Improvement of Congo Red Photodegradation Performance Through Zn/Al-TiO$_2$ and Zn/Al-ZnO Preparation

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Abstract
Layered double hydroxide (LDH) is an anionic clay material known to be effective as a catalyst for the photodegradation of dye organic pollutants. Zn/Al LDH was synthesized by coprecipitation then impregnated with metal oxides and calcined at 300°C to form Zn/Al-TiO$_2$ and Zn/Al-ZnO as photodegradation catalysts of congo red (CR). The characterization of the catalysts after preparation using SEM and UV-DRS while the catalyst that have been used in 5 regeneration cycles was characterized by XRD and FTIR. Photodegradation of CR was carried out by optimizing pH, catalyst weight, and time radiation. Zn/Al LDH which was modified into Zn/Al-TiO$_2$ and ZnAl-ZnO had a better degradation percentage, rate constant, and stability than Zn/Al LDH pristine structure. Zn/Al LDH, Zn/Al-TiO$_2$ and Zn/Al-ZnO catalyzed CR photodegradation for 120 minutes with percent degradation 68.39%, 81.24% and 71.09%, respectively.

Keywords
Zn/Al LDH, Congo Red, ZnO, TiO$_2$, Photodegradation

1. INTRODUCTION
Pollution problems are often associated with global industrial waste. Waters polluted by textile dyeing can account for nearly twenty percent of the pollutants (Guzmán-Vargas et al., 2016). Layered double hydroxide (LDH) has been reported by many studies as a photocatalyst for the degradation of organic pollutants including textile dyes (Muhammad and Garzali, 2019). LDH is the best clay that is semiconducting, has a cation of 2+ valence at the center of an octahedral of 6 hydroxyl groups and forms lamellar. The cations with valence 2+ are partially substituted by cations with valence 3+ resulting in a positive charge on the layer of the cation structure. Negatively charged anions can be intercalated between the structure of the cation layer. LDH materials can be prepared by varying the content of cation elements to produce different physical and chemical properties as needed (Li et al., 2020; Contreras-Ruiz et al., 2019).

Zn/Al LDH LDH materials have often been used to degrade pollutants owing to their good semiconductor properties as photocatalysts (Li et al., 2020). Not a few studies on the degradation of organic pollutants catalyzed by Zn/Al LDH. Hydrothermal methods have been used in preparing Zn/Al-based catalysts, including Zn/Al-C$_3$N$_4$ (Gandamalla et al., 2021), Zn/Al-TiO$_2$ (Mourid et al., 2020), Zn/Al-ZnO (Trujillano et al., 2020), ZnBa/Al-ZnO (Elhaliil et al., 2018), Zn/Al-TiO$_2$ (Aoudjit et al., 2019), Zn/Al-ZnO (Zhang et al., 2016), and ZnCu/Al (Kim et al., 2017) as photocatalysts in the degradation of organic pollutants, respectively ciprofloxacin, sulfamethoxazole, nitrophenol, caffeine, dodecyl sulphate, methylene blue, and orange II dyes.

LDH preparation technique by coprecipitation is often done because it is not expensive and simple to synthesize, this technique includes the deposition of two or more metal ions using a precipitating agent (Song et al., 2019). Studies have been carried out using the coprecipitation method using urea as a precipitate in the preparation of Zn/Al LDH-based materials, these studies were as follows: ZnMn/Al (Morales-Mendoza et al., 2015), ZnNi/Al (Wang et al., 2014a) and Zn/Al-TiO$_2$ (Wang et al., 2014b) which were used in the degradation of chlorophenol, orange G and methylene blue, respectively, while Zn/AlCe (Suárez-Quezada et al., 2016) and Zn/AlLa (Tsompantzi et al., 2014) were used for phenol degradation. The coprecipitation method using sodium hydroxide precipitate was carried out in the preparation of ZnNi/Al (Qi et al., 2018) and ZnCo/Al (Li et al., 2018) as a catalyst for rhodamine-
B degradation, while ZnCu/Al-TiO$_2$ (Seftel et al., 2015) in phenol degradation. The studies of coprecipitation technique that used calcination temperatures include the preparation of Zn/AlGa calcined 500°C (Amor et al., 2018), Zn/Al LDH calcined 500°C and 900°C (Abderrazek et al., 2016), Zn/Al-TiO$_2$ calcined 500°C (Hadnadjev-Kostic et al., 2017), Zn/Al LDH calcined 800°C (Huo et al., 2013) which were respectively used during the degradation of methylene blue, preparation of Zn/AlCe calcined 450-900°C (Zhu et al., 2016) and Zn/Al-TiO$_2$ calcined 500°C (Rudic et al., 2014) as catalysts in the degradation of rhodamine-B and the preparation of Zn/AlGa calcined at 550°C used for phenol degradation (Prince et al., 2015). From the studies of LDH-based composites that have been carried out, it appears that not a few have prepared LDH-type metal oxides LDH-TiO$_2$ and LDH-ZnO as photocatalysts for organic pollutant degradation.

LDH composites with TiO$_2$ or ZnO has advantages. LDH is a dispersant for TiO$_2$ owing to the surface hydroxyl group, thereby simultaneously generating and enhancing photocatalytic properties (Contreras-Ruiz et al., 2019). Meanwhile, ZnO is often considered an alternative to TiO$_2$ because it can absorb a wider spectrum of energy than TiO$_2$ (Amor et al., 2018). This study is intended to prepare Zn/Al-TiO$_2$ and Zn/Al-ZnO from the basic structure of Zn/Al LDH which has been synthesized, the catalysts used in the degradation of congo red (CR) dye. The calcination temperature of the composite preparation was used at 300°C, which was relatively low compared to the calcination temperature of several previous studies.

### 2. EXPERIMENTAL SECTION

#### 2.1 Materials and Instrumental

The starting chemicals used in this study were aluminum nitrate by Merck, zinc nitrate by Smart-Lab, titanium dioxide by Merck, zinc oxide by LOBA Chemie, sodium carbonate by Merck hydrogen chloride by Mallinkrodt LabGuard, and sodium hydroxide by Merck. Congo red was measured by UV-Vis Biobase BK-UV 1800 PC. Characterization of morphology materials was performed by SEM Quanta-650 Oxford Instrument, band gap energy provided by UV-DRS Jasco V-760, diffractogram by Rigaku XRD Miniflex-6000, and analysis of functional groups by FTIR Alpha Bruker.

#### 2.2 Synthesis of LDH

The synthesis of Zn/Al LDH was adjusted according to the modified procedure of Yan et al. (2016) namely by coprecipitation. 50 mL of zinc nitrate and aluminum nitrate each with a mole ratio of Zn/Al= 3 to form a total concentration of 1 M stirred for 2 hours. To this solution, 0.3 M alkaline and 0.25 M Na$_2$CO$_3$ were added, each 60 mL for pH 10 conditioning. The mixture was stirred for 10 hours at 80°C. The precipitate formed was filtered, washed and dried at a temperature of 110°C, then the precipitate was weighed.

#### 2.3 Preparation of Composite

The Zn/Al-TiO$_2$ photocatalyst was prepared according to the Zn/Al LDH synthesis procedure. A mixture of zinc nitrate and aluminum nitrate was added with alkaline and Na$_2$CO$_3$, after being stirred for 10 hours, it was impregnated with TiO$_2$ using a weight ratio of LDH/TiO$_2$ = 1. The best ratio of Zn/Al to TiO$_2$ is 1 (Djeda et al., 2020). The mixture was stirred for 3 hours and added 150 mL of 0.87 M base, then stirred again for 10 hours at a temperature of 70°C. The mixture produced a precipitate and was filtered, washed, and then calcined at a temperature of 300°C for 7 hours. The Zn/Al-ZnO photocatalyst was prepared according to the same process but the oxide used was zinc oxide.

#### 2.4 Photodegradation by Catalysts

Prior to the photodegradation process, the catalyst was contacted with a dye model in the form of a solution of 100 mg/L CR as much as 20 mL and shaken in the dark for 60 minutes. Variations in photodegradation conditions under UV 352 nm included variations in pH, catalyst weight, and time. The rate constant ($k$) was determined according to the equation of the integral result of the Langmuir-Hinshelwood formulation, the equation $\ln \left(C_0/C_t\right) = k \cdot t$ produced the value of $k$ as the slope.

#### 3. RESULT AND DISCUSSION

#### 3.1 Effect of Photodegradation Conditions

One of the things that have an effect on the photodegradation of dye is the pH condition of the media. Figure 1(a) shows the variation of pH to CR photodegradation. At pH 4, it appears that Zn/Al LDH produces the lowest C/C$_0$ or the highest CR photodegradation. CR is an anion so it can be more compatible with the positive charge of the photocatalyst under acidic conditions. Figure 1(a) also shows Zn/Al-TiO$_2$ has an optimum pH at pH 3 while Zn/Al-ZnO is optimum at pH 6. This study found that the optimum pH when using Zn/Al-ZnO was higher than using Zn/Al-TiO$_2$, this was found to occur because according to Elhalil et al. (2018) under acidic conditions ZnO is at risk of being decomposed by electron holes h$^+$ and also dissolution into Zn$^{2+}$.

Variations in the weight of the catalyst used can affect the photodegradation process. The more catalysts used within certain limits will provide optimum conditions. Figure 1(b) shows the effect of catalyst weight on CR photodegradation. It can be seen in the figure that the heavier the catalyst used, the better the photodegradation tendency. The decrease in the photodegradation performance of the Zn/Al-ZnO catalyst at 0.1 g according to Elhalil et al. (2018) and Yuliasari et al. (2022b) could be due to light being more difficult to penetrate into the media.
The varied time can show the optimum condition of photodegradation. Figure 2(a) shows the photodegradation of CR for each catalyst over time. The photodegradation results showed that after Zn/Al LDH was modified to LDH-metal oxide, it could improve the photodegradation performance. Zn/Al catalyzed degradation of CR for 120 minutes as much as 68.39%, Zn/Al-TiO$_2$ as much as 81.24%, and Zn/Al-ZnO as much as 71.09%.

Photodegradation progresses better with an increasing rate constant. Figure 2(b) shows the rate constant for CR degradation in the presence of catalysts. The rate constant was increased when using a composite catalyst compared to the pristine structure LDH. The rate constants ($k$) using Zn/Al-TiO$_2$ and Zn/Al-ZnO composites were 0.0158 and 0.0120, respectively, while using Zn/Al LDH was 0.0083. Several rate constants obtained by photodegradation studies of organic pollutants include catalyst Zn/Al-TiO$_2$ with $k$ 0.0060 (Contreras-Ruiz et al., 2019); catalyst Zn/AlCe with $k$ 0.0043 (Suárez-Quezada et al., 2016) and catalyst ZnMn/Al with $k$ 0.0013 (Morales-Mendoza et al., 2015).

3.2 Characterization of Fresh and Reuse Materials
Catalyst performance can be explained by surface conditions or surface morphology. The morphological conditions of the fresh catalysts can be seen from the 500 times magnification of SEM images in Figure 4 shows the surface of catalysts. SEM images of Zn/Al-TiO$_2$ appear to be more homogeneous than Zn/Al-ZnO.

LDH materials can be modified with metal oxides to form composites that have better structural strength (Djeda et al., 2020; Yuliasari et al., 2022a). Figure 3 shows that LDH-metal oxides, namely Zn/Al-TiO$_2$ and Zn/Al-ZnO are better in the reuse of the material up to 5 regeneration photodegradation cycles than Zn/Al LDH as the pristine structure.
that the TiO\textsubscript{2} peak of LDH-TiO\textsubscript{2} appears at 2\(\theta\) around 25\(^\circ\), 54\(^\circ\), 55\(^\circ\), and 69\(^\circ\). While Zn/Al-ZnO still shows the typical peak of LDH at an angle of 32\(^\circ\), 34\(^\circ\), 36\(^\circ\), 48\(^\circ\), 56\(^\circ\), 63\(^\circ\), and 68\(^\circ\) which is in line with Bhuvaneswari et al. (2019).

Analysis of the functional groups contained in the catalyst materials that have degraded the dye was carried out by FTIR. Figure 6(b) shows the FTIR spectra of catalyst materials that were used in the degradation of CR. Zhao et al. (2018) stated that the band around 3400 cm\(^{-1}\) is the stretching OH vibration of LDH and conveyed that the band around 1360 cm\(^{-1}\) is a vibration of CO\textsubscript{3}\textsuperscript{2-}. The band between 400 cm\(^{-1}\) to 800 cm\(^{-1}\) that appears according to Aoudjit et al. (2019) is a vibration of the bond between metal elements and oxygen. The 1620 cm\(^{-1}\) band is probably the vibration of the azo group or the primary amine group (Nandiyanto et al., 2019), where CR has these functional groups.

4. CONCLUSION
Modification of Zn/Al LDH into LDH-Metal oxides, namely Zn/Al-TiO\textsubscript{2} and Zn/Al-ZnO increases the photodegradation performance. The percentage degradation and rate constant of congo red (CR) using Zn/Al LDH increased when using Zn/Al-metal oxides. The regeneration cycle showed that Zn/Al-TiO\textsubscript{2} and Zn/Al-ZnO had better stability than Zn/Al-LDH. After the materials degraded CR in the fifth regeneration cycle, the XRD diffractogram still shows that the catalyst has a cationic layer structure for Zn/Al LDH catalyst and TiO\textsubscript{2} or ZnO. Similarly, the FTIR spectra after the materials were used up to the fifth regeneration cycle still show the presence of hydroxy groups, interlayer anions, and metal oxides. This characterization data can show that the stability of the catalyst material is well.

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