

UNIVERSITI TEKNOLOGI MARA

**OPTIMISATION OF METHYL
ORANGE ADSORPTION FROM
AQUEOUS SOLUTION ONTO
MAGNESIUM-ALUMINIUM
LAYERED DOUBLE HYDROXIDE
USING RESPONSE SURFACE
METHODOLOGY**

**NURUL AISYAH BINTI MAT
ZAMZURI**

MSc

August 2026

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NURUL AISYAH BINTI MAT ZAMZURI

Thesis submitted in fulfilment
of the requirements for the degree of
**Master of Science
(Chemistry)**

Faculty of Applied Sciences

August 2026

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ABSTRACT

The discharge of dye containing wastewater from textile industries poses serious environmental and health concerns due to the persistence and toxicity of synthetic dyes such as methyl orange. Methyl orange was selected as the target pollutant because it is a common anionic azo dye that widely used in textile related industries. This study investigates the adsorption performance of magnesium–aluminium layered double hydroxide (MgAl-LDH) for the removal of methyl orange from aqueous solutions. MgAl-LDH was synthesised via the co-precipitation method and characterised using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM–EDX). The characterisation results confirmed the successful formation of a hydrotalcite-like layered structure with characteristic functional groups, plate-like morphology and homogeneous elemental composition. The adsorption process was optimised using response surface methodology (RSM) to evaluate the effects of adsorbent dosage, pH solution, initial methyl orange concentration and contact time on methyl orange removal efficiency. Statistical analysis showed that the developed quadratic model was significant and reliable, with optimum adsorption conditions obtained at approximately 0.65 g of MgAl-LDH, 7.78 of pH, 40 mg/L of initial methyl orange concentration and 20 minutes of contact time, achieving a maximum predicted methyl orange removal of 92.19%. Experimental validation conducted under the optimum conditions resulted in a higher methyl orange removal efficiency of 94.4% with relative standard error values below 5%, demonstrating good agreement with the model prediction and confirming the reliability of the RSM approach. Adsorption isotherm analysis indicated that the equilibrium data were better described by the Freundlich model, suggesting heterogeneous and multilayer adsorption behaviour. Kinetic studies showed that the adsorption process followed the pseudo-second-order model, indicating chemisorption as the dominant mechanism. The applicability of MgAl-LDH was further evaluated using textile wastewater where highest removal efficiency was 78.80% were achieved under the optimised conditions. Desorption and reusability studies revealed a gradual decrease in adsorption efficiency over repeated cycles, highlighting the need for improved regeneration strategies. Overall, this study demonstrates that MgAl-LDH is an effective adsorbent for methyl orange removal and has promising potential for wastewater treatment applications.

ACKNOWLEDGEMENT

Alhamdulillah, all praise and gratitude are due to Allah SWT, the Most Merciful and the Most Compassionate. Firstly, I wish to thank Allah for giving me the opportunity to embark on my Master journey and for granting me the strength, patience and perseverance to complete this challenging journey successfully. Without His guidance and blessings, this achievement would not have been possible. I would like to express my deepest gratitude and sincere appreciation to my supervisor, Dr. Nurul Auni binti Zainal Abidin, for her continuous guidance, encouragement, patience and invaluable advice throughout this research. Her dedication, constructive feedback and motivation have greatly contributed to the successful completion of this study. My heartfelt thanks are also extended to my co-supervisor, Dr. Sofiah binti Abu Kassim, for her insightful suggestions, support and guidance, which have been instrumental in improving the quality of this research. My deepest appreciation and love go to my beloved parents, Mat Zamzuri bin Zakaria, and Mat Zamzuri binti Mat Zamzuri, for their endless prayers, unconditional love, sacrifices and continuous support throughout my academic journey. Their encouragement and belief in me have been my greatest source of strength. I am also deeply grateful to my siblings, especially my sister Nurul Najihah binti Mat Zamzuri, for always being there, offering emotional support and encouraging me during both difficult and joyful moments. A special thanks goes to my special friend, Shazwan Hadi bin Abdul Ghani, for his constant encouragement, understanding, patience and moral support. His presence, motivation and kindness have made this journey less stressful and more meaningful. My heartfelt thanks are extended to my lab mates and postgraduate friends, Muizzuddin, Azhan, Jannah, Najihah, Syahnaz, Irfan, Kak Adrina and Kak Alia, who have always been willing to help, share knowledge, offer encouragement and support me throughout this journey. Thank you for the late-night discussions, laboratory assistance and for always being there to pick me up whenever and wherever needed. Your friendship, kindness and teamwork have truly made my postgraduate journey more meaningful and memorable. I would also like to extend my sincere appreciation to the laboratory staff for their assistance, technical support and cooperation throughout my experimental work. Their help and guidance were invaluable in ensuring that my research work was conducted smoothly. Finally, I would like to thank everyone who has directly or indirectly contributed to the completion of this thesis. May Allah SWT reward all of you with the best in this world and the hereafter. Aminn.

TABLE OF CONTENTS

	Page
CONFIRMATION BY PANEL OF EXAMINERS	ii
AUTHOR'S DECLARATION	iii
ABSTRACT	iv
ACKNOWLEDGEMENT	v
TABLE OF CONTENTS	vi
LIST OF TABLES	x
LIST OF FIGURES	xi
LIST OF SYMBOLS	xiii
LIST OF ABBREVIATIONS	xiv
LIST OF NOMENCLATURE	xv
CHAPTER 1 INTRODUCTION	1
1.1 Research Background	1
1.2 Problem Statement	2
1.3 Research Objectives	3
1.4 Significance of Study	4
1.5 Scope and Limitation	5
CHAPTER 2 LITERATURE REVIEW	7
2.1 Methyl Orange	7
2.1.1 Structure and Properties of Methyl Orange	8
2.1.2 Toxicity and Regulatory Concern	9
2.2 Conventional Treatment Methods for Dye Removal	12
2.3 Adsorption Process	13
2.3.1 Principle of Adsorption	16
2.3.2 Effect of Adsorbent Dosage	19
2.3.3 Effect of pH Solution	20
2.3.4 Effects of Methyl Orange Concentration	21

2.3.5	Effects of Contact Time	22
2.3.6	Desorption and Regeneration in Adsorption Studies	23
2.4	Adsorbent Used for Methyl Orange Removal	24
2.5	Layered Double Hydroxides	26
2.5.1	Synthesis of LDHs	27
2.5.2	Application of LDHs in Adsorption	29
2.5.3	Desorption and Regeneration of LDHs	30
2.6	Ultraviolet Visible Spectroscopy	31
2.6.1	Analytical Detection of Methyl Orange Using UV-Vis	32
2.7	X-ray Diffraction	34
2.8	Fourier Transform Infrared Spectroscopy	35
2.9	Scanning Electron Microscopy with Energy Dispersive X-ray	36
2.10	Response Surface Methodology in Adsorption Studies	37
2.11	Application of LDHs in Treating Wastewater	39
CHAPTER 3 RESEARCH METHODOLOGY		42
3.1	Materials and Instruments	42
3.2	List of Apparatus	42
3.3	Synthesis of MgAl- Layered Double Hydroxide (MgAl-LDH)	42
3.4	Characterisation Methods	43
3.4.1	X-ray Diffraction	43
3.4.2	Fourier Transform Infrared Spectroscopy	44
3.4.3	Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy	44
3.5	Design of Experiment by Central Composite Design	44
3.5.1	Standard Calibration Method of Methyl Orange	45
3.5.2	Experimental Method	46
3.5.3	Removal Efficiency	48
3.6	Statistical Analysis using Analysis of Variance (ANOVA)	48
3.7	Model Validation method	49
3.7.1	Verification Model	49
3.7.2	Optimum Conditions Validation	50
3.7.3	Relative Standard Error	50

3.8	Adsorption Study	50
3.8.1	Isotherm Study	51
3.8.2	Kinetic Study	52
3.9	Real Sample	53
3.10	Reusability Study	54
3.11	Method Flowchart	56
CHAPTER 4 RESULTS AND DISCUSSION		57
4.1	Synthesis of MgAl-Layered Double Hydroxide (MgAl-LDH)	57
4.2	Characterisation of LDH	58
4.2.1	X-ray Diffraction	59
4.2.2	Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy	61
4.2.3	Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy	63
4.3	Standard Curve for Methyl Orange Removal	66
4.4	Optimisation of Adsorption	67
4.4.1	Determination of Selected Model	68
4.4.2	Accuracy of Model	71
4.4.3	Analysis of Model (ANOVA)	75
4.4.4	Interaction between Variables	77
4.4.5	Verification Model	84
4.4.6	Optimum Condition of Methyl Orange Removal	85
4.5	Adsorption Study of the MgAl-LDH onto Methyl Orange	87
4.5.1	Mechanism of the adsorption	87
4.5.2	Isotherm Study	90
4.5.3	Kinetic Study	92
4.6	Desorption and Reusability Performance	95
4.7	Real Samples Application	96
CHAPTER 5 CONCLUSION		99
5.1	Conclusion	99
5.2	Recommendation	100

REFERENCES	103
APPENDICES	114
AUTHOR’S PROFILE	118

LIST OF TABLES

Tables	Title	Page
Table 2.1	Summary on Previous Studies for Adsorbent of Methyl Orange Removal	25
Table 3.1	Parameter and Level for CCD of Removal Methyl Orange	45
Table 3.2	Dilution of Methyl Orange Standard Solution for Calibration Curve	46
Table 3.3	Experimental Run and Factor from RSM for Removal Methyl Orange	47
Table 4.1	EDX Data of Element in MgAl-LDH	65
Table 4.2	Experimental Run, Factors and Response for the Removal Methyl Orange by MgAl-LDH	68
Table 4.3	Sequential Model Sum of Square for Removal Methyl Orange by MgAl-LDH	69
Table 4.4	Lack of Fit Test of Removal Methyl Orange by MgAl-LDH	70
Table 4.5	Model Summary Statistic of Removal Methyl Orange by MgAl-LDH	70
Table 4.6	Analysis of Model of Removal of Methyl Orange by MgAl-LDH	76
Table 4.7	Actual and Predicted for Verification Model of Methyl Orange Removal	84
Table 4.8	Optimum Validation Condition of Methyl Orange Removal	85
Table 4.9	Actual and Predicted Data for Optimum Conditions of Methyl Orange Removal	86
Table 4.10	Parameters of Isotherm Models for Methyl Orange Removal	92
Table 4.11	Parameter of Kinetics Model for Methyl Orange Removal	93
Table 4.12	Adsorption-Desorption Result for Methyl Orange Removal	95
Table 4.13	Percentage of Methyl Orange Removal on Real Sample Application	97

LIST OF FIGURES

Figures	Title	Page
Figure 2.1	Chemical Structure of Methyl Orange	8
Figure 2.2	Schematic Adsorption Process in Wastewater Treatment	14
Figure 2.3	Possible Adsorption Mechanism of Layered Double Hydroxide	15
Figure 2.4	Structure of Layered Double Hydroxide	27
Figure 2.5	Possible Method Synthesis of Layered Double Hydroxide.	29
Figure 3.1	Flowchart Methodology of Removal Methyl Orange	56
Figure 4.1	Schematic Illustration on the Formation of MgAl-LDH	58
Figure 4.2	XRD Spectrum of MgAl-LDH	60
Figure 4.3	FTIR Spectrum of MgAl-LDH	62
Figure 4.4	SEM Photograph of MgAl-LDH at a) 20 μ m, b) 2 μ m, c) 500 nm and d) 500 nm	63
Figure 4.5	EDX Data of MgAl-LDH (a) Site 1 and (b) Site 2	65
Figure 4.6	Standard Calibration Curve of Methyl Orange	66
Figure 4.7	Normal Plot of Removal Methyl Orange by MgAl-LDH	71
Figure 4.8	Residuals vs Predicted Plot of Removal Methyl Orange by MgAl-LDH	72
Figure 4.9	Predicted vs Actual Plot of Removal Methyl Orange by MgAl-LDH	72
Figure 4.10	Residual vs Run Plot of Removal Methyl Orange by MgAl-LDH	73
Figure 4.11	Cook's Distance plot of Removal Methyl Orange by MgAl-LDH	74
Figure 4.12	Box-Cox plot of Removal Methyl Orange by MgAl-LDH	74
Figure 4.13	3D Surface Plot Methyl Orange Removal: a) Interaction between Adsorbent Dosage and pH Solution (AB); b) Interaction between Adsorbent Dosage and Initial Methyl Orange Concentration (AC)	79
Figure 4.14	3D Surface Plot of Methyl Orange Removal: a) Interaction between Adsorbent Dosage and Contact Time (AD); b) Interaction between pH Solution and Initial Methyl Orange Concentration (BC)	80

Figure 4.15	3D Surface Plot of Methyl Orange Removal: a) Interaction between Contact Time and pH Solution (DB); b) Interaction between Initial Methyl Orange Concentration and Contact Time (CD)	82
Figure 4.16	Optimum Condition Model of Methyl Orange Removal	86
Figure 4.17	Schematic Illustration of the Adsorption of Methyl Orange Using MgAl-LDH	89
Figure 4.18	Langmuir Plot of Methyl Orange Removal	91
Figure 4.19	Freundlich Plot of Methyl Orange Removal	91
Figure 4.20	Pseudo-Second Order Plot of Methyl Orange Removal	93
Figure 4.21	Adsorption-Desorption Cycle Performance for Methyl Orange Removal	95

LIST OF SYMBOLS

Symbols

g	Mass of Adsorbent
L	Volume of Solution
min	Contact Time
ppm	Initial and Equilibrium Concentration of Methyl Orange
%R	Percentage Dye Removal Efficiency
R^2	Coefficient of Determination

LIST OF ABBREVIATIONS

Abbreviations

ANOVA	Analysis of Variance
CCD	Central Composite Design
EDX	Energy Dispersive X-ray Spectroscopy
FTIR	Fourier Transform Infrared Spectroscopy
LDH	Layered Double Hydroxide
MgAl-LDH	Magnesium-Aluminium Layered Double Hydroxide
MO	Methyl Orange
PFO	Pseudo-First Order
PSO	Pseudo-Second Order
RSE	Relative Standard Error
RSM	Response Surface Methodology
SEM	Scanning Electron Microscopy
UV-Vis	Ultraviolet Visible Spectroscopy
XRD	X-ray Diffraction

LIST OF NOMENCLATURE

Nomenclatures

Adsorbate	Substance Being Adsorbed (Methyl Orange)
Adsorbent	Material Used to Remove Adsorbate (MgAl-LDH)
Adsorption Capacity	Amount of Adsorbate Adsorbed Per Unit Mass of Adsorbent
Adsorption Isotherm	Relationship Between Adsorbate Concentration and Adsorption at Equilibrium
Adsorption Kinetics	Study of Adsorption Rate and Time Dependence
Brucite-Like Layers	Positively Charged Hydroxide Layers in LDH Structure
Co-Precipitation	Synthesis Method Involving Simultaneous Precipitation of Metal Ions
Desorption	Release of Adsorbed Molecules from Adsorbent Surface
Equilibrium	State Where Adsorption and Desorption Rates are Equal
Interlayer Anions	Anions Located Between LDH Layers
Isotherm Model	Mathematical Model Describing Adsorption Equilibrium
Kinetic Model	Mathematical Model Describing Adsorption Rate
Regeneration	Process of Restoring Adsorbent for Reuse
Reusability	Ability of Adsorbent to be Reused Over Multiple Cycles
Response Surface Methodology	Statistical Technique for Process Optimisation

CHAPTER 1

INTRODUCTION

1.1 Research Background

Industrial development in sectors such as textiles, leather, paper and plastics has led to the widespread use of synthetic dyes in various manufacturing processes. The discharge of dye containing effluents into the environment has become a major concern due to their high chemical stability, strong colour intensity and resistance to biodegradation (Abdolhossein Rejali and Dinari, 2025). Among these dyes, methyl orange, an anionic azo dye, is one of the most used compounds in textile and analytical industries as a pH indicator and colorant. However, its release into water bodies even at low concentrations can cause serious environmental and health problems, such as aesthetic pollution, toxicity to aquatic organisms and inhibition of photosynthesis due to reduced light penetration (Al-khuder and Karam, 2025).

Several treatment techniques have been applied for dye containing wastewater in many studies, including coagulation-flocculation, membrane filtration, biological treatment (Kathawala et al., 2021), photocatalysis, advanced oxidation processes and electrochemical treatment (Palani et al., 2021). However, some of these methods may require high operating costs, generate secondary sludge, involving complex operating conditions or shows limited efficiency for stable synthetic dyes such as methyl orange (Rong et al., 2024). Adsorption was selected in this study because it is a simple, cost effective and efficient treatment method that can remove dye pollutants from aqueous solution under mild operating conditions (Taheri et al., 2022). In addition, adsorption does not require complicated instrumentation and can be improved through the development of suitable adsorbent materials (Sonawane et al., 2025). Therefore, MgAl-LDH was chosen as the adsorbent due to its positively charged layered structure, exchangeable interlayer anion and strong affinity toward anionic dyes such as methyl orange.

Among the various adsorbents developed, layered double hydroxides (LDHs) have attracted significant attention because of their unique layered structure, tuneable chemical composition, high surface area and excellent anion exchange capacity (Daud

et al., 2019). In this study, magnesium and aluminium ions are used to synthesise magnesium-aluminium layered double hydroxide (MgAl-LDH) through a co-precipitation method, which is simple and effective for producing high-purity LDHs.

Since the adsorption efficiency depends on multiple factors including pH of the solution, adsorbent dosage, contact time and dye concentration, an optimisation technique is necessary to determine the best conditions for maximum removal. Response Surface Methodology (RSM) is a statistical and mathematical tool that efficiently analyses the combined effects of multiple parameters and identifies optimal operating conditions with fewer experimental runs. Hence, this study focuses on optimising the adsorption of methyl orange onto MgAl-LDH using RSM.

1.2 Problem Statement

The continuous discharge of dye containing wastewater from textile, dyeing, paper and printing industries remain a serious environmental concern worldwide. Synthetic dyes such as methyl orange are extensively used due to their chemical stability, strong colour intensity and resistance to biodegradation. However, these same properties make methyl orange difficult to remove using conventional wastewater treatment processes. The presence of methyl orange in aquatic systems contributes to aesthetic pollution, reduces light penetration, disrupts photosynthetic activity and poses potential toxic effects to aquatic organisms and human health (Kainth et al., 2024). Therefore, the development of effective, economical and environmentally sustainable treatment methods for dye removal remains an urgent priority.

Among available treatment techniques, adsorption has emerged as one of the most promising approaches due to its operational simplicity, high removal efficiency and minimal generation of harmful by products (Elbager et al., 2025). In recent years, layered double hydroxides (LDHs) particularly magnesium-aluminium LDH (MgAl-LDH) have gained increasing attention as potential adsorbents for anionic dyes (Ahmad et al., 2025). Their unique brucite-like layered structure, tuneable metal composition, high surface area and excellent anion exchange capacity make it suitable for the removal of negatively charged dye molecules such as methyl orange (Taoufik et al., 2022).

Secondly, adsorption isotherm models, such as Langmuir and Freundlich, provide valuable information on the surface properties and adsorption capacity of the

adsorbent, while kinetic models help explain the rate and controlling steps of the adsorption process (Kanwal et al., 2024). These analyses are often performed independently without integration with statistical optimisation, equilibrium modelling and kinetic analysis within a single study is still lacking.

Furthermore, limited attention has been given to assessing the applicability of pristine MgAl-LDH in real wastewater matrices contains competing ions and complex constituents that may influence adsorption efficiency. Without testing under realistic conditions, the practical feasibility of MgAl-LDH for industrial wastewater treatment remains uncertain. In addition, although LDHs possess anion exchange capability, their long term structural stability and adsorption performance over repeated adsorption-desorption cycles are not sufficiently reported.

Therefore, a systematic and comprehensive investigation is required to address these gaps. This study aims to synthesise and characterise MgAl-LDH, optimise methyl orange adsorption conditions using response surface methodology, evaluate adsorption behaviour through isotherm and kinetic modelling and assess its applicability textile wastewater as well as its reusability performance. By addressing these limitations, this research seeks to provide a clearer understanding of the adsorption mechanism and the practical potential of MgAl-LDH as an effective adsorbent for dye contaminated wastewater treatment.

1.3 Research Objectives

- a) To synthesise the MgAl-LDH adsorbents and characterise their structural properties and surface functional group using Fourier-transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy with Energy Dispersive X-ray (SEM-EDX) and X-ray Diffraction (XRD).
- b) To optimise the adsorption conditions for methyl orange removal, including MgAl-LDH adsorbent amount, pH solution, initial methyl orange concentration and contact time using response surface methodology.
- c) To evaluate the adsorption efficiency of the MgAl-LDH adsorbents towards methyl orange using adsorption mechanism, isotherm and kinetics models, reusability and real wastewater treatment.

1.4 Significance of Study

The discharge of dye containing wastewater from textile industries remains a major environmental concern due to the high colour intensity, chemical stability and potential toxicity of synthetic dye such as methyl orange. Even at low concentrations, methyl orange can significantly reduce light penetration in water bodies, disrupt aquatic ecosystems and pose risks to human health (A et al., 2022). Conventional wastewater treatment methods often show limited efficiency in removing anionic dyes, highlighting the need for effective, economical and environmentally friendly treatment technologies (Abdolhossein Rejali and Dinari, 2025).

LDHs particularly MgAl-LDH, have attracted increasing attention as promising adsorbents for dye removal due to their unique layered structure, high surface area, tuneable composition and strong affinity toward anionic contaminants (Taoufik et al., 2022). Unlike many chemically modified or composite adsorbents, unmodified MgAl-LDH offers advantages such as simpler synthesis, lower chemical consumption and reduced environmental impact. The use of adsorption in this study is significant because it offers a simple and practical approach for methyl orange removal (Taheri et al., 2022), while the application of MgAl-LDH provides additional advantages through electrostatic attraction and anion-exchange properties toward anionic dye molecule. Therefore, investigating the adsorption performance of non-modified MgAl-LDH provides valuable insight into its practical feasibility for real wastewater treatment applications (Zaghloul et al., 2020). This study is significant because it systematically evaluates the adsorption efficiency of MgAl-LDH toward methyl orange using both synthetic and textile wastewater. The application of response surface methodology (RSM) allows for the optimisation of key operational parameters, including adsorbent amount, pH solution, initial methyl orange concentration and contact time. By employing RSM, the study minimises experimental runs while providing statistically reliable results, which is crucial for scaling up adsorption processes and improving industrial applicability.

Furthermore, the study contributes to a deeper understanding of the adsorption mechanism through isotherm and kinetic modelling. The evaluation of Langmuir and Freundlich isotherm models provides insight into the nature of dye-adsorbent interactions, surface homogeneity and adsorption capacity of MgAl-LDH. Meanwhile,

kinetic studies using pseudo-first order and pseudo-second-order models help identify the rate-controlling steps of the adsorption process (Waluyo et al., 2025). Such information is essential for predicting adsorption behaviour under different operating conditions and designing efficient treatment systems. Another important contribution of this research lies in the validation of optimised conditions using textile wastewater samples. Many adsorption studies are limited to synthetic dye solutions, which do not accurately represent complex industrial effluents. By applying MgAl-LDH to actual wastewater and comparing predicted and experimental dye removal efficiencies, this study demonstrates the robustness and reliability of the developed adsorption model. This enhances the practical relevance of the findings and supports the potential implementation of MgAl-LDH in real world wastewater treatment.

The novelty of this study lies in its comprehensive evaluation of methyl orange adsorption onto MgAl-LDH by integrating several important adsorption aspects within one systematic study. Instead of focusing only on dye removal efficiency, this work combines material synthesis and characterisation, statistical optimisation using RSM, adsorption mechanism interpretation, isotherm and kinetic modelling, reusability assessment and application to real textile wastewater. This integrated approach provides a more complete understanding of the adsorption performance of MgAl-LDH and its practical potential for wastewater treatment. In particular, the application of the optimised adsorption conditions to real textile wastewater strengthens the environmental relevance of the studies, as it demonstrates the performance of MgAl-LDH under more realistic conditions rather than only in synthetic methyl orange solutions.

1.5 Scope and Limitation

This study focuses on the synthesis and application of MgAl-LDH for the removal of methyl orange from aqueous solution. MgAl-LDH was synthesised using the co-precipitation method with a Mg:Al ratio of 3:1 and carbonate as the interlayer anion. The synthesised material was characterised using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX) to confirm the formation, functional groups, surface morphology and elemental composition of the adsorbent.

The adsorption study was limited to methyl orange as the target anionic dye. The effects of selected adsorption parameters which including adsorbent dosage, solution pH, initial methyl orange concentration and contact time were investigated and optimised using Response Surface Methodology (RSM). The adsorption performance was further evaluated through isotherm, kinetic, reusability, desorption and real textile wastewater application studies. The real wastewater test was conducted to assess the applicability of MgAl-LDH under more realistic conditions compared with synthetic methyl orange solution.

However, several limitations were identified in this study. Firstly, only unmodified MgAl-LDH was investigated which the effect of surface modification, magnetic functionalisation or incorporation of the other metal oxides on adsorption performance was not evaluated. Secondly, carbonate was used as the interlayer anion and the effect of other interlayer anion such as nitrate, sulphate or organic functional anion was not studied. Since carbonate has a strong affinity toward LDH layers, the accessibility of large dye molecules into the interlayer region may be limited. Thirdly, the study focused only on methyl orange as single model dye whereas industrial wastewater may contain mixed dyes, heavy metals, pharmaceutical, surfactants and other competing contaminants. Therefore, competitive adsorption in multi-component systems was beyond the scope of this study. In addition, the point of zero charge, pH_{pzc} of MgAl was not determined in this study. The absence of pH_{pzc} analysis limits the direct explanation of the relationship between solution pH, adsorbent surface charge and methyl orange behaviour. Thermodynamic studies were also not conducted which the spontaneity and energetic nature of the adsorption process were not evaluated. These limitations provide useful directions for future studies to improve the understanding and practical application of MgAl-LDH in wastewater treatment.

CHAPTER 2

LITERATURE REVIEW

2.1 Methyl Orange

Methyl orange is one of the most widely used synthetic azo dyes in laboratories and industries due to its intense coloration, chemical stability and strong pH sensitivity. It belongs to the class of anionic azo dyes that contain one or more azo groups ($-\text{N}=\text{N}-$) linking aromatic rings, which are responsible for their vivid colours. Methyl orange is commonly utilised as an acid–base indicator in titration experiments and in the textile, paper, leather and food industries for dyeing purposes (A et al., 2022). However, despite its usefulness, the discharge of methyl orange into aquatic environments has raised serious environmental and health concerns due to its toxicity, persistence and non-biodegradable nature (El Bourachdi et al., 2025).

Chemically, methyl orange is an anionic dye, meaning it tends to dissociate in aqueous solutions and can strongly interact with oppositely charged surfaces. Its molecular formula is $\text{C}_{14}\text{H}_{14}\text{N}_3\text{NaO}_3\text{S}$, with a molecular weight of approximately 327.33 g/mol. It displays a bright orange colour in acidic medium and turns yellow in alkaline medium due to changes in its molecular structure upon protonation or deprotonation. This colour transition makes it a popular pH indicator in acid–base titrations, which can be detected by a UV-Vis spectrophotometer (Sulistyaningsih et al., 2024).

Methyl orange was selected as the target dye in this study because it represents a suitable model pollutant for evaluating the adsorption performance of MgAl-LDH. Structurally, methyl orange which containing an azo bond and a sulfonate group that contribute to its stability, water solubility and persistence in aqueous environments (A et al., 2022). The presence of the negatively charged sulphonate group makes methyl orange highly relevant for adsorption studies involving layered double hydroxides, since MgAl-LDH possesses positively charged hydroxide layers and exchangeable interlayer anions. Therefore, methyl orange is not only environmentally significant as a dye pollutant, but also chemically suitable for investigating electrostatic attraction and anion exchange interactions with MgAl-LDH (Yang et al., 2023). Compared with cation

dyes, methyl orange provides a more appropriate model for studying the anion removal ability of MgAl-LDH. In addition, its strong colour and measurable absorbance in the visible region make it suitable for quantitative analysis using UV-Vis spectrophotometry (Zaghloul et al., 2020). Thus, the selection of methyl orange in this study was based on its environmental relevance, chemical stability, anionic nature and compatibility with the adsorption properties of MgAl-LDH.

2.1.1 Structure and Properties of Methyl Orange

Methyl orange (sodium 4-[(4-dimethylamino)phenylazo] benzenesulfonate) is composed of two aromatic rings connected by an azo ($-N=N-$) linkage. One aromatic ring bears a dimethyl amino group ($-N(CH_3)_2$), which acts as an electron-donating substituent, while the other ring contains a sulfonate group ($-SO_3^-$), which provides water solubility and ionic character to the dye molecule.

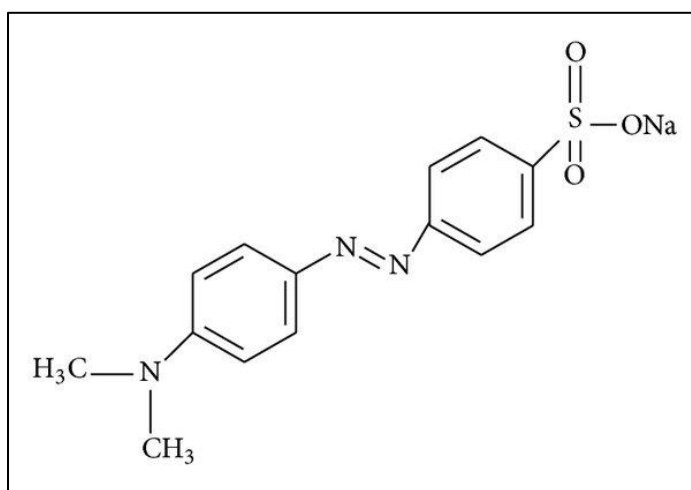


Figure 2.1 Chemical Structure of Methyl Orange

Figure 2.1 shows a chemical structure of methyl orange where C represents carbon atoms forming the aromatic backbone of dye molecule, H represent hydrogen atoms attached to the aromatic rings and substituent groups, N corresponds to the nitrogen atoms present in azo ($-N=N-$) linkage and amino group, Na represents the sodium ion associated with the sulfonate group, O indicates oxygen atoms within the sulfonate ($-SO_3^-$) functional group and S represents the sulphur atom in the sulfonate (Alzain et al., 2023). The presence of conjugated double bonds and aromatic rings

allows extensive π - π electron delocalisation, which is responsible for the dye's strong coloration. This delocalisation gives rise to characteristic adsorption peaks in the visible region of the electromagnetic spectrum. The azo linkage ($-\text{N}=\text{N}-$) is the chromophore group that provides the distinctive colour of methyl orange (Al-khuder and Karam, 2025).

Methyl orange exhibits pH-dependent behaviour, acting as a weak acid. In acidic medium ($\text{pH} < 3.1$), it exists primarily in its protonated form, which absorbs light around 510 nm, giving an orange-red colour. In alkaline medium ($\text{pH} > 4.4$), deprotonation occurs and the dye shifts to a yellow-coloured anionic form that absorbs around 464 nm (Daud et al., 2019). The reversible colour change forms the basis of its function as a pH indicator.

Physiochemically, methyl orange is highly soluble in water, poorly biodegradable and chemically stable under normal environmental conditions. Its molecular structure is relatively resistant to oxidation and microbial degradation, making it a persistent pollutant once discharged into water systems (Ali et al., 2025). The negatively charged sulfonate group allows it to interact with positively charged surfaces, such as metal oxides or LDHs, through electrostatic attraction, making adsorption a suitable technique for its removal.

2.1.2 Toxicity and Regulatory Concern

The toxicity of methyl orange and other azo dyes has become a major environmental and public health concern. As reported in American Chemical Society Study (Ali et al., 2019), methyl orange is classified as a toxic, carcinogenic, tumorigenic, mutagenic and genotoxic compound. The azo ($-\text{N}=\text{N}-$) functional group present in methyl orange can undergo reductive cleavage under anaerobic conditions, producing aromatic amines that are known to be more toxic and potentially carcinogenic than the parent compound.

Once released into the environment, methyl orange can undergo partial degradation through photolysis or microbial activity. However, these processes are often incomplete, leading to the formation of toxic aromatic amines such as sulfonic acid and N,N-dimethyl aniline known to be carcinogenic and mutagenic which posing significant risks to aquatic organisms and humans exposed to contaminated water (Badri

et al., 2022). Furthermore, methyl orange is resistant to biodegradation due to its complex aromatic structure and azo linkage ($-N=N-$), which limits microbial breakdown under aerobic conditions (Al-khuder and Karam, 2025). As a result, the compound tends to persist in the environment, increasing its potential for bioaccumulation and transfer through the food chains.

In addition, methyl orange contamination can negatively affect water quality by altering pH and increasing chemical oxygen demand (COD). High COD levels indicate that the water contains large amounts of oxidisable organic material (Alzain et al., 2023), which can consume oxygen needed for aquatic organisms. Persistent dye contamination can also cause unpleasant odors, affect aesthetic value and reduce the usability of water for domestic or agricultural purposes. Even at low concentrations, the intense coloration of methyl orange can block sunlight penetration into water bodies, reducing photosynthetic activity in aquatic plants and algae. This interference in photosynthesis disrupts oxygen production and causes a decrease in dissolved oxygen levels, leading to the loss of biodiversity (Zheng et al., 2019).

From an ecological perspective, exposure to methyl orange has been linked to growth inhibition, oxidative stress and reproductive toxicity in aquatic species such as *Daphnia magna* and *Danio rerio*. Prolonged exposure to azo dyes in wastewater has also been associated with disruption of enzymatic functions and DNA damage in aquatic microorganisms (Alzain et al., 2023). These adverse effects demonstrate the far-reaching implications of dye pollution beyond visible coloration, highlighting its biochemical and genetic toxicity in the environment.

In human health perspective, exposure to methyl orange and related azo dyes has been associated with allergic reactions, liver dysfunction, kidney damage, nervous system disorders and potentially damage to internal organs after long-term exposure (El Bourachdi et al., 2025). The aromatic amine intermediates formed during partial degradation can interact with DNA which led to mutagenic and carcinogenic effects (Li et al., 2025). Even at low concentrations, these azo dyes may cause chronic toxicity due to their bioaccumulation potential and prolonged environmental persistence. Therefore, continuous discharge of untreated dye represents a significant public health concern, particularly in regions with insufficient wastewater treatment (Hosseinpour and Rahbar-Kelishami, 2025).

Because of these hazards, regulatory authorities worldwide have implemented strict controls on azo dyes due to their potential to release hazardous aromatic amines. The studies conducted by Li et al. (2025) and Alzain et al. (2023) stated that the United State Environmental Protection Agency (US EPA), through its Benzidine Dyes Action Plan (2010), highlighted that although azo dyes may be present in textiles and various consumer products, regulatory attention should primarily focus on occupational exposure and the control of industrial emissions. In the European Union, strict regulation governs the use of azo dyes in consumer goods including textiles, cosmetics, toys and food related products. Under the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), regulation sheet No 1907/2006, 24 aromatic amines are classified as hazardous to human health. The legislation prohibits the use of azo dyes that can release these aromatic amines in concentrations equal to or exceeding 30 mg kg^{-1} in products that may come into direct and prolonged contact with human skin. This regulation aims to reduce consumer exposure to potentially carcinogenic and mutagenic compounds.

Several European countries including Germany, the Czech Republic and Switzerland, have also established national regulations that limit the permissible levels of azo dyes and the aromatic amines released from them. In Asia, India was among the first countries to regulate synthetic dyes which introducing restriction in 1997 that covered 112 dyes including methyl orange. The list of prohibited aromatic amines in India corresponds to the list specified under the EU REACH Regulation. Subsequently, other Asian countries implemented similar restriction including China (2005), South Korea (2010), Taiwan (2011) and Egypt (2012) (Alzain et al., 2023).

Despite these regulations, the removal of azo dyes like methyl orange remains challenging due to its high chemical stability and non-biodegradable. Therefore, it is crucial to develop advanced treatment technologies, such as adsorption using efficient and eco-friendly adsorbents, to minimise its environmental impact. The use of materials like LDHs offers a sustainable and cost-effective approach for the removal of methyl orange and other anionic dyes from aqueous systems.

2.2 Conventional Treatment Methods for Dye Removal

The removal of such dyes from wastewater is challenging because of their synthetic origin, complex aromatic molecular structures and resistance to biodegradation (Im et al., 2025). Therefore, the selection of an effective and environmentally sustainable treatment method is essential to minimise their impact on aquatic ecosystems and human health.

Various treatment technologies have been developed and applied for the removal of dyes from aqueous solutions, which can generally be categorised into biological, chemical and physical processes. Each method exhibits distinct advantages and limitations depending on operational conditions, dye type, concentration and wastewater characteristics (Karyab et al., 2023).

Biological treatments, such as aerobic and anaerobic biodegradation, fungal decolourisation and enzyme-based degradation, utilise microorganisms or enzymes to metabolise dyes into simpler compounds. Although these methods are considered eco-friendly and cost-effective, they are time-consuming and highly sensitive to environmental conditions, such as pH, temperature and the presence of toxic dye components. Most azo dyes, including methyl orange, are non-biodegradable or only partially degraded under aerobic conditions because of the stability of their azo ($-N=N-$) bonds. Anaerobic degradation may reduce azo dyes to aromatic amines, but these intermediates are often toxic and carcinogenic, which further complicates wastewater treatment. Therefore, biological methods are generally less effective for treating highly stable synthetic dyes (Kathawala et al., 2021).

Chemical oxidation and reduction processes, such as ozonation, Fenton and photo-Fenton reactions, electrochemical oxidation and advanced oxidation processes (AOPs) can break down complex dye molecules into smaller and less toxic compounds (Palani et al., 2021). However, these methods often require expensive reagents, strict pH control and high energy consumption, which limit their large-scale applicability. For instance, ozonation can effectively degrade dyes, but ozone generation is costly and the process efficiency decreases with the presence of inorganic ions in wastewater. Fenton oxidation, although powerful, generates iron sludge that requires proper disposal. Similarly, photocatalysis using TiO_2 and UV light can degrade dyes but depends heavily

on light intensity, pH and catalyst dosage, making it less practical for continuous treatment systems (Rong et al., 2024).

Physical methods, such as membrane filtration, coagulation–flocculation, precipitation and adsorption, rely on the physical separation or retention of dye molecules. Membrane technologies, including ultrafiltration, nanofiltration and reverse osmosis, can efficiently remove dyes but suffer from membrane fouling, high maintenance cost and limited lifespan (Tabish et al., 2024). Coagulation–flocculation processes are inexpensive and suitable for large-scale operations; however, they produce a large amount of chemical sludge that requires disposal and may cause secondary pollution (Bustos-Terrones et al., 2021).

Among all the physical methods, adsorption has been recognised as the most effective, simple and widely applicable approach for dye removal due to its high efficiency, versatility and environmental compatibility. Adsorption is a surface phenomenon in which dye molecules are transferred from the aqueous phase onto the surface of a solid material (adsorbent) through physical or chemical interactions. The process does not require complex instruments and the adsorbent materials can often be regenerated and reused (Tabish et al., 2024).

2.3 Adsorption Process

Adsorption is one of the most widely applied physicochemical processes for removing contaminants from aqueous solutions due to its simplicity, high efficiency and cost-effectiveness (Aissa et al., 2024). It involves the accumulation of molecules, ions, or atoms (adsorbates) from a liquid or gas phase onto the surface of a solid material (adsorbent) until an equilibrium concentration is achieved as shown in Figure 2.2 (Al-Amrani et al., 2022). In wastewater treatment, adsorption has been recognised as a particularly effective technique for removing organic dyes, heavy metals and other persistent pollutants that are resistant to conventional biological or chemical degradation methods (Taheri et al., 2022).

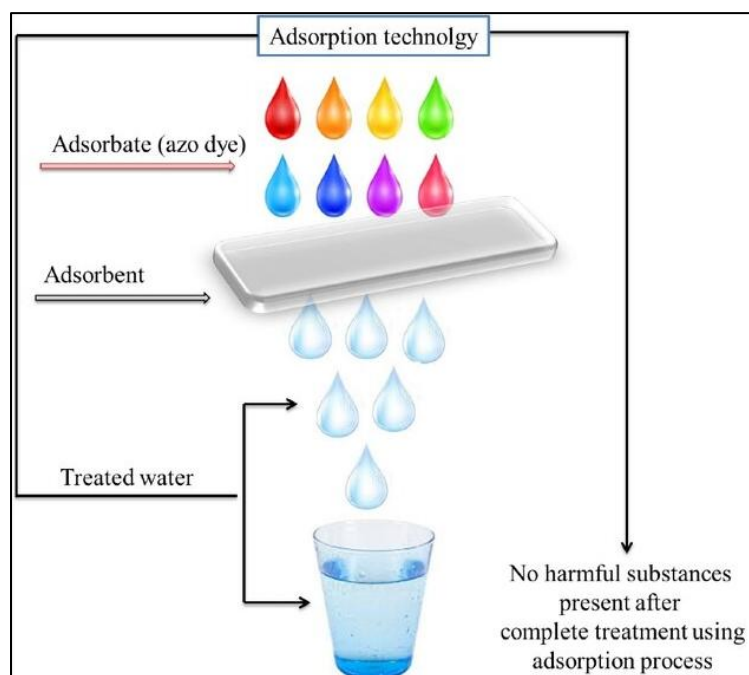


Figure 2.2 Schematic Adsorption Process in Wastewater Treatment

Because of its versatility and performance, adsorption has been successfully applied for the treatment of a wide range of contaminants, including anionic and cationic dyes, heavy metals and pharmaceutical residues. In the context of dye removal, adsorption offers several practical advantages, such as operation under mild conditions, minimal sludge production and ease of adsorbent regeneration (Sonawane et al., 2025). Moreover, it allows for the recovery of valuable materials and can be adapted for both batch and continuous treatment systems. Various materials have been explored as adsorbents, including activated carbon, clays, zeolites, biochar and layered double hydroxides (LDHs)(Al-Amrani et al., 2022).

The efficiency of the adsorption process is influenced by several parameters, including the mass adsorbent, concentration, pH, contact time and temperature. These factors collectively affect the availability of active sites, surface charge of the adsorbent and the interaction strength between adsorbate and adsorbent. The adsorption process is also governed by the nature of the adsorbate molecules, such as their molecular size, polarity, solubility and functional groups (Aissa et al., 2025).

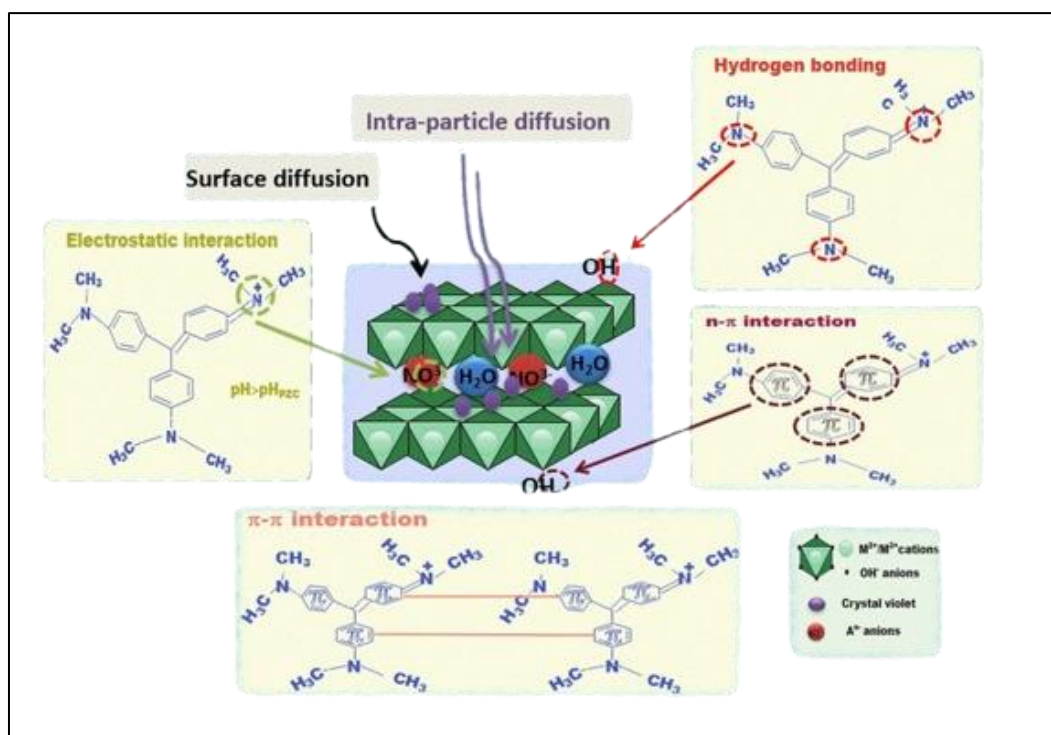


Figure 2.3 Possible Adsorption Mechanism of Layered Double Hydroxide

The adsorption of methyl orange onto LDHs involves a combination of surface interactions and diffusion processes, as illustrated in Figure 2.3 (Abdel-Hady et al., 2023). LDHs possess positively charged brucite-like layers and interlayer anions, making them highly effective for binding anionic dyes such as methyl orange. The mechanism begins with surface diffusion, where methyl orange molecules migrate across the LDH surface before entering the porous structure. This step is followed by intra-particle diffusion, during which the dye molecules penetrate deeper into the LDH layers or pores, contributing to the overall adsorption rate and capacity (Abdel-Hady et al., 2023).

One of the dominant interactions involved in methyl orange adsorption is electrostatic attraction. Because methyl orange exists predominantly in an anionic form under neutral and basic conditions, it readily interacts with the positively charged LDHs surface or the protonated hydroxyl groups. This interaction is especially strong at lower pH values, where surface charge becomes more positive, thereby enhancing binding through Coulombic forces (Ding et al., 2025). The adsorption process is also supported by n- π interactions, particularly between the aromatic rings of methyl orange and the metal-oxygen surface sites or carbon-containing functional groups. These interactions stabilise the dye molecules on the surface by forming electron donor-acceptor

complexes. Additionally, π - π stacking can occur between neighbouring methyl orange molecules, allowing multilayer adsorption in some cases (Lu et al., 2016).

Hydrogen bonding represents another crucial mechanism. The SO_3^- groups and azo ($\text{N}=\text{N}$) functional groups of methyl orange can form hydrogen bonds with surface hydroxyl groups (OH) of LDHs or with water molecules coordinated to metal cations. This enhances the stability of the adsorbed layer and contributes significantly to adsorption at intermediate pH levels. Furthermore, interactions such as π - π interactions may occur between the lone pair electrons on oxygen or nitrogen atoms in methyl orange and vacant orbitals on the metal centre within LDH structures. This type of electron donor acceptor interaction further strengthens adsorption, especially in modified LDHs with transition metal substitution (Abdolhossein Rejali and Dinari, 2025).

2.3.1 Principle of Adsorption

The principle of adsorption is based on the interaction between a solid surface (adsorbent) and solute molecules (adsorbate) present in a liquid or gas phase. When a solid surface is exposed to a fluid containing dissolved molecules, some of these molecules accumulate on the surface due to intermolecular forces such as electrostatic attraction, hydrogen bonding, van der Waals forces, or chemical bonding. This phenomenon results in a higher concentration of adsorbate on the solid surface than in the bulk solution, leading to the formation of an adsorbed layer.

Adsorption can be broadly classified into two categories which is physisorption which is physical adsorption and chemisorption which is chemical adsorption. Physisorption involves weak van der Waals or electrostatic forces, occurs at low temperatures and is usually reversible, allowing easy regeneration of the adsorbent. Chemisorption, on the other hand, involves the formation of stronger covalent or ionic bonds between the adsorbate and the surface sites, resulting in higher binding energy and often irreversible adsorption. Chemisorption typically forms a monolayer, while physisorption may lead to multilayer formation (Sibiya-Dlomo et al., 2025).

The driving force for adsorption arises from the difference in concentration between the bulk solution and the solid surface. At the beginning of the process, the concentration of the solute is high in the liquid phase and low on the solid surface,

creating a concentration gradient. Molecules diffuse toward the adsorbent surface until an equilibrium state is reached, where the rate of adsorption equals the rate of desorption. The amount of solute adsorbed per unit mass of adsorbent at equilibrium is a function of the solute concentration in the solution and is typically described by adsorption isotherms.

Initially, adsorption isotherms describe how solute molecules distribute between the liquid phase and the solid phase at equilibrium under constant temperature. They provide critical information about the adsorption capacity, surface properties and interaction mechanism between adsorbate and adsorbent (Sumardiono et al., 2025). The most applied isotherm models in dye adsorption studies are the Langmuir and Freundlich isotherms.

The Langmuir model assumes monolayer adsorption on a homogeneous surface with a finite number of identical adsorption sites and no interaction between adsorbed molecules (Khazaie et al., 2026). The linear form of the Langmuir equation is expressed as:

$$\frac{C_e}{q_e} = \frac{1}{Q_m b} + \frac{C_e}{Q_m} \quad (2.1)$$

Where C_e is equilibrium concentration of adsorbate in solution (mg/L), q_e is amount of adsorbate adsorbed per unit mass of adsorbent at equilibrium (mg/g), Q_m is maximum monolayer adsorption capacity (mg/g) and b is Langmuir constant related to adsorption energy (L/mg).

The Freundlich model assumes adsorption on a heterogeneous surface with sites of varying affinities and multilayer adsorption (Khazaie et al., 2026). The linearised form of Freundlich equation is given as:

$$\text{Log } q_e = \text{log } K_F + \frac{1}{n} \text{log } C_e \quad (2.2)$$

Where C_e is equilibrium concentration of adsorbate in solution (mg/L), q_e is amount of adsorbate adsorbed per unit mass of adsorbent at equilibrium (mg/g), K_F is

Freundlich constant indicative of adsorption capacity and $1/n$ is heterogeneity factor representing adsorption intensity.

If the value of $1/n < 1$, it indicates favourable adsorption, while $1/n > 1$ suggests cooperative adsorption (Sibiya-Dlomo et al., 2025). The Freundlich model is particularly useful for describing adsorption on non-uniform such as porous materials and composite adsorbents. Other isotherm models such as Temkin, Dubinin-Radushkevich and Redlich-Peterson may also be employed to provide additional insight into the adsorption mechanism, energy distribution and surface heterogeneity but the Langmuir and Freundlich models remain the most widely used for methyl orange adsorption studies.

The second one is kinetics. Adsorption kinetics describe the rate of adsorbate uptake on an adsorbent surface and provide insights into the mechanism and rate-controlling steps of the process, such as external film diffusion, intraparticle diffusion, or surface adsorption (Khazaie et al., 2026). The two most applied kinetic models are the pseudo-first order and pseudo-second-order models.

The pseudo-first-order model assumes that the rate of occupation of adsorption sites is proportional to the number of unoccupied sites. The linearised form is expressed as:

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (2.3)$$

Where q_t is the amount of adsorbate adsorbed at time, t (mg/g), q_e is the adsorption capacity at equilibrium, (mg/g) and k_1 is pseudo-first-order rate constant (1/min). A plot of $\log(q_e - q_t)$ versus t gives a straight line if the model fits the adsorption process, allowing determination of k_1 and q_e .

For pseudo-second order, this model assumes that the adsorption process is controlled by chemisorption involving valence forces through sharing or exchange of electrons between adsorbate and adsorbent (Sibiya-Dlomo et al., 2025). The linearised form is:

$$q_t = \frac{q_e^2 k^2 t}{1 + q_e k^2 t} \quad (2.4)$$

Where q_t is the amount of adsorbate adsorbed at time, t (mg/g), q_e is the adsorption capacity at equilibrium, (mg/g) and k_2 is pseudo-second-order rate constant (g/mg.min). A plot of t/q_t versus t should yield a straight line if the model applies and q_e can be calculated from the slope. Generally, if the experimental data fit better with the pseudo-second-order model, it indicates that chemisorption is the rate-limiting step.

Lastly, thermodynamics of adsorption who further explain the nature and feasibility of the adsorption process (Kanwal et al., 2024). The standard Gibbs free energy change (ΔG°), enthalpy change (ΔH°) and entropy change (ΔS°) are determined using the following equations:

$$\ln K_c = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \quad (2.5)$$

Where K_c is equilibrium constant, R is universal gas constant (8.314 J/mol.K) and T is absolute temperature (K). A negative value of ΔG° indicates a spontaneous process, a positive ΔH° denotes endothermic adsorption and a positive ΔS° suggests increased randomness at the solid-liquid interface (Alshammari et al., 2025).

2.3.2 Effect of Adsorbent Dosage

The amount of adsorbent dosage used in an adsorption process significantly influences the efficiency of pollutant removal. Its dosage determines the availability of active adsorption sites. When the adsorbent dosage increases, the number of adsorption sites also rises, leading to improved pollutant removal efficiency. This trend occurs because more adsorbent material enhances the surface area and active functional groups available for interactions with the adsorbate, such as electrostatic attractions, hydrogen bonding, or π - π interactions (Zhao et al., 2021). The relationship between adsorbent dosage and adsorption efficiency is often modelled using adsorption isotherms, such as the Langmuir and Freundlich models. These models help predict the maximum

adsorption capacity and identify the optimal adsorbent dosage for specific applications. By balancing dosage and efficiency, researchers can minimise material costs while maximising pollutant removal (Kanwal et al., 2024).

However, after a certain dosage, the removal efficiency may plateau due to saturation of the adsorbent or equilibrium between adsorbate molecules and the adsorption sites (Akanda et al., 2024). Excessive amounts of adsorbent dosage can lead to agglomeration, where particles clump together and reduce the effective surface area. This aggregation hinders the availability of active sites and may decrease adsorption efficiency. Other than that, overdosing the adsorbent dosage can introduce operational challenges, such as increased solution viscosity, making it harder to achieve uniform dispersion. This phenomenon highlights the importance of optimising the adsorbent dosage to achieve maximum adsorption efficiency without wastage (Huang et al., 2024).

In addition, in practical applications, the optimal adsorbent dosage is determined experimentally by conducting batch adsorption studies. These studies evaluate pollutant removal efficiency at varying adsorbent dosages while keeping other factors, such as pH, contact time and pollutant concentration, constant. The results typically show an initial rapid increase in adsorption efficiency, followed by a plateau or slight decline at higher adsorbent dosage dosages. This behaviour underscores the need to balance the adsorbent dosage to achieve cost-effectiveness and high pollutant removal performance (Shabani and Dinari, 2021).

2.3.3 Effect of pH Solution

The pH of the solution plays a vital role in controlling the adsorption process, as it directly influences both the surface charge of the adsorbent and the degree of ionisation of the adsorbate. For dye adsorption studies, pH determines the electrostatic interactions between the adsorbent surface and dye molecules, thereby affecting the adsorption efficiency (Shabani and Dinari, 2021). Methyl orange, being an anionic azo dye, exhibits different adsorption behaviour depending on the pH of the solution due to its dissociation properties and the surface chemistry of the adsorbent.

At low pH (acidic conditions), the concentration of H^+ ions is high, leading to protonation of the adsorbent surface. In this state, the surface carries a positive charge,

which facilitates strong electrostatic attraction between the positively charged adsorbent sites and negatively charged sulfonate groups ($-\text{SO}_3^-$) of methyl orange molecules. As a result, the adsorption capacity is typically higher in acidic media. Conversely, at high pH (alkaline conditions), the adsorbent surface becomes negatively charged due to deprotonation of surface hydroxyl groups, which leads to electrostatic repulsion between the negatively charged dye ions and the adsorbent surface, reducing the adsorption efficiency (Wang et al., 2022).

Several studies have confirmed that pH significantly influences the adsorption capacity of MgAl-LDH and other layered double hydroxides. For instance, Abdel-Hady et al., (2023), found that the removal efficiency of dye using ZnMgAl-LDH increased dramatically when pH decreased from 10 to 6, indicating a strong dependence on electrostatic attraction.

The relationship between pH and adsorption efficiency can be illustrated graphically, where a bell-shaped or decreasing curve is typically observed, showing a maximum adsorption at an optimal pH range. This emphasises that maintaining an appropriate pH condition during adsorption is essential for maximising performance (Gao et al., 2024). Hence, optimisation of pH solution is a critical step in adsorption studies to ensure effective dye removal, as it governs both the surface charge behaviour of the adsorbent and the ionisation degree of the adsorbate.

2.3.4 Effects of Methyl Orange Concentration

The initial concentration of methyl orange in solution is an important factor that strongly affects the efficiency of the adsorption process. It determines the driving force for mass transfer between the liquid and solid phases. At lower concentrations, the number of dye molecules available in solution is relatively small compared to the number of available adsorption sites on the adsorbent surface, allowing for rapid and efficient adsorption (Li et al., 2022). As a result, a high percentage of dye removal is usually achieved at low initial concentrations due to the sufficient availability of active sites on the adsorbent surface.

However, as the initial methyl orange concentration increases, the competition among dye molecules for the limited active sites also increases, leading to a decrease in removal efficiency even though the total amount of dye adsorbed per unit mass of

adsorbent (adsorption capacity, q_e), increases. This is because at higher concentrations, the adsorbent surface becomes saturated more quickly, leaving unabsorbed dye molecules in the solution (Akanda et al., 2024). Therefore, the adsorption process typically exhibits a trend where the percentage removal decreases with increasing dye concentration, but the adsorption capacity increases until equilibrium is reached.

The influence of initial methyl orange concentration also affects the rate of adsorption. At higher concentrations, the concentration gradient between the solution and adsorbent surface is greater, which enhances the diffusion of dye molecules onto the adsorbent during the early stages of adsorption (Sukla Baidya and Kumar, 2021). However, as equilibrium is approached, the adsorption rate slows down due to site saturation and reduced mass transfer driving force. Hence, identifying the optimal initial concentration is crucial to ensure high efficiency while minimising the risk of overloading the adsorbent.

Several researchers have reported similar findings. For example, Hosseinpour and Rahbar-Kelishami (2025) observed that increasing the initial dye concentration from 10 mg/L to 250 mg/L resulted in an increase in adsorption capacity of from 7.46 mg/g to 50.46 mg/g, while the overall removal efficiency decreases from 74.59% to 20.18%. Similarly, Zaghloul et al. (2020) reported that at low concentrations, adsorption proceeded rapidly due to an abundance of available sites, but at higher concentrations, site saturation occurred, leading to slower adsorption kinetics and incomplete removal.

2.3.5 Effects of Contact Time

The contact time between the adsorbent and adsorbate is one of the key operational parameters that determines the rate and extent of the adsorption process. It directly influences how quickly the adsorbate molecules interact with the active sites of the adsorbent and reach adsorption equilibrium. In general, dye adsorption occurs in two distinct stages which is a rapid initial phase, followed by a slower equilibrium phase (Krishni et al., 2014).

During the initial stage, many vacant and accessible active sites are available on the adsorbent surface, leading to rapid uptake of dye molecules from the solution. This stage is mainly controlled by external surface adsorption, where dye molecules adhere to the outer layer of the adsorbent. As time progresses, these surface sites

become occupied and dye molecules must diffuse deeper into the internal pores of the adsorbent structure. Consequently, the adsorption rate slows down and gradually reaches equilibrium when no further adsorption occurs due to saturation of available sites or establishment of dynamic equilibrium between adsorption and desorption processes (Gkika et al., 2025).

The optimum contact time varies depending on factors such as adsorbent surface area, porosity, particle size and dye concentration. An excessively short contact time may result in incomplete adsorption, whereas excessively long durations do not significantly improve removal efficiency once equilibrium has been achieved, thus reducing process efficiency. Determining the equilibrium time is therefore crucial for designing cost-effective and time-efficient adsorption systems.

Several studies have demonstrated that methyl orange adsorption using MgAl-LDH follows this general behaviour. For instance, Zaghloul et al. (2020), reported that approximately 90% of methyl orange removal occurred within the first 10 minutes of contact, with equilibrium being reached after 20–30 minutes.

The relationship between contact time and adsorption capacity is typically analysed using kinetic models such as the pseudo-first order and pseudo-second order models to determine the mechanism controlling the adsorption rate. A short equilibrium time combined with high removal efficiency indicates a high affinity between the dye and adsorbent surface, as well as good accessibility of adsorption sites. In contrast, longer contact times may suggest diffusion limitations within the pores or slower chemical interactions.

2.3.6 Desorption and Regeneration in Adsorption Studies

Desorption is an essential component of adsorption studies as it provides critical insight into the reversibility of adsorption processes, adsorbent regeneration and long-term applicability of adsorption systems in water and wastewater treatment. While adsorption focuses on the removal of contaminants from aqueous solution, desorption examines the ability of the adsorbent to release the retained pollutants under specific conditions, thereby allowing the adsorbent to be reused.

In general, desorption studies are conducted to evaluate the sustainability of adsorption-based treatment technologies. An adsorbent that exhibits high adsorption

efficiency, but poor desorption performance may not be suitable for large scale applications due to frequent replacements (Badri et al., 2022). Therefore, regeneration and reuse through effective desorption are considered key criteria in assessing the practical potential of adsorbent materials.

The desorption process is strongly influenced by the nature of interactions between the adsorbate and adsorbent. Adsorption mechanisms such as physisorption, electrostatic attraction and hydrogen bonding are typically reversible and allow for high desorption efficiency. In contrast, chemisorption involving strong covalent or complexation interactions often results in low desorption efficiency, indicating irreversible adsorption. As a result, desorption behaviour is frequently used as indirect evidence to distinguish between physical and chemical adsorption mechanism (Sonawane et al., 2025).

Various desorbing agents have been reported in the literature, including acidic solutions, alkaline solutions, salt solutions and organic solvents (Kameda et al., 2021). The selection of an appropriate desorbing agent depends on the chemical nature of the adsorbate, the surface properties of the adsorbent and the dominant adsorption mechanism. For anionic contaminants, alkaline solutions are often effective due to competitive anion exchange and electrostatic repulsion, whereas acidic solutions are commonly employed for the desorption of cationic species.

In addition to desorption efficiency, repeated adsorption-desorption cycles are commonly performed to assess adsorbent stability and durability. A gradual decline in adsorption capacity over multiple cycles may occur due to partial pore blockage, loss of active sites or structural degradation of the adsorbent. Nevertheless, adsorbents that retain a significant portion of their initial adsorption capacity after several regeneration cycles are considered promising for practical applications (Li and Lu, 2025).

2.4 Adsorbent Used for Methyl Orange Removal

A wide range of adsorbents have been investigated for the removal of dyes such as methyl orange from aqueous solutions. These include natural materials, synthetic polymers, activated carbon, clay minerals, metal oxides and adsorbent dosages. Each adsorbent type exhibits different adsorption capacities and efficiencies, depending on its surface area, porosity, functional groups and surface chemistry. Table 2.1

summarises several adsorbents that have been studied for methyl orange removal, along with their maximum adsorption capacities and specific features.

Table 2.1
Summary on Previous Studies for Adsorbent of Methyl Orange Removal

Adsorbent	Type	Results	Previous study
Activated carbon	Commercial activated carbon	High surface area but costly to regenerate	(Wang et al., 2024)
Chitosan	Biopolymer derived from chitin	Biodegradable and eco-friendly but poor stability in acidic	(Liu et al., 2020)
Zeolite	Natural aluminosilicate mineral	Moderate capacity and limited active site	(Ranjana et al., 2022)
TiO ₂ nanoparticles	Metal oxide nanomaterial	Good photocatalytic property but costly synthesis	(Bahrudin et al., 2020)
Graphene oxide	Carbon-based nanomaterial	High adsorption efficiency due to π - π interaction	(Jeyaseelan et al., 2024)
Fe ₃ O ₄ /SiO ₂ nanocomposite	Magnetic composite	Easy separation using magnet, high reusability	(Ahmad et al., 2024)
MgAl-LDH	Layered double hydroxide	High anion exchange capacity and reusability	(Zaghloul et al., 2020)
NiFe-LDH	Layered double hydroxide	Good structural stability and selective dye removal	(Zheng et al., 2019)
Geopolymer powder	Aluminosilicate-based synthetic material	Low-cost and environmentally friendly but moderate efficiency	(Bassam et al., 2024)

The results summarised in Table 2.1 indicate that the adsorption performance of methyl orange varies greatly depending on the physicochemical properties of the adsorbent. Activated carbon remains one of the most widely used materials due to its large surface area and well-developed porosity, allowing for effective dye adsorption. However, its high cost and regeneration difficulty limit its large-scale use (Wang et al., 2024). On the other hand, natural materials such as zeolites and clays are abundant and inexpensive, but they exhibit lower adsorption capacities due to limited surface functional groups and pore accessibility (Ranjana et al., 2022).

Biopolymer-based adsorbents, such as chitosan, offer eco-friendly and biodegradable alternatives, yet their mechanical instability and solubility in acidic solutions can reduce long-term performance (Liu et al., 2020). Nanomaterials like graphene oxide and metal oxide nanoparticles (TiO₂, Fe₃O₄) have shown remarkable adsorption capabilities due to their high surface area and active functional groups.

Moreover, magnetic nanocomposite like $\text{Fe}_3\text{O}_4/\text{SiO}_4$ allow for easy magnetic separation after adsorption, improving reusability and process efficiency (Ahmad et al., 2024).

Among the various adsorbents, LDHs stand out as one of the most promising materials for dye removal. LDHs, also known as hydrotalcite-like compounds, possess positively charged layers and exchangeable interlayer anions, which make them particularly effective for anionic dye adsorption such as methyl orange (Zaghloul et al., 2020). The LDH like MgAl-, NiFe-, ZnCr-LDH, has high anion exchange capacity, tuneable composition, large surface area and chemical stability that helps to achieve both high adsorption efficiency and good regeneration performance.

2.5 Layered Double Hydroxides

Layered Double Hydroxides (LDHs) are a class of anionic clays characterised by a unique lamellar structure that allows for versatile applications across environmental, biomedical and industrial fields. Structurally, LDHs consist of positively charged brucite-like layers intercalated with anions and water molecules like in the Figure 2.4 . The general formula for LDHs is $[\text{M}^{2+}_{1-x}\text{M}^{3+}_x(\text{OH})_2]^{x+} [\text{A}^{n-}_{x/n} \cdot m\text{H}_2\text{O}]$ where M^{2+} and M^{3+} are divalent and trivalent metal cations, A^{n-} represents interlayer anions and x defines the ratio of trivalent cations to the total metal content (Tang et al., 2020).

The layered structure provides LDHs with a high surface area, tuneable interlayer spacing and exceptional anion exchange capacity. These properties allow for the incorporation of various functional groups, making LDHs suitable for applications such as pollutant adsorption, catalysis and drug delivery. The interlayer anions can be exchanged with other anions, enabling LDHs to act as carriers for targeted delivery or as adsorbents for removing contaminants from water (Shan et al., 2014). One of the most notable features of LDHs is their "memory effect," which refers to the ability of calcined LDHs to regenerate their original layered structure upon rehydration in the presence of suitable anions. This property enhances their reusability and makes them highly effective for environmental remediation (Li et al., 2024)

Applications of LDHs are diverse and growing. In environmental science, LDHs are used for water purification, particularly for the adsorption of heavy metals, dyes and antibiotics. Their high anion exchange capacity and tuneable surface

properties make them effective for removing pollutants from aqueous systems (Mittal, 2021). In biomedical fields, LDHs serve as drug delivery vehicles due to their biocompatibility, controlled release properties and ability to intercalate therapeutic agents within their layers (Li et al., 2023). In analytical chemistry, LDHs have been employed as sorbents in sample preparation, electrode materials in electrochemical sensing and colorimetric detection indicators. Their ability to form composites with nanomaterials further enhances their applicability in advanced sensing technologies (Tang et al., 2020).

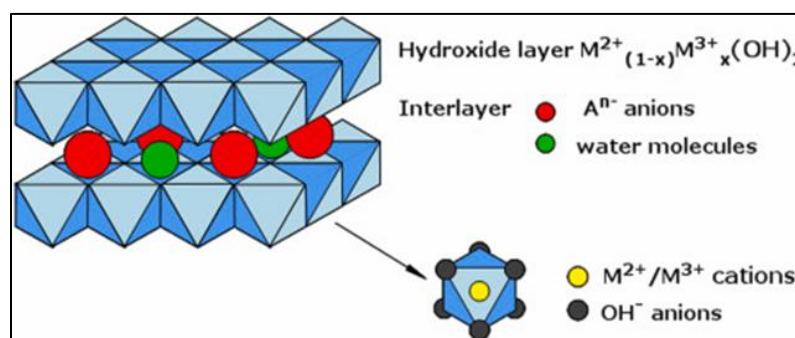


Figure 2.4 Structure of Layered Double Hydroxide

2.5.1 Synthesis of LDHs

The synthesis of LDHs involves a variety of direct and indirect methods tailored to achieve specific structural and functional properties as shown at Figure 2.5 (Daud et al., 2019). The figure shows some common methods that used to synthesis LDH which are (a) Hydrothermal method, (b) Layer-by-layer assembly, (c) Microwave-assisted hydrothermal, (d) Electrostatic self-assembly and (e) Co-precipitation method. Among the indirect methods, techniques such as anion exchange, exfoliation-restacking, memory effect and layer-by-layer assembly are commonly employed. Anion exchange involves replacing the host anions like NO_3^- , CO_3^{2-} , Cl^- with desired guest anions such humate, EDTA, glutamate, polysulfides by stirring pristine LDH in a concentrated solution of target anions. This process is driven by electrostatic interactions between the positively charged lamellar layers of LDHs and the guest anions, with pH typically maintained above 4 to preserve the hydroxide layers (Taoufik et al., 2022). Exfoliation-restacking, another effective method, involves the delamination of LDH layers using

solvents like sodium dodecyl sulphate (SDS), formamide, or water, which reduce the electrostatic forces between layers. The delaminated layers are then deposited onto substrates through electrostatic interactions, followed by restacking through stirring for extended periods like 12 hours, centrifugation and washing with deionised water and ethanol to achieve high-purity adsorbent dosages (Fu et al., 2025).

The hydrothermal technique is another widely used method for synthesising LDHs, where the precursor solution undergoes crystallisation at elevated temperatures and pressures in an autoclave. This technique is particularly advantageous for producing highly crystalline LDHs with uniform particle sizes and enhanced stability (Dutta et al., 2025). Similarly, the layer-by-layer (LbL) assembly method involves sequential deposition of LDH layers onto a substrate, forming multilayered films with precise thickness control. Microwave-assisted synthesis is a more recent advancement, leveraging rapid heating and uniform energy distribution to produce LDHs with high phase purity and crystallinity in significantly reduced reaction times. These methods offer flexibility in tuning the structural and chemical properties of LDHs for specific applications, such as adsorption, catalysis and drug delivery (Fu et al., 2025).

Direct synthesis methods, such as co-precipitation and the memory effect, are also extensively employed. Co-precipitation involves the simultaneous precipitation of divalent and trivalent metal cations in an alkaline solution under controlled pH and temperature conditions. This method allows for the precise control of metal ratios and the incorporation of various anions into the interlayer spaces (Daud et al., 2019). The memory effect, unique to LDHs, involves calcining the material to form mixed metal oxides and then rehydrating it in the presence of target anions, restoring the original layered structure. These direct methods are simple, scalable and highly effective for producing LDHs with tailored functionalities, making them suitable for a wide range of industrial and environmental applications (Taoufik et al., 2022).

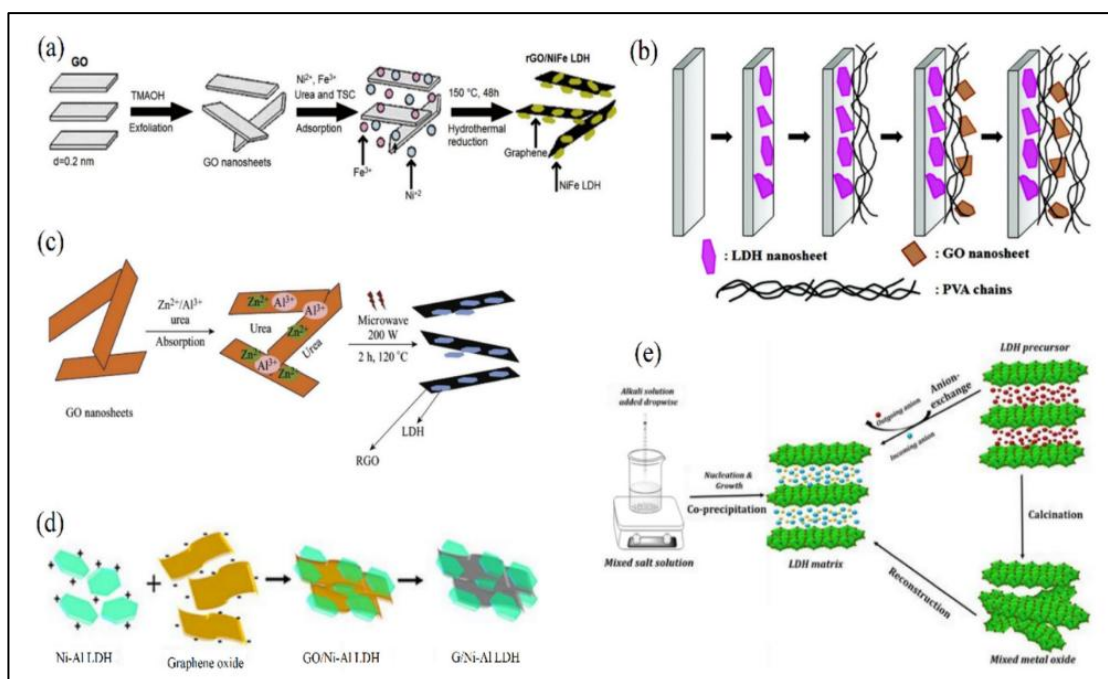


Figure 2.5 Possible Method Synthesis of Layered Double Hydroxide.

2.5.2 Application of LDHs in Adsorption

Layered Double Hydroxides (LDHs) have gained considerable attention as efficient adsorbents for the removal of a wide range of pollutants from aqueous environments, including dyes, heavy metals and organic compounds. Their unique lamellar structure, high anion exchange capacity, tuneable chemical composition and surface charge properties make them highly versatile materials for adsorption applications (Yang et al., 2023). LDHs are composed of positively charged brucite-like layers with interlayer anions and water molecules, which enable the exchange of anions or organic species within the interlayer region. This property allows LDHs to interact with both anionic and cationic pollutants, depending on the surface modification and synthesis method used.

The adsorption performance of LDHs is influenced by several factors, including the type of metal cations like Mg^{2+} , Al^{3+} , Zn^{2+} , Fe^{3+} , interlayer anion like (CO_3^{2-} , NO_3^- , Cl^-), surface area, crystallinity and the synthesis route. For instance, Mg-Al LDH has been widely used due to its high surface area and chemical stability, whereas Fe-Al and Zn-Al LDHs have been explored for their redox activity and photocatalytic properties. The presence of hydroxyl groups on the LDH surface also contributes to hydrogen

bonding and electrostatic interactions, further enhancing adsorption capacity (Dutta et al., 2025).

LDHs can remove pollutants through various mechanisms such as anion exchange, surface complexation, electrostatic attraction and intercalation. For example, in the case of anionic dyes like methyl orange, the removal process primarily involves electrostatic attraction between the negatively charged dye molecules and the positively charged LDH layers. Additionally, partial intercalation of dye molecules into the interlayer galleries has been reported, contributing to higher adsorption capacities (Yu et al., 2021).

Numerous studies have demonstrated the efficiency of LDHs and their modified forms in adsorptive removal processes. For instance, Mg-Al LDH synthesised via the coprecipitation method showed high adsorption efficiency for methyl orange due to its strong electrostatic interactions and layered structure that facilitates dye molecule penetration (Mittal et al., 2021). The reusability and regeneration of LDHs are also important aspects in adsorption studies. LDHs generally maintain good adsorption performance after multiple cycles of adsorption–desorption, especially when mild desorbing agents such as NaOH or NaCl are used. This indicates that LDHs are not only effective but also sustainable adsorbents for water purification (Yang et al., 2023).

2.5.3 Desorption and Regeneration of LDHs

Desorption and regeneration studies are essential in adsorption research as they provide important information on the reusability and stability of an adsorbent. An efficient adsorbent should not only exhibit high adsorption capacity but also maintain its performance after multiple adsorption-desorption cycles. In dye adsorption systems, desorption studies help to elucidate the adsorption mechanism and assess whether the interaction between the adsorbate and adsorbent is reversible (Sonawane et al., 2025)

LDHs have attracted significant attention as reusable adsorbents due to their anion exchange capability, surface charge properties and structural stability. In the case of anionic dye such as methyl orange, adsorption onto LDHs is mainly governed by electrostatic attraction, surface adsorption and anion exchange within the interlayer region (Badri et al., 2022). Consequently, desorption can be effectively achieved by introducing competitive anion or altering the pH of the solution.

Several studies have reported that alkaline solutions, particularly sodium hydroxide (NaOH), are highly effective desorbing agents for methyl orange and other anionic dyes adsorbed onto MgAl-LDH. The presence of excess hydroxide ion (OH^-) weakens the electrostatic interactions between the negatively charged dye molecules and the positively charged LDH layers, thereby facilitating dye release back into the solution (Grover et al., 2022; Isidoro Ribeiro et al., 2023; Kameda et al., 2021). This observation suggested that electrostatic attraction plays a dominant role in the adsorption mechanism.

Isidoro Ribeiro (2023) reported that MgAl-LDH exhibited high desorption efficiency for methyl orange when treated with NaOH solution, with only a slight reduction in adsorption capacity after several regeneration cycles. Similarly, (Li and Lu, 2025) demonstrated that LDH based adsorbents retained more than 80% of their initial adsorption capacity after multiple adsorption-desorption cycles, indicating good structural stability and reusability. These findings highlight the suitability of LDHs for repeated use in wastewater treatment applications.

Furthermore, desorption studies provide indirect evidence of the reversibility of adsorption processes. High desorption efficiency generally indicates that physical adsorption and electrostatic interactions dominate which low desorption may suggest stronger chemical bonding or structural changes in the adsorbent (Li and Lu, 2025). Therefore, desorption analysis not only supports regeneration performance but also strengthens the interpretation of the adsorption mechanism.

2.6 Ultraviolet Visible Spectroscopy

Ultraviolet Visible (UV-Vis) Spectroscopy is a commonly used analytical method that measures the adsorption of ultraviolet and visible light by molecules. This technique relies on electronic transitions within a molecule, typically involving $\pi-\pi^*$ or $n-\pi^*$ transitions. Molecules with conjugated systems or specific chromophores absorb light at characteristic wavelengths, making UV-Vis spectroscopy a powerful tool for identifying and quantifying compounds. Its applications span various fields, including pharmaceuticals, environmental monitoring and materials science (Rong et al., 2024).

One of the significant advantages of UV-Vis spectroscopy is its simplicity and rapid analysis. The method involves passing a beam of light through a sample and

measuring the absorbed light. The absorbance, governed by Beer-Lambert's law, is directly proportional to the concentration of the absorbing species. This makes UV-Vis spectroscopy a reliable tool for quantitative analysis, as seen in its use for monitoring water quality by detecting heavy metals and organic pollutants (Cao et al., 2024). In pharmaceuticals, UV-Vis spectroscopy plays a crucial role in quality control and drug development. It is used to determine the concentration of active pharmaceutical ingredients (APIs) in formulations and to study drug-receptor interactions. Its high sensitivity and non-destructive nature make it an indispensable tool in ensuring the consistency and potency of pharmaceutical products (Ghosh and Nandi, 2024).

One of the primary advantages of UV-Vis in adsorption studies is its ability to distinguish between adsorbed and free species in complex matrices. This capability is enhanced when coupled with multivariate analysis techniques, which can deconvolute overlapping spectral features and provide detailed insights into the molecular interactions at play. Such approaches have been successfully applied to evaluate adsorption isotherms and optimise adsorbent materials for environmental and industrial applications (Rong et al., 2024). UV-Vis spectroscopy is also highly adaptable, supporting studies of adsorption under various conditions, such as different pH levels, temperatures and ionic strengths. This flexibility allows researchers to investigate the thermodynamics and kinetics of adsorption processes in real time, providing valuable information about adsorption capacities, binding affinities and energy changes associated with adsorption events. Furthermore, UV-Vis spectroscopy can be combined with advanced materials like nanoparticles or functionalised adsorbents to enhance sensitivity and selectivity. For example, studies using UV-Vis to evaluate adsorption on functionalised metal-organic frameworks (MOFs) have demonstrated its effectiveness in identifying optimal adsorbents for pollutants, providing insights into adsorption mechanisms at the molecular level (Rong et al., 2024).

2.6.1 Analytical Detection of Methyl Orange Using UV-Vis

UV-Vis spectroscopy is widely used analytical techniques for the quantitative determination of dyes such as methyl orange in aqueous solutions. It is based on the principle that molecules absorb light of specific wavelengths, causing electronic transitions between molecular orbitals. The extent of adsorption is directly related to the

concentration of the absorbing species (Ghosh and Nandi, 2024), following Beer-Lambert's Law, which is expressed as:

$$A = \epsilon l C \quad (2.6)$$

Where A is the absorbances (dimensionless), ϵ is molar absorptivity ($\text{L} \cdot \text{mol}^{-1} \cdot \text{cm}^{-1}$), l is the path length of the cuvette (cm) and C is the concentration of the analytes ($\text{mol} \cdot \text{L}^{-1}$). This linear relationship forms the basis for constructing curves used to determined unknown concentrations of methyl orange in solutions.

Methyl orange is an azo dye that exhibits strong adsorption in the visible region due to the presence of conjugated π -electron systems and azo ($-\text{N}=\text{N}-$) chromophores. The molecule exists in two distinct forms depending on the pH solution, the protonated form (red) in acidic medium and the deprotonated form (orange yellow) in alkaline medium. The absorbance maximum (λ_{max}) for methyl orange typically appears around 464nm in its deprotonated form and around 510 nm in the protonated form (Badri et al., 2022).

During adsorption experiments, the UV-Vis spectrophotometer is used to monitor the decrease in absorbance of methyl orange solution at its λ_{max} before and after contact with the adsorbent. The reduction in absorbance corresponds to the amount of dye removed from the solution, which can be used to calculate the concentration of residual dye using the calibration curve (Meira et al., 2023). The removal efficiency (%R) and equilibrium adsorption capacity (q_e) are then calculated using the following equation:

$$\%R = \frac{C_o - C_e}{C_o} \times 100 \quad (2.7)$$

$$q_e = \frac{(C_o - C_e)V}{m} \quad (2.8)$$

Where C_o and C_e ($\text{mg}\cdot\text{L}^{-1}$) are the initial and equilibrium concentrations of methyl orange, respectively; V is the volume of the solution (L) and m is the mass of adsorbent used (g).

The reliability and sensitivity of UV–Vis spectroscopy make it ideal for adsorption studies, as it enables rapid and non-destructive analysis of dye concentration with minimal sample preparation. The technique also allows real-time monitoring of adsorption kinetics, enabling researchers to study the adsorption mechanism and rate-controlling steps (Sarkar and Upadhyay, 2025). Furthermore, UV–Vis analysis can be used to evaluate desorption processes during adsorbent regeneration, providing insights into the reusability of the adsorbent material.

2.7 X-ray Diffraction

X-ray Diffraction (XRD) is a powerful analytical technique widely used to investigate the crystallographic structure, phase composition and interlayer spacing of solid materials. The technique is based on the diffraction of monochromatic X-rays when they interact with the periodic lattice of crystalline substances. According to Bragg's Law, $n\lambda = 2d \sin \theta$, diffraction occurs when the X-ray wavelength matches the spacing between crystal planes. This produces characteristic diffraction peaks at specific 2θ values, allowing identification of materials based on their unique crystallographic fingerprints (Zhang et al., 2019).

In adsorption and materials science studies, XRD plays a crucial role in confirming the successful synthesis of adsorbent such as layered double hydroxides (LDHs), metal oxides, zeolites, biochar composites and other porous adsorbents. The presence, absence, or shifting of diffraction peaks can reveal important information regarding crystallinity, structural defects and phase purity. For LDHs specifically, XRD is used to determine their basal spacing (d-spacing) and verify the layered structure by identifying the characteristic (003), (006), (012) and (015) reflections. Changes in peak position or intensity provide insight into structural modifications such as intercalation, calcination, swelling, or ion exchange.

Furthermore, XRD is essential in adsorption studies to confirm whether the adsorbate (such as methyl orange) interacts with or modifies the crystal structure of the adsorbent. For instance, peak shifts, peak broadening and changes in crystallinity can

indicate the incorporation, intercalation, or surface attachment of dye molecules onto the adsorbent (Badri et al., 2022). Thus, XRD functions not only as a structural characterisation tool but also as a supporting technique to elucidate adsorption mechanisms.

2.8 Fourier Transform Infrared Spectroscopy

Fourier Transform Infrared Spectroscopy (FTIR) is a widely used analytical technique for identifying the functional groups and chemical bonds present in a material by measuring the adsorption of infrared radiation at different wavenumbers. The interaction between infrared radiation and molecular vibrations such as stretching, bending and twisting produces characteristic adsorption bands that are unique to specific functional groups (Wu et al., 2019). As a result, FTIR provides valuable qualitative information regarding the chemical composition and structural features of materials.

In adsorption studies, FTIR is commonly applied to investigate surface functional groups, bonding environments and interactions between adsorbents and adsorbates. The technique is particularly useful for confirming the presence of hydroxyl groups, interlayer anions and metal–oxygen bonds, which play crucial roles in adsorption mechanisms. Additionally, FTIR analysis before and after adsorption can provide insights into the adsorption mechanism by revealing peak shifts, intensity changes, or the appearance of new bands, indicating interactions such as electrostatic attraction, hydrogen bonding, or ion exchange. FTIR has several advantages, including rapid analysis, minimal sample preparation, high reproducibility and the ability to analyse both crystalline and amorphous materials (Altalhi et al., 2024). These advantages make FTIR an essential characterisation tool in the study of layered double hydroxides (LDHs), especially for confirming successful synthesis and understanding their surface chemistry.

Typical FTIR spectra of MgAl-LDH exhibit several characteristic adsorption bands. A broad band in the range of 3400–3500 cm^{-1} is attributed to the stretching vibration of hydroxyl (–OH) groups from both the brucite-like layers and interlayer water molecules. The band observed around 1600–1650 cm^{-1} corresponds to the bending vibration of interlayer water molecules, further confirming the hydrated nature

of LDHs. A strong adsorption band appearing near 1350–1380 cm^{-1} is commonly associated with the stretching vibration of carbonate ions (CO_3^{2-}), which often occupy the interlayer region due to their high affinity for LDH structures. The presence of this band indicates successful formation of the LDH layered structure and atmospheric carbonate incorporation during synthesis. In the lower wavenumber region, bands observed below 1000 cm^{-1} , typically around 500–900 cm^{-1} , are attributed to metal-oxygen (M-O) and metal-hydroxyl (M-OH) lattice vibrations, such as Mg-O and Al-O bonds (Altalhi et al., 2024; Zaghloul et al., 2020). These characteristic FTIR bands serve as clear evidence for the successful synthesis of MgAl-LDH and are consistent with previous reports in the literature. Any changes in peak position or intensity after adsorption experiments are often interpreted as evidence of interactions between LDH surfaces and adsorbate molecules, supporting adsorption mechanisms such as electrostatic attraction and hydrogen bonding.

2.9 Scanning Electron Microscopy with Energy Dispersive X-ray

Scanning Electron Microscopy (SEM) is a powerful surface characterisation technique widely used to examine the surface morphology, particle size, texture and microstructural features of solid materials. SEM operates by scanning a focused beam of high-energy electrons over the surface of a sample. The interaction between the electron beam and the sample surface produces secondary electrons, backscattered electrons and characteristic X-rays, which are detected to generate high-resolution images of the material surface. SEM provides detailed information on surface topography and structural homogeneity, making it particularly useful for studying porous materials and adsorbents (Altalhi et al., 2024). The technique allows visualisation of particle agglomeration, layer stacking, surface roughness and morphological changes before and after adsorption. These features are critical in adsorption studies, as surface morphology directly influences the availability of active sites and mass transfer behaviour during pollutant removal processes. Energy-Dispersive X-ray Spectroscopy (EDX), when coupled with SEM, enables elemental composition analysis of the material. EDX works by detecting characteristic X-rays emitted from elements in the sample when excited by the electron beam. This technique provides qualitative and semi-quantitative information about the elemental distribution

and atomic ratios present in the sample (Bassam et al., 2024). The combined SEM-EDX analysis is therefore widely applied to confirm material composition and correlate morphology with chemical composition.

SEM-EDX analysis is extensively employed to characterise MgAl-LDH due to its ability to provide complementary morphological and elemental information. SEM images of MgAl-LDH typically reveal plate-like, lamellar, or flaky structures, which are characteristic of layered materials formed through co-precipitation synthesis. These platelet-like morphologies arise from the stacking of positively charged hydroxide layers and are often observed as aggregated or partially exfoliated particles (Wu et al., 2019). The surface morphology observed in SEM micrographs provides insight into the degree of crystallinity, particle agglomeration and surface roughness of MgAl-LDH. A rough and porous surface morphology is generally associated with enhanced adsorption performance, as it increases surface area and exposes more active adsorption sites. In some cases, SEM analysis after adsorption may show surface coverage or structural changes, indicating successful interaction between the LDH surface and dye molecules. EDX analysis is used to confirm the elemental composition of MgAl-LDH by detecting the presence of magnesium (Mg), aluminium (Al), oxygen (O) and, in some cases, carbon (C) originating from interlayer carbonate anions. The presence of Mg and Al peaks in the EDX spectrum confirms the successful incorporation of divalent and trivalent metal cations into the LDH structure (Altalhi et al., 2024). Furthermore, the Mg/Al atomic ratio obtained from EDX analysis provides important information regarding the stoichiometry of the synthesised LDH and its agreement with the intended synthesis design (Hosseini and Ghasemi, 2020).

2.10 Response Surface Methodology in Adsorption Studies

Response Surface Methodology (RSM) is a statistical and mathematical tool used to optimise processes by evaluating the relationships between multiple independent variables and a response variable. Developed initially for industrial process optimisation, RSM has found widespread application in scientific research, including adsorption studies, due to its ability to model complex systems. RSM involves designing experiments, fitting mathematical models and generating graphical representations such as response surfaces and contour plots to identify optimal

conditions (Elbager et al., 2025). Its strength lies in its ability to evaluate not just the individual effects of variables but also their interactions, providing a comprehensive understanding of the process.

In adsorption studies, RSM is particularly valuable for optimising key parameters such as pH, adsorbent dosage, contact time, initial pollutant concentration and temperature. For example, study by Muleta (2025), the adsorption of cadmium using calcium oxide nanoparticles in which RSM help to determine the optimal pH, contact time also cadmium ion concentration that balances the charge state of the pollutant and the surface characteristics of the adsorbent.

Similarly, RSM can assess the combined effects of contact time and adsorbent dosage to maximise adsorption efficiency while minimising resource use. The advantage of RSM lies in its systematic approach. Instead of evaluating one variable at a time, it uses factorial or central composite designs to simultaneously vary multiple parameters within specified ranges. This reduces the number of experiments required and provides richer data for statistical analysis (Moosaei et al., 2024). For instance, in a study optimising lead adsorption, RSM was used significantly improving removal efficiency compared to conventional one-factor-at-a-time methods (Elbager et al., 2025). RSM employs mathematical models, typically second-order polynomial at (2.9) which to describe the relationship between variables and the response. The general form of the model is:

$$Y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \beta_{ij} X_i X_j \quad (2.9)$$

Where Y is the response, X_i and X_j are the independent variables, β_0 is the intercept, β_i are the linear coefficients, β_{ii} are the quadratic coefficients and β_{ij} are the interaction coefficients. After generating experimental data, regression analysis is used to estimate the coefficients and assess the model's fit. Statistical parameters such as R^2 , adjusted R^2 and analysis of variance (ANOVA) are used to evaluate the model's accuracy and significance. A well-fitted model not only predicts the response under tested conditions but also interpolates or extrapolates to predict performance under untested conditions. This capability is invaluable for adsorption studies, where

identifying ideal operational ranges can significantly enhance efficiency (Sarani et al., 2024)

One of the most powerful features of RSM is its graphical representation of data through response surface plots and contour diagrams. These visual tools help researchers quickly identify optimal conditions and understand the interplay between variables. For example, a 3D response surface plot for pesticides adsorption efficiency might reveal that maximum removal occurs at an intermediate pH and a moderate adsorbent dosage. Contour plots, which are 2D projections of response surfaces, can further highlight the precise combinations of variables that yield the best results (Ali and Mohammed, 2023). Such visualisation aids not only in data interpretation but also in communicating findings to broader audiences.

RSM offers several advantages in the context of adsorption studies. First, it reduces the experimental workload and resource consumption by optimising variables efficiently. Second, it provides a deeper understanding of variable interactions, which are often critical in systems involving complex pollutants like methyl orange that exhibit pH-dependent behaviour and speciation. Finally, RSM facilitates the identification of robust operating conditions, ensuring high adsorption efficiency even under variable environmental conditions (Boulaabail et al., 2024).

For instance, in studies involving bentonite layered double hydroxide, RSM can help optimise synthesis parameters as well as adsorption conditions (Ali and Mohammed, 2023). By systematically analysing the effects of these variables, researchers can develop highly efficient and cost-effective adsorbents for methyl orange removal.

2.11 Application of LDHs in Treating Wastewater

Layered double hydroxides (LDHs) have been investigated as promising materials for wastewater treatment due to their unique layered structure, high surface area, tuneable metal composition and excellent anion exchange capacity. These properties make LDHs particularly suitable for removing a wide range of contaminants from aqueous environments, including dyes, heavy metals, nutrients, pharmaceuticals and other emerging pollutants. In recent years, increasing environmental concerns

related to industrial wastewater discharge have driven extensive research into LDHs as cost-effective and efficient adsorbents (Kameda et al., 2021).

One of the most widely reported applications of LDHs is in the removal of dyes from wastewater, especially from textile and dyeing industries. Anionic dyes such as methyl orange, congo red and reactive dyes pose serious environmental risks due to their high stability, toxicity and resistance to biodegradation (Altalhi et al., 2024). MgAl-LDH have shown excellent performance in removing anionic dyes because the positively charged hydroxide layers can strongly attract and bind negatively charged dye molecules through electrostatic interaction, surface adsorption and interlayer anion exchange. Several studies have reported high dye removal efficiencies using MgAl-LDH under mild operating conditions, demonstrating its potential for practical wastewater treatment applications (Hosseini and Ghasemi, 2020; Qin et al., 2025; Wu et al., 2019).

In addition to dye removal, LDHs have also been extensively applied for the adsorption of heavy metals such as lead (Pb^{2+}), cadmium (Cd^{2+}), chromium (Cr^{6+}) and arsenic (As^{5+}) from wastewater. The removal mechanisms include surface complexation, precipitation and ion exchange, depending on the metal species and solution conditions. Studies have shown that MgAl-LDH exhibits strong affinity toward toxic metal ions and maintains good structural stability after adsorption, making it suitable for repeated use (Fu et al., 2025).

More importantly, recent research has increasingly focused on the application of LDHs in real wastewater systems rather than only synthetic solutions. Real wastewater contains competing ions, organic matter and fluctuating pH, which can significantly affect adsorption performance. Several studies have demonstrated that LDHs, particularly MgAl-based LDHs, can effectively remove pollutants from textile wastewater, showing good adsorption efficiency even in complex matrices (Shan et al., 2014; Yang et al., 2023). These findings highlight the robustness and practical applicability of LDHs for industrial wastewater treatment.

Despite the growing body of literature on LDH-based adsorption, many studies still focus on material modification or laboratory-scale tests without systematic optimisation of operating parameters. Furthermore, limited studies integrate statistical optimisation techniques, such as response surface methodology (RSM), with adsorption isotherm, kinetic analysis and real wastewater validation in a single framework.

Therefore, further investigation is required to optimise adsorption conditions, understand adsorption behaviour and verify LDH performance under realistic wastewater conditions. In this context, the present study evaluates the application of MgAl-LDH for methyl orange removal by combining RSM optimisation, adsorption isotherm and kinetic studies and validation using textile wastewater, thereby contributing valuable insights toward the practical implementation of LDHs in wastewater treatment.

CHAPTER 3

RESEARCH METHODOLOGY

3.1 Materials and Instruments

The chemicals that were used in this study include magnesium nitrate hexahydrate ($\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) (Merck), aluminium nitrate nonahydrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) (Sigma-Aldrich), sodium hydroxide (NaOH) (Merck), sodium carbonate (Na_2CO_3) (Sigma-Aldrich), methanol (HPLC grade, Merck), hydrochloric acid (HCl) (Sigma-Aldrich), 95% methyl orange reagent (Sigma-Aldrich) and sodium hydroxide solution (0.1M NaOH) (Merck).

The instruments include for characterisation, X-ray Diffraction (XRD) was performed using a D/MAX 2200 diffractometer (Rigaku, Tokyo), Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FTIR) (Perkin-Elmer, United States) and Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM-EDX). Statistical analysis will be performed using Design-Expert software (Version 13) (Stat-Ease, USA). UV–Vis Spectrophotometer (Shimadzu, Japan) was used to measure the concentration of analytes studied in this research.

3.2 List of Apparatus

The apparatus that used was magnetic stirrer with hot plate (IKA, Germany), pH meter (Mettler Toledo, USA), micropipettes (Sigma-Aldrich), beakers, measuring cylinders, volumetric flasks, conical flask, filter paper, filter funnel, oil bath (Elma, Germany), mortar and pestle and oven.

3.3 Synthesis of MgAl- Layered Double Hydroxide (MgAl-LDH)

MgAl-Layered Double Hydroxide was synthesis using a co-precipitation approach at Mg:Al molar ratio 3:1 in 100 ml solution (7.69g $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 3.75g $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, $V_{\text{methanol}}/V_{\text{water}} = 1/1$). The solution was stirred continuously until it was completely dissolved, forming a clear homogenous solution (Lin et al., 2017; Modrogan et al., 2020).

A separate alkaline solution was prepared by dissolving 0.4 g NaOH and 0.5g Na₂CO₃ in 100 ml solution ($V_{\text{methanol}}/V_{\text{water}} = 1/1$), followed by continuous stirring until fully dissolved. The alkaline solution was slowly added dropwise in MgAl solution under vigorous stirring. The pH was carefully adjusted to pH 10 by controlling the addition of NaOH/Na₂CO₃. Then, the mixture was then stirred for 3 hours room temperature to allow complete precipitation. The resulting slurry was sealed and aged in an oil bath at 70 °C for 24 hours to promote crystallisation of the LDH layers. After aging, the suspension was centrifuged and the solid product was washed 3-4 times with deionised water to remove unreacted ions and impurities. Lastly, the washed precipitate was then dried in an oven at 70 °C for 12 hours. The dried solid was gently ground to obtain a fine, uniform MgAl-LDH powder, which was stored in airtight containers for further characterisation and adsorption experiments (Lin et al., 2017; Modrogon et al., 2020).

3.4 Characterisation Methods

The synthesised MgAl-LDH and its modified forms were characterised using X-ray diffraction (XRD), Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FTIR) and Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM-EDX) to confirm their structural, chemical and functional properties. These analyses provided essential information regarding crystallinity, functional groups and the successful synthesis of the layered double hydroxide.

3.4.1 X-ray Diffraction

X-ray diffraction (XRD) analysis was conducted to determine the crystalline structure, phase composition and basal spacing of the synthesised MgAl-LDH. The XRD patterns were obtained using a D/MAX 2200 X-ray diffractometer (Rigaku, Tokyo) equipped with Cu K α radiation ($\lambda = 0.154$ nm) operated at 40 kV and 300 mA.

The samples were scanned over a 2θ range of 5°–70° with a step size of 0.03°. This scan range allowed identification of the characteristic reflections of hydrotalcite-like LDH, especially the (003), (006), (009) and (110) planes. The resulting diffraction

patterns were analysed to confirm the layered structure, evaluate crystallinity and detect any structural changes after modification or adsorption.

3.4.2 Fourier Transform Infrared Spectroscopy

Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FTIR) was used to identify the functional groups present in the LDH structure. FTIR measurements were recorded using a Spectrum One ATR-FTIR spectrometer (Perkin–Elmer, United States) within the wavelength range of 4000–650 cm^{-1} .

3.4.3 Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy

SEM analysis was carried out at an accelerating voltage ranging from 5 to 20 kV, depending on image clarity and resolution requirements. Micrographs were captured at various magnifications to observe particle morphology, surface texture, agglomeration behaviour and layered structure characteristics of the MgAl-LDH. These observations were used to evaluate the surface features relevant to adsorption performance. Energy-Dispersive X-ray Spectroscopy (EDX) analysis also was conducted simultaneously with SEM imaging to determine the elemental composition of the synthesised LDH. The presence of magnesium (Mg), aluminium (Al), oxygen (O) and carbon (C) was analysed to confirm the successful formation of MgAl-LDH and the incorporation of interlayer carbonate anions. Elemental mapping and atomic percentage analysis were also performed to assess the distribution and relative ratios of Mg and Al within the LDH structure.

3.5 Design of Experiment by Central Composite Design

The Design Expert Software (version 13) was used for the statistical design of experiments and data analysis. In the present study, the central composite design (CCD) and response surface methodology (RSM) are applied to optimise and assess the relationship among four significant independent variables (1) adsorbent dosage (g), (2) pH solution, (3) initial methyl orange concentration (ppm) and (4) contact time (min).

According to CCD, the number of experimental runs conduct in this research were designed based on the following equation (3.1):

$$2^k + 2k + n_o \quad (3.1)$$

Where k = the number of independent variables and n_o = the number of center point replication(Ahmad et al., 2020). Table 3.1 shows the coded and actual variables level for each parameter that determined by RSM.

Table 3.1
Parameter and Level for CCD of Removal Methyl Orange

Independent Variables	Symbol	Actual Variables Level				
		-2	-1	0	1	2
Adsorbent Dosage (g)	A	0.05	0.25	0.45	0.65	0.85
pH Solution	B	3	5	7	9	11
Initial Methyl Orange Concentration (mg L ⁻¹)	C	10	20	30	40	50
Contact Time (min)	D	10	20	30	40	50

CCD is an experimental design, which is useful in RSM, for building a second order (quadratic) model for the response variable without needing to use a complete three level factorial experiment (Sharafinia and Rashidi, 2023). Therefore, according to the equation, for 4 factor ($k = 4$) the design included 2^k (16 experiments) factorial points, $2k$ (8 experiments) axial points and 6 repeated experiments for the central point to estimate the experimental error. The total number of the achieved experiments from these factors was 30 CCD experimental design.

3.5.1 Standard Calibration Method of Methyl Orange

A stock solution of methyl orange with a concentration of 1000 mg L⁻¹ was prepared by dissolving 0.025 g of methyl orange powder in distilled water and diluting it to 25 mL in a volumetric flask. The solution was stirred until completely dissolved to obtain a homogeneous stock solution. From this stock solution, a series of methyl orange standard solutions with concentrations of 10, 20, 30, 40, 50 and 60 mg L⁻¹ were

prepared by dilution using distilled water as in Table 3.2. The dilution was performed using the equation $C_1V_1=C_2V_2$, where C_1 is the concentration of the stock solution, V_1 is the volume of stock solution required, C_2 is the desired standard concentration and V_2 is the final volume of the standard solution.

The absorbance of each standard solution was measured using a UV-Vis spectrophotometer at the selected wavelength for methyl orange. Distilled water was used as a blank before analysis. A calibration curve was then constructed by plotting the absorbance values against the corresponding methyl orange concentrations. The linear regression equation obtained from the calibration curve was used to determine the residual concentration of methyl orange in the supernatant after adsorption treatment.

Table 3.2
Dilution of Methyl Orange Standard Solution for Calibration Curve

Standard concentration, C_2 (mg L ⁻¹)	Volume of 1000 mg L ⁻¹ stock, V_1 (mL)	Dilute to final volume (mL)
10	0.25	25
20	0.50	25
30	0.75	25
40	1.00	25
50	1.25	25
60	1.50	25

3.5.2 Experimental Method

Adsorption experiments was performed on a laboratory scale and at room temperature for each analyte separately. The effect of variables which is initial methyl orange concentration, pH solution, adsorbent and contact time on methyl orange removal efficiency using MgAl-LDH was investigated using the models provided in the Design Expert software. For this purpose, experiments was performed by mixing different amounts of MgAl-LDH (0.05-0.85 g) as an adsorbent in 15 mL of methyl orange solution with various concentrations (10-60 mg L⁻¹) and at different pH levels (3-11). The reaction mixture was be placed in an ultrasonic bath at different contact times (10–50 min), also subjected to ultrasonic waves at laboratory temperature. After shaking, the suspension was centrifuged to separate the solid adsorbent. Finally, the supernatant was analysed using a UV-Vis spectrophotometer (Zaghloul et al., 2020) at

the maximum adsorption wavelength of methyl orange ($\lambda_{\max} \approx 464$ nm) where, in acidic medium ($\text{pH} < 3.1$), methyl orange exists primarily in its protonated form, which absorbs light around 510 nm, giving an orange-red colour. In alkaline medium ($\text{pH} > 4.4$), deprotonation occurs and the methyl orange shifts to a yellow-coloured anionic form that absorbs around 464 nm (Zhang et al., 2012). The remaining concentrations (Ce) was determined using the calibration curve.

Table 3.3
Experimental Run and Factor from RSM for Removal Methyl Orange

Run	Adsorbent Dosage (g)	pH Solution	Methyl Orange Initial Concentration (mg L ⁻¹)	Contact Time (min)
1	0.25	9	20	20
2	0.65	5	20	40
3	0.25	5	40	20
4	0.25	9	40	40
5	0.25	5	20	20
6	0.45	3	30	30
7	0.65	9	20	20
8	0.45	7	30	30
9	0.65	5	20	40
10	0.45	11	30	30
11	0.25	5	40	40
12	0.65	9	40	20
13	0.85	7	30	30
14	0.45	7	30	10
15	0.45	7	30	50
16	0.05	7	30	30
17	0.65	9	40	40
18	0.45	7	50	30
19	0.25	9	20	40
20	0.45	7	30	30
21	0.65	5	40	40
22	0.65	5	40	20
23	0.45	7	30	30
24	0.45	7	10	30
25	0.25	9	40	20
26	0.25	5	20	40
27	0.45	7	30	30
28	0.65	5	20	20
29	0.45	7	30	30
30	0.45	7	30	30

3.5.3 Removal Efficiency

The percentage of removal efficiency of methyl orange can be generally explained as a percentage representing the number of methyl orange which methyl orange removed or destroyed in comparison to the number of metal ion that entered the system. Therefore, the removal efficiency can be determined by the following equation (Abdolhossein Rejali and Dinari, 2025) (3.2):

$$\%R = \frac{(C_i - C_e)}{C_i} \times 100 \quad (3.2)$$

Where R is the removal efficiency (%), C_i is the initial methyl orange concentration (mg L^{-1}) and C_e is the equilibrium concentration of the methyl orange (mg L^{-1}).

3.6 Statistical Analysis using Analysis of Variance (ANOVA)

The experimental data obtained from CCD-RSM design were statistically analysed using analysis of variance (ANOVA) to evaluate the significance and adequacy of the developed model. The percentage removal of methyl orange was selected as the response while adsorbent dosage, solution pH, initial methyl orange concentration and contact time were selected as the independent variables. The experimental data were fitted to different polynomial models, including linear, two-factor interaction (2F1), quadratic and cubic models to determine the most suitable model for describing the adsorption process.

The significance of the model and model terms was evaluated based on the F-value and p-value at a 95% confidence level. A p-value of less than 0.05 was considered statistically significant, indicating that the model or factor had a significant effect on the study. The adequacy of the model was further evaluated using the coefficients of determination (R^2), adjusted R^2 , predicted R^2 , adequate precision and lack-of-fit test. A high R^2 value indicates good agreement between the experimental and predicted values while close agreement between adjusted R^2 and predicted R^2 indicates good predictive ability of the model.

The lack-of-fit test was used to determine whether the selected model adequately represented the experimental data. A non-significant lack of fit ($p > 0.05$) was preferred, as it indicates that the model error was not significant compared with the pure experimental error. Therefore, the best model was selected based on a significant model p-value, non-significant lack of fit, high R^2 , good agreement between adjusted and predicted R^2 and suitability for predicting the optimum adsorption conditions.

3.7 Model Validation method

Model validation was conducted to evaluate the adequacy, reliability and predictive accuracy of the developed RSM model for methyl orange adsorption using MgAl-LDH. Validation is an essential step to confirm that the optimised model can accurately predict experimental responses under selected conditions and is suitable for practical application. In this study, model validation was performed through experimental verification at the optimised adsorption conditions suggested by the numerical optimisation tool in Design-Expert software.

3.7.1 Verification Model

The verification experiment was carried out by conducting adsorption experiments under the optimised conditions predicted by the RSM model. These experiments were repeated three times to ensure reproducibility and reliability of the experimental data. The optimised parameters obtained from the numerical optimisation were applied and the percentage of experimental methyl orange removal efficiency (%) was determined using UV–Vis spectrophotometry at a wavelength range of 464–510 nm.

The experimentally obtained methyl orange removal percentages were then compared with the predicted value generated by the RSM model. The accuracy of the model was assessed using the relative standard error (RSE), which evaluates the deviation between predicted and experimental values.

3.7.2 Optimum Conditions Validation

The optimum adsorption conditions for methyl orange removal using MgAl-LDH were determined through numerical optimisation using the desirability function approach in Design-Expert software. The goal of the optimisation was to maximise the percentage of methyl orange removal (%) while maintaining all independent variables within their specified experimental ranges.

3.7.3 Relative Standard Error

The accuracy of the RSM model was quantitatively evaluated using the relative standard error (RSE). The RSE is a statistical parameter used to measure the deviation between experimentally obtained results and the values predicted by the model. A lower RSE value indicates better agreement between predicted and experimental data which confirming the reliability of the developed model. The RSE was calculated by comparing the experimental methyl orange removal efficiency obtained from the validation experiments with the predicted methyl orange removal value generated by the RSM model. The RSE is expressed as a percentage and was calculated using the following equation (3.3):

$$RSE(\%) = \frac{(\text{actual value} - \text{predicted value})}{\text{predicted value}} \times 100 \quad (3.3)$$

Where the actual value represents the experimentally measured methyl orange removal efficiency (%) under the optimum conditions and the predicted value corresponds to the methyl orange removal efficiency (%) predicted by the RSM model. In general, an RSE value below 5% is considered acceptable and indicates good agreement between experimental and predicted value (Khairuddin et al., 2023).

3.8 Adsorption Study

Adsorption studies were carried out to investigate the interaction between methyl orange molecules and the synthesised MgAl-layered double hydroxide (MgAl-

LDH) adsorbent under optimised operating conditions. The adsorption behaviour was evaluated in terms of equilibrium characteristics and adsorption rate to provide a comprehensive understanding of the adsorption mechanism and performance of MgAl-LDH. In this study, adsorption experiments were divided into isotherm and kinetic investigations. The adsorption isotherm study was conducted to describe the equilibrium distribution of methyl orange between the aqueous phase and the solid adsorbent surface at a constant temperature. Isotherm models were applied to determine the adsorption capacity and surface characteristics of MgAl-LDH. Meanwhile, adsorption kinetic studies were performed to examine the rate of methyl orange uptake and to identify the controlling adsorption mechanism.

All adsorption experiments were conducted using the optimised conditions obtained from Response Surface Methodology (RSM) to ensure consistency and reliability of the experimental data. The concentration of methyl orange before and after adsorption was determined using UV–Vis spectrophotometry and the adsorption performance was evaluated based on adsorption capacity and methyl orange removal efficiency.

3.8.1 Isotherm Study

The adsorption isotherm experiments were conducted to evaluate the equilibrium behaviour of methyl orange on the synthesised LDH adsorbent. The optimum operating conditions obtained from the Response Surface Methodology (RSM) were applied in all isotherms experiments to ensure consistency and reliability of the equilibrium data.

To generate isotherm data, a series of methyl orange solutions with varying initial concentrations from 5 to 50 mg/L were prepared. A fixed amount of LDH adsorbent 0.65g was added to each solution. The suspensions were agitated in a shaker at the 170 rpm shaking speed and 70 °C temperature in 20 min at pH 7.78. After adsorption, the samples were centrifuged and the supernatant was analysed using UV–Vis spectrophotometry to determine the equilibrium concentration (C_e). The amount of methyl orange adsorbed at equilibrium (q_e) was calculated using:

$$q_e = \frac{(C_o - C_e)V}{m} \quad (3.4)$$

Where C_o was initial methyl orange concentration (mg/L), C_e was equilibrium methyl orange concentration (mg/L), V was volume of solution (L) and m was mass of LDH adsorbent (g). The equilibrium data were fitted to the Langmuir and Freundlich isotherm models. The Langmuir model assumes monolayer adsorption on a homogeneous surface, while the Freundlich model describes heterogeneous surface adsorption (Sumardiono et al., 2025).

Langmuir equation:

$$q_e = \frac{q_{max} KLC_e}{1 + KLC_e} \quad (3.5)$$

Freundlich equation:

$$q_e = \frac{KFC_e^{1/n}}{n} \quad (3.6)$$

Linear and nonlinear regression analyses were used to determined isotherm constants and evaluate the best-fitting model based on R^2 and error analysis (Isidoro Ribeiro et al., 2023).

3.8.2 Kinetic Study

Kinetic studies were performed to investigate the rate of methyl orange adsorption onto the LDH adsorbent and to identify the mechanism controlling the adsorption process.

A fixed concentration which is 40 ppm methyl orange solution (based on optimised C_o) was prepared and the optimised amount of LDH which are 0.65g was added. The mixture was stirred using a water bath shaker at the 70°C temperature at pH7.78 and 170 rpm agitation speed. Samples were withdrawn at predetermined time intervals (5, 10, 20, 30, 40, 45 min), immediately centrifuged and analysed using UV–Vis spectrophotometry to determine the remaining methyl orange concentration. The adsorption capacity at time t (q_t) was calculated using:

$$q_e = \frac{(C_o - C_t)V}{m} \quad (3.7)$$

Where C_t was methyl orange concentration at time t (mg/L). The time-dependent adsorption data were analysed using two commonly used kinetic models which is pseudo-first order (PFO) and pseudo-second order (PSO) models (Kanwal et al., 2024):

$$\text{Pseudo-first-order model:} \quad \log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (3.8)$$

Where k_1 is pseudo-first-order rate constant (1/min).

$$\text{Pseudo-second-order model:} \quad q_t = \frac{q_e^2 k^2 t}{1 + q_e k^2 t} \quad (3.8)$$

Where k_2 is pseudo-second-order rate constant (g/mg.min). Nonlinear regression was performed to determine model parameters and assess model suitability. Model selection was based on R^2 , sum of squared errors (SSE) and agreement between experimental and theoretical q_e values (Isidoro Ribeiro et al., 2023).

3.9 Real Sample

To evaluate the applicability of the optimised adsorption conditions, textile wastewater sample were collected from the Textile Laboratory at University Teknologi MARA (UiTM) Kuala Pilah, Negeri Sembilan. Prior to adsorption, 50 mL of the wastewater samples were collected from three different batch originating from separate practical class sessions conducted in the same laboratory and each batch were generated from different groups of students performing similar dyeing experiments. The initial methyl orange concentration in the wastewater was determined using a UV–Vis spectrophotometer at a wavelength of 190–1100 nm.

Adsorption experiments were performed under the optimised conditions obtained from the response surface methodology (RSM), which is 0.65 g of MgAl-LDH

adsorbent, pH solution of 7.78, initial methyl orange concentration of approximately 40 ppm and a contact time of 20 minutes. A fixed solution volume of 15 mL was used for each experiment. The mixture was agitated at room temperature to ensure uniform contact between the adsorbent and the wastewater. After adsorption, the suspension was filtered and the residual methyl orange concentration was measured using UV–Vis spectrophotometry. Each experiment was conducted in triplicate to ensure reproducibility. The percentage of methyl orange removal efficiency (%) was calculated based on the difference between the initial and final methyl orange concentrations.

3.10 Reusability Study

The adsorption–desorption study was carried out to evaluate the reusability and regeneration performance of MgAl-layered double hydroxide (MgAl-LDH) for methyl orange removal. The adsorption experiments were conducted under the optimised conditions obtained from the Response Surface Methodology (RSM), namely pH solution of 7.78, initial methyl orange concentration of 40 mg/L, contact time of 20 min and an MgAl-LDH amount of 0.65 g for the first cycle. For subsequent cycles, a fixed adsorbent mass of 0.65 g was used to maintain sufficient material for repeated handling.

In each adsorption cycle, 15 mL of methyl orange solution was mixed with the 0.65 g of MgAl-LDH and agitated using an orbital shaker at 70°C for 20 mins. After adsorption, the suspension was centrifuged to separate the adsorbent from the solution. The supernatant was collected and analysed using UV–Vis spectrophotometry to determine the equilibrium methyl orange concentration (C_e).

The dye loaded MgAl-LDH obtained after adsorption was subjected to desorption using 15 mL of 0.1 M NaOH solution as the desorbing agent. The mixture was shaken for 30 min to promote desorption of methyl orange from the adsorbent surface. Sodium hydroxide was selected due to its effectiveness in weakening the electrostatic attraction between the negatively charged methyl orange molecules and the positively charged MgAl-LDH layers. After desorption, the suspension was centrifuged and the supernatant was analysed by UV–Vis spectrophotometry to determine the desorbed methyl orange concentration (C_d). Following desorption, the regenerated MgAl-LDH was washed repeatedly with deionised water to remove residual NaOH, dried at room temperature and reused in the next adsorption cycle. This adsorption–

desorption procedure was repeated for four consecutive cycles to assess the reusability and stability of the MgAl-LDH adsorbent (Badri et al., 2022; Isidoro Ribeiro et al., 2023). The desorption efficiency was calculated using the following equations:

$$\text{Desorption efficiency (\%)} = \frac{C_d \times V_d}{(C_o - C_e) \times V_a} \times 100 \quad (3.9)$$

Where C_d is the concentration of methyl orange in the desorption solution (mg/L), V_d is the volume of the desorption solution (L), C_o is the initial concentration of methyl orange before adsorption (mg/L), C_e is the equilibrium concentration of methyl orange after adsorption (mg/L) and V_a is the volume of the adsorption solution (L).

3.11 Method Flowchart

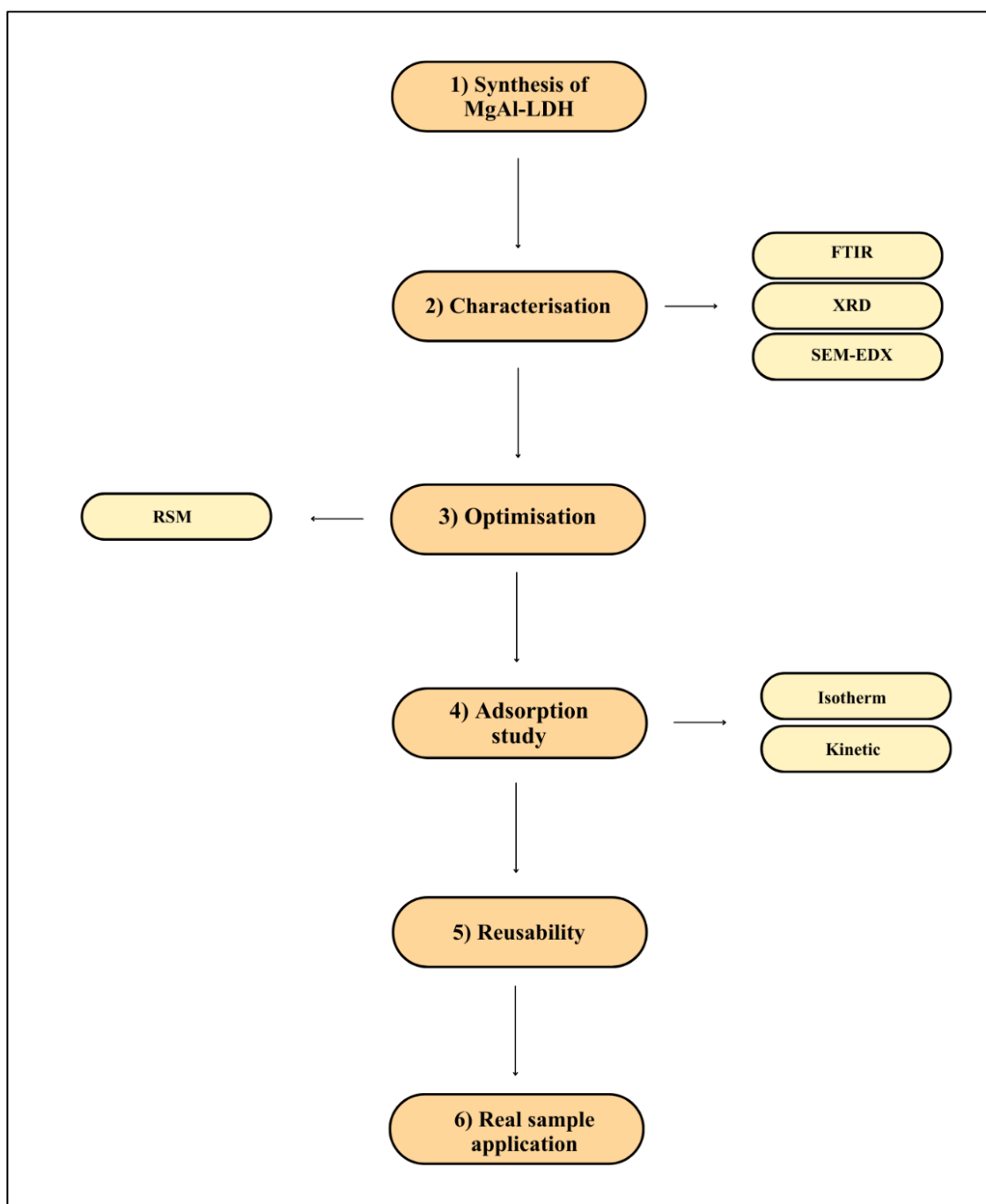


Figure 3.1 Flowchart Methodology of Removal Methyl Orange

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Synthesis of MgAl-Layered Double Hydroxide (MgAl-LDH)

MgAl-LDH was successfully synthesised via the co-precipitation method using magnesium nitrate hexahydrate, $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and aluminium nitrate nonahydrate, $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ as the metal precursors while sodium hydroxide, NaOH and sodium carbonate, Na_2CO_3 were used as the precipitating and carbonate sources, respectively. During the synthesis process, Mg^{2+} and Al^{3+} ions were first dissolved in the methanol and water medium to form a homogeneous metal salt solution (Lin et al., 2017; Modrogan et al., 2020). The addition of NaOH increased the pH of the system and supplied hydroxide ions, which promoted the precipitation of metal hydroxide species. At alkaline pH, Mg^{2+} and Al^{3+} ions reacted with OH^- ions to form magnesium and aluminium hydroxide species, which then underwent nucleation and growth to form the layered double hydroxide structure.

The formation of the MgAl-LDH can be explained based on the development of a brucite-like hydroxide layer as shown in Figure 4.1. In a brucite structure, Mg^{2+} ions are octahedrally coordinated with hydroxide ions to form neutral $\text{Mg}(\text{OH})_2$ layers (Mittal, 2021). However, in MgAl-LDH, part of Mg^{2+} ions is substituted by Al^{3+} ions. Since Al^{3+} has a higher positive charge than Mg^{2+} , this substitution generates an excess positive charge within the hydroxide layers. This positive layer charge must be balanced by the incorporation of interlayer anions. In this study, carbonate ions (CO_3^{2-}) supplied by Na_2CO_3 acted as the charge which balancing the anions and were intercalated between the positively charged MgAl-LDH layers together with water molecules. Therefore, the general structure of the synthesised LDH can be represented as $[\text{M}^{2+}_{1-x}\text{M}^{3+}_x(\text{OH})_2]^{x+} [\text{A}^{n-}_{x/n} \cdot m\text{H}_2\text{O}]$ (Riaz et al., 2025).

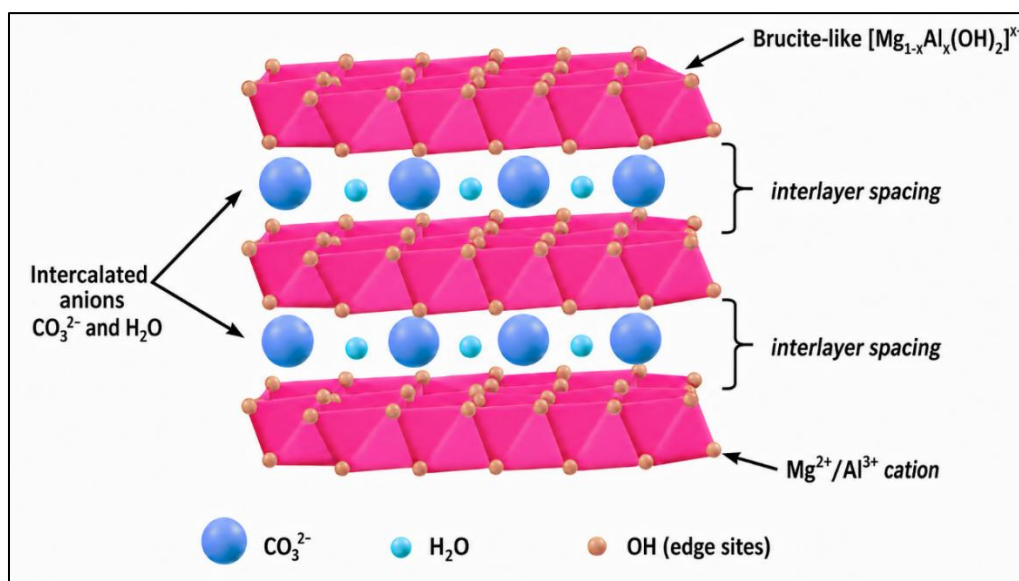


Figure 4.1 Schematic Illustration on the Formation of MgAl-LDH

The role of sodium carbonate is important because carbonate ions strong affinity toward the positively charged LDH layers. The presence of carbonate ions helps to stabilize the interlayer region and maintain the ordered layered structure of MgAl-LDH . Meanwhile, nitrate ions originating from the magnesium and aluminium nitrate salts remain mainly in the solution and are removed during the washing process. The washing step also removes excess NaOH, sodium nitrate and unreacted species while drying produces the final MgAl-LDH powder.

The aging process at 70 °C for 24 hours further promotes the crystallization and structural ordering of MgAl-LDH. During aging, the initially formed precipitate undergoes reorganization which allowing better stacking of the hydroxide layers and more uniform arrangement of carbonate ions and water molecule within the interlayer space, as illustrated in Figure 4.1. This process contributes to the formation of a stable layered structure with define interlayer spacing.

4.2 Characterisation of LDH

Characterisation of the synthesised MgAl-layered double hydroxide (MgAl-LDH) is an essential step to confirm the successful formation of the layered structure and to understand its physicochemical properties prior to adsorption application. Comprehensive characterisation allows evaluation of the crystallographic structure, functional groups, surface morphology and elemental composition of the LDH, all of

which play important roles in determining its adsorption performance toward methyl orange. In this study, X-ray diffraction (XRD) was employed to identify the crystal structure and phase purity of the synthesised MgAl-LDH and to confirm the formation of a hydrotalcite-like layered structure. Fourier transform infrared spectroscopy (FTIR) was used to identify the characteristic functional groups, metal–oxygen bonds and interlayer species such as hydroxyl groups and carbonate anions. Meanwhile, scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM–EDX) was used to examine the surface morphology, particle aggregation and elemental composition of the synthesised LDH.

4.2.1 X-ray Diffraction

The X-ray diffraction (XRD) pattern of the synthesised magnesium–aluminium layered double hydroxide (MgAl-LDH) is shown in Figure 4.2. The diffraction peaks were observed at approximately $2\theta = 11.5^\circ, 23^\circ, 34^\circ, 39^\circ, 46^\circ$ and 61° , which correspond to the (003), (006), (009), (015), (018) and (110) planes, respectively. These characteristic reflections are in good agreement with the standard pattern of hydrotalcite like MgAl-LDH (JCPDS card no. 38-0486), confirming the successful formation of the layered double hydroxide structure.

The strong and sharp peak at around 11.5° indicates a well-developed layered structure, representing the (003) plane which is the most important reflection for LDH materials because it represents the basal spacing between the layered hydroxide sheets. The presence of this low angle reflection indicates the regular stacking of brucite-like layers along the c-axis. Based on Bragg's law, the calculated basal spacing (d_{003}) was approximately 7.69 Å, which is typical for carbonate-intercalated MgAl-LDH (Qin et al., 2025). This confirms that carbonate ions from sodium carbonate were successfully incorporated into the interlayer region during the co-precipitation process. The carbonate ions play an important role in balancing the positive charge generated by the substitution of Mg^{2+} with Al^{3+} within the hydroxide layers.

The presence of higher-order reflections which is (006) and (009) further supports the high crystallinity and periodic stacking of the LDH layers. The reflections at other higher angles, particularly around $2\theta = 39^\circ, 46^\circ$ and 61° , correspond to in-plane (018) until (110) planes, indicating the presence of a well-ordered brucite-like

octahedral sheet composed of Mg^{2+} and Al^{3+} cations coordinated with hydroxide ions (Badri et al., 2022; Qin et al., 2025).

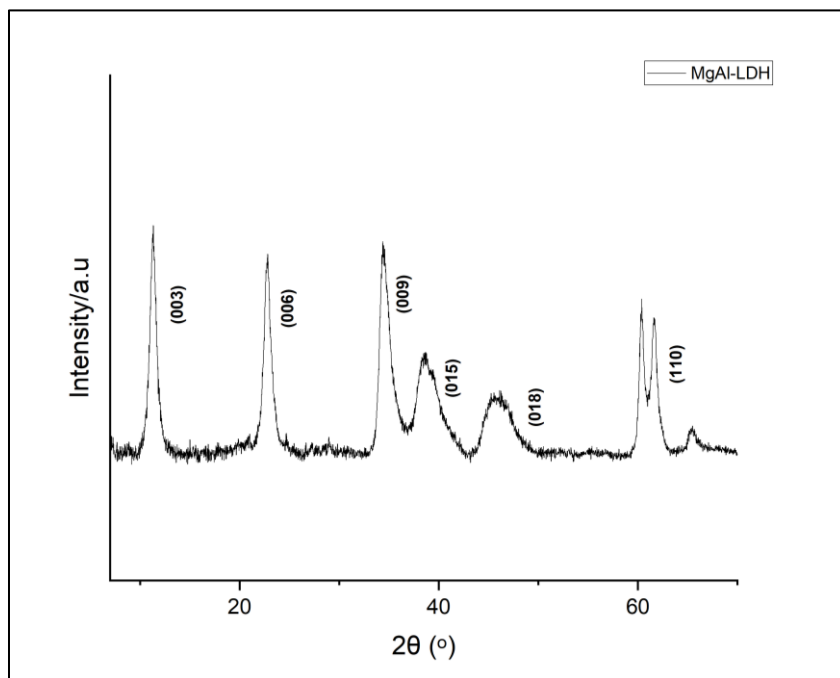


Figure 4.2 XRD Spectrum of MgAl-LDH

The formation of the MgAl-LDH structure is closely related to the synthesis mechanism discussed earlier. During co-precipitation, the addition of NaOH promotes the formation of Mg and Al hydroxide species while partial substitution of Mg^{2+} by Al^{3+} produces positively charged brucite-like layers. The presence of carbonate ions in the synthesis allows charge compensation between the layers, leading to the formation of a stable layered structure (Sarkar and Upadhyay, 2025). Therefore, the XRD pattern supports the proposed formation pathway of the MgAl-LDH through hydroxide precipitation, layer growth, carbonate intercalation and structural order during the aging process.

The XRD result is important in supporting the proposed adsorption mechanism of methyl orange onto MgAl-LDH. The confirmed layered structure indicates the presence of positively charged hydroxide layers and interlayer carbonate ions, which are both important for adsorption of anionic methyl orange. The positively charged LDH layers can promote electrostatic attraction with the negatively charged sulfonate

groups of methyl orange, while the interlayer carbonate ions provide possible exchangeable sites for partial anion exchange.

4.2.2 Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy

The ATR-FTIR spectrum of the synthesised MgAl layered double hydroxide (MgAl-LDH) is presented in Figure 4.3. Several characteristic adsorption bands were observed, indicating successful formation of the LDH structure. A broad and intense band appears around 3467 cm^{-1} , which is attributed to the stretching vibrations of hydroxyl groups (-OH) from both brucite-like layers and the interlayer water molecules. The broad nature of this band indicates extensive hydrogen bonding between hydroxyl groups and water molecules within the layered structure of MgAl-LDH. This is a typical feature of layered double hydroxide where water molecules are commonly located within the interlayer region and on the surface of the material (Hosseini and Ghasemi, 2020; Shan et al., 2014).

The absorption band observed at approximately 1648 cm^{-1} corresponds to the bending vibration of interlayer water molecules. The presence of this band further supports the formation of a hydrated layered structure which is consistent with the general LDH formula containing interlayer water molecules (Hosseini and Ghasemi, 2020). This indicates that water molecules were retained between the layers during the co-precipitation and aging process.

A sharp and strong absorption band at approximately 1359 cm^{-1} is assigned to the asymmetric stretching vibration of carbonate ions (CO_3^{2-}). This confirms that carbonate ions were successfully incorporated into the interlayer region of MgAl-LDH during synthesis which the carbonate ions are originated from sodium carbonate (Na_2CO_3) which was added during the co-precipitation process. Their presence is important because carbonate ions act as charge balancing anions to compensate for the positive charge generated by the partial substitution of Mg^{2+} with Al^{3+} in the hydroxide layers (Sarkar and Upadhyay, 2025). Therefore, the carbonate band strongly supports the proposed formation mechanism of MgAl-LDH, where hydroxide precipitation is followed by the formation of the positively charged layers and incorporation of carbonate ions within the interlayers.

In the lower wavenumber region, absorption bands at approximately at 893 cm^{-1} and 691 cm^{-1} are assigned to metal-oxygen and metal-hydroxide lattice vibrations which particularly Mg-O and Al-O vibrations within the brucite-like octahedral layers consisting of Mg^{2+} and Al^{3+} coordinates with hydroxide ions. This supports the successful development of the MgAl-LDH stacking rather than only the precipitation of separate metal hydroxide species (Zaghloul et al., 2020).

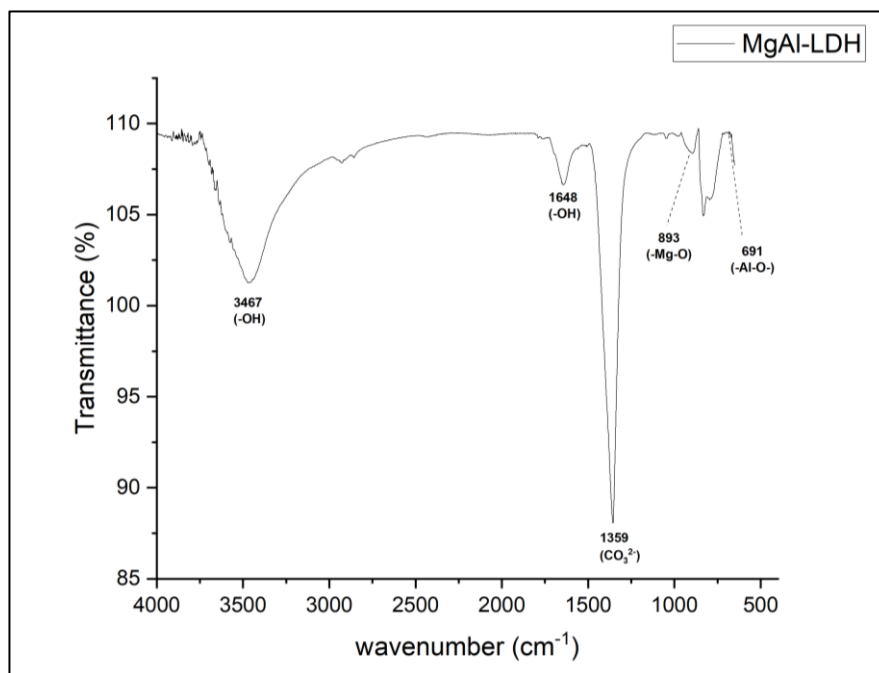


Figure 4.3 FTIR Spectrum of MgAl-LDH

This FTIR result also relevant to the proposed adsorption behaviour of methyl orange onto MgAl-LDH. The presence of hydroxyl groups on the LDH surface provides possible interaction sites for methyl orange through surface adsorption and hydrogen bonding. In addition, the confirmed presence of interlayer carbonate ions supports the possibility of partial anion exchange during adsorption, where negatively charged methyl orange species may interact with the positively charged LDH layers and exchange with accessible carbonate ions. Therefore, the FTIR spectrum confirms the presence of the important structural and functional components including hydroxyl group, interlayer water, carbonate ions and metal-oxygen bonds which contribute to the adsorption performance of MgAl-LDH (Qin et al., 2025).

4.2.3 Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy

The surface morphology of the synthesised MgAl-LDH was examined using scanning electron microscopy (SEM). Figure 4.4 shows at low (20 μm), intermediate (20 μm) and high (500 nm) magnifications.

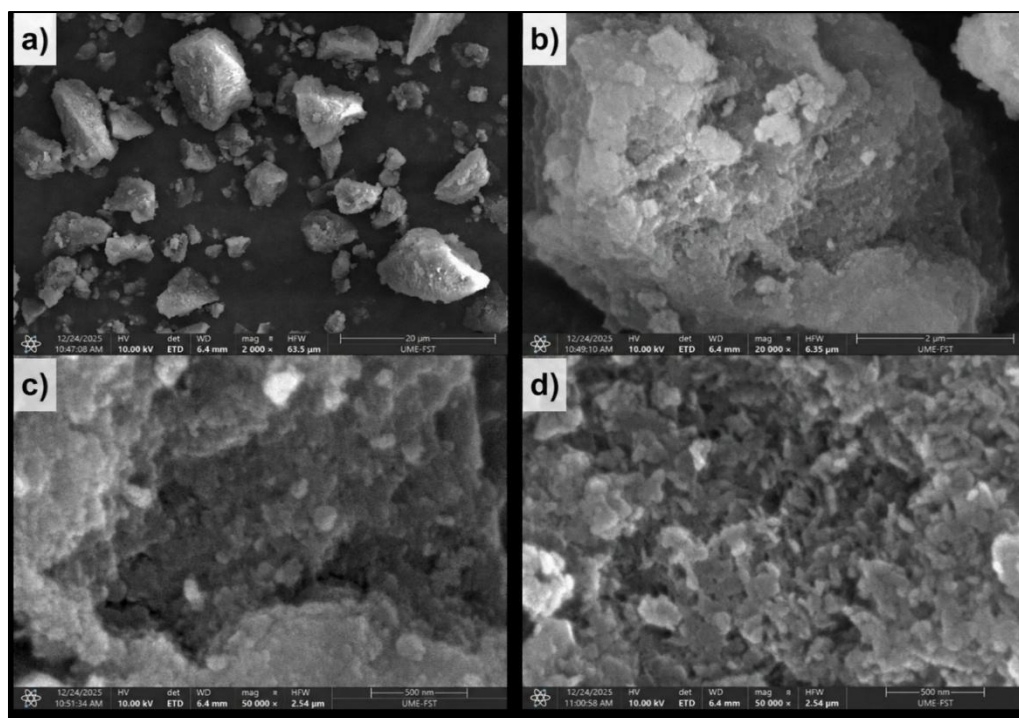


Figure 4.4 SEM Photograph of MgAl-LDH at a) 20 μm , b) 2 μm , c) 500 nm and d) 500 nm

At low magnification (20 μm), the MgAl-LDH sample exhibits irregularly shaped agglomerates composed of fine particles clustered together. This agglomeration is commonly observed in LDH materials due to interparticle interaction such as hydrogen bonding, electrostatic attraction and the stacking of layered hydroxide sheets during drying process. The formation of agglomerates suggests a high surface energy, which is beneficial for adsorption applications as it increases the availability of active sites (Badri et al., 2022). At intermediate magnification (2 μm), the morphology becomes more distinguishable, revealing plate-like and flaky structures, which are characteristic of LDH materials synthesised via the co-precipitation method. These plate-like crystallites originate from stacked brucite-like layers, confirming the successful formation of the layered double hydroxide structure. The overlapping and stacking of these plates create interparticle voids that may facilitate methyl orange

diffusion during adsorption. At higher magnification (500 nm), the nanoscale features of MgAl-LDH are clearly observed. Thin layered sheets with rough and uneven surfaces are visible, indicating a high density of exposed edges and surface defects. Such surface irregularities are advantageous for adsorption, as they provide additional active sites for electrostatic interaction, hydrogen bonding and anion exchange with methyl orange molecules. The absence of smooth, compact surfaces further suggests that the LDH possesses a porous texture, enhancing mass transfer during the adsorption process (Hosseini and Ghasemi, 2020; Zaghloul et al., 2020).

The observed rough surface and layered morphology are important for the adsorption process. The irregular surface texture increases the contact area between MgAl-LDH and methyl orange solution while the exposed edges and surfaces provide active sites for interaction with dye molecules. These sites may contribute to electrostatic attraction between the positive charged LDH layers and the negatively charged sulfonate group of molecules to diffuse toward interlayer diffusion mechanisms.

The element composition of the synthesised MgAl-LDH was further analysed using energy-dispersive X-ray spectroscopy (EDX) as shown in Figure 4.5 and Table 4.1. The EDX spectra confirmed the presence of magnesium (Mg), aluminium (Al), oxygen (O) and carbon (C) on the synthesised material. The detection of Mg and Al indicates that both metal cations were successfully incorporated into the LDH framework while the high oxygen content is attributed to the hydroxide layers, interlayer water molecules and carbonate species (Modrogon et al., 2020). The presence of carbon is related to carbonate ions located in the interlayer region which originated from sodium carbonate used during synthesis.

The EDX analysis was performed at two different sites to evaluate the elemental distribution and homogeneity of the synthesised MgAl-LDH. Both sites showed similar element composition, confirming that Mg and Al were consistently distributed throughout the sample (Hosseini and Ghasemi, 2020). The calculated Mg and Al atomic ratios were approximately 3.05 and 3.13 for site 1 and site 2, respectively. These values are close to the theoretical Mg:Al ratio of 3:1 used during synthesis which indicate that co-precipitation method successfully produced MgAl-LDH with the intended metal composition. This ratio is important because it provides a suitable balance between positive layer charge, structural stability and anion exchange capacity.

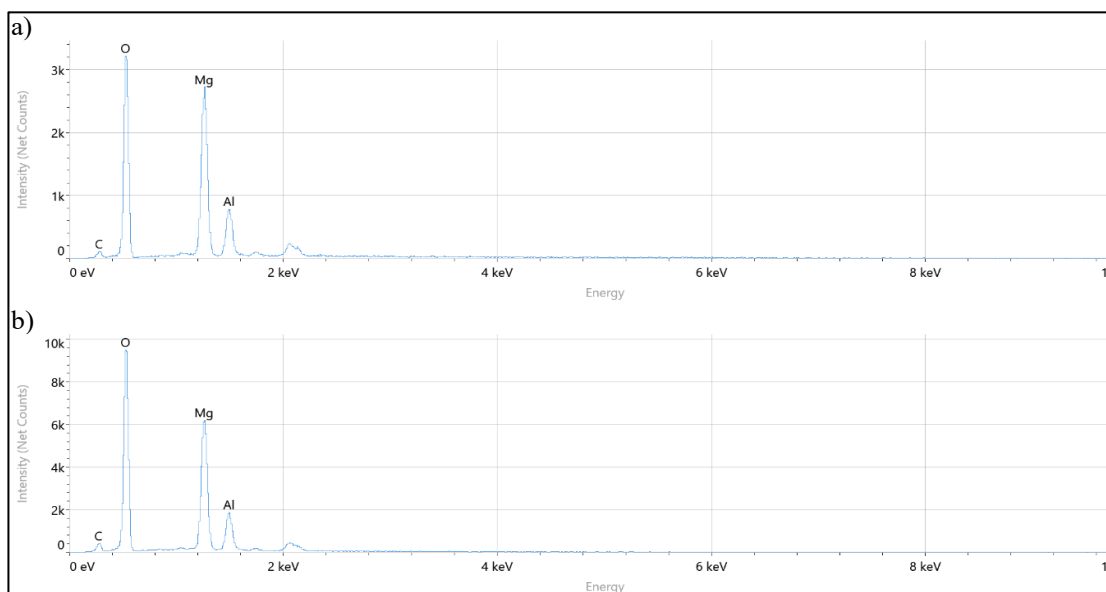


Figure 4.5 EDX Data of MgAl-LDH (a) Site 1 and (b) Site 2

Table 4.1

EDX Data of Element in MgAl-LDH

Element	Atomic (%)		Weight (%)	
	Site 1	Site 2	Site 1	Site 2
C	3.4	4.7	2.1	3.0
O	58.5	62.3	48.6	53.1
Mg	28.7	25.0	36.2	32.4
Al	9.4	8.0	13.1	11.5

The presence of Mg, Al, O and C also supports the proposed formation mechanism of MgAl-LDH. During synthesis, Mg^{2+} and Al^{3+} reacted with hydroxide ions to form brucite-like hydroxide layers while carbonate ions were incorporated into the interlayer region to balance the positive layer charge. Therefore, the EDX result complements the SEM observation by confirming that the plate-like particles observed on the micrographs are composed of the expected elements of carbonate intercalated MgAl-LDH (Qin et al., 2025). The absence of major impurity elements further indicates that the washing process effectively removed soluble by-product such as residual nitrate and sodium salts.

SEM-EDX analysis confirms that MgAl-LDH was successfully synthesised with aggregated plate-like morphology and a homogeneous elemental composition. The layered and rough surface structure provides favourable adsorption site while the presence of Mg, Al, O and carbonate related carbon supports the formation of carbonate

intercalated MgAl-LDH. These characteristics are important for methyl orange adsorption because they provide positively charged layers, surface active sites and possible interlayer regions for electrostatic attraction, surface adsorption and anion exchange assisted adsorption.

4.3 Standard Curve for Methyl Orange Removal

Figure 4.6 shows the standard calibration curve of methyl orange that was constructed based on absorbance value of methyl orange at six different concentrations in the range of 0 to 60 mg/L. The linear equation of the curve was used in determining the concentration of residual methyl orange after adsorption by LDH where y is the concentration to be determined and x is the absorbance value of methyl orange. A measure of the fit can be found in the coefficient determination (R^2) displayed in Figure 4.6. High precision and accuracy of the data used to build the calibration curve are indicated by an R^2 value of 1.0. The linear graph was found to well-fitted, as indicated by the high coefficient determination value ($R^2 = 0.9985$) produced based on the standard curve, with the value being around 1.0. The high degree of data accuracy for this standard curve made it dependable to use in calculating the amount of methyl orange in wastewater after adsorption by LDH. The outcome shown that the absorbance value of methyl orange increased as the concentration methyl orange increased.

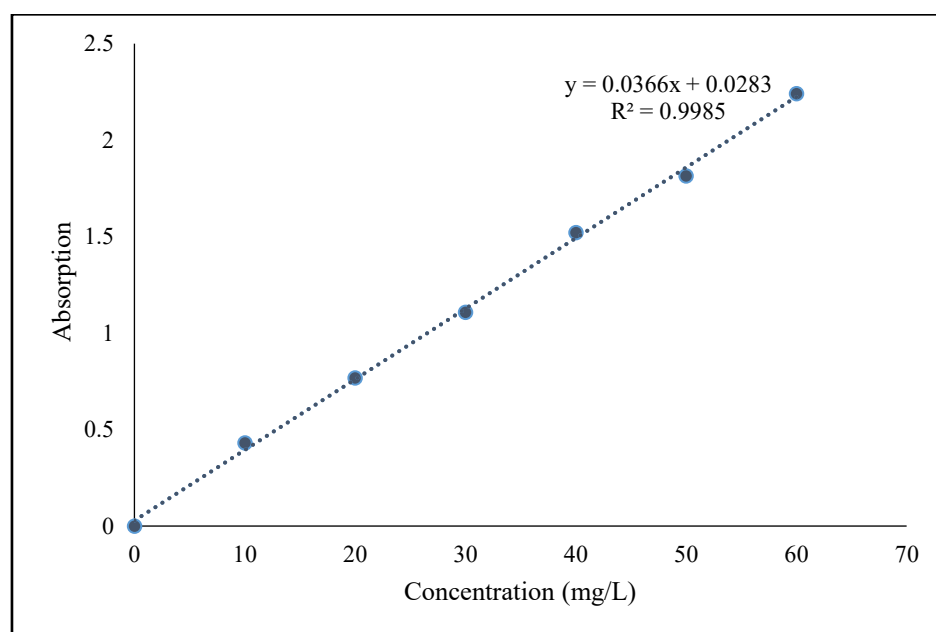


Figure 4.6 Standard Calibration Curve of Methyl Orange

4.4 Optimisation of Adsorption

In this study, the central composite design (CCD) was carried out for the optimisation of methyl orange adsorption by LDH in wastewater. The operating variables chosen for evaluation were mass of adsorbent, pH solution, initial methyl orange concentration and contact time. These variables were used to study their effects in the adsorption process towards the percentage removal of methyl orange. A total of 30 runs were executed in this study. Absorbance readings of each sample run were inserted into Equation (3.2) using final concentrations of residual methyl orange to calculate the methyl orange percentage. Table 4.2 shows the removal percentage of methyl orange obtained under different testing conditions. Based on the experimental results, the predicted values generated by the quadratic model closely matched the actual experiment results with only minor deviations. This close agreement demonstrates the reliability and accuracy of the developed model in predicting methyl orange efficiencies under various experimental conditions.

The highest percentage of methyl orange removal was 93.94% obtained at 0.45g of LDH amount, pH 7, initial concentration of 50 ppm and 30 min of contact time. This condition provided a strong mass transfer driving force and a surface environment that maximised electrostatic attraction between the anionic form of methyl orange and the positively charged LDH surface. Meanwhile, the lowest methyl orange was recorded at 0.45g of LDH amount, pH 3, 30 ppm initial concentration and 30 min of contact time with the percentage value of 60.66%. The small standard deviation and %RSD values (< 5%) obtained for all experiments indicate excellent precision and reproducibility of the adsorption measurements. In adsorption and environmental analysis studies, %RSD values below 5% are generally considered acceptable, while values below 2% indicate excellent analytical reliability (Lucia Carmen and Afrodita Doina, 2018). Therefore, the low variation observed in this study confirms the consistency and robustness of the experimental procedure.

Table 4.2
Experimental Run, Factors and Response for the Removal Methyl Orange by MgAl-LDH

Run	Factors				Response (% Removal MO)	
	Adsorbent Dosage (gram)	pH Solution	Methyl Orange Initial Concentration (mg L ⁻¹)	Contact Time (min)	Actual (%) (Mean $\pm$ SD, n=3)	Predicted (%)
1	0.25	9	20	20	85.91 $\pm$ 0.01	86.37
2	0.65	5	20	40	79.03 $\pm$ 0.01	78.68
3	0.25	5	40	20	74.69 $\pm$ 0.02	75.22
4	0.25	9	40	40	88.06 $\pm$ 0.01	88.15
5	0.25	5	20	20	70.22 $\pm$ 0.04	69.96
6	0.45	3	30	30	60.66 $\pm$ 0.02	61.19
7	0.65	9	20	20	84.99 $\pm$ 0.02	85.46
8	0.45	7	30	30	84.28 $\pm$ 0.02	84.62
9	0.65	5	20	40	74.60 $\pm$ 0.01	74.22
10	0.45	11	30	30	78.46 $\pm$ 0.03	77.85
11	0.25	5	40	40	82.16 $\pm$ 0.04	81.59
12	0.65	9	40	20	90.82 $\pm$ 0.01	90.71
13	0.85	7	30	30	82.14 $\pm$ 0.02	82.57
14	0.45	7	30	10	83.46 $\pm$ 0.04	82.97
15	0.45	7	30	50	82.15 $\pm$ 0.03	82.56
16	0.05	7	30	30	86.02 $\pm$ 0.03	85.51
17	0.65	9	40	40	79.63 $\pm$ 0.03	79.80
18	0.45	7	50	30	93.94 $\pm$ 0.01	94.13
19	0.25	9	20	40	90.95 $\pm$ 0.01	91.24
20	0.45	7	30	30	84.19 $\pm$ 0.02	84.62
21	0.65	5	40	40	79.83 $\pm$ 0.03	79.55
22	0.65	5	40	20	85.22 $\pm$ 0.02	84.83
23	0.45	7	30	30	84.42 $\pm$ 0.02	84.62
24	0.45	7	10	30	88.02 $\pm$ 0.04	87.75
25	0.25	9	40	20	87.14 $\pm$ 0.02	87.42
26	0.25	5	20	40	80.17 $\pm$ 0.02	80.46
27	0.45	7	30	30	85.31 $\pm$ 0.01	84.62
28	0.65	5	20	20	75.28 $\pm$ 0.03	75.37
29	0.45	7	30	30	84.18 $\pm$ 0.01	84.62
30	0.45	7	30	30	85.33 $\pm$ 0.01	84.62

4.4.1 Determination of Selected Model

The Sequential Model Sum of Squares analysis was performed to determine the most appropriate model for describing the adsorption behaviour of methyl orange onto

the MgAl-LDH. This statistical test evaluates the contribution of increasingly complex models which is linear, two-factor interactions (2FI), quadratic and cubic. As shown in Table 4.3, the linear model showed a statistically significant improvement over the mean model ($F = 3.91$, $p = 0.0133$), indicating that the linear terms of the factors contribute meaningfully to explaining the response. However, the addition of the interaction terms in the 2FI model did not significantly improve the model ($F = 1.57$, $p = 0.2092$), suggesting that interaction between variables alone were insufficient for accurately predicting methyl orange removal. In contrast, the quadratic model produced a highly significant improvement over the 2FI model, with a large F -value of 386.43 and a p -value of <0.0001 , confirming that inclusion of quadratic terms substantially increased the model's explanatory power (Moosaei et al., 2024). The result indicates that curvature effects are important in describing the adsorption process, likely due to the non-linear relationship between the process parameters and the removal efficiency. The cubic model was found to be aliased and did not provide a significant improvement over model the quadratic model ($F = 1.85$, $p = 0.2157$), meaning it cannot be interpreted reliably. Therefore, the quadratic model was deemed the most suitable and was recommended for further analysis. The small residual sum of squares further supports the adequacy of the quadratic model in fitting the experimental data.

Table 4.3
Sequential Model Sum of Square for Removal Methyl Orange by MgAl-LDH

Source	Sum of Square	dF	Mean Square	F -value	p -value	
Mean vs Total	2.036E+05	1	2.036E+05			
Linear vs Mean	490.51	4	122.63	3.91	0.0133	
2FI vs Linear	259.92	6	43.32	1.57	0.2092	
Quadratic vs 2FI	518.30	4	129.58	386.43	< 0.0001	Suggested
Cubic vs Quadratic	3.42	8	0.4270	1.85	0.2157	Aliased
Residual	1.61	7	0.2305			
Total	2.048E+05	30	6828.15			

The Lack of Fit (LOF) results indicate that the quadratic model is the most appropriate for describing the adsorption behaviour because its LOF p -value is greater than 0.05, showing an insignificant lack of fit, meaning the model adequately captures the relationship between the variables and the response without systematic error (Sarani et al., 2024). In contrast, the linear and 2FI models show highly significant lack of fit

($p < 0.0001$), demonstrating that they fail to represent the curvature and interactions present in the experimental data. Although the cubic model also exhibits an insignificant lack of fit ($p = 0.8533$) which is aliased, making its term unsuitable for use. Therefore, based on the LOF test, the quadratic model provides the best statistical fit and is selected for further optimisation and predictive analysis.

Table 4.4
Lack of Fit Test of Removal Methyl Orange by MgAl-LDH

Source	Sum of Square	dF	Mean Square	F-value	p-value	
Linear	781.74	20	39.09	129.06	< 0.0001	
2FI	521.82	14	37.27	123.07	< 0.0001	
Quadratic	3.52	10	0.3515	1.16	0.4615	Suggested
Cubic	0.0992	2	0.0496	0.1638	0.8533	Aliased
Pure Error	1.51	5	0.3029			

As for Model Summary Statistics in Table 4.5, its clearly support the quadratic model as the most suitable model for describing the adsorption process, as it demonstrates the strongest statistical performance across all key indicators. The quadratic model shows an R^2 (0.9961), indicating that more than 99% of the variation in the response, while the adjusted R^2 (0.9924) confirms that the model remains highly robust even after adjusting for the number of predictors. Additionally, the predicted R^2 (0.9824) is very close to the adjusted R^2 which the difference is less than 0.2, showing excellent predictive capability and minimal overfitting(Karyab et al., 2023). The quadratic model also exhibits the lowest PRESS value (22.43), indicates superior predictive accuracy compared to linear and 2FI models, which PRESS values are much higher. Although the cubic model shows similarly high R^2 values, it is aliased which making it statistically unreliable. Therefore, based on the combination of high R^2 , strong predictive performance and acceptable residual error, the quadratic model is identified as the best-fitting and most reliable model for this study.

Table 4.5
Model Summary Statistic of Removal Methyl Orange by MgAl-LDH

Source	Std. Dev.	R^2	Adjusted R^2	Predicted R^2	PRESS	
Linear	5.60	0.3851	0.2867	0.0514	1208.23	
2FI	5.25	0.5891	0.3729	0.3475	831.15	
Quadratic	0.5791	0.9961	0.9924	0.9824	22.43	Suggested
Cubic	0.4801	0.9987	0.9948	0.9871	16.47	Aliased

4.4.2 Accuracy of Model

The accuracy of model was evaluated using several diagnostic plots including the normal probability plot of residual (Figure 4.7), residual vs predicted plot (Figure 4.8), predicted vs actual plot (Figure 4.9) residual vs run plot (Figure 4.10), cook's distance plot (Figure 4.11) and box-cox plot (Figure 4.12). The normal probability plot of residuals in Figure 4.7 shows that the data points align closely along the reference straight line, indicating that the residuals are approximately normal distributed. This pattern shows the quadratic RSM model fits the experimental data well. The absence of strong curvature, clustering or systematic deviation from the line implies that there are no significant outliers or abnormal error structures affecting the model. The plot validates that the residual behaviour is acceptable and supports the reliability of the developed quadratic model for predicting methyl orange removal performance.

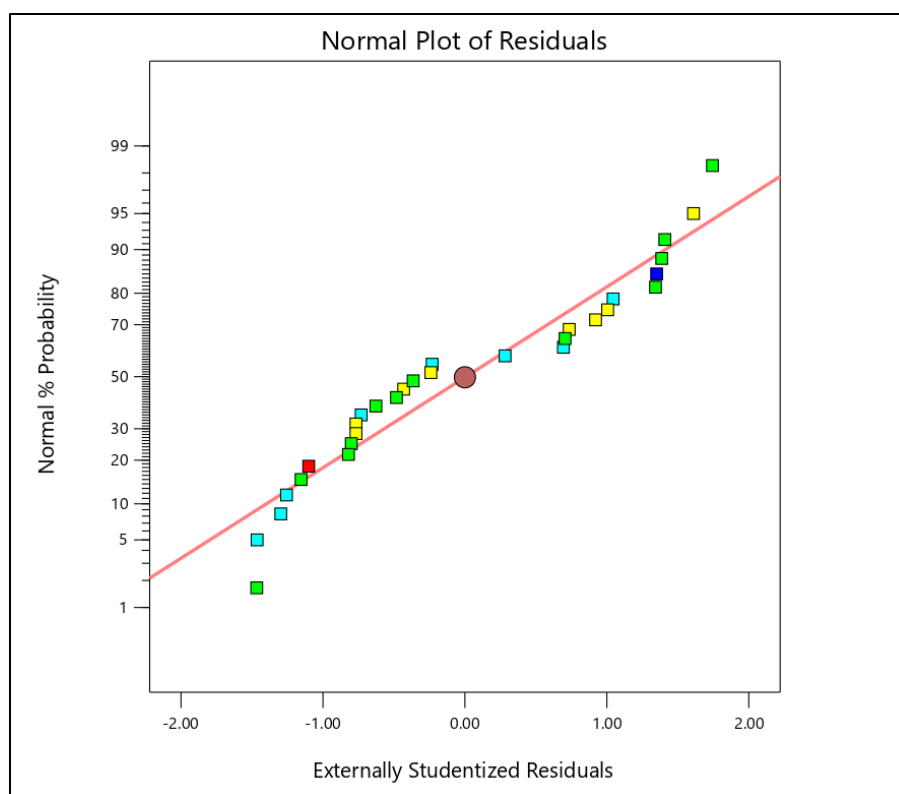


Figure 4.7 Normal Plot of Removal Methyl Orange by MgAl-LDH

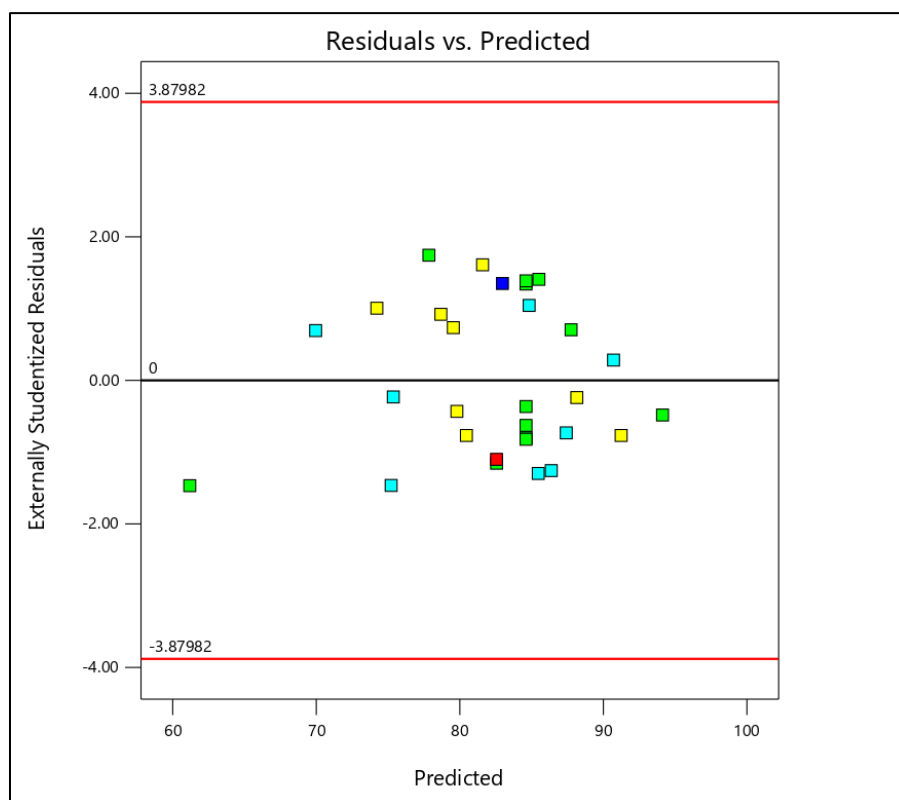


Figure 4.8 Residuals vs Predicted Plot of Removal Methyl Orange by MgAl-LDH

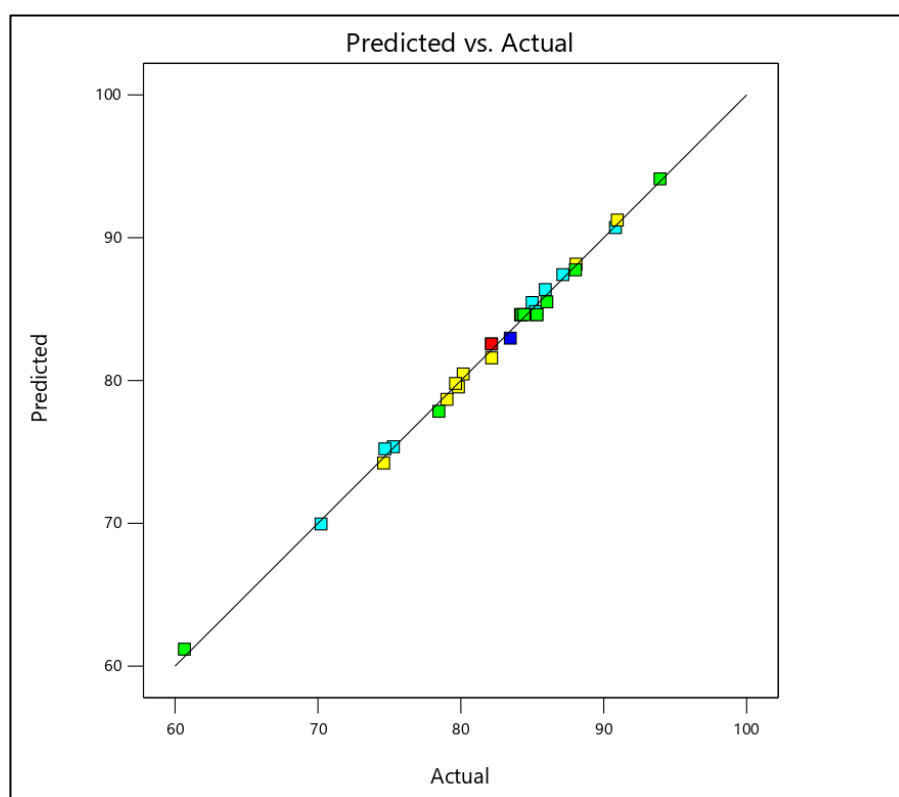


Figure 4.9 Predicted vs Actual Plot of Removal Methyl Orange by MgAl-LDH

Figure 4.8 shows the Residuals vs. Predicted plot that all data points fall within the acceptable range, indicating that no outliers or influential observations are present. The residuals are randomly scattered around zero without forming any obvious pattern which suggesting that the model assumptions linearity and independence are reasonably met. Overall, the randomness and bounded distribution of the residuals confirm that the developed RSM model provides a reliable fit for predicting the final methyl orange concentration.

The Predicted vs. Actual plot in Figure 4.9 demonstrated a strong agreement between the experimental values estimated by the RSM model. All data points fall closely around the line which indicating that model accurately predicts the final methyl orange concentration across the entire experimental range. The absence of large deviations suggests that the model captures the relationship between the variables effectively.

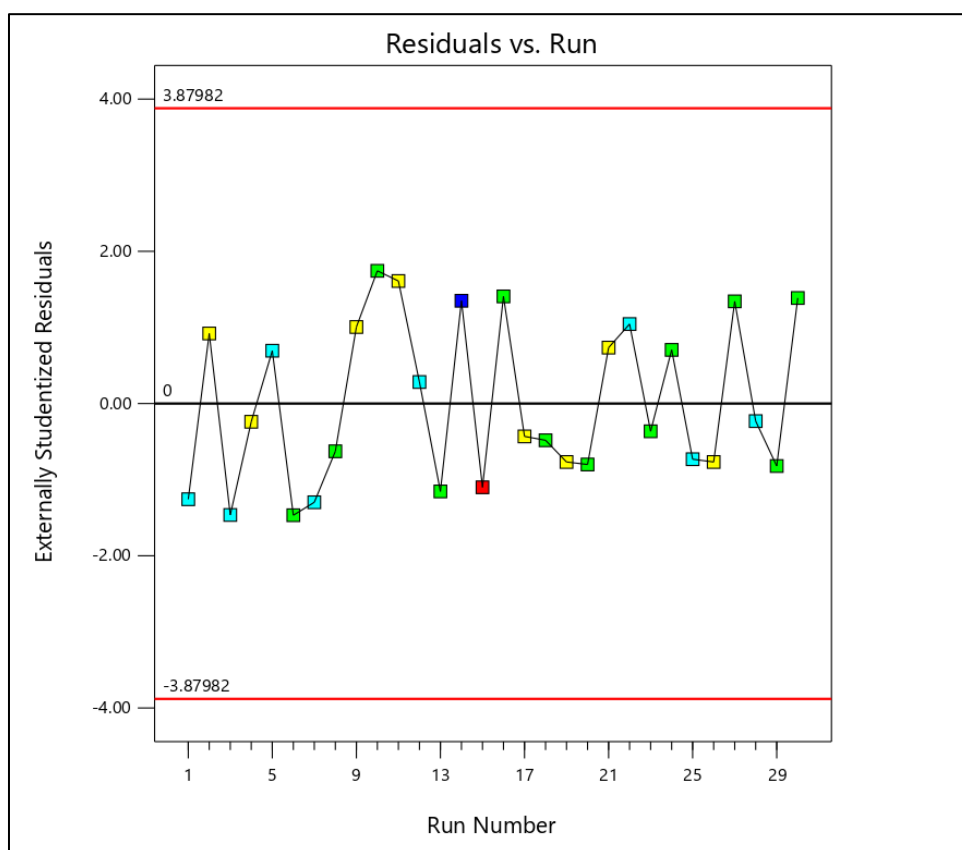


Figure 4.10 Residual vs Run Plot of Removal Methyl Orange by MgAl-LDH

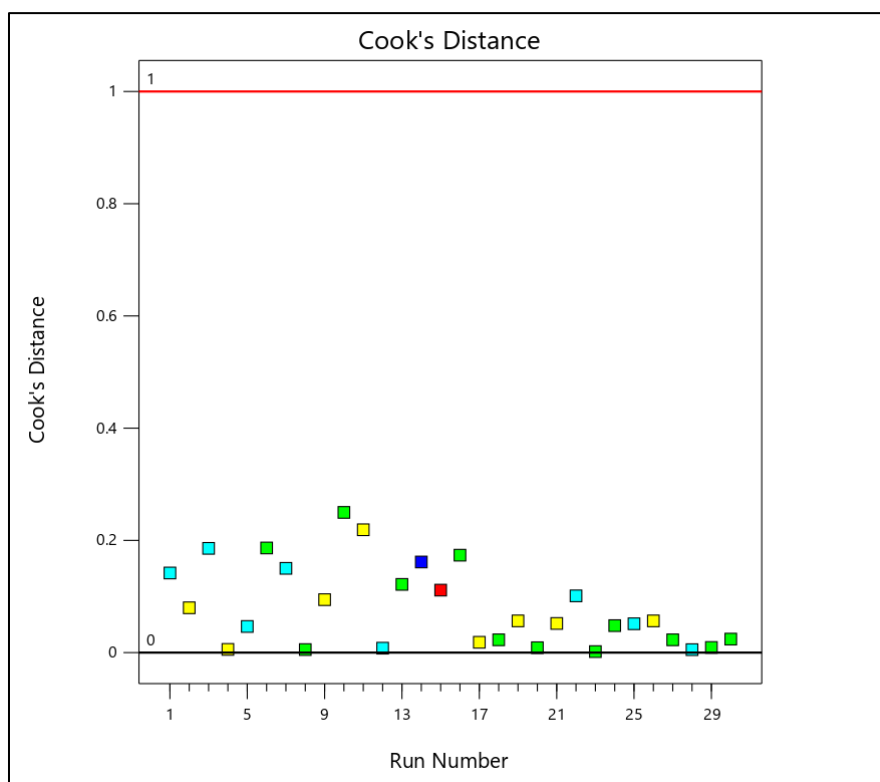


Figure 4.11 Cook's Distance plot of Removal Methyl Orange by MgAl-LDH

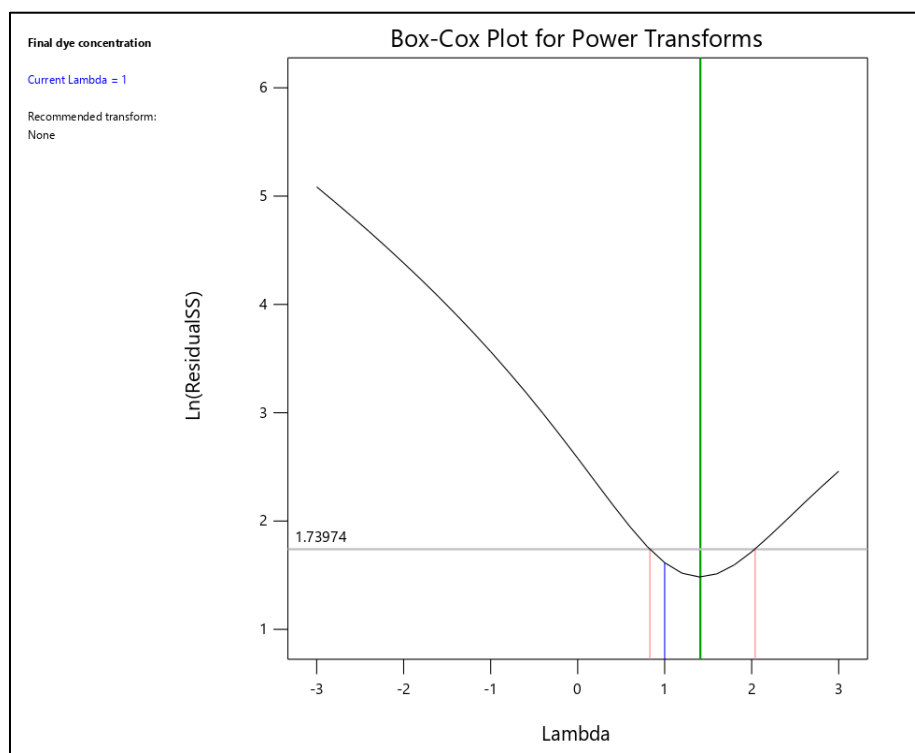


Figure 4.12 Box-Cox plot of Removal Methyl Orange by MgAl-LDH

Figure 4.10 shows the Residual vs Run plot are randomly distributed around zero with no observable pattern, clustering or drift over time. All residuals fall well within the control limits of ± 3.87982 , confirming that no run produced an outlier or unusually large deviation from the model predictions. The alternating sequence of positive and negative residuals further suggests stable experimental performance and the plot verifies that the experimental data were collected consistently and that the quadratic model accurately represent the system across the full sequence of runs.

The Cook's distance plot in Figure 4.11 shows that all experimental runs fall well below the critical threshold value of 1, indicating that none of the data points exert an undue influence on fitted quadratic model. This suggest that no single run disproportionately affects the regression coefficients. Most values remain close to zero which confirming that their influence is minimal and within acceptable limits. The absence of high-leverage points demonstrates that the dataset is stable and the model's predictive performance is not dependent on any observation.

The Box-Cox plot evaluates whether a transformation of the response variable is necessary to improve model performance by stabilising variance of correcting non-normality. By referring to the Figure 4.12 in this study, the curve reaches its minimum residual sum of square at a lambda value of 1 which indicates by the green vertical line and this value lies well within the 95% confidence interval represented by the blue and red boundary lines. Because $\lambda=1$ corresponds to using the original data and since the optimal lambda falls comfortably within the acceptable range, the software explicitly recommends no transformation or repetition needed for the response variable. This means the current data already satisfy the assumptions required for ANOVA and regression modelling, including constant variance and approximate normality of residuals.

4.4.3 Analysis of Model (ANOVA)

The ANOVA result for the quadratic model based on Table 4.6 demonstrate that the model is highly significant, as indicated by the large model F-value of 270.26 and a p-value of <0.0001 , confirming that the quadratic model successfully explains the variability in methyl orange removal rather than occurring by random noise. Among the four main factors which is adsorbent dosage (A), pH solution (B), initial methyl orange

concentration (C) and contact time (D), factor A, B and C show strong significant ($p < 0.0001$) while contact time (D) is not significant individually ($p = 0.3995$). This indicates that the methyl orange removal process is primarily governed by adsorbent dosage, pH and initial concentration. Most two-factor interaction terms which is AB, AC, AD, BC, BD and CD are also significant, highlighting the presence of interactive effects between variables, meaning the influence of one factor strongly depends on the level of another. For the quadratic terms B^2 , C^2 and D^2 are significant contributors to the model, suggesting the presence of curvature in the response surface for these parameters while A^2 is not significant. The small residual error of sum of square (5.03) confirming that the model fits the data well because the unexplained variation is comparable to pure experimental error (Muleta, 2025). Overall, the ANOVA confirms that this quadratic model is statistically valid, reliable and capable of accurately predicting methyl orange adsorption under the tested conditions.

Table 4.6
Analysis of Model of Removal of Methyl Orange by MgAl-LDH

Source	Sum of Square	dF	Mean Square	F-value	p-value	
Model	1268.74	14	90.62	270.26	< 0.0001	significant
A-Adsorbent Dosage	12.99	1	12.99	38.75	< 0.0001	
B-pH Solution	416.33	1	416.33	1241.62	< 0.0001	
C-Initial Methyl Orange Concentration	60.93	1	60.93	181.71	< 0.0001	
D-Contact Time	0.2522	1	0.2522	0.7520	0.3995	
AB	39.94	1	39.94	119.12	< 0.0001	
AC	17.64	1	17.64	52.61	< 0.0001	
AD	135.72	1	135.72	404.76	< 0.0001	
BC	17.77	1	17.77	52.98	< 0.0001	
BD	31.75	1	31.75	94.70	< 0.0001	
CD	17.10	1	17.10	50.99	< 0.0001	
A^2	0.5767	1	0.5767	1.72	0.2094	
B^2	390.87	1	390.87	1165.69	< 0.0001	
C^2	68.47	1	68.47	204.20	< 0.0001	
D^2	5.90	1	5.90	17.59	0.0008	
Residual	5.03	15	0.3353			
Lack of Fit	3.52	10	0.3515	1.16	0.4615	not significant
Pure Error	1.51	5	0.3029			
Cor Total	1273.77	29				

$$Y = 84.62 - 0.7358A + 4.16B + 1.59C - 0.1025D - 1.58AB + 1.05AC - 2.91AD - 1.05BC - 1.41BD - 1.03CD - 0.1450A^2 - 3.77B^2 + 1.58C^2 - 0.4637D^2 \quad (4.1)$$

The final regression equation (4.1) expressed in terms of coded factors provides a quantitative description of how each variable influences the final methyl orange concentration within the experimental range. The intercept value of 84.62 represents the predicted response at the centre point of all factors. The coefficients associated with the linear terms show the direction and magnitude of each factor's effect, where pH solution (B) and initial methyl orange concentration (C) have the largest positive coefficients, indicating a strong increasing effect on the response, while adsorbent dosage (A) has a negative coefficient, reflecting that increasing dosage will increase percent removal. The interaction terms (AB, AC, AD, BC, BD, CD) reveal how the combined effects of two variables deviate from their individual contributions, with negative coefficients indicating synergistic enhancement in methyl orange removal and positive coefficients showing antagonistic effects. Quadratic terms, particularly B² and C², exhibit large coefficients, confirming the curvature in the response surface and supporting the earlier ANOVA results showing significant quadratic effects. This equation allows a straightforward comparison of factor influence magnitudes and is useful for predicting responses under different operational conditions.

4.4.4 Interaction between Variables

The 3D surface plot in Figure 4.13 illustrates the combined effect of adsorbent dosage (A) and pH solution (B) on the methyl orange removal efficiency (%). The surface plot reveals that increasing the adsorbent dosage at moderate pH enhances the methyl orange removal efficiency. As the dosage of MgAl-LDH increases, the number of available active sites and total surface area also increase. This provides more adsorption sites for methyl orange molecules, leading to a higher removal percentage. A greater amount of adsorbent increases the probability of interactions between dye molecules and the adsorbent surface, thereby improving adsorption performance. This trend is clearly observed at moderate pH conditions where the removal efficiency

reaches higher values. At moderate pH which approximately pH 6-8, the adsorption process becomes more favourable due to suitable surface charge conditions. Since methyl orange is an anionic dye, its negatively charged sulfonate group ($-\text{SO}_3^-$) interacts effectively with the positively charged layers of MgAl-LDH. Under extremely acidic conditions, excess H^+ ions may compete with methyl orange molecules for adsorption sites, while at higher pH value, surface deprotonation may reduce electrostatic attraction. Therefore, moderate pH provides a balanced condition for optimal adsorption.

Figure 4.13 also presents the interaction between adsorbent dosage (A) and initial methyl orange concentration (C) on methyl orange removal efficiency (%). The plot demonstrates that decreasing the adsorbent dosage while increasing the initial methyl orange concentration increases the percentage of methyl orange removal. Generally, increasing adsorbent dosage increases the removal efficiency because more active sites become available for adsorption. However, in this case, the 3D surface suggests that at lower dosages the adsorption sites are more effectively utilised. At higher dosages, particle aggregation may occur, which reduces the effectiveness of surface area and limits the accessibility of active sites. As a result, not all adsorption sites participate efficiently in the adsorption process which leads to a less significant in removal percentage. On the other hand, increasing the initial methyl orange concentration enhances the driving force for mass transfer between the solution and the adsorbent surface. A higher concentration increases the probability of collision between methyl orange molecules and the available adsorption sites, thereby improving adsorption efficiency. The interaction between adsorbent dosage and initial methyl orange concentration suggests that adsorption efficiency depends not only on the amount of adsorbent used but also on how effectively the adsorption sites are utilised under different concentration conditions.

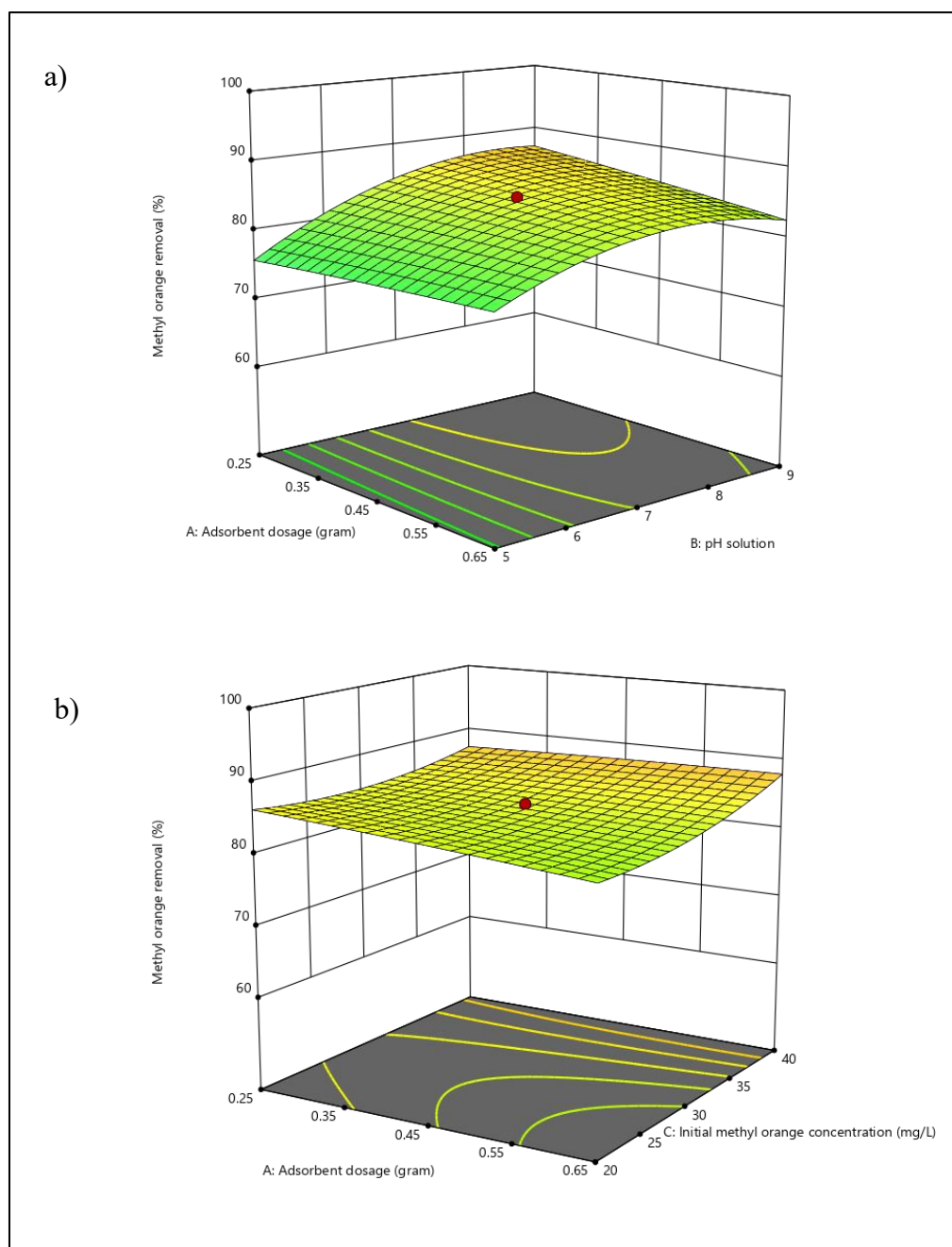


Figure 4.13 3D Surface Plot Methyl Orange Removal: a) Interaction between Adsorbent Dosage and pH Solution (AB); b) Interaction between Adsorbent Dosage and Initial Methyl Orange Concentration (AC)

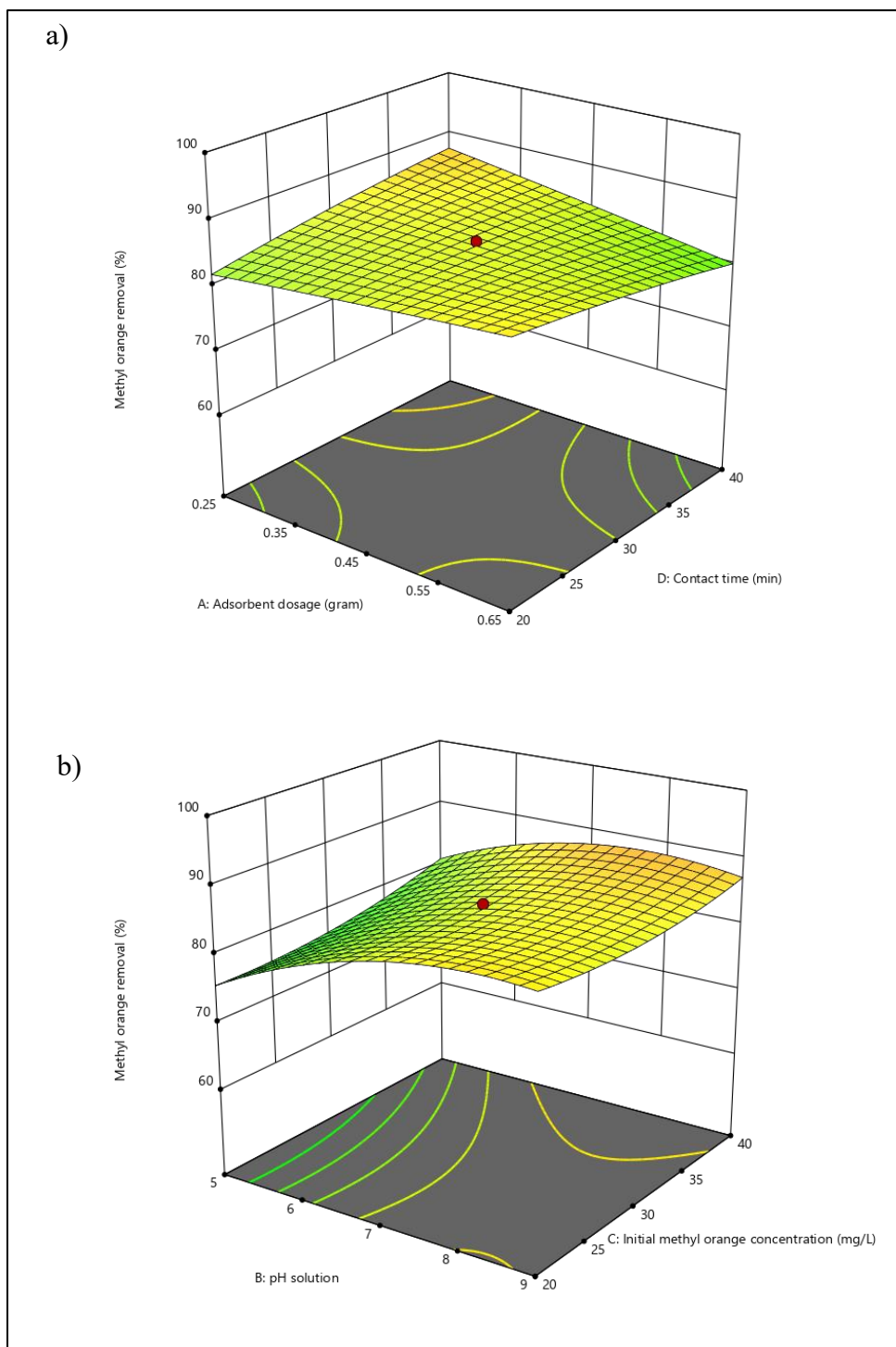


Figure 4.14 3D Surface Plot of Methyl Orange Removal: a) Interaction between Adsorbent Dosage and Contact Time (AD); b) Interaction between pH Solution and Initial Methyl Orange Concentration (BC)

Figure 4.14 shows the interaction between adsorbent dosage (A) and contact time (D) on methyl orange removal efficiency (%). The response surface indicates that increasing the adsorbent dosage while decreasing the contact time enhances the

percentage of methyl orange removal within the studied experimental range. An increase in adsorbent dosage provides a larger number of available active sites for adsorption. With more adsorption sites present in the system, a greater number of methyl orange molecules can be rapidly captured from the solution. This significantly enhances removal efficiency, especially during the initial stage of adsorption. At shorter contact times, adsorption typically occurs rapidly due to the high concentration gradient between methyl orange molecules in solution and the active sites on the adsorbent surface. When the dosage is increased, this initial rapid adsorption phase becomes more dominant because sufficient active sites are immediately available. Therefore, even with reduced contact time, high removal efficiency can still be achieved. This behaviour suggests that the adsorption process approaches equilibrium quickly under higher dosage conditions. Once equilibrium is reached, extending the contact time does not significantly improve removal efficiency. This also explains why contact time may not appear as a highly significant factor individually in the ANOVA results, as the equilibrium was likely achieved within the selected time range. This interaction between the adsorbent dosage and contact time indicates that increasing dosage can compensate for shorter contact time which leads to efficient methyl orange removal in a shorter duration.

Figure 4.14 also illustrates the combined effects of pH solution (B) and initial methyl orange concentration (C) on methyl orange removal efficiency (%). The response surface shows that increasing the initial methyl orange concentration at a moderate pH enhances the percentage methyl orange removal. At moderate pH conditions, the surface of MgAl-LDH carries a relatively positive charge due to partial protonation of hydroxyl groups. Since methyl orange is an anionic dye, strong electrostatic attraction occurs between the positively charged adsorbent surface and the methyl orange molecule. When initial methyl orange concentration increases, the driving force for mass transfer also increases. A higher concentration gradient between the bulk solution and the adsorbent surface enhances diffusion of methyl orange toward active sites. As long as sufficient adsorption sites are available, this increased driving force results in higher removal efficiency. The interaction between pH and initial methyl orange demonstrates that adsorption performance is maximised when both parameters are balanced which is a moderate pH ensure favourable surface charge condition while higher initial methyl orange concentration increases the mass transfer driving force.

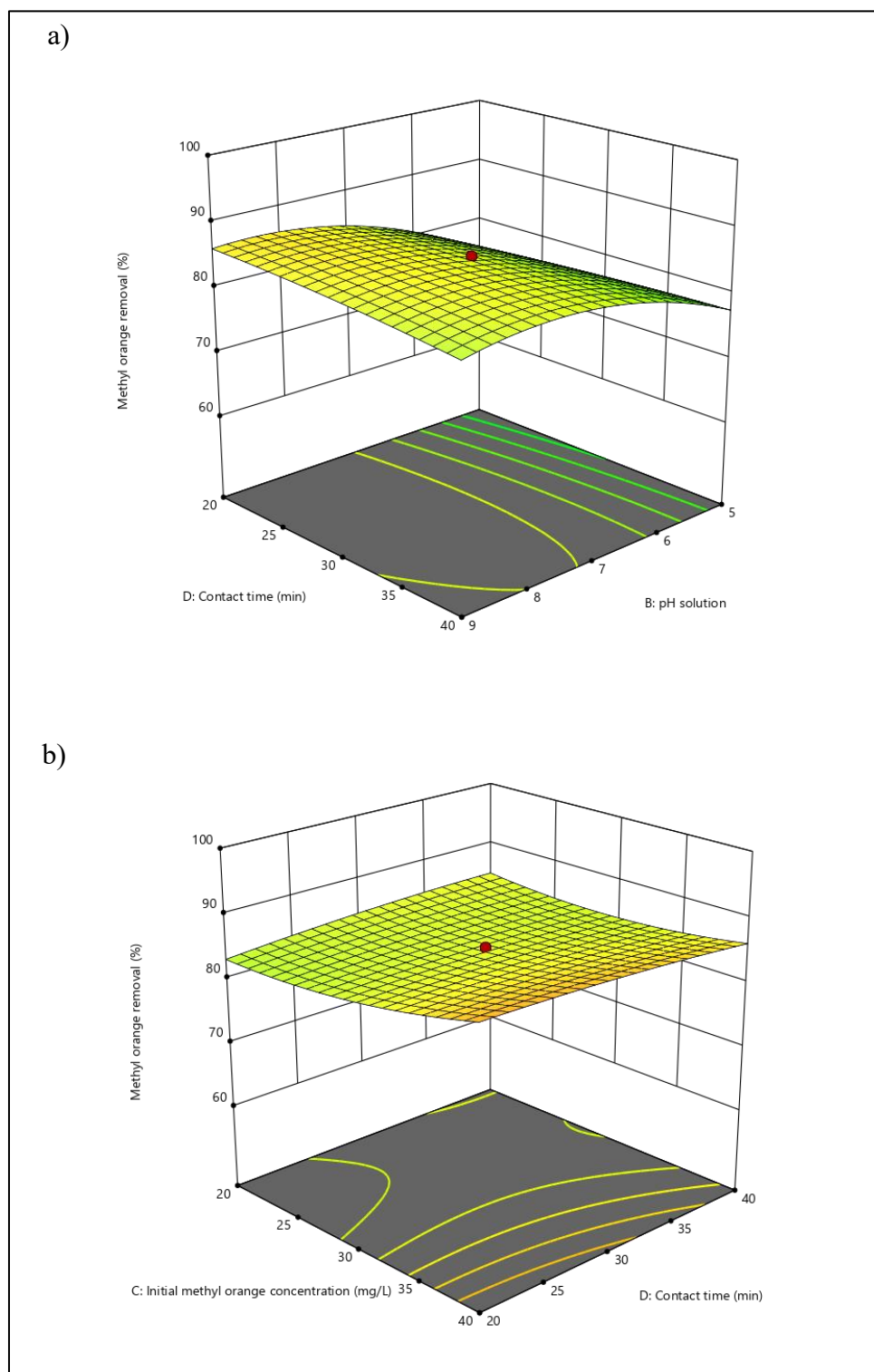


Figure 4.15 3D Surface Plot of Methyl Orange Removal: a) Interaction between Contact Time and pH Solution (DB); b) Interaction between Initial Methyl Orange Concentration and Contact Time (CD)

Figure 4.15 presents the interaction between pH solution (B) and contact time (D) on methyl orange removal efficiency (%). The plot clearly indicates that the percentage removal efficiency increases at moderate pH with shorter contact time. This behaviour suggest that adsorption occurs rapidly under favourable pH conditions. At moderate pH (pH 6-8), the strong electrostatic attraction occurs between the negatively charge sulfonate groups of the methyl orange and the positive charged adsorbent surface. This enhances adsorption efficiency even within a shorter contact time. The observation that removal is increasing at shorter contact time indicates that the adsorption process is very fast. This is because many active sites are still available which resulting rapid in mass transfer. As contact time increases, the adsorption rate slows down because most of active sites become occupied and equilibrium is approached. Therefore, further increase in time does not significantly improve removal efficiency. This interaction shows that under optimal pH conditions, equilibrium can be achieved quickly and extending the contact time beyond this point does not substantially enhance the methyl orange removal. Hence, moderate pH combined with shorter contact time is sufficient to achieve high removal efficiency.

Figure 4.15 also shows the interaction between initial methyl orange concentration (C) and contact time (D) on methyl orange removal efficiency (%). The surface plot demonstrates that the percentage removal increases when initial methyl orange concentration increases at shorter contact time. At higher initial methyl orange concentration, the driving force for mass transfer becomes stronger. This is due to the large concentration gradient between the bulk solution and the surface of MgAl-LDH. As a result, methyl orange molecules diffuse more rapidly toward the available active sites on the adsorbent surface which lead to higher removal efficiency during the initial stage of adsorption. At shorter time, many adsorption sites remain unoccupied and the adsorption occurs very rapidly. Since the adsorption process is fast at the beginning, increasing methyl orange concentration under these conditions enhances the probability of methyl orange adsorbent collisions which increasing the overall removal percentage. However, as contact time increases, the rate of adsorption slow down because most active sites become progressively occupied and the system approaches equilibrium. Once equilibrium is reached, additional time does not significantly improve removal efficiency. This interaction indicates that under sufficient initial methyl orange, high methyl orange removal can be achieved even within a shorter contact time.

4.4.5 Verification Model

To validate the adequacy and reliability of the developed RSM quadratic model, verification experiments were conducted under selected operating conditions within the experimental range. Three different sets of conditions were chosen by varying adsorbent dosage, pH solution, initial methyl orange concentration and contact time, as summarised in Table 4.7. These conditions were selected to represent different regions of the design space rather than only the optimum point to ensure the model's predictive capability.

Table 4.7
Actual and Predicted for Verification Model of Methyl Orange Removal

Run	Adsorbent Dosage (g)	pH Solution	Initial Methyl Orange Conc. (ppm)	Contact Time (min)	Methyl Orange Removal (%)		
					Predicted	Actual	RSE
1	0.30	8	20	40	90	89	1.11
2	0.25	7	40	38	89	91	2.25
3	0.30	9	20	20	86	82	4.65

The experimental methyl orange removal efficiencies obtained from the verification runs were 89%, 91% and 82%, while the corresponding values predicted by the RSM model were 90%, 89% and 86%, respectively. A close agreement between the experimental and predicted results was observed for all verification runs, with only small deviations between the two sets of values. This indicates that the developed model is capable of accurately predicting the adsorption performance of MgAl-LDH for methyl orange removal. The minor differences between experimental and predicted methyl orange removal percentages may be attributed to experimental uncertainties such as slight pH fluctuations, heterogeneity of the adsorbent particles, or mass transfer limitations during adsorption. Nevertheless, the deviations remain within an acceptable range, demonstrating that the quadratic model provides a reliable representation of the adsorption system.

The calculated RSE values for the verification experiments ranged from 1.11% to 4.65%, indicating a small deviation between the experimental and predicted methyl orange removal efficiencies. Generally, an RSE value of less than 5% is considered acceptable for validating response surface models (Khairuddin et al., 2023), confirming

good agreement between model predictions and experimental observations. Run 1 exhibited the lowest RSE (1.11%), demonstrating excellent consistency between the predicted and experimental results. Slightly higher deviations observed in Runs 2 and 3 may be attributed to experimental uncertainties such as minor variations in pH solution, adsorption equilibrium conditions, or mass transfer effects during agitation. However, these deviations remain within an acceptable range and do not significantly affect the predictive reliability of the model.

4.4.6 Optimum Condition of Methyl Orange Removal

The validation experiment was conducted to confirm the reliability of the optimum adsorption conditions predicted by the RSM. The optimised conditions consisted of an adsorbent dosage (MgAl-LDH) of 0.65 g, pH solution 7.78, initial methyl orange concentration of 40 mg/L and contact time of 20 min. All selected factor levels were within the predefined experimental ranges like in Table 4.8, ensuring that the optimisation was performed within a statistically valid and experimentally feasible region. The optimisation goal was set to maximise methyl orange removal efficiency, while maintaining all operational variables within their respective limits.

Table 4.8
Optimum Validation Condition of Methyl Orange Removal

Variables	Goal	Lower limit	Upper limit
Adsorbent Dosage	In range	0.25	0.65
pH Solution	In range	5	9
Initial Methyl Orange Concentration	In range	20	40
Contact Time	In range	20	40
Final Methyl Orange Concentration	Maximise	60.66	93.94

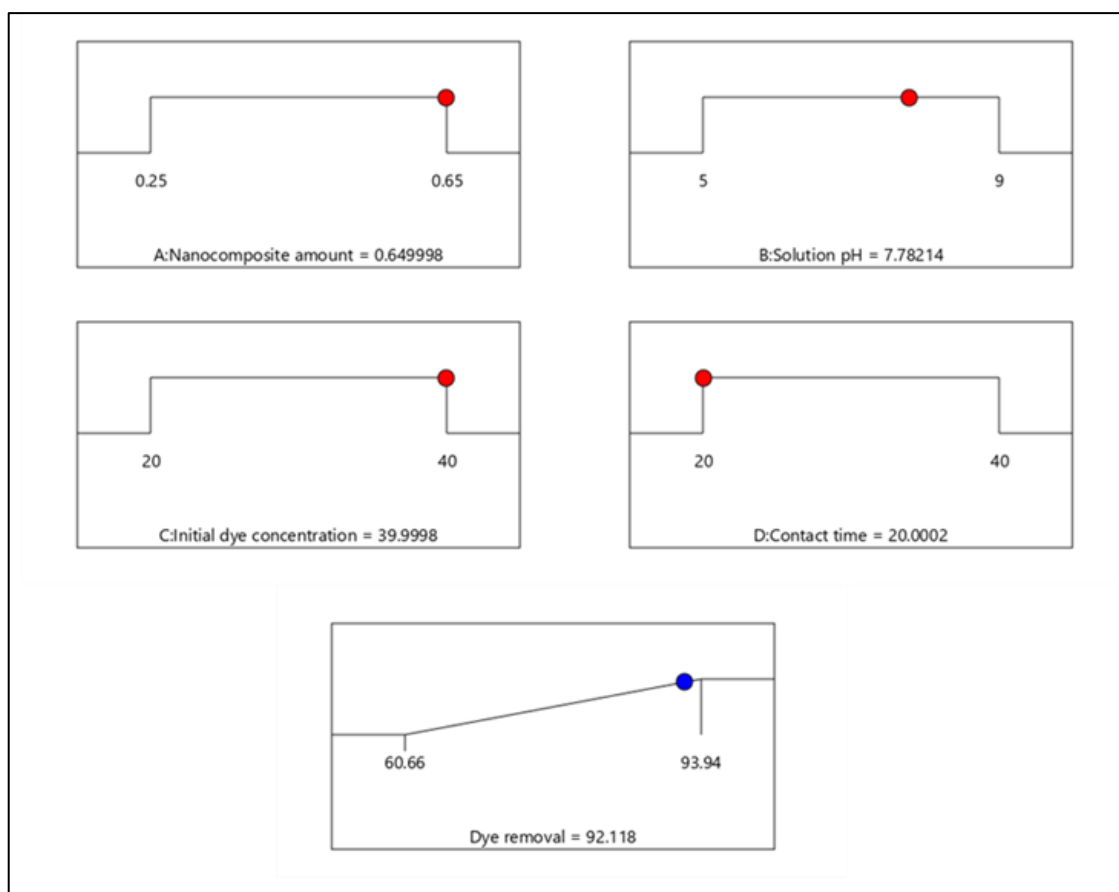


Figure 4.16 Optimum Condition Model of Methyl Orange Removal

Under these optimised conditions, the RSM model predicted a methyl orange removal efficiency of 92.19% with the desirability of 1, while the three repeated experimentally obtained value was 94.44%. The Relative Standard Error (RSE) between the predicted and actual values was calculated to be 2.48%, indicating a small deviation between the model prediction and experimental result. An RSE value below 5% is generally considered acceptable for model validation, confirming good agreement between the predicted and experimental responses (Khairuddin et al., 2023).

Table 4.9
Actual and Predicted Data for Optimum Conditions of Methyl Orange Removal

Adsorbent Dosage (g)	pH Solution	Initial Methyl Orange Conc.	Contact Time (min)	Methyl Orange Removal (%)		
				Predicted	Actual	RSE
0.65	7.78	40	20	92.19	94.44	2.48

4.5 Adsorption Study of the MgAl-LDH onto Methyl Orange

Adsorption study presents a comprehensive evaluation of the adsorption performance of MgAl-LDH for the removal of methyl orange from aqueous solution under optimised conditions. The adsorption study was carried out to elucidate the interaction mechanism between methyl orange molecules and the MgAl-LDH surface, as well as to understand the equilibrium and time-dependent behaviour of the adsorption process. To achieve this, equilibrium adsorption data were analysed using adsorption isotherm models, while adsorption rate and controlling mechanisms were investigated using kinetic models. In addition, the applicability of the optimised adsorption system to real wastewater samples was assessed to evaluate the practical feasibility of MgAl-LDH for methyl orange removal. The adsorption isotherm analysis focuses on the distribution of methyl orange between the liquid phase and the solid adsorbent at equilibrium and provides insight into the surface properties and adsorption capacity of MgAl-LDH. Meanwhile, the kinetic study examines the adsorption rate which are essential for understanding the adsorption mechanism and designing efficient treatment processes.

4.5.1 Mechanism of the adsorption

Figure 4.17 illustrates the schematic representation of the MgAl-layered double hydroxide (MgAl-LDH) structure and its role in the adsorption of methyl orange from aqueous solution. MgAl-LDH belongs to the hydrotalcite-like family of layered double hydroxide which in this study Mg^{2+} ions function as divalent cations and Al^{3+} ions as trivalent cations. The partial substitution of Mg^{2+} by Al^{3+} within the brucite-like hydroxide sheets generates an overall positive charge on the layers. The positive layer charge is electrostatically balanced by the presence of interlayer anions which is carbonate (CO_3^{2-}), together with water molecules located between the stacked hydroxide layers. The layered structure consists of octahedrally coordinated hydroxide sheets containing Mg^{2+} and Al^{3+} ions, forming positively charged layers separated by hydrated interlayer regions. The presence of interlayer water molecules enhances hydration behaviour and facilitates ions mobility within the structure (Mittal, 2021). Furthermore, the abundance of surface hydroxyl group (-OH) on the LDH layers provides active

adsorption site capable of interacting with the methyl orange molecules. This unique structural configuration, combining with positively charged layers, exchangeable interlayer anions and accessible surface functional groups which makes MgAl-LDH particularly suitable for the adsorption of anionic dyes like methyl orange.

The lower part of the figure demonstrates the adsorption process where the MgAl-LDH particles are introduced into an aqueous methyl orange solution. The adsorption of methyl orange onto MgAl-LDH occurs through several complementary mechanisms that come from the unique structural and surface properties of layered double hydroxides. The first and most dominant mechanism is electrostatic attraction. MgAl-LDH possesses positively charged hydroxide layers due to the partial substitution of divalent Mg^{2+} ions with trivalent Al^{3+} ions within the brucite-like structure (Chen et al., 2021). This substitution generates a positive charge on the layers which is balanced by interlayer anions that is carbonate. In aqueous solution, methyl orange exists predominantly in its anionic form due to the presence of negatively charged sulfonate group ($-\text{SO}_3^-$). The electrostatic interaction between the negatively charged methyl orange molecules and the positively charged LDH layers leads to strong attraction and rapid adsorption onto the external surface of the particles (Santamaría et al., 2020). This mechanism is highly influenced by pH solution as pH affects both the surface charge density of MgAl-LDH and the degree of ionisation of methyl orange. At favourable pH conditions, electrostatic forces are maximised (Mittal, 2021) thereby enhancing methyl orange removal efficiency.

Beyond the electrostatic attraction, surface adsorption or surface complexation may also contribute significantly to the adsorption process. The surface of MgAl-LDH is rich in hydroxyl groups ($-\text{OH}$) that can participate in hydrogen bonding with functional groups present in methyl orange molecules. In addition, weak coordination interactions may occur between the azo group ($-\text{N}=\text{N}-$), aromatic rings or sulfonate groups of the methyl orange and the metal centres (Mg^{2+} or Al^{3+}) located within the hydroxide layers (Daud et al., 2019). These interactions involve partial electron sharing of localised bonding which leads to stronger attachment of methyl orange molecules to the LDH surface compared to purely physical adsorption. Such interactions are consistent with the kinetic behaviour observed in this study where the pseudo-second-order model suggests that surface interactions play an important role in controlling the adsorption rate.

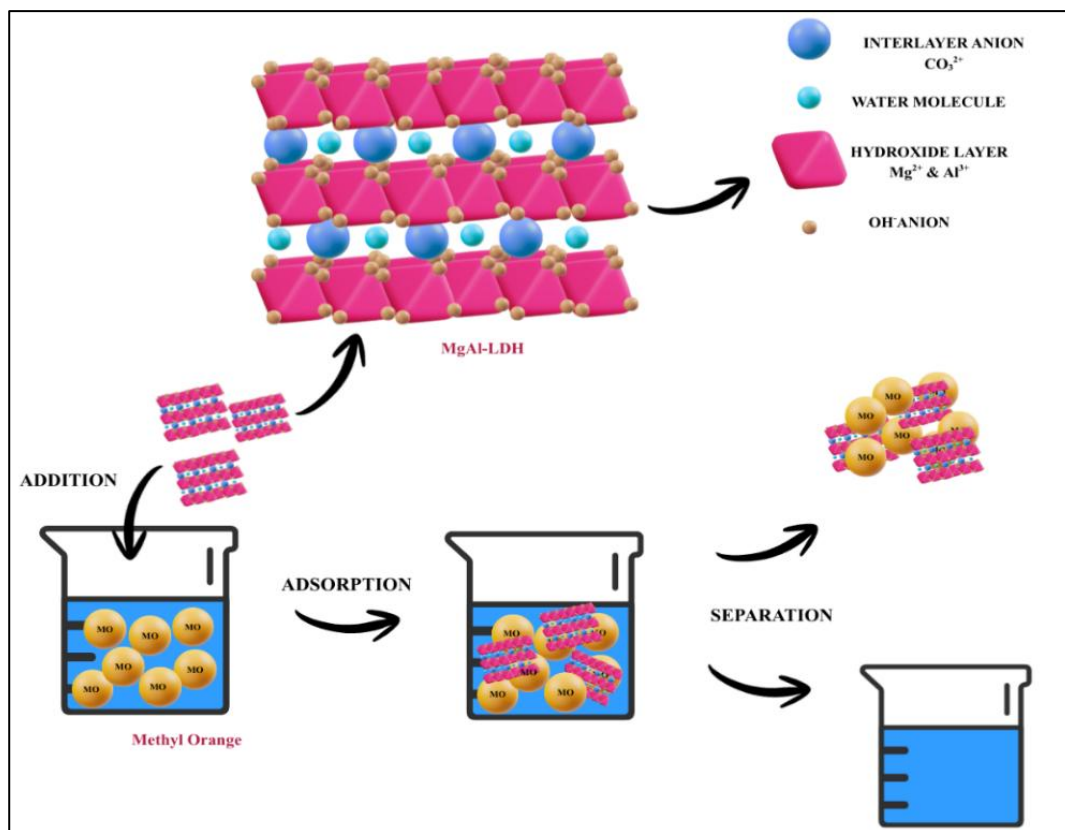


Figure 4.17 Schematic Illustration of the Adsorption of Methyl Orange Using MgAl-LDH

Another important mechanism is anion exchange. One of the defining characteristics of layered double hydroxides is their ability to exchange interlayer anions due to their positively charged layers (Hosseini and Ghasemi, 2020). In MgAl-LDH, carbonate ions are typically located within the interlayer region to maintain charge neutrality. During the adsorption process, methyl orange anions present in solution may compete with and partially replace these interlayer carbonate ions. This exchange process allows methyl orange molecules to penetrate the interlayer parts rather than remaining only on the external surface (Santamaría et al., 2020). As a result, adsorption is not limited to surface binding but may involve intercalation within the LDH structure. This mechanism enhances the overall adsorption capacity and contributes to the relatively strong retention of methyl orange molecule observed during desorption studies.

In addition to this mechanism, diffusion processes also influence the overall adsorption behaviour, initially, methyl orange molecule rapidly adsorb onto the available surface sites due to strong electrostatic forces (Mittal, 2021). As adsorption

progresses, methyl orange molecules must diffuse from the bulk solution to internal diffusion may act as secondary rate controlling steps, particularly at longer contact times. The presence of interlayer water molecules facilitates this diffusion process by providing a hydrated pathway for dye transport into internal adsorption sites.

4.5.2 Isotherm Study

The adsorption isotherm analysis was conducted to evaluate the interaction between the methyl orange molecules and the adsorbent dosage adsorbent at equilibrium using the Langmuir and Freundlich models. The linearised Langmuir and Freundlich plots obtained from the experimental data are presented in Figure 4.18 and Figure 4.19, respectively. The Langmuir isotherm model assumes of monolayer adsorption onto a homogeneous surface with a finite number of identical active sites and no interaction between adsorbed molecules (Sumardiono et al., 2025). The linear form of the Langmuir model showed a strong linear relationship with a high correlation coefficient ($R^2 = 0.9976$), indicating that the adsorption process can be partially described by monolayer coverage.

The slope and intercept of the Langmuir plot suggest that the adsorption capacity increases proportionally with equilibrium concentration, implying favourable adsorption under the studied conditions. The Freundlich isotherm model, which describes adsorption on heterogeneous surfaces and allows for multilayer adsorption, exhibited an even higher correlation coefficient ($R^2 = 0.9989$) compared to the Langmuir model. This superior fit indicates that the adsorption of methyl orange onto the adsorbent dosage surface is better represented by the Freundlich model. The Freundlich constant, $1/n$, obtained from the slope of the plot was found to be close to unity and less than 1, confirming favourable adsorption and suggesting a heterogeneous surface with varying adsorption energies (Khazaie et al., 2026). The value of n greater than 1 further implies a strong interaction between the methyl orange molecules and the adsorbent surface.

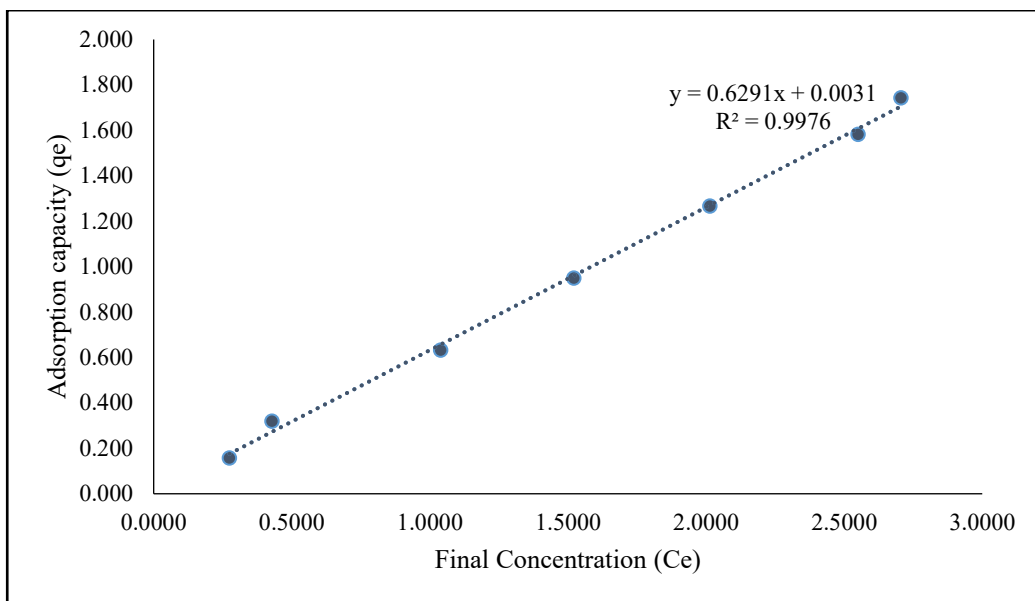


Figure 4.18 Langmuir Plot of Methyl Orange Removal

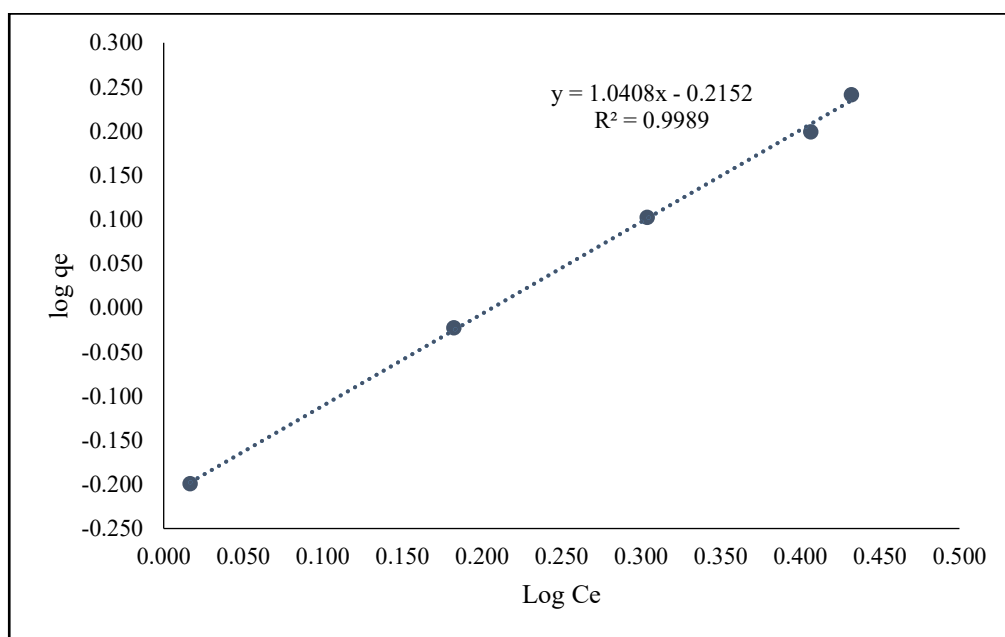


Figure 4.19 Freundlich Plot of Methyl Orange Removal

Although both isotherm models demonstrated excellent agreement with the experimental data, the Freundlich model provided a slightly better fit, indicating that the adsorption process is predominantly governed by multilayer adsorption on a heterogeneous surface rather than ideal monolayer adsorption. This behaviour is consistent with the structural characteristics of layered double hydroxide-based adsorbent dosages, which possess diverse surface functional groups, variable pore structures and non-uniform adsorption sites. Therefore, the adsorption mechanism is

likely influenced by a combination of electrostatic interactions, surface complexation and physical adsorption (Sibiya-Dlomo et al., 2025). Overall, the isotherm analysis confirms that the adsorbent dosage exhibits high adsorption efficiency toward the methyl orange with the Freundlich isotherm being the most suitable model to describe the equilibrium adsorption behaviour.

Table 4.10
Parameters of Isotherm Models for Methyl Orange Removal

Isotherm models	Parameters	Values
Langmuir	q_m (mg/g)	0.954
	K_L (L/mg)	205.7
	R_L	8.84×10^{-5}
	R^2	0.9976
Freundlich	K_F ($\text{mg}^{1-1/n} \text{L}^{1/n} \text{g}^{-1}$)	1.016
	$1/n_F$	1.0408
	n_F	0.96
	R^2	0.9989

The Langmuir and Freundlich isotherm parameters for methyl orange adsorption onto MgAl-LDH are summarised in Table 4.10. The Langmuir maximum adsorption capacity ($q_m = 0.954$ mg/g) and high Langmuir constant ($K_L = 205.7$ L/mg) indicate a strong affinity between methyl orange molecules and the adsorbent surface. The separation factor ($R_L = 8.84 \times 10^{-5}$) lies between 0 and 1, confirming that the adsorption process is highly favourable. For the Freundlich model, the Freundlich constant ($K_F = 1.016 \text{ mg}^{1-1/n} \text{L}^{1/n} \text{g}^{-1}$) reflects the adsorption capacity of the adsorbent, while the intensity factor ($n_F = 0.96$) suggests strong adsorbate with adsorbent interactions on a heterogeneous surface. Although both models exhibit high correlation coefficients, the Freundlich model shows a slightly higher R^2 value (0.9989) compared to the Langmuir model (0.9976) indicating a marginally better fit. This suggests that methyl orange adsorption onto MgAl-LDH occurs on a heterogeneous surface with strong interaction mechanisms.

4.5.3 Kinetic Study

The kinetic study in this study was conducted to evaluate the rate of methyl orange adsorption onto MgAl-LDH, determine the time required to reach equilibrium and

provide insight into the possible adsorption mechanism (Kanwal et al., 2024). Kinetic modelling helps to assess whether the adsorption process is primarily governed by physical diffusion, surface interaction or a combination of both mechanisms.

In this study, the pseudo-first-order kinetic model could not adequately describe the adsorption behaviour. The experimental data did not produce a consistent linear relationship between $\ln (q_e - q_t)$ and contact time. Moreover, the calculated equilibrium adsorption capacity (q_e) obtained from the pseudo-first-order model deviated significantly from the experimentally determined q_e value. At longer contact time, the term $(q_e - q_t)$ approaches zero, resulting in instability and large errors in the logarithmic calculation and further deviation from linearity. These findings indicate that the pseudo-first order is not suitable for describing the adsorption kinetics of methyl orange onto MgAl-LDH under the studied conditions.

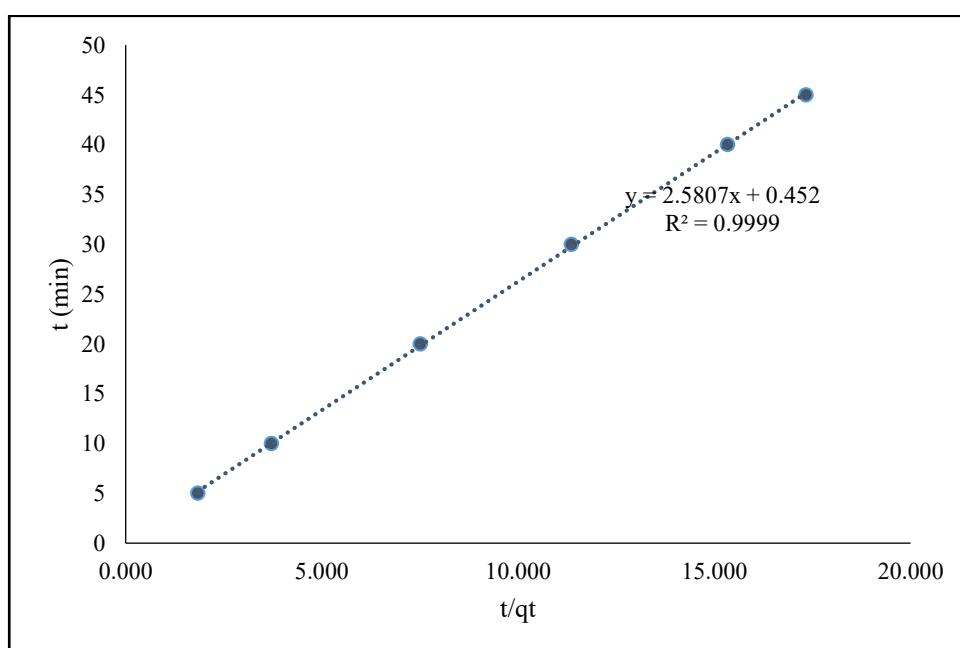


Figure 4.20 Pseudo-Second Order Plot of Methyl Orange Removal

Table 4.11

Parameter of Kinetics Model for Methyl Orange Removal

Kinetic models	Parameters	Values
Pseudo-second order	q_e (mg/g)	0.387
	k_2 (g mg ⁻¹ min ⁻¹)	14.8
	R^2	0.9999

In contrast, the pseudo-second-order kinetic plot for methyl orange adsorption onto MgAl-LDH shows an excellent linear relationship, with a very high coefficient of determination ($R^2 = 0.9999$). The close agreement between the calculated equilibrium adsorption capacity ($q_e = 0.387 \text{ mg/g}$) and the experimentally observed value further confirms the reliability of this model. The high pseudo-second-order rate constant ($k_2 = 14.8 \text{ g mg}^{-1} \text{ min}^{-1}$) indicates that the adsorption process occurs rapidly and is strongly dependent on the availability of active adsorption sites on the MgAl-LDH surface (Kim Ngan Tran et al., 2023). The excellent fitting of the pseudo-second-order model suggests that the adsorption rate is controlled predominantly by surface interactions rather than external mass transfer resistance. This implies that the rate limiting step is associated with interactions between methyl orange molecules and functional groups present on the MgAl-LDH surface.

Although kinetic modelling alone cannot conclusively prove the nature of adsorption, several findings from this study collectively suggest that chemisorption plays a dominant role in the adsorption mechanism. Firstly, the pseudo-second-order kinetic model provided an excellent fit ($R^2 = 0.9999$) and showed strong agreement between calculated and experimental q_e values. This model assumes that the adsorption rate is proportional to the square of the number of unoccupied sites, which is commonly associated with surface interactions involving valence forces. Secondly, the adsorption process was strongly influenced by pH, indicating that electrostatic interactions and surface charge effects significantly contribute to dye uptake. MgAl-LDH possesses positively charged hydroxide layers that facilitate interaction with negatively charged sulfonate groups ($-\text{SO}_3^-$) of methyl orange. Such interactions are consistent with electrostatic attraction and possible anion exchange mechanisms. Thirdly, the relatively low desorption efficiency observed during regeneration studies suggests that the methyl orange molecule is strongly bound to the LDH surface. If the adsorption were purely physical in nature, desorption would be more easily achieved. The partial irreversibility of adsorption indicates stronger interactions beyond weak van der Waals forces. Based on these combined observations, the adsorption mechanism of methyl orange onto MgAl-LDH is likely dominated by chemisorption involving electrostatic interaction, surface complexation and anion exchange.

4.6 Desorption and Reusability Performance

The reusability of MgAl-LDH was evaluated through successive adsorption–desorption cycles using 0.1 M NaOH as the desorbing agent under optimised adsorption conditions (0.65 g MgAl-LDH, pH 7.78, 40 mg/L initial concentration and 20 min contact time). The adsorption performance over four consecutive cycles is summarised in illustrated in Table 4.12 and Figure 4.21.

Table 4.12
Adsorption-Desorption Result for Methyl Orange Removal

	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5
Percentage Removal (mean $\pm$ SD, n=3)	93.94 $\pm$ 0.02	75.54 $\pm$ 0.03	56.41 $\pm$ 0.03	16.02 $\pm$ 0.03	-4.42 $\pm$ 0.03
Percentage Desorption (mean $\pm$ SD, n=3)	3.81 $\pm$ 0.03	3.57 $\pm$ 0.01	3.63 $\pm$ 0.02	10.68 $\pm$ 0.01	-34.98 $\pm$ 0.01

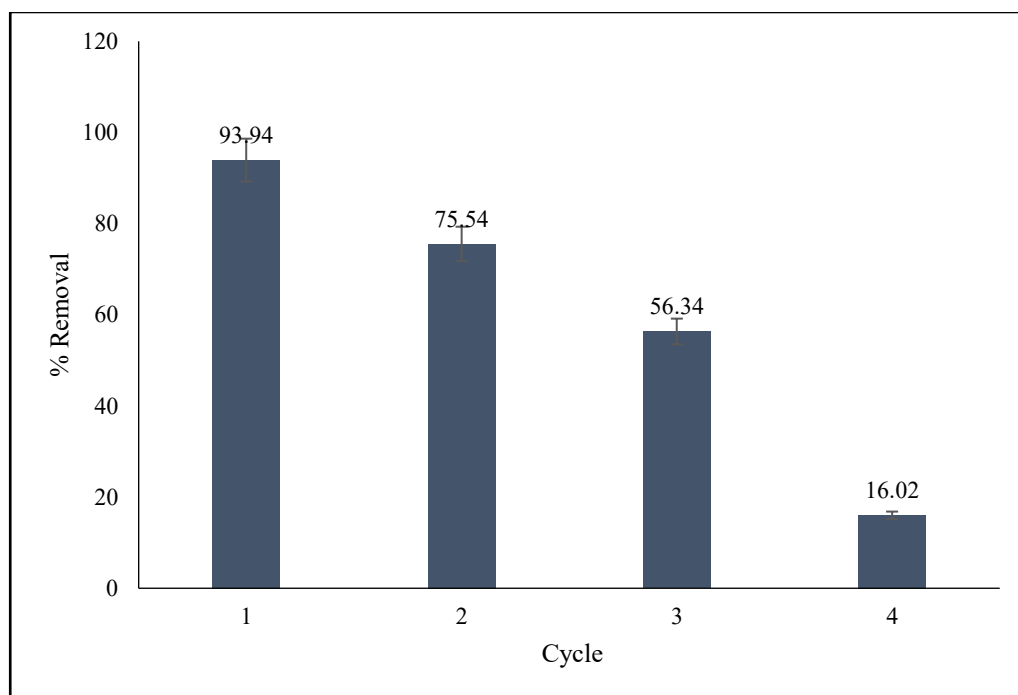


Figure 4.21 Adsorption-Desorption Cycle Performance for Methyl Orange Removal

In the first adsorption cycle, MgAl-LDH exhibited excellent removal efficiency of 93.94%, confirming its strong affinity toward methyl orange. This high initial performance can be attributed to the positively charged brucite-like layers of MgAl-LDH, which favour electrostatic attraction with the anionic sulfonate groups of methyl

orange, as well as possible interlayer anion exchange and hydrogen bonding interactions. Upon regeneration with NaOH, a gradual decline in adsorption efficiency was observed in subsequent cycles. The removal efficiency decreased to 75.54%, 56.34% and 16.02% in cycles 2, 3 and 4, respectively. This progressive reduction indicates partial loss of active adsorption sites and incomplete desorption of methyl orange molecules from the LDH surface. Similar trends have been widely reported for LDH-based adsorbents subjected to repeated alkaline regeneration, where strong dye surface interactions limit full reversibility (Zhang et al., 2023; Zubair et al., 2021).

The relatively low desorption efficiencies (3.57–3.80%) in the early cycles suggest that methyl orange was strongly bound to the LDH surface. Although NaOH increases solution alkalinity and weakens electrostatic interactions, complete desorption was not achieved. Repeated exposure to alkaline conditions may also induce partial dissolution of Mg^{2+} and Al^{3+} ions or collapse of the layered structure, thereby reducing adsorption capacity in later cycles. An abnormal behaviour was observed in cycle 5, where the equilibrium methyl orange concentration exceeded the initial concentration, resulting in negative removal and adsorption values. This behaviour indicates structural degradation of the MgAl-LDH (Ahmad et al., 2025) and possible release of previously adsorbed methyl orange into the solution. Therefore, cycle 5 was excluded from performance evaluation, as negative adsorption is not physically meaningful and has been similarly excluded in previous adsorption studies involving LDH regeneration. The very small standard deviation values across all cycles indicate good experimental reproducibility and consistency among replicates (Isidoro Ribeiro et al., 2023).

4.7 Real Samples Application

For textile wastewater experiment, the optimised adsorption parameter obtained from RSM which adsorbent dosage of 0.65g, solution pH of 7 and contact time of 20 min. However, the optimised initial methyl orange concentration which is 40 ppm was not adjusted for real sample. Instead of spiking or modifying the methyl orange concentration, the wastewater was used as collected to evaluate the practical applicability under realistic environmental conditions.

Table 4.13
Percentage of Methyl Orange Removal on Real Sample Application

Real Sample	Concentration (ppm)		% Removal (mean $\pm$ SD, n = 3)	% RSD
	Before	After		
1	33.93	7.44	78.06 $\pm$ 0.05	0.06
2	37.66	7.98	78.80 $\pm$ 0.02	0.03
3	29.29	7.75	73.53 $\pm$ 0.01	0.01

As shown in Table 4.13, the initial methyl orange concentrations in the wastewater ranged from 29.29 to 37.66 ppm. After treatment, the residual methyl orange concentrations were reduced to approximately 7-8 ppm corresponding to removal efficiencies between 73.53% and 78.80%. Although the methyl orange removal efficiency in real wastewater was slightly lower than that obtained using synthetic methyl orange solutions, the results demonstrate that MgAl-LDH remains effective in complex wastewater matrices.

The lower removal efficiency observed in textile wastewater compared to the synthetic methyl orange solution can be attributed to the contaminant present, allowing direct interaction between the methyl orange molecules and the active adsorption sites on the MgAl-LDH surface. Under optimum conditions, the removal efficiency was achieved at 94.44% due to the absence of competing species. In contrast, textile wastewater contains a mixture of dissolving salts such as Na^+ , Ca^{2+} , Mg^{2+} , competing anions such as SO_4^{2-} , NO_3^- , Cl^- , organic matter and other dye residues. Since MgAl-LDH primarily removes anionic species through electrostatic attraction, anion exchange and surface complexation mechanisms, it is not entirely selective towards methyl orange. Other negatively charged species present in the wastewater can compete with methyl orange molecules for the same active adsorption sites (Sarkar and Upadhyay, 2025). This competitive adsorption reduces the availability of active sites for the target dye which lead to lower overall removal efficiency. Additionally, the higher ionic strength of real wastewater may weaken electrostatic interactions between the negatively charged methyl orange molecules and the positively charged LDH surface. Increased ionic strength compresses the electrical double layer, thereby reducing the effective attractive forces between the adsorbent and adsorbate. Matrix effects may also influence the analytical measurement and adsorption behaviour. Natural organic matter and suspended solids in wastewater can partially block the pores of MgAl-LDH which limiting access to internal adsorption sites. Furthermore, organic constituents may form

complexes with methyl orange, altering its effective charge, solubility or availability for adsorption (Hussain et al., 2024).

Nevertheless, the consistently high removal efficiencies above 70% with low %RSD values (<1%) which fall well within the generally accepted performance range for adsorption-based treatment systems which is 65%-105% (Navalgund et al., 2026) which demonstrate that MgAl-LDH retains good adsorption capability even in complex wastewater systems. This indicates that although the material is not highly selective, it remains sufficiently robust for practical applications under optimised conditions.

CHAPTER 5

CONCLUSION

5.1 Conclusion

This study investigated the feasibility and effectiveness of magnesium–aluminium layered double hydroxide (MgAl-LDH) as an adsorbent for the removal of methyl orange from aqueous solutions. The research covered the synthesis and characterisation of MgAl-LDH, optimisation of adsorption conditions using response surface methodology (RSM), evaluation of adsorption behaviour through isotherm and kinetic studies, model validation, application to textile wastewater and assessment of desorption and reusability performance.

MgAl-LDH was successfully synthesised using the co-precipitation method. The structural and chemical characteristics of the synthesised material were confirmed through X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy coupled with energy-dispersive X-ray (SEM-EDX) analysis. The XRD patterns verified the formation of a well defined hydrotalcite-like layered structure, while FTIR spectra confirmed the presence of hydroxyl group, interlayer carbonate anions and metal-oxygen bonds. SEM image revealed the typical plate-like morphology of LDH and EDX analysis confirmed the presence of Mg and Al elements. These results validated the successful preparation of MgAl-LDH.

The adsorption conditions were successfully optimised using Response Surface Methodology (RSM). The developed quadratic model was statistically significant, with a high coefficient of determination and insignificant lack-of-fit, demonstrating good agreement between predicted and experimental data. The optimum adsorption conditions were determined to be approximately 0.65 g of MgAl-LDH, pH solution of 7.78, initial methyl orange concentration of 40 mg/L and a contact time of 20 minutes. Under these conditions, a maximum predicted methyl orange removal of 92.19% was obtained, while experimental validation achieved 94.4% removal with relative standard error (RSE) values below 5%, confirming the reliability and accuracy of the RSM model.

Adsorption isotherm studies were carried out to understand the interaction between methyl orange molecules and the MgAl-LDH surface at equilibrium. Adsorption behaviour was evaluated using isotherm and kinetic models. The equilibrium data were better described by the Freundlich isotherm model which indicate heterogeneous surface adsorption with possible multilayer formation. Kinetic analysis showed that the adsorption process followed the Pseudo-second-order model, suggesting that chemisorption is the dominant rate controlling mechanism.

The practical applicability of MgAl-LDH was further evaluated using textile wastewater samples containing methyl orange. Under optimised conditions, the removal efficiency obtained for real sample were 78.06%, 78.80% and 73.53% respectively. Although these values were slightly lower than the removal efficiency achieved in synthetic solutions, the results demonstrated that MgAl-LDH remain effective in complex wastewater matrices. The slight reduction in efficiency can be attributed to the presence of competing ions and other organic or inorganic constituents in the real wastewater which may interfere with the adsorption process.

Reusability studies were conducted using adsorption-desorption using 0.1M NaOH as the desorbing agent. The removal efficiency decreased progressively over five cycles. This decline indicates partial loss of active sites, possible structural changes and incomplete desorption of methyl orange after repeated use. Despite the reduction in performance, MgAl-LDH demonstrated a good adsorption efficiency during the cycles which suggesting that the material can be reused to a certain extent. Overall, this study contributes valuable insight into the application of layered double hydroxide for methyl orange contaminated wastewater treatment and supports their potential use in environment remediation.

5.2 Recommendation

Based on the findings obtained in this study, several recommendations are proposed to further enhance the performance of layered double hydroxides (LDHs) and to expand their application in wastewater treatment. At the same time, certain limitations of the present study are acknowledged, which provide direction for future research.

Firstly, future studies are recommended to focus on the surface modification or functionalisation of MgAl-LDH to improve its adsorption capacity, selectivity and

reusability. In the present work, unmodified MgAl-LDH demonstrated good removal efficiency for methyl orange however, the desorption and reusability results indicated a gradual decrease in performance over repeated cycles. This suggests that the structural stability and regeneration efficiency of unmodified LDHs may be limited under repeated adsorption-desorption conditions. Surface modification using organic surfactants, polymers, bio-based materials, or metal oxides could enhance surface area, increase active adsorption sites and improve structural stability during regeneration. Modified LDHs may also strengthen electrostatic interactions, hydrogen bonding, or π - π interactions with methyl orange molecules, leading to higher adsorption efficiency and better regeneration performance.

Secondly, the intercalation of different anions into the LDHs interlayer is recommended. In this study, carbonate was the dominant interlayer anion which may limit the accessibility of large dye molecule into interlayer region. Since LDHs possess a tuneable interlayer structure, replacing carbonate with nitrate, sulphate or organic functional anions may improve interlayer accessibility and adsorption capacity. This would also provide deeper insight into the role of the interlayer chemistry in adsorption mechanism.

Thirdly, this study focused only on methyl orange as model anionic dye under batch experimental conditions. Although methyl orange is commonly used to represent textile dyes, real industrial wastewater typically contains multiple contaminants including mixed dyes, heavy metals, pharmaceuticals and emerging pollutants. Therefore, future research should be extending the application of LDHs to multiple pollutants systems to better simulate real wastewater conditions. Future work should also include point of zero charge, pH_{pzc} analysis to further clarify the relationship between solution pH, MgAl-LDH surface charge and methyl orange adsorption. This analysis would determine whether the adsorbent surface is positively or negatively charge under different pH conditions and provide stronger support for their proposed electrostatic attraction mechanism. Additionally, the present study was conducted at a single temperature and thermodynamic parameter were not evaluated. A detailed thermodynamic study involving Gibbs free energy, enthalpy and analysis is recommended to determine the spontaneity and energetic nature of the adsorption process. In summary, although MgAl-LDH exhibited strong adsorption performance

and good optimisation results, improvement in many way are required to fully establish its industrial applicability.

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APPENDICES

APPENDIX A

Experimental run, factors and response for the Removal Methyl Orange by LDHs

Run	pH	LDH Amount	Initial Conc. Dye (Ppm)	Contact Time (Min)	Absorption	Final Conc. Dye (Ppm)	% Removal
1	9	0.25	5	20	0.076	1.099	78.011
2	9	0.65	5	40	0.062	0.771	84.586
3	5	0.25	9	20	0.08	1.268	85.912
4	9	0.65	9	40	0.095	1.682	81.308
5	5	0.25	5	20	0.085	1.406	71.878
6	3	0.65	5	30	0.05	0.439	91.215
7	9	0.25	9	20	0.076	1.157	87.139
8	7	0.65	9	30	0.064	0.826	90.823
9	5	0.25	5	40	0.07	0.992	80.166
10	11	0.65	5	30	0.135	2.787	44.254
11	5	0.25	9	40	0.089	1.517	83.149
12	9	0.65	9	20	0.07	0.992	88.981
13	7	0.25	5	30	0.062	0.771	84.586
14	7	0.65	5	10	0.066	0.881	82.376
15	7	0.25	9	50	0.073	1.075	88.060
16	7	0.65	9	30	0.12	2.373	73.634
17	9	0.05	7	40	0.067	0.909	87.017
18	7	0.85	7	30	0.088	1.489	78.729
19	9	0.45	3	40	0.079	1.240	58.656
20	7	0.45	11	30	0.088	1.489	86.464
21	5	0.45	7	40	0.062	0.771	88.990
22	5	0.45	7	20	0.052	0.494	92.936
23	7	0.45	7	30	0.076	1.157	83.465
24	7	0.45	7	30	0.054	0.550	92.147
25	9	0.45	7	20	0.084	1.378	80.308
26	5	0.45	7	40	0.051	0.467	93.331
27	7	0.45	7	30	0.08	1.268	81.886
28	5	0.45	7	20	0.066	0.881	87.411
29	7	0.45	7	30	0.079	1.240	82.281
30	7	0.45	7	30	0.065	0.854	87.806

APPENDIX B

Table data of Isotherm and Kinetic Study

ISOTHERM

Run	1	2	3	4	5	6	7
Mass (G)	0.45	0.45	0.45	0.45	0.45	0.45	0.45
pH	7	7	7	7	7	7	7
Time	30	30	30	30	30	30	30
Initial Conc.	5	10	20	30	40	50	55
Abs 1	0.039	0.043	0.061	0.084	0.103	0.121	0.128
Abs 2	0.038	0.045	0.069	0.082	0.101	0.121	0.127
Abs 3	0.038	0.044	0.069	0.086	0.102	0.123	0.127
Average	0.038	0.044	0.066	0.084	0.102	0.122	0.127
Final Conc. (Ce)	0.2741	0.4290	1.0392	1.5219	2.0137	2.5510	2.7058
Adsorption Capacity (Qe)	0.158	0.319	0.632	0.949	1.266	1.582	1.743
Log Ce	-0.562	-0.368	0.017	0.182	0.304	0.407	0.432
Log Qe	-0.803	-0.496	-0.199	-0.023	0.103	0.199	0.241
Ke	0.575	0.744	0.608	0.624	0.629	0.620	0.644

KINETIC

Run	1	2	3	4	5	6
Mass (G)	0.45	0.45	0.45	0.45	0.45	0.45
pH	7	7	7	7	7	7
Time	5	10	20	30	40	55
Initial Conc.	50	50	50	50	50	50
Abs 1	0.063	0.081	0.103	0.118	0.14	0.148
Abs 2	0.064	0.081	0.103	0.118	0.139	0.148
Abs 3	0.064	0.081	0.101	0.118	0.141	0.148
Average	0.064	0.081	0.102	0.118	0.140	0.148
Final Conc. (Ce)	0.9663	1.4399	2.0228	2.4508	3.0519	3.2705
Adsorption Capacity (Qe)	2.724	2.698	2.665	2.642	2.608	2.596
qt	2.596	2.596	2.596	2.596	2.596	2.596
t/qt	7.244	4.861	3.461	2.856	2.294	2.140
ln (qe-qt)	-2.055	-2.285	-2.668	-3.087	-4.404	-9.387

APPENDIX C

Table data of Desorption

Optimum condition	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5
Amount of LDH	0.45	0.5	0.5	0.5	0.5
pH	7	7	7	7	7
Initial Conc (Co)	50	50	50	50	50
Contact Time	30	30	30	30	30
Final conc (Ce)	3.03	12.23	21.83	41.99	52.21
Adsorp vol (Va)	0.025	0.025	0.025	0.025	0.025
Desorp vol (Vd)	0.025	0.025	0.025	0.025	0.025
Desorp (uvvis)	0.037	0.021	0.009	0.003	0
Desorp conc (Cd)	1.78	1.35	1.02	0.86	0.77
Amounts adsorb	1.174	0.944	0.704	0.200	-0.055
Amounts desorb	0.045	0.034	0.025	0.021	0.019
% Removal	93.94	75.54	56.34	16.02	-4.42
% Desorption	3.80	3.57	3.62	10.68	-34.99
	0.037985	0.035663	0.036178	0.106765	-0.34988

AUTHOR'S PROFILE



Nurul Aisyah obtained her Bachelor of Science (Hons.) in Chemistry with Management in 2024 from Univesiti Teknologi Mara, Kuala Pilah. She is currently completing her MSc in Chemistry, where her research focuses on the application of magnesium-aluminium layered double hydroxide (MgAl-LDH) as an adsorbent for the removal of methyl orange dye from aqueous solutions. Her work emphasises the optimisation of adsorption parameter using response surface methodology (RSM), evaluation of adsorption behaviour through isotherm and kinetic models and assessment of adsorbent performance using real wastewater samples. Her research interests include adsorption-based wastewater treatment.

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Nurul Aisyah Mat Zamzuri, N. A., Zainal Abidin, N. A., Abu Kassim, N. S., Ibrahim, W. N. W., Dzakaria, N., Sheikh Mohd Ghazali, S. A. I., & Pungot, N. H. (2026). Optimization of activated carbon as adsorbent using response surface methodology and its application for cadmium removal from wastewater. *Pure and Applied Chemistry*.

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1. IUPAC 51st World Polymer Congress (MACRO 2026). Tittle: Optimisation Of

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