2] full factorial design was used to investigate the effects of pH and dosage on dye removal (%). Tables 1 and 2 show the factorial designs to be employed for the ALAV and MGAV experiments, respectively. A total of 7 runs were conducted for each factorial design. Factor levels coded as -1 and +1 represent the low level and the high level for factor range, respectively. The centre point is coded as 0. Centre points were added to detect the curvature. The output of the factorial design was analyzed using Minitab 14 statistical software to determine the effects of the independent factors and their interaction. Axial points –α, +α and central axial points were added to augment the factorial design to central composite design as shown in Tables 3 and 4 for the optimization of ALAV and MGAV, respectively.

Table 1 Factorial design for ALAV

Run order	pH	Dosage	Dye removal (%)
1	5 (-1)	1000 (-1)	29.7240
2	6 (+1)	1000 (-1)	48.6640
3	5 (-1)	3000 (+1)	50.6910
4	6 (+1)	3000 (+1)	52.7650
5	5.5(0)	2000 (0)	45.0690
6	5.5 (0)	2000 (0)	45.5300
7	5.5 (0)	2000 (0)	45.5760

Table 2 Factorial design for MGAV

Run order	pH	Dosage	Dye removal (%)
1	11.5 (-1)	1000 (-1)	37.4941
2	12.5 (+1)	1000 (-1)	53.8720
3	11.5 (-1)	3000 (+1)	57.8510
4	12.5 (+1)	3000 (+1)	64.8991
5	12 (0)	2000 (0)	48.9442
6	12 (0)	2000 (0)	49.6481
7	12 (0)	2000 (0)	49.8827

Table 3 Central composite design for ALAV

Run Order	pH	Dosage	Dye removal (%)
1	5 (-1)	1000 (-1)	29.7240
2	6 (-1)	1000 (-1)	48.6640
3	5 (-1)	3000 (+1)	50.6910
4	6 (+1)	3000 (+1)	52.7650
5	5.5 (0)	2000 (0)	45.0690
6	5.5 (0)	2000 (0)	45.5300
7	5.5 (0)	2000 (0)	45.5760
8	4.8 (-α)	2000 (0)	39.5390
9	6.2 (+α)	2000 (0)	53.8710
10	5.5 (0)	586 (-α)	36.3590
11	5.5 (0)	3414(+α)	54.6080
12	5.5 (0)	2000 (0)	45.5760
13	5.5 (0)	2000 (0)	45.6220
14	5.5 (0)	2000 (0)	45.3920

Table 4 Central composite design for MGAV

Run Order	pH	Dosage	Dye removal (%)
1	11.5 (-1)	1000 (-1)	37.4941
2	12.5 (+1)	1000 (-1)	53.8720
3	11.5 (-1)	3000 (+1)	57.8510
4	12.5 (+1)	3000 (+1)	64.8991
5	12 (0)	2000 (0)	48.9442
6	12 (0)	2000 (0)	49.6481
7	12 (0)	2000 (0)	49.8827
8	11.3 (-α)	2000 (0)	55.0446
9	12.7 (+α)	2000 (0)	70.7180
10	12 (0)	586 (-α)	34.2680
11	12 (0)	3414 (+α)	58.7490
12	12 (0)	2000 (0)	49.4603
13	12 (0)	2000 (0)	48.6157
14	12 (0)	2000 (0)	51.1028

3. Results and discussion

3.1. Screening of factors for the dye removal (%)

To determine the factors that are likely to be significant in removing dye from the wastewater, a 2^2 full factorial design was used. Independent factors, i.e. pH and dosage were taken into account to investigate their effects as well as their interaction on the dye removal (%). The significance of the factors and their interaction were evaluated based on the normal probability plots of standardized effect with p=0.05 as shown in Fig. 1 and 2. All the independent factors and their interaction for ALAV and MGAV show non zero means, whereby A, B and AB which denote pH, dosage and pH* dosage respectively deviate from the blue straight line, imply that they are significant in affecting dye removal (%). The further the factor deviates from the blue straight line imply the more significant of the factor. Therefore, it is noted that dosage of ALAV and MGAV has the strongest effect on the dye removal (%) follow by pH and their interaction.

Main effects and interaction plots consisting mean response values at different levels of the factors were used to determine the relative strength of the factors in affecting dye removal (%). Addition of centre point (red point) is to detect the curvature between the levels. The main effects and interaction plots of ALAV are shown in Fig. 3 and 4, respectively. With the increase of pH and dosage, the dye removal (%) using ALAV increases correspondingly. The main effects and interaction plots of MGAV are shown in Fig. 5 and 6, respectively. It is noted that the dye removal (%) using MGAV increases at a relatively low rate below the centre point but it increases drastically thereafter. It is because magnesium-based MGAV has a relatively narrow range for pH and dosage. Therefore, it requires certain pH and dosage for the dye removal to take place during the coagulation-flocculation process [6]. The interaction plots show the interaction effect between the factors. The unparallel effect plots imply that there is an interaction between pH and dosage.

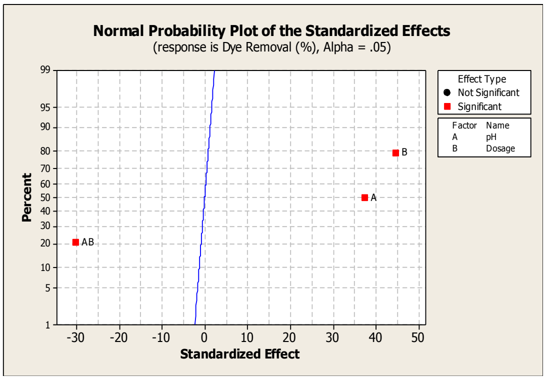

Fig. 1. Normal probability plot of standardized effect for dye removal (%) using ALAV

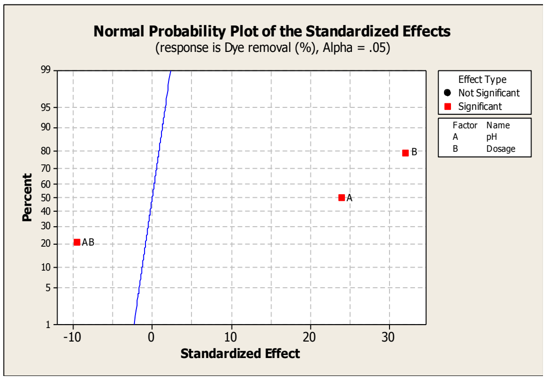

Fig. 2. Normal probability plot of standardized effect for dye removal (%) using MGAV

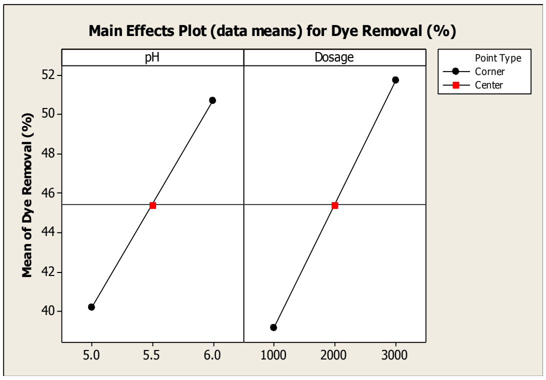

Fig. 3. Main effects plot for dye removal (%) using ALAV

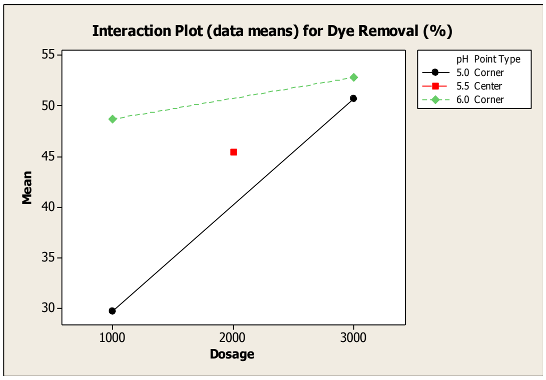

Fig. 4. Interaction plot for dye removal (%) using ALAV

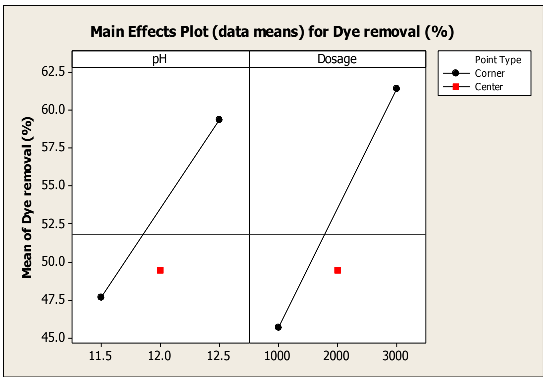

Fig. 5. Main effects plot for dye removal (%) using MGAV

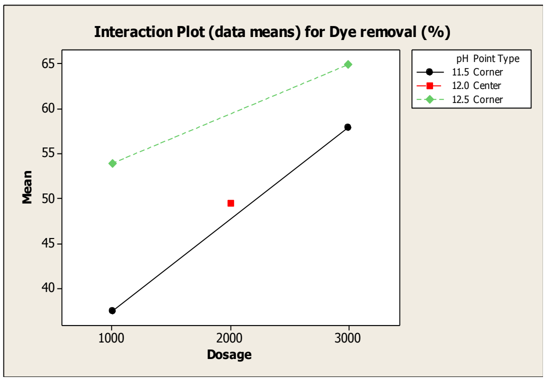

Fig. 6. Interaction plot for dye removal (%) using MGAV

3.2. Optimization of factors for the dye removal (%)

To optimize the dye removal (%), the 2^2 full factorial design used earlier was augmented into central composite design by adding axial points and axial centre points. Fig. 7 and 8 shows the contour plots of dye removal (%) using ALAV and MGAV, respectively. Regression models for dye removal (%) using ALAV and MGAV were fitted using quadratic equations as shown in Equations 1 and 2.

$$\text{Dye removal (\%) using ALAV} = 45.4608 + 5.1603A + 6.3595B + 0.4638A^2 - 4.2165AB \quad (1)$$

$$\text{Dye removal (\%) using MGAV} = 49.6090 + 5.6989A + 8.2507B + 6.3447A^2 - 1.8417B2 - 2.3324AB \quad (2)$$

The corresponding ANOVA analysis for the reduced models of dye removal (%) using ALAV and MGAV are shown in Tables 5 and 6, respectively. The insignificant p-values (p>0.05) of lack-of-fit imply that the fitted models are adequate in predicting dye removal (%) using ALAV and MGAV. A comparison between the dye removal (%) of ALAV and MGAV was made, ALAV gave dye removal of 50–55 % at the optimal pH=6 and dosage=3000mg/L, meanwhile MGAV gave dye removal of 60-70 % at the optimal pH=12.5 and dosage=3000mg/L. Therefore, MGAV is better in terms of dye removal (%) as compared to that of ALAV.

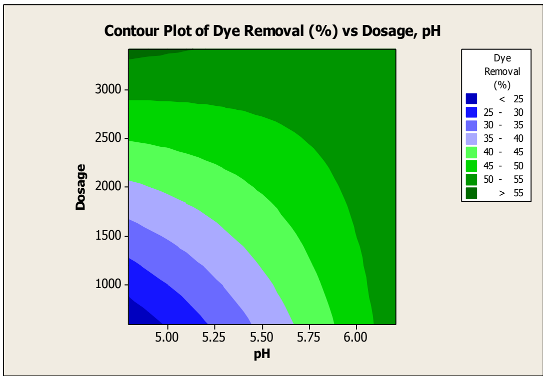

Fig. 7. Contour plot of dye removal (%) using ALAV

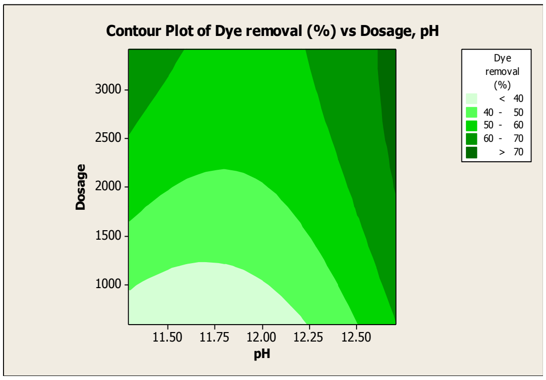

Fig. 8. Contour plot of dye removal (%) using MGAV

Table 5 ANOVA analysis of dye removal (%) using ALAV

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Blocks	1	0.621	0.621	0.621	8.13	0.025
Regression	5	609.528	609.528	121.906	1595.86	0.000
Linear	2	536.576	536.576	268.288	3512.15	0.000
Square	2	1.836	1.836	0.918	12.02	0.005
Interaction	1	71.115	71.115	71.115	930.97	0.000
Residual error	7	0.535	0.535	0.076		
Lack of fit	3	0.348	0.348	0.116	2.48	0.200
Pure error	4	0.187	0.187	0.047		
Total	13	610.684				

Table 6 ANOVA analysis of dye removal (%) using MGAV.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Blocks	1	2.06	2.058	2.058	2.43	0.163
Regression	5	1163.76	1163.76	232.753	274.84	0.000
Linear	2	804.41	804.41	402.206	474.94	0.000
Square	2	337.59	337.59	199.32	12.02	0.000
Interaction	1	21.76	21.76	21.76	25.70	0.001
Residual error	7	5.93	5.928	0.847		
Lack of fit	3	2.25	2.252	0.751	0.82	0.548
Pure error	4	3.68	3.676	0.919		
Total	13	1171.75				

4. Conclusion

The primary treatment of methylene blue (MB) dye wastewater was conducted using aluminium sulphate-*Aloe vera* (ALAV) and magnesium sulphate-*Aloe vera* (MGAV) hybrid coagulants. The effects of pH, dosage and their interaction were investigated using 2^2 full factorial designs. All the independent factors and their interaction were significant in affecting dye removal (%). The order of significance was: dosage > pH > pH*dosage. The dye removal (%) for both ALAV and MGAV were then optimized through central composite design. Regression models for dye removal (%) using ALAV and MGAV were fitted to simulate the primary treatment of dye wastewater. ALAV was able to remove 50–55% of dye at the optimal pH=6 and dosage=3000mg/L; MGAV was able to remove 60-70% of dye at the optimal pH=12.5 and dosage=3000mg/L. As a conclusion, MGAV is proven to be a more effective hybrid coagulant in removing dye.

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