

Research Article

Polyoxometalate Intercalated M^{2+}/Al ($M^{2+}=Ni, Mg$) Layered Double Hydroxide for Degradation of Methylene Blue

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Received: 28th March 2023; Revised: 14th May 2023; Accepted: 15th May 2023

Available online: 18th May 2023; Published regularly: July 2023



Abstract

The synthesis and characterization of M^{2+}/Al ($M^{2+}=Ni, Mg$) layered double hydroxide (LDH) and intercalated polyoxometalate is presented. We have reported the growth of polyoxometalate on Ni/Mg layered double hydroxide for degradation methylene blue (MB). By considering variables such as pH of dye solution, dye concentration, and time as degradation variables, the efficiency of organic dye degradation and degradation parameters of M^{2+}/Al ($M^{2+}=Ni, Mg$) LDH and both composite LDH-polyoxometalate has been identified. X-Ray Diffraction (XRD), Fourier Transform Infra Red (FTIR), Scanning Electron Microscope (SEM), and Ultra Violet Diffuse Reflectance Spectroscopy (UV-DRS) spectroscopy confirmed the layered double hydroxide structure. XRD and FTIR analysis confirmed the single-phase of the as-made and polyoxometalate intercalated LDH. SEM images show the formation of aggregates of small various sizes. The material's photodegradation was assessed through methylene blue (MB) degradation process. The result showed that NiAl-Si has a good degradation capacity for MB as compared to NiAl-Pw, MgAl-Si, and MgAl-PW. The result shows that LDH composite presents stability and has good photocatalytic activities toward the reduction of methylene blue. The FTIR measurement confirming the LDH composite structure reveals the materials used in the fifth regeneration. The activity of MB photodegradation pristine were NiAl (45%), MgAl (43%), NiAl-Pw (78%), NiAl-Si (85%), MgAl-Pw (58%), and MgAl-Si (75%), respectively. The LDH-polyoxometalate composite material's capacity to successfully photodegrade, as measured by the percentage of degradation, revealed an increase in photodegradation catalysis and the ability of the LDH to regenerate.

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Keywords: LDH; LDH-polyoxometalate; photocatalyst; methylene blue

How to Cite: Y. Hanifah, R. Mohadi, M. Mardiyanto, N. Ahmad, S. Suheryanto, A. Lesbani (2023). Polyoxometalate Intercalated M^{2+}/Al ($M^{2+}=Ni, Mg$) Layered Double Hydroxide for Degradation of Methylene Blue. *Bulletin of Chemical Reaction Engineering & Catalysis*, 18 (2), 210-221 (doi: 10.9767/bcrec.17789)

Permalink/DOI: <https://doi.org/10.9767/bcrec.17789>

1. Introduction

According to its chemical makeup, the methylene blue dye (3,7-bis(dimethylamino)-phenothiazine-5-iumchloride;MB) is an azo dye,

and depending on how it is used, it is either a cationic dye or a basic dye [1]. Figure 1 shows the structure of methylene blue. Water containing organic pollutants and an organic dye used in industrial applications are the principal contaminants, specifically in aquaculture due to its uses as an antibacterial agent. It is also used as

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a dyeing agent for wool, paper, leather, etc. However, it also has a wide range of negative side effects, particularly for mammals, including mutagenesis, carcinogenicity, and deformity [2].

Dye and organic molecules cause severe consequences for the ecosystem. The two main classes of the organic components are autochromes and chromophores [3]. Organic dyes are frequently found as harmful pollutants in leather, paper. Biowaste and industrial activated carbon products, plastics, textiles, food, and the cosmetics industry [4]. The dye used in the textile industry alters the lakes, oceans, and rivers to the point where they resemble watercolors, causing biological and chemical changes that could harm the fish. Significant health issues for humans result from harming living things and taking lives. To remove the organic pigment from water, conventional techniques such filtration, coagulation, biological processes, and precipitation have been used. One of the drawbacks of the adsorption, coagulation, and precipitation method is that the dye waste still has a fixed structure in the process so its toxicity is still high. Recently, treatment was used to degrade dyes from wastewater via photocatalysis [5]. Photocatalysis is a green treatment technology, due of its low cost, good performance, and great efficiency, one of the most promising technologies, according to experts [6,7]. The photodegradation process is still regarded as being most beneficial to remove dyes from water are thus easily and effectively eliminated [8], porous carbon, and other materials have all been used with the photocatalyst to remove colors from aqueous solutions [9,10]. According to Yang *et al.* [11], implementing ZnAl-LDH successfully degrades methylene blue. Tabatabaeian *et al.* [12] confirmed that BNC-Cl₂ modified with NiAl-LDH successfully removed methylene blue dye reached to 84%. Guo *et al.* [13] successfully degraded methylene blue with TiO₂.

Layered double hydroxide (LDH), which has brucite-like layers with positively charged multimetal cations and interlayer compensating anions, has been employed for the removal of

pollutants. LDH has the general formula $M_{1-x}^{2+}M_x^{3+}(\text{OH})_2]^{x+}(\text{A}^n)_{x/n} \cdot m\text{H}_2\text{O}$ where M^{2+} and M^{3+} are trivalent metals (Al, Fe, Cr) and divalent metal cations (Ca, Mg, Co, Fe, Cu, Ni, Mn). A is the anion that is between the layers of valence n (Cl^- , NO_3^- , ClO_4^- , CO_3^{2-} , SO_4^{2-} and so on), and the value of x is equal to the molar ratio of $M^{3+}/(M^{2+} + M^{3+})$, where the value is between 0.20 and 0.33 [14]. Recent LDH research has focused on energy storage, heavy metal ions, adsorbents, and other techniques to remove pollutants from wastewater (both organic and inorganic) [15,16]. Due to its shape, a single LDH is not thought to be an effective catalyst [17]. The structural properties of LDHs allow for the significant improvement of catalytic performance by using a variety of control synthesis methods, such as intercalated and immobilized, to modify the active centers and morphology (such as flaws, crystal faces, and electrical states) [18–20]. However, in its utilization, the hydroxy double layered is still out used effectively as a catalyst because it has a small interlayer spacing and low density in the activity side of its surface area, so it needs to be modified so that the surface area increases and the activity increase [21].

As a result, the use of LDH as a photocatalyst to decompose organic contaminants, according to a recent study has been established with a variety of components. LDH composite has greater performance than LDH pristine, according to Yuliasari *et al.* [22] which claimed that metal oxide affected LDH (ZnAl and MgAl) for decomposing MG [23]. Bi₂O₃ successfully degraded methyl orange [24]. Bi₂O₃ synthesis with H₂C₂O₄ resulted higher on degrading methyl orange compared with Bi₂O₃ synthesis using Na₂CO₃ [25]. NiCo-LDH composited with transition metal phosphide (Ni₂P) was effectively produced and applied to degraded tetracycline to increase photodegradation process [20]. According to Guo *et al.* [13], they are composite synthesized, exhibit more severe degradation, and exhibit composite CoAl-primary CeO₂'s photocatalytic activity. We successfully synthesized [26], respectively. The cation-anion exchange capacity, high adjustable band gap, and adsorption capacity of LDH materials enable them to create spatially separated redox reaction sites. Ni and Mg LDH are capable on photocatalysts on degradation dye. NiAl-LDH [27] and MgAl-LDH [28] modified with polyoxometalate successfully degraded malachite green. La-doped NiFe-layered double hydroxides containing polyoxometalate interlayer were synthesized and applied as catalyst for the removal of peroxymonosulfate [29]. Fur-

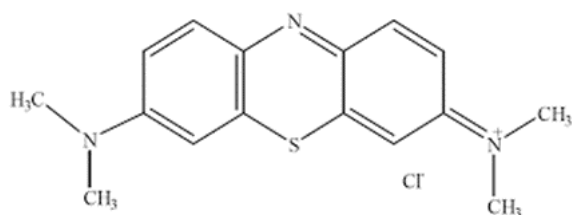


Figure 1. The structures of the methylene blue (MB).

thermore, LDHs feature a wide range of morphological benefits and adaptable compositions, including an interlayered arrangement of metal ions, varied chemical compositions, and the capacity to interchange anions on layers [30] as well as LDH materials, which have a highly adjustable adsorption capacity, band gap, and the ability to create spatially isolated redox reaction sites [31].

In the present study, Mg and Ni LDH that was mainly composited of two different polyoxometalates (POM) type Keggin $K_3[\alpha\text{-PW}_{12}\text{O}_{40}]$ and $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$ were created using the co-precipitation method, and the effects of pH, contact duration, catalyst loading, and material regeneration on their photocatalytic performances were examined. Because POM anions have a large negative charge and can improve the performance of cationic dye, which can perform photodegradation methylene blue (MB) dye, we choose POM as intercalate to composite with Mg and Ni LDH. Both materials modified were un yet used for the photodegradation of methylene blue. The substance is used as a photocatalyst to decompose MB dye. FTIR, XRD, SEM, and UV-DRS were all utilized to describe the produced material. Also, this study used the application as a photocatalyst to carry out the process of photodegradation, particularly pH affects, contact duration time, catalyst loading, and reusable of material.

2. Materials and Methods

2.1 Chemicals

Aluminum nitrate ($\text{Al}(\text{NO}_3)_3$), magnesium nitrate ($\text{Mg}(\text{NO}_3)_2$), sodium phosphate (Na_3PO_4), nickel nitrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), sodium carbonate (Na_2CO_3), sodium tungstate (Na_2WO_4), sodium hydroxide (NaOH), and concentrated hydrogen chloride (HCl) were used without any furtherer purification and were purchased from Sigma Aldrich and Merck. One of the synthetic dyes with the formula methylene blue (MB) is $\text{C}_{16}\text{H}_{18}\text{ClN}_3\text{S}$ and maximum absorbency at λ_{max} 665 nm.

2.2 Characterization

The materials LDH pristine and LDH composite catalyst were characterized with Shimadzu FTIR ALPHA Bruker (Platinum-ATR) is used. By using Rigaku XRD Miniflex-6000 diffractometer, $\text{Cu-K}\alpha$ as the radiation source, 30 kV voltage, 10 mA electricity, and 2θ ranging from 10° to 90° , XRD was analyzed. The UV-Vis Biobase BK-UV 1800 PC spectrophotometer was used to measure the degradation

of MB dye between 660-668 nm. SEM FEI Quanta 650 was used to carry out SEM analysis. The UV-Vis JASCO V-760 was used to do a band gap analysis utilizing a UV-Vis diffuse reflectance spectrometer.

2.3 Synthesis of M^{2+}/Al ($\text{M}^{2+}=\text{Ni}, \text{Mg}$)

Aluminum nitrate 0.25 M and magnesium nitrate 0.75 M were dissolved in water (100 mL) and stirred for two hours. Through the use of a modified co-precipitation process, the catalyst was created. Sodium hydroxide was added to the mixture to bring the pH below to 10. To create MgAl-LDH, this combination was agitated for six hours at 85°C . NiAl-LDH was created using the same method as MgAl-LDH; mixing nickel nitrate 0.75 M and aluminum nitrate 0.25 M dissolved in water 200 mL and stirring for two hours. To achieve a pH of 8, 2 M sodium hydroxide was then agitated for 4 hours at 343 K to produce NiAl-LDH.

2.4 Preparation of Composite

Solution A is 2 g of polyoxometalate compound and 50 mL of distilled water were used to create the composite. Solution B is 2 g of material catalyst (pristine and composite LDH) in 25 mL of sodium hydroxide 1 M. In order to create the composite LDH, the suspension was mixed right away with N_2 gas for 24 hours. The product of composite LDH will be characterized using XRD analysis, FTIR spectrophotometer, SEM analysis, and UV-DRS to analyze the composite LDH product. $K_3[\alpha\text{-PW}_{12}\text{O}_{40}] \cdot n\text{H}_2\text{O}$ and $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}] \cdot n\text{H}_2\text{O}$ polyoxometalate, according to previously publish literature, was created [32].

2.5 Photocatalytic Study

The composite was used dissolve solution of the methylene blue. To achieve a desorption equilibrium, methylene blue was first immersed in the darkened and magnetically agitated for 30 min with an ideal catalyst weight of 20 mg/L. Then, 20 mg/L of methylene blue were used in the photocatalytic test that was conducted at regular intervals. For catalyst loading at 75, 100, 250, 500, and 750 mg respectively, and for degradation contact time at 10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 110, and 120 minutes, optimization degradation incorporates variations in pH ranges (3, 5, 7, 9, and 11). This degradation process is carried out using UV light W (4×20 watt) on wavelength 665 nm. The following equation formula is used to define the percentage of degradation:

Degradation rate in percentage (%) = $(C_0 - C_t) / C_0 \times 100$, where C_0 is the dye concentration at the beginning of the degradation process and C_t is the dye concentration after the process has finished [33].

2.6 Regeneration Experiment

The initial reaction's suspension created a solid powder that was then centrifuged and used to test the photocatalyst's repeatability of the photocatalyst. The solution and the suspension were separated by a precipitate. The precipitate powder was then dried for 24 h at 70 °C. The following photocatalytic reaction was carried out on the solid powder. The confirmation of the reproducibility of both the LDH pristine and LDH composite photocatalyst, the above method was carried out a fifth time.

3. Results and Discussion

3.1 LDH Pristine and Composite

On Figure 2, XRD analysis of LDH pristine and LDH composites were made. The X-Ray diffraction pattern of pure polyoxometalate

compounds which are $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$, $K_3[\alpha\text{-PW}_{12}\text{O}_{40}]$, LDH pristine NiAl-LDH, MgAl-LDH, and LDH composite are shown in Figure 2. The LDH crystal plane is shown by LDH pristine diffraction peaks at 11.58°, 23.18°, 35.01°, 39.41°, 46.70°, 60.94° and 62.26° which correspond to the (003), (006), (012), (015), (018), (110) and (113) correspondingly, indicate the LDH crystal plane (JCPNDF No. 22-0452) [34]. The diffraction of polyoxometalate compound $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$, $K_3[\alpha\text{-PW}_{12}\text{O}_{40}]$ was shown at 2θ angles of 8-20°, 25-30° [35]. Due to the utilization of composite LDH and polyoxometalate on MgAl-LDH. According to Figure 2, each polyoxometalate peak in MgAl-PW₁₂O₄₀ that was at angles 7.73°, 28.6°, 35.6° which correspond to the (003), (006) and (009) is still visible. $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$ is present in LDH, however, as shown by the 2 angles of 8.61°, 24.27°, 34.96° and 66.34°. The synthesis of the composites was successful. The successfully intercalated of NiAl composite material by showing angles at 10.76°, 26.59°, 30.8° and 63.11°. Each material was shown the LDH pristine by peak 003 indicating that the material contains carbonate

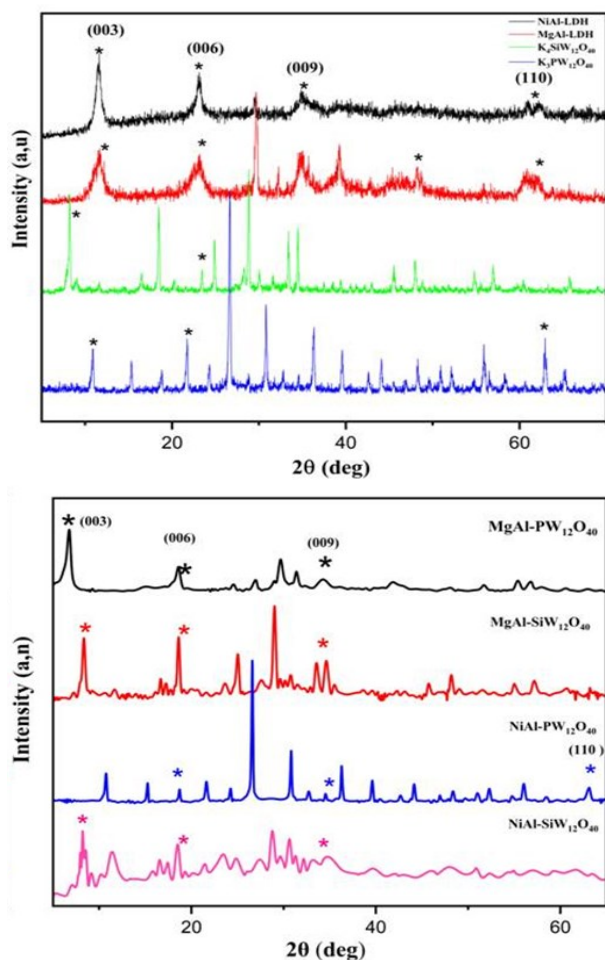


Figure 2. Diffractogram of catalyst.

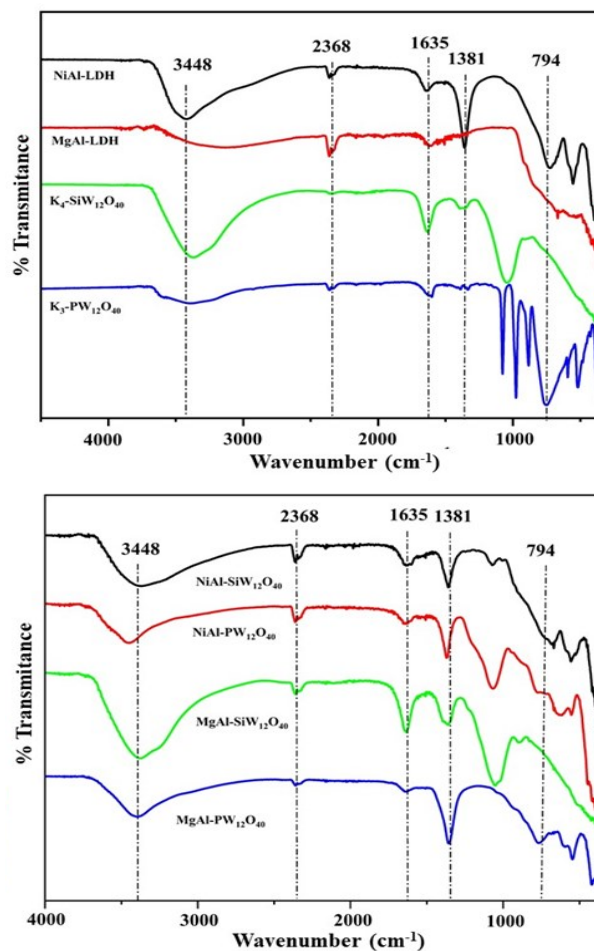


Figure 3. FTIR analysis of catalyst.

and nitrate anions. Nevertheless the typical of polyoxometalate compound in this study showed diffraction at angles 8.26° , 11.34° , 29° and 35.1° .

Figure 3 shows the infrared spectrum of LDH pristine and LDH composite. Spectrum of polyoxometalate compound was measured. The alfa-Keggin SiW_{12} exhibits four distinctive asymmetric vibration bands that correspond to 927 , 973 , 882 , and 813 cm^{-1} in the range $780\text{--}980\text{ cm}^{-1}$ indicated of W–O [36]. By using FTIR analysis, the surface groups of LDH were identified. The composites synthesis was successful. The finding of FTIR characterization of LDH pristine and LDH composite is displayed in Figure 2. The FTIR spectrum band between $3420\text{--}3500\text{ cm}^{-1}$ was shown brucite-like layers

and interlayer water molecules. This band was shown for all material pristine and material composite. The absorption of medium intensity (H_2O) from interlayer water is also close to 1630 cm^{-1} . The 1381 cm^{-1} band represents the CO_3 vibrations [37]. In the bands of $983\text{--}870\text{ cm}^{-1}$ and 804.84 cm^{-1} , respectively, represent the W–O and W–O–W bands [35]. These bands can still be seen in their typical LDH-polyoxometalate catalyst state, as seen in Figure 3.

SEM analysis was used to analyse the morphology of the subsequently synthesized MgAl-LDH, as seen in Figure 4. The MgAl-LDH samples showed morphology, demonstrating the validity of the LDH microsphere to the layered framework. LDH composite and LDH pristine

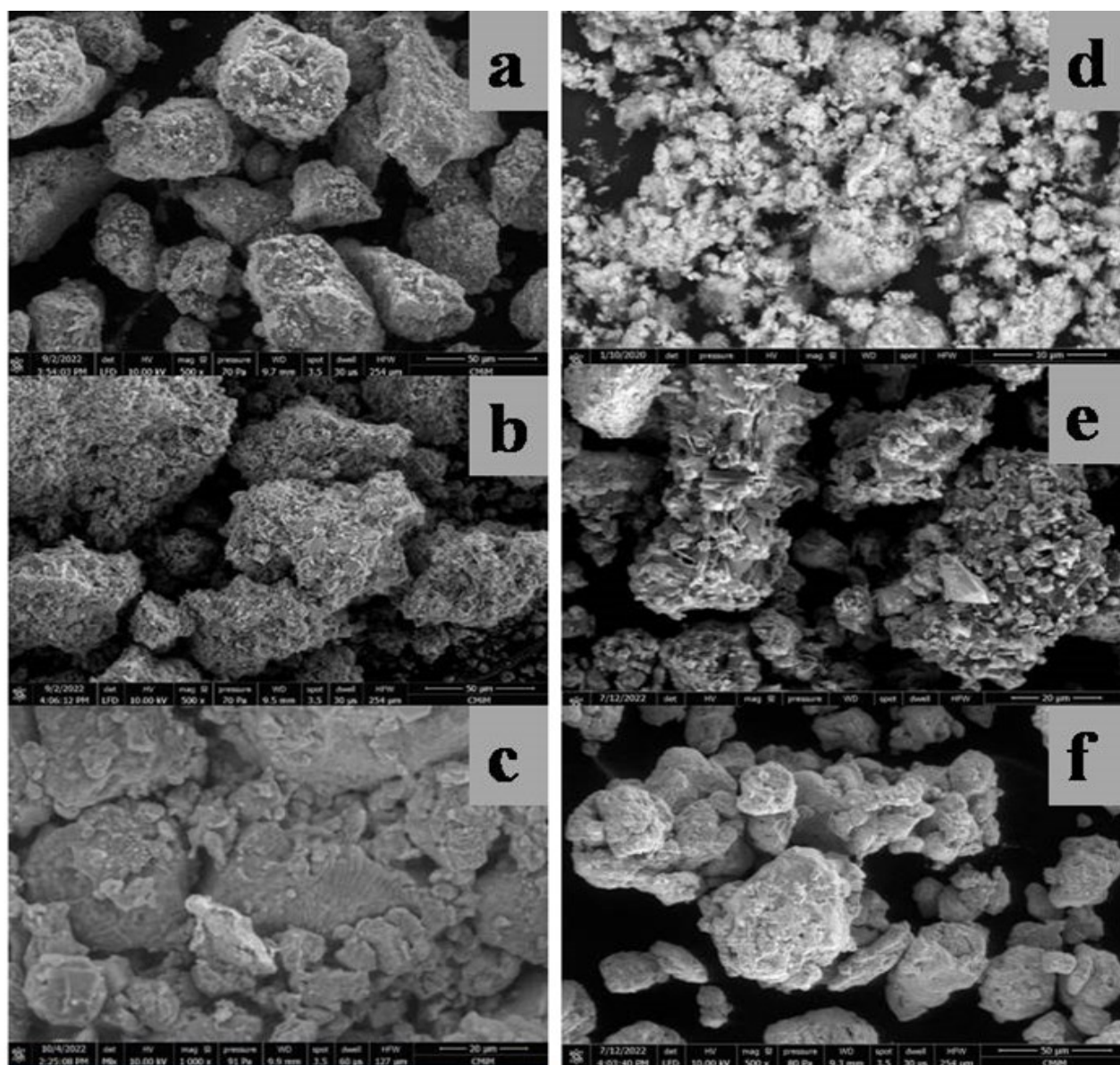


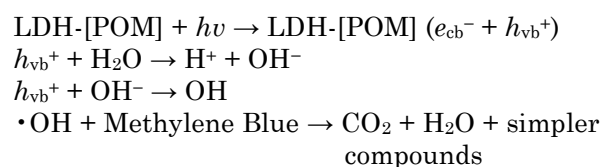
Figure 4. (a) MgAl-LDH, (b) MgAl-[$\text{SiW}_{12}\text{O}_{40}$]. nH_2O , (c) MgAl-[$\text{PW}_{12}\text{O}_{40}$]. nH_2O , (d) NiAl-LDH, (e) NiAl-[$\text{PW}_{12}\text{O}_{40}$]. nH_2O , and (f) NiAl-[$\text{PW}_{12}\text{O}_{40}$]. nH_2O SEM.

have different heterostructures. The morphology which appears to be created by the assembly the two-layered materials is proof that LDH intercalated heterostructures are present. When [SiW₁₂O₄₀], the aggregate appeared on the surface material, according to the morphological structure. It is feasible to understand that platelet-shaped microcrystals exist. If the LDH composite is compared to the LDH pristine, was polypore which meant introducing polyoxometalate added to the blending solution has the potential to bond to the composite's surface. The percentage of Ni, Al, W, P, Si, K, Cl, C, O, and Na atoms are shown in Table 1 of the EDX research. After being composited into MgAl-[SiW₁₂O₄₀] and MgAl-[PW₁₂O₄₀].

3.2 Impact of Optimization Parameters on Methylene Blue Degradation

The stability of the LDH structure may be impacted by the impact of polyoxometalate on MB degradation pH in MgAl-[SiW₁₂O₄₀] and MgAl-[PW₁₂O₄₀]. The capacity to generate positively charged electron holes can be improved by increasing the number of anionic species, especially in media pH levels below 7, which are capable of producing OH• radicals that can degrade dyes and also cause the polyoxometalate

to photodegrade into ions. The decomposition of polyoxometalate into their ions is also possible when the pH is low. There is a photocatalytic reaction of LDH-POM, a UV-irradiated catalyst LDH-POM produces a hole (h_{vb}^+). The hole in the valence reacts with hydroxyl (OH⁻) anion that form a hydroxyl radical (•OH) which can degrade methylene blue and become simply intermediate [38]. The following stage of the process uses LDH-POM to demonstrate the photocatalysis mechanism (Figure 5):



This causes in this study MgAl-LDH and NiAl-LDH were obtained at pH 11 and pH 7 while, pH 11 for K₃[α-PW₁₂O₄₀] and K₄[α-SiW₁₂O₄₀], while for composite material, such as MgAl-[PW₁₂O₄₀], is optimum at pH 1, MgAl-[SiW₁₂O₄₀] is optimum at pH 5, NiAl-[PW₁₂O₄₀] and NiAl-[PW₁₂O₄₀] at pH 11, respectively. Figure 6 demonstrates the impact of pH on the degradation of methylene blue by both pristine and composite LDH catalysts.

In this study, the impact of catalyst weight on the degradation process was also investigated by applying it for 100 min at the ideal pH. Figure 7 shows the actual effect of different loading catalysts on the degrading MB by both LDH pristine and LDH composite, where it is evident that the degradation percentages of the two materials are equivalent. The impact of MB degradation contact time variations by both material catalyst LDH composite and LDH pristine is demonstrated in Figure 7. After 100 min of the treatment, the value of C/Co increased, and as time went on, more MB was

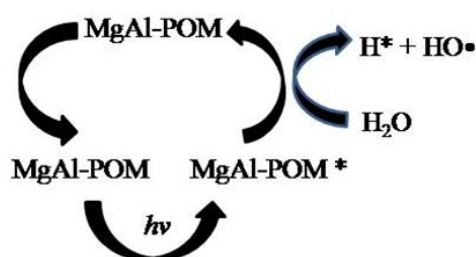


Figure 5. MgAl-POM-based process mechanism for photocatalysis.

Table 1. Catalysts EDX.

Element	MgAl-LDH (%wt)	MgAl-Pw (%wt)	MgAl-Si (%wt)	NiAl-LDH (%wt)	NiAl-Pw (%wt)	NiAl-Si (%wt)
Ni	-	-	-	88.3	4.9	3.7
Mg	8.14	6.32	2.51	-	-	-
Al	5.20	6.85	1.62	1.3	1.7	1.0
W	-	2.15	32.54	-	44.7	19.5
P	-	-	7.82	-	0.7	-
Si	-	-	-	-	-	0.9
K	-	-	3.53	-	2.7	4.9
Cl	-	-	9.17	-	-	7.9
C	13.57	7.98	8.20	-	14.5	24.2
O	51.39	45.85	28.78	9.1	25.6	27.8
Na	2.58	4.1	5.83	-	4.9	9.6
N	9.12	-	-	1.4	-	-

degraded. The percent MB degradation for MgAl-LDH, NiAl-LDH, MgAl-[PW₁₂O₄₀], MgAl-[SiW₁₂O₄₀], NiAl-[PW₁₂O₄₀], NiAl-[SiW₁₂O₄₀] were 43.14%, 45.53%, 58.73%, 75.94%, and 85.86%, respectively. The percent degradation

for K₃[α-PW₁₂O₄₀] and K₄[α-SiW₁₂O₄₀] were 73.14% and 78.79%, respectively. According to the percentage decomposition results, the catalyst of LDH composite appears to have a greater capacity to degrade MB than LDH pristine.

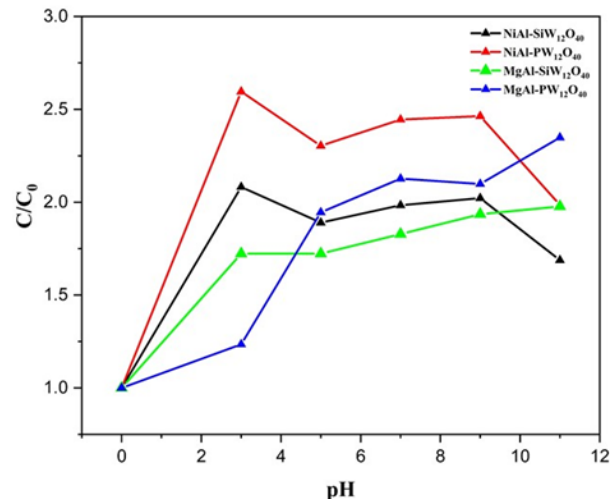
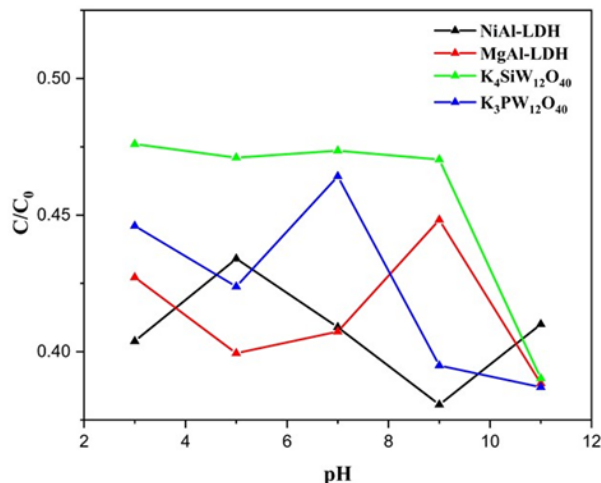


Figure 6. Impact of pH on catalyst mediated MB degradation.

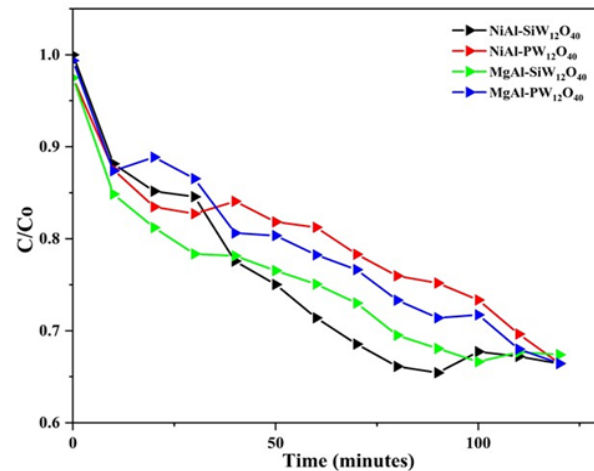
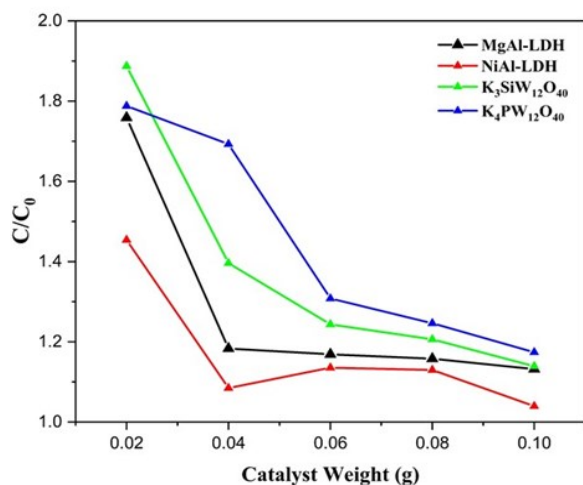
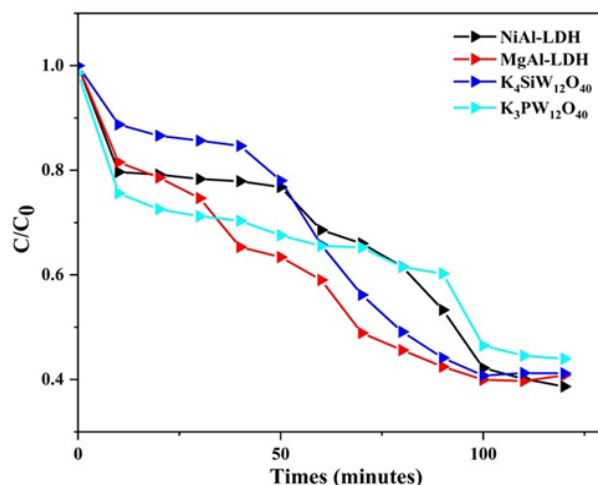
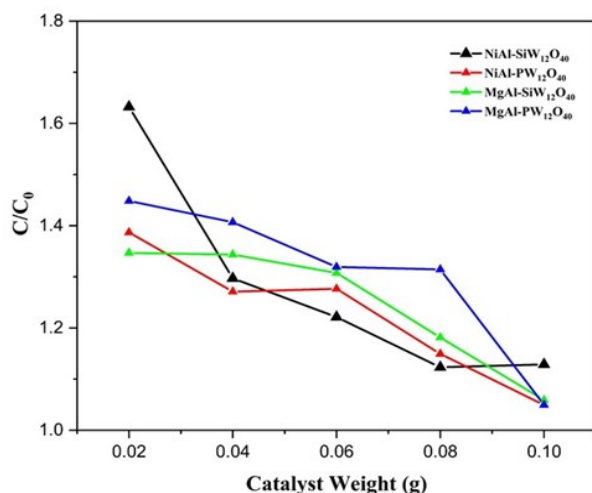


Figure 7. Effect of catalyst weight on MB degradation.

Figure 8. Effect of time on catalytic degradation of MB.

Figure 8 shows for all materials increased the %degradation process by increased contact time effect.

3.3 Regeneration of Catalyst

Indicating that the catalyst's use is anticipated to be effective so that it can be used frequently to degrade organic pollutants, its ca-

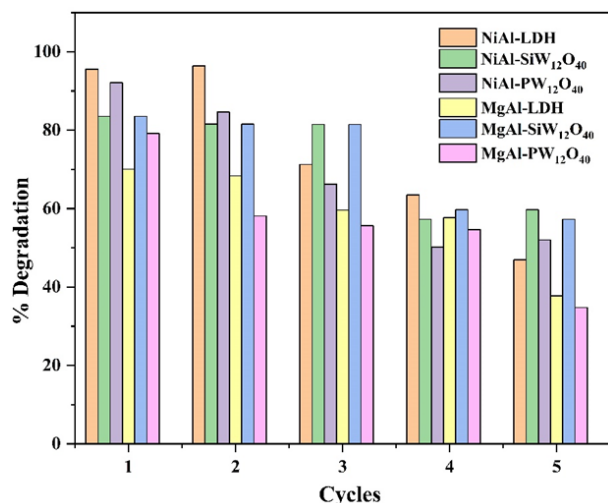
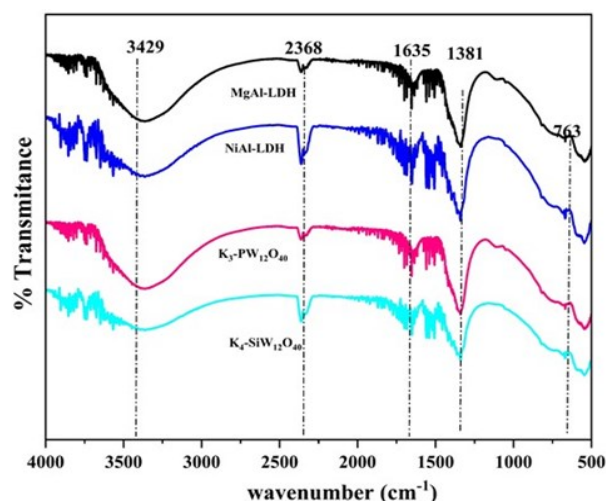


Figure 9. Catalyst regeneration in fifth cycle.



capacity is determined by the decline in the percentage of degradation after several cycles of repeated usage. After being used again as a photocatalyst, the deteriorating ability is shown to decreased in Figure 9. After five cycles, the degradation rate using the MgAl-LDH and NiAl-LDH catalyst was reduced by 46.1% and 55.3%. After 5 cycles of usage, MgAl-[PW₁₂O₄₀], MgAl-[SiW₁₂O₄₀], NiAl-[PW₁₂O₄₀] and NiAl-[SiW₁₂O₄₀] reduced LDH composite degradation by 63.4%, 70.4%, 72%, and 83%, respectively. The apparent rate value and percentage of deterioration matched the reusability of both pristine and composite LDH. The %degradation and apparent rate value matched the reusability of all materials. Table 2 shows the value of %degradation methylene blue with different material. It is evident from the reusability data that the LDH-[SiW₁₂O₄₀] exceeded the LDH-[PW₁₂O₄₀].

Figure 10 shows FTIR spectra of all materials MgAl-LDH, NiAl-LDH, K₄[α-SiW₁₂O₄₀], K₃[α-PW₁₂O₄₀].nH₂O, MgAl-[SiW₁₂O₄₀], MgAl-[PW₁₂O₄₀], NiAl-[SiW₁₂O₄₀], and NiAl-[PW₁₂O₄₀]. The interpretation of stretching and bending vibration of FTIR spectra revealed the

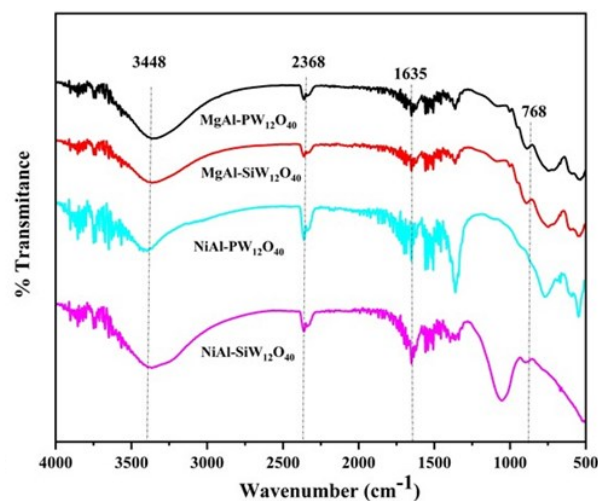


Figure 10. FTIR spectrum following a fifth cycle regeneration.

Table 2. The value of %degradation methylene blue.

Catalyst	%degradation methylene blue	Ref.
Mn ₂ Cr-LDH	81	[39]
Mn-ZnO	83	[40]
Zn/Al	75	[41]
BNC-CLs	84	[42]
MnMgFe-LDH	80	[43]
MgAl-[PW ₁₂ O ₄₀]	58	In this study
MgAl-[SiW ₁₂ O ₄₀]	75	In this study
NiAl-[PW ₁₂ O ₄₀]	78	In this study
NiAl-[SiW ₁₂ O ₄₀]	85	In this study

good formation of MgAl-LDH, NiAl-LDH, $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$, $K_3[\alpha\text{-PW}_{12}\text{O}_{40}]$, and MgAl- $[\text{SiW}_{12}\text{O}_{40}]$, MgAl- $[\text{PW}_{12}\text{O}_{40}]$, NiAl- $[\text{SiW}_{12}\text{O}_{40}]$, and NiAl- $[\text{PW}_{12}\text{O}_{40}]$. LDH composite that needed to degrade MB was characterized using an FTIR spectrum to show the effect upon regeneration. The unlimited range a bands with wavelengths between $3420\text{--}3500\text{ cm}^{-1}$ and around $1630\text{--}1583.6\text{ cm}^{-1}$ included the stretching vibration of the hydroxyl groups coupled with the stretching vibration of interlayer water molecules. The intercalated NO_3^- bending vibration is represented by the peak at 1359 cm^{-1} [44]. The W-O and W-O-W bands, which are typical of polyoxometalate, are in the 985.4 cm^{-1} and 856.3 cm^{-1} ranges, respectively. The band on MgAl- $[\text{PW}_{12}\text{O}_{40}]$ are also demonstrated to be 795.4 [45]. Ta from the IR spectra $[\text{PW}_{12}\text{O}_{40}]$ are still stored in the LDH. Figure 10 demonstrates that for the catalyst that has degraded MB, where these band are still visible.

4. Conclusion

Successful preparation and usage of the LDH pristine and LDH composite for photodegradation MB. Based on the characterization findings, the materials that were synthesized in the best way had layer structures that were well-oriented. The capacity to photodegrade cationic dyes MB was improved by modifying LDH material, which forms composites with two polyoxometalates $[\alpha\text{-PW}_{12}\text{O}_{40}]$ and $[\alpha\text{-SiW}_{12}\text{O}_{40}]$. In comparison to MgAl-LDH, composite MgAl- $[\text{PW}_{12}\text{O}_{40}]$ and MgAl- $[\text{SiW}_{12}\text{O}_{40}]$ composites both enhanced MB degradation regeneration performance. Materials that have degraded MB were characterized by FTIR after the fifth cycle of regeneration, and the results showed that all materials LDH pristine and LDH composite both still had their original LDH and polyoxometalate structures. The investigation's finding on pH, catalyst loading, and ideal contact time revealed percentages of degradation and reusability.

Acknowledgment

The authors thank to Research Center of Inorganic Materials and Complexes FMIPA Universitas Sriwijaya for valuable discussion, apparatus, and chemical analysis.

CRedit Author Statement

Author Contribution: Yulizah Hanifah: Experiment, Methodology, Investigation, Resorces, data Curation, Writing, Review and

Editing, Supervision; Risfidian Mohadi: Review, Formal Analysis and Data Curation; M. Mardiyanto: Review and Data Curation; Nur Ahmad: Review and Data Curation; S. Suheryanto: Review and Data Curation; Aldes Lesbani : Review, Formal Analysis, Editing and Data Curation. All authors have read and agreed to the published version of the manuscript.

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