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Research Paper

Catalytic Activity of LDH-TiO_2 and LDH-ZnO in Photodegradation of Procion Red

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Abstract

Pristine layered double hydroxides (LDHs) which have been modified into LDHs-metal oxide composite were applied as procion red (PR) photodegradation catalysts. The preparation was performed using a low calcination temperature of 300°C. Optimization variations include pH, catalyst weight and catalysis time. The degradation percentages of Mg/Al LDH, Mg/Al-TiO_2 and Mg/Al-ZnO were 51.8%, 72.0% and 89.5%, respectively. The percentage of degradation of Zn/Al LDH, Zn/Al-TiO_2 and Zn/Al-ZnO were 44.0%, 61.2% and 58.4%, respectively. The study results showed that all composites produced a greater percentage of PR degradation and better reusability than pristine LDH. Mg/Al LDH-based catalysts tend to catalyze PR better than Zn/Al LDH-based catalysts. PR which is an anionic dye when photodegraded using Mg/Al-ZnO tends to be better than Mg/Al-TiO_2, meanwhile Zn/Al-TiO_2 and Zn/Al-ZnO have competitive performance.

Keywords

LDH, Photodegradation, Procion Red, TiO_2, ZnO

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1. INTRODUCTION

Procion red (PR) is one of the anionic organic dyes used in the cellulose, silk, nylon or wool textile industries. The heavy molecular weight and complex chemical composition cause this dye to have high solubility in industrial wastewater (Nazifa et al., 2018). Removal of dyes from wastewater bodies requires methods that do not pose a threat, such as reusing pollutant adsorbents so that they are relatively not wasted into the environment. Several types of adsorbents can be further used as catalysts for pollutant degradation. Layered double hydroxide (LDH) is an adsorbent that can be used as a catalyst for pollutant photodegradation (Shen et al., 2020).

LDH material is brucite-like hydroxide layers composed of cations 2^+ (M^{II}), cations 3^+ (M^{III}) and anions (A^{n-}) with the composition $[M^{II}{}_{1-x}\ M^{III}{}_x\ (OH)_2](A^{n-})_{x/n}.mH_2O$ (Jaerger et al., 2021; Zhao et al., 2018). This photodegradation catalyst material is often synthesized through the coprecipitation technique because it only goes through one easy step with a good yield (Rahman and Pullabhotla, 2022). Co-precipitation also produces a uniform product. Hydrothermal is effective when the LDH interlayer species is an organic compound. The sol gel technique produces product properties that depend on condensation, hydrolysis and solvent (Song et al., 2019). The microemulsion synthesis method requires large amounts of surfactants (Xu et al., 2017).

Metal oxide semiconductor materials such as TiO_2 and ZnO can be impregnated on LDH to form a photodegradation catalyst. The use of LDH composited with TiO_2 and ZnO will inhibit recombination in the band gap, besides facilitating electron transfer (Jaerger et al., 2021; Bhuvaneswari et al., 2019). The optimum LDH/TiO_2 ratio composition is at 1:1 (Djeda et al., 2020). LDH-TiO_2 which degrades phenol produces a similar degradation percentage between LDH/TiO_2 with a ratio of 1:1 and 1:10 (Contreras-Ruiz et al., 2016). Several studies have stated that ZnO is more capable of degrading several organic compounds than TiO_2 (Yan et al., 2016; Tzompantzi et al., 2014). This is possible because ZnO can absorb a wider spectrum of energy quanta than TiO_2 (Amor et al., 2018). However, there is a risk of dissolution and photodecomposition of ZnO in acidic media (Elhalil et al., 2018).

Based on other studies, Mg/Al LDH-based materials catalyze better than Mg/Fe LDH. Zn/Al LDH which has better semiconducting properties, is able to produce a relatively higher percentage of degradation than Mg/Al LDH (Contreras Ruiz et al., 2019). On the contrary, Djeda et al. (2020) found that Zn/Al LDH catalyzed photodegradation better than Zn/Cr LDH, but Mg/Al LDH was able to catalyze better than Zn/Al LDH.

Several studies on LDH materials as catalysts for the photodegradation of organic pollutants have been carried out by calcination at relatively high temperatures. Mg/Fe-TiO_2 was prepared by sol gel and calcined at a temperature of 550°C (Contreras Ruiz et al., 2019). The coprecipitation technique followed by calcination at a temperature of 500°C was carried out on the preparations of Zn,Mg/Al (Zarate Morales et al., 2018), Mg,Zn/Al-ZnO (Yan et al., 2016), Mg/Fe-TiO_2 (Phuong et al., 2016) and Zn/Al-TiO_2 (Hadnadjev Kostic et al., 2017). Hydrothermal techniques have also been used in the preparation of Zn/Al-ZnO photodegradation catalysts followed by calcination at 650°C (Trujillano et al., 2020) and Ba,Zn/Al-ZnO which were calcined at 500°C (Elhalil et al., 2018). Di et al. (2017) obtained the optimum calcination temperature at 300°C while synthesizing Zn/Fe LDH, this is a temperature that is not high and has the potential to save calcination energy. In recent years, there has been a tendency to study the preparation of LDH-based catalysts synthesized by coprecipitation to avoid calcination temperatures, increasing photodegradation performance can be carried out by adding persulfate (Dung et al., 2021), adding H_2O_2 (Xue et al., 2023) or using a catalyst which can release H_2O_2 to liquid media (Aragaw et al., 2022).

The novelty of this study includes preparing Mg/Al-ZnO, Mg/Al-TiO_2, Zn/Al-ZnO and Zn/Al-TiO_2 catalysts by coprecipitation at a relatively low calcination temperature of 300°C. The LDH/metal oxide ratio used was 1:1. The catalysts were applied to the photodegradation of procion red dyes.

2. EXPERIMENTAL SECTION

2.1 Chemicals and Instrumentations

The chemicals used in this study include titanium dioxide, sodium carbonate, sodium hydroxide, aluminum nitrate and magnesium nitrate were produced by Merck, zinc nitrate obtained from Smart-Lab, zinc oxide supplied by LOBA Chemie and hydrogen chloride by Mallinckrodt LabGuard. Characterization of materials were provided by Rigaku XRD Miniflex-6000, SEM Quanta-650 Oxford Instrument and FTIR Bruker. Analyses PR were performed by UV-Vis Biobase BK-UV 1800 PC at 537 nm.

2.2 Synthesize of Pristine LDH and Preparation of Composites

LDH-pristine was synthesized by reacting 0.25 M $Al(NO_3)_3 \cdot 9H_2O$ with 0.75 M $Mg(NO_3)_2 \cdot 6H_2O$ or $Zn(NO_3)_2 \cdot 6H_2O$ of 50 mL each. After the solution was stirred for 2 hours, 60 mL of 0.25 M Na_2CO_3 and 60 mL of 0.5 M NaOH were added as a base. This mixture was stirred for 10 hours at 80°C. This process produced precipitate, then the resulting sludge is dried in the oven and weighed (Yuliasari et al., 2022d).

Pristine LDH synthesis is also used as an initial procedure for forming composites. After the cationic salts were reacted under alkaline conditions at pH 10 and stirred at 80°C, the composite was prepared with metal oxide impregnation. ZnO or TiO_2 is dispersed into the mixture with the same weight ratio as the weight of pristine LDH previously formed. This mixture was stirred for 3 hours and required alkaline conditions with 150 mL of 0.37 M NaOH, then stirred for 10 hours at 70°C. The resulting precipitate was washed with water and then calcined at 300°C for 7 hours (Yuliasari et al., 2022d; Djeda et al., 2020).

2.3 Catalytic Fotodegradation

The catalysts formed were used in the photodegradation of PR with an initial concentration before contact with the catalyst of 400 mg/L. The PR solution was contacted with the catalyst in a dark room for 60 minutes, after which the photodegradation was carried out using UV light. The variables used during photodegradation were pH optimization, catalyst weight, and catalysis time. The reusability ability was reported based on the *5th* regeneration cycle with the desorption method using sonication for 10 minutes. The rate of photodegradation takes place based on Equation 1, if this equation is integrated it will be the formulation of first order pseudo kinetics according to Equation 2 (Contreras Ruiz et al., 2019). The C_0 value is the PR concentration after contact with the catalyst for 60 minutes in the dark room, C is the residual PR concentration according to the catalysis time. The kinetic constant is denoted by K and t is the catalytic time.

$$\frac{\text{-dC}}{\text{dt}} = \text{K.C} \tag{1}$$

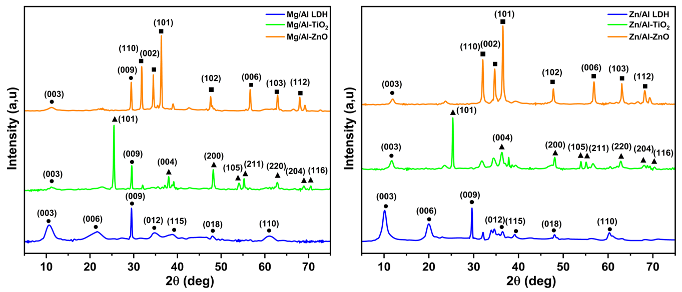
$$\ln\frac{(C_o)}{C} = \text{K.t} \tag{2}$$

3. RESULTS AND DISCUSSION

3.1 Characterization of Fresh Prepared Catalysts

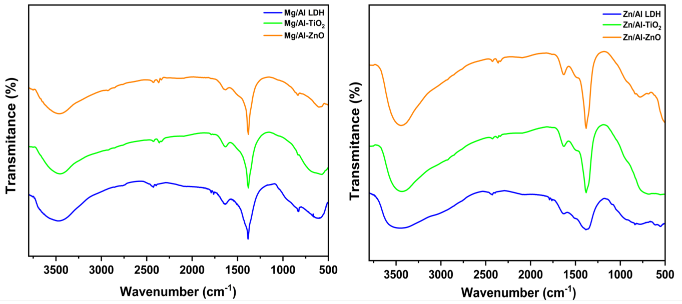
The pristine LDH was characterized by a unique peak indicating a cationic layer structure at an angle of 2θ around 11° for the Miller index plane 003, the presence of interlayer anions around 60o for the index plane 110 and metal oxide peaks at 20°-35° for the index planes 006 and 012 (Bernard et al., 2022). The results of the study by Elhalil et al. (2018) reported that the ZnO peaks of the calcined Ba,Zn/Al-ZnO material were at an angle 2θ 31.8°, 34.5°, 36.3°, 47.6°, 56.6°; 62.9°, 68.0° dan 69.1° which respectively indicate the Miller index 110, 002, 101, 102, 006, 103, 112 and 201. Research byContreras Ruiz et al. (2019) synthesized Mg/Al-TiO_2 and Zn/Al-TiO_2 via the sol gel method, obtained characteristic peaks of TiO_2 at an angle of 2θ at 25°, 38°, 48°, 55°, 63° respectively showing index of 101, 004, 200, 105 and 220. Figure 1 presents the diffractogram of the prepared catalysts. The results of this study also produced the LDH peaks in the synthesized pristine LDH as well as the peaks of TiO_2 and ZnO modified composites.

Material characterization using FTIR instruments shows distinctive bands indicating the presence of vibrations of certain functional groups of chemical compounds. The stretching vibration band of the OH^- group of LDH lies between 3200



(a) (b)

Figure 1. Diffractogram of Fresh Mg/Al-LDH Based Catalyst (a) Zn/Al-LDH Based Catalyst (b)



(a) (b)

Figure 2. FTIR Spectra of Mg/Al-LDH Based Catalyst (a) Zn/Al-LDH Based Catalysts (b)

cm^{-1}-3600 cm^{-1}. OH^- bending vibration approx 1640 cm^{-1} (Aragaw et al., 2022). Hosseini and Akbari (2016) and Yuliasari et al. (2022a) obtained the band that appears on 1381 cm^{-1} was vibration CO_3^{2-} from LDH, while Zhao et al. (2018) stated band 1384 cm^{-1} shows the stretching vibration of CO_3^{2-}. Bernard et al. (2022) claimed the asymmetric stretching vibration of CO_3^{2-} located at 1380 cm^{-1}. Aragaw et al. (2022) reported a metal-oxide vibration band around 453-619 cm^{-1}. Aoudjit et al. (2019) states the existence of metal-oxygen bond is indicated by the vibration band between 400 cm^{-1}-800 cm^{-1}. Jaerger et al. (2021) states that the wave numbers between 455 cm^{-1}-719 cm^{-1} are metal-oxygen stretching vibrations and metal-hydroxyl bending vibrations. Figure 2 shows the vibration bands of the prepared Mg/Al LDH-based, Zn/Al LDH-based materials. The IR spectra of this study also showed OH^- stretching vibration bands, OH^- bending, anions between layers of CO_3^{2-} appearing in all pristine LDH, LDH-metal oxide composites and the spectra exhibiting metal oxide vibrations.

3.2 Composites Morphology

The prepared catalyst product was characterized by its surface morphology. The rough surface of the catalyst is advantageous for the photodegradation process, where the catalyst with greater surface roughness results in a greater percentage of degradation (Contreras Ruiz et al., 2019). Figure 3 shows a 10,000 magnification photo of the surface morphology of the rough catalyst after preparation and before contact with the PR as dye model.

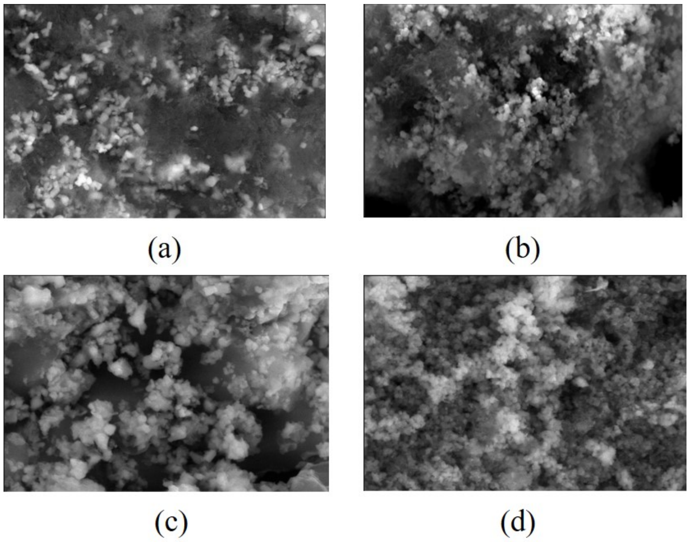
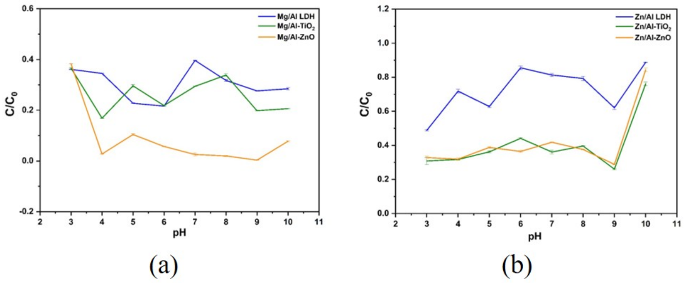


Figure 3. Morphology of Mg/Al-ZnO (a), Mg/Al-TiO_2 (b), Zn/Al-ZnO (c) Zn/Al-TiO_2 (d)

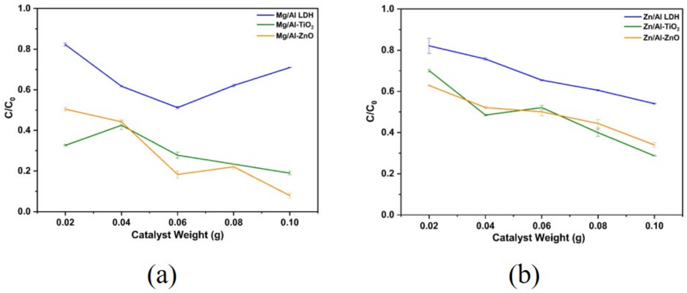
3.3 Effect of pH on Procion Red Fotodegradation

The acidity or pH of the media affects the photodegradation process because both the compounds being degraded and the catalyst each have a certain charge. Research conducted by Ma et al. (2017) chose pH 9 to degrade anionic dyes with the help of MgZn/Cr-TiO_2. This is understandable because at alkaline pH there are more OH• so that there are more hydroxyl oxidizing pollutants. Meanwhile, other studies obtained the optimum pH for photodegradation of anionic dyes at pH below 7, including photodegradation catalyzed by NiCoFe reaching 40% at pH 3 and only 7.7% at pH 7.5 (Narde et al., 2017); Cr_2O_3 catalyzes photodegradation at pH 3 of 34% and at pH 10 of 14% (Shaban et al., 2017); likewise, photodegradation reached 97.2% at pH 3 and 46.4% at pH 10 (Shaban et al., 2020). PR dyes have negatively charged groups which cause a tendency to interact with catalysts which are positively charged or at a media pH below pH 7. Figure 4 shows the effect of pH variations on PR photodegradation for 120 minutes, the PR concentration before contact with the catalyst was 50 mg/L. Catalysis by Mg/Al LDH, Mg/Al-TiO_2, Zn/Al-LDH occurred at pH 6, pH 4 and pH 3 respectively, while the catalysis by Mg/Al-ZnO and Zn/Al-ZnO was optimum at pH 9. Elhalil et al. (2018) stated that there is a risk of ZnO dissolving and photodecomposing into cations in acidic conditions. In this study Zn/Al-TiO_2 is also optimum at pH 9 because the calcination process can cause the formation of ZnO as well.



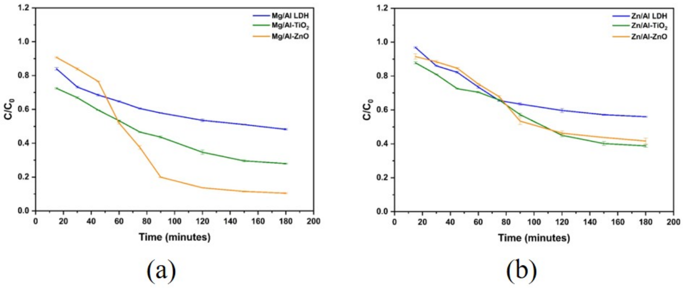
(a) (b)

Figure 4. Effect of pH on the Photodegradation of Procion Red



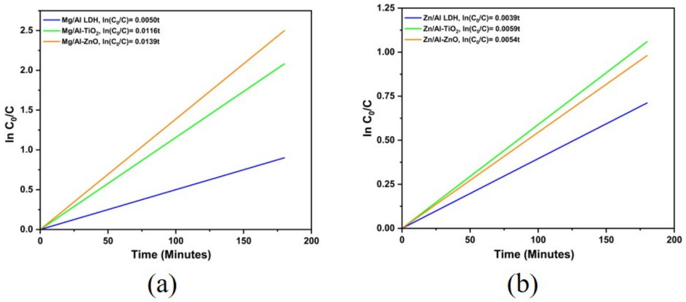
(a) (b)

Figure 5. Effect of Catalyst Weight on Procion Red Photodegradation



(a) (b)

Figure 6. The Effect of Catalysis Time on the Photodegradation of Procion Red



(a) (b)

Figure 7. Kinetic Curve of Pseudo First Order Degradation of Procion Red photodegradation

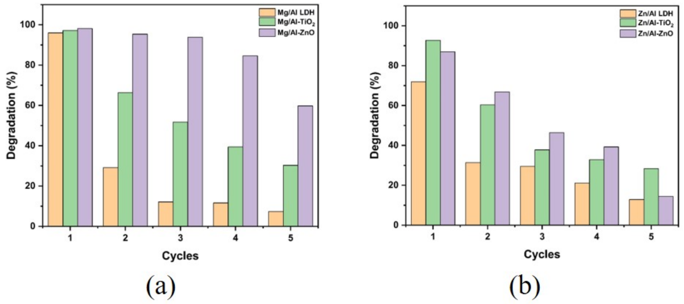
3.4 Effect of Catalyst Weigh on Procion Red Fotodegradation

The effect of catalyst weight on the photodegradation of procion red for 120 minutes is shown in Figure 5, the PR before contact with catalyst concentration 100 mg/L. In general, the greater the amount of catalyst, the more dye can be degraded. The decrease in photodegradation performance by Mg/Al-LDH when the catalyst weight is above 0.06 g/20 mL can be caused by the effect of light refraction that occurs in the liquid medium. Elhalil et al. (2018) reported that there was a possibility of the effect of light refraction which caused a reduction in the percent degradation when carrying out experiments on variations in catalyst weight.

3.5 Effect of Catalysis Time on Procion Red Fotodegradation

The effect of length of time on the photodegradation of procion red is shown by Figure 6, the PR before contact with catalyst concentrate 400 mg/L. The data shows that the longer the catalytic time, the more the dye is degraded. The degradation percentages of Mg/Al LDH, Mg/Al-TiO_2 and Mg/Al-ZnO were 51.8%, 72.0% and 89.5%, respectively. The degradation percentages of Zn/Al LDH, Zn/Al-TiO_2 and Zn/Al-ZnO were 44.0%, 61.2% and 58.4%, respectively. These data are in line with the results of Yuliasari et al. (2022c) who obtained the percent degradation of the Mg/Al-TiO_2 and Mg/Al-ZnO composites better than pristine LDH during the photodegradation of congo red anionic dyes. The data in the figure shows the tendency of the Mg/Al-ZnO composite to have a relatively better percentage of degradation than Mg/Al-TiO_2. This is likely to occur because according to Amor et al. (2018) ZnO can absorb a wider spectrum energy quanta than TiO_2. In addition, the optimum pH conditions obtained and carried out when using Mg/Al-ZnO applied at pH 9 providing more OH• radicals to degraded. Meanwhile, the competitive ability of photodegradation performance between Zn/Al-TiO_2 and Zn/Al-ZnO can be caused by the same optimum pH conditions at pH 9 and the calcination of Zn/Al-TiO_2 allows the formation of ZnO as well. Table 1 presents several percent degradation results of PR photodegradation studies.

The effect of catalysis time can be processed into a pseudo-first order kinetic curve. The kinetic equation used is ln (C_0/C)=



(a) (b)

Figure 8. The Procion Red Photodegradation Regeneration Cycle Curve

Table 1. Comparison of the Percentage of PR Degradation Against Other Studies

Material	Degradation (%)	Dye	Dye Concentration (mg/L)	References
Pseudomonas Stutzeri	30	PR	300	Bera and Tank (2021)
Active Carbon-TiO_2	71	PR	300	Ruliza et al. (2018)
ZnO-Zeolit	83.96	PR	150	Salam et al. (2018)
Aspergillus Terreus	68	PR	200	Almeida and Corso (2014)
Film TiO_2	60	PR	50	Almuminin et al. (2017)
ZnO-TiO_2	55.4	PR	50	Haryati et al. (2014)
Mg/Al-TiO_2	72.0	PR	400	This study
Mg/Al-ZnO	89.5	PR	400	This study
Zn/Al-TiO_2	61.2	PR	400	This study
Zn/Al-ZnO	58.4	PR	400	This study

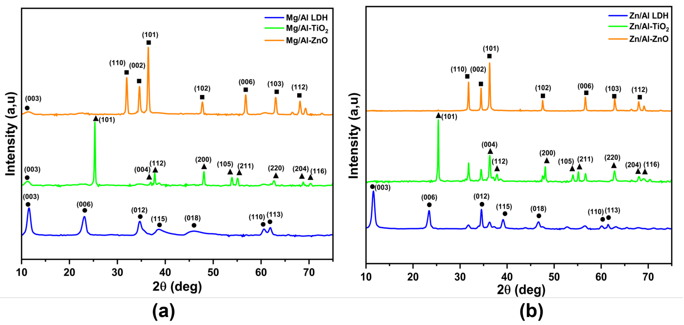


Figure 9. Diffractogram of Mg/Al-LDH Based Catalyst (a) Zn/Al-LDH Based Catalyst (b) After 5^{th} Cycle Regeneration

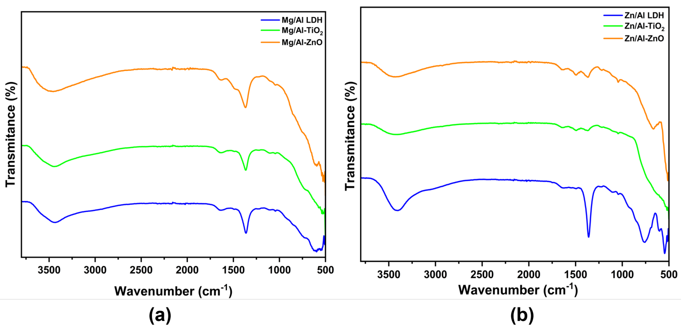


Figure 10. FTIR Spectra of Mg/Al-LDH Based Catalyst (a) Zn/Al-LDH Based Catalysts (b) After 5^{th} Cycle Regeneration

K.t according to the Equation 2. The value of the kinetic constant (K) is directly proportional to the photodegradation performance. The use of Al^{3+} cations also resulted in a better percent degradation and kinetic rate than Fe^{3+} in the pristine LDH Mg^{2+}/M^{3+} (Contreras Ruiz et al., 2019). Figure 7 displays the kinetics curves of the catalysts in PR photodegradation. It can be seen from the figure that the performance of the modified composites outperforms pristine LDH. Kinetic constants of Mg/Al LDH, Mg/Al-TiO_2 and Mg/Al-ZnO were 0.0050 min^{-1}, 0.0116 min^{-1} and 0.0139 min^{-1}, respectively. The kinetic constants of Zn/Al LDH, Zn/Al-TiO_2 and Zn/Al-ZnO were 0.0039 min^{-1}, 0.0059 min^{-1} and 0.0054 min^{-1}, respectively. From the data it can be seen that the modified composites do have a higher kinetic constant than pristine LDH. These data agree with the results of Yuliasari et al. (2022b) who obtained the catalytic kinetics constant of the Zn/Al-TiO_2 and Zn/Al-ZnO composites better than pristine LDH during the photodegradation of congo red anionic dyes.

3.6 Regeneration Cycle on Procion Red Fotodegradation

The reusability of the catalysts used for 120 minutes until the fifth cycle of PR photodegradation is presented in Figure 8. The results show that all modified composites produce a material that has better stability than pristine LDH. The $CO_3{}^{2-}$ anion produces the best LDH structural stability compared to $SO_4{}^{2-}$, Cl^-, and $NO_3{}^-$ (Bernard et al., 2022). Al^{3+} in the LDH-ZnO composite structure has good stability (Elhalil et al., 2018). Zhao et al. (2018) stated that modification of the basic LDH structure to a LDH-metal oxide composite will have the advantages of reusability and stability.

3.7 Characterization after Regeneration Cycle

Catalyst materials that have undergone a regeneration cycle are characterized to study the stability of the catalyst structure. Figure 9 shows the diffractogram of the catalyst that has undergone the 5^{th} cycle of PR photodegradation regeneration. The data show that pristine LDH still exhibits an LDH structure with a Miller index of 003 at an angle of 2θ at about 10°, similarly it still shows an ionic interlayer structure at an angle of 2θ around 60° which shows a Miller index of 110 and 113. Composites that have undergone calcination during preparation has lower cationic layer diffractogram peaks, but still shows clear peaks of TiO_2 and ZnO.

The FTIR spectra of the catalysts that have interacted with procion red until the 5^{th} cycle of regeneration is shown in Figure 10. FTIR data shows that all catalysts still have a hydroxide structure at wave numbers around 3400 cm^{-1} and 1600 cm^{-1}. The carbonate band as an intercalation anion is shown at a wave number around 1360 cm^{-1} and metal oxide band appears at a wave number around 500 cm^{-1}.

4. CONCLUSION

This study concluded that modifying pristine LDH into LDH composites would increase the catalytic activity of procion red (PR) photodegradation as evidenced by increasing the percentage of PR degradation. The preparation of this LDH-metal oxide composite also increases the stability of the material so that it had better reusability than pristine LDH. Mg/Al LDH based catalyst tends to catalyze PR better than Zn/Al LDH based catalyst. PR photodegradation of an anionic dye when using Mg/Al-ZnO tends to be better than Mg/Al-TiO_2 because it is optimum at alkaline pH, while Zn/Al-ZnO and Zn/Al-TiO_2 have competitive performance because both are optimum at alkaline pH.

5. ACKNOWLEDGMENT

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