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Research Paper

Catalytic Oxidative Desulfurization of Dibenzothiophene by Composites Based Ni/Al-Oxide

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Abstract

In the present study, composite layer double hydroxide-metal oxide (Ni/Al-TiO_2 and Ni/Al-ZnO) was successfully prepared and used as catalyst of oxidative desulfurization of dibenzothiophene. Characterization of catalyst was used XRD, FTIR, and SEM-EDS. The structure of Ni/Al-LDH, TiO_2, and ZnO in composite Ni/Al-TiO_2 and Ni/Al-ZnO was consistent, which also indicated that the preparation of composite did not change the form of precursors. FTIR spectra of Ni/Al-TiO_2 and Ni/Al-ZnO absorption band at 3398, 1639, 1339, 832, 731, and 682 cm^{-1}. The catalysts have an irregular structure, TiO_2 and ZnO adhere to the surface of Ni/Al-LDH. The percent mass of Ti and Zn on the composite at 29.3% and 18.2%, respectively. The acidity of Ni/Al-LDH increased after being composited with TiO_2 and ZnO. The optimum reaction time, dosage catalyst, and temperature were 30 min, 0.25 g, and 50°C, respectively, and n-hexane as a solvent. The percentage conversion of dibenzothiophene on Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO were 99.44%, 91.92%, 95.36%, 99.88%, and 99.90%, respectively. The catalysts are heterogeneous system and the advantage is that can be used for reusability. After 3 times catalytic reactions, the conversion of dibenzothiophene still retains more than 80%, even Ni/Al-TiO_2 and Ni/Al-ZnO composites still 97.79% and 98.99%, respectively.

Keywords

Desulfurization, Dibenzothiophene, Layered Double Hydroxide, Composite

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1. INTRODUCTION

Fuel oil is an energy source that has a vital role in the global economy (Zeng et al., 2017). The fuel is oil from exploration on the earth, known as petroleum. Although the availability of petroleum continues to run low, its supply is still sufficient for the needs of human life on earth. However, the intensive use of petroleum, especially in transportation, causes gas emissions to be produced, namely SO_x (Mahmoudi et al., 2021). SO_x significantly contributes to air pollution, acid rain, and damage to the gas emission section of vehicles (Abedini et al., 2021; Kang et al., 2018; Mousavi-Kamazani et al., 2020).

The specifications issued by Japan and European Commission show that the sulfur content in the oil is constantly changing and revising due to the effects caused when the sulfur content is found in large quantities in fuel oil. In 2005, the sulfur content in fuel oil allowed in Europe was 10 ppm (Ren et al., 2016). The same thing happened in America, which revised the permissible sulfur content in fuel oil to 15 ppm in 2006 (Bazyari et al., 2016). The current trend shows that the sulfur content in fuel oil is a maximum of 10 ppm (Luna et al., 2022). Sulfur compounds in fuel oil are organosulfur such as dibenzothiophene (DBT) and derivatives (Mujahid et al., 2020; Ye et al., 2020). The chemical structure of dibenzothiophene is shown in Figure 1.

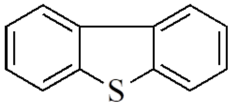

Figure 1. Chemical Structure of Dibenzothiophene

Reducing sulfur content in dibenzothiophene is known as the desulfurization process using the catalytic principle of oxidation. The success of the desulfurization process is highly dependent on the catalyst used, considering the process applied in catalytic oxidation. Malani et al. (2021) used microorganisms in the desulfurization process known as biodesulfurization. However, this method has difficulties in controlling the use of microorganisms and in the treatment of microorganisms, so chemical methods with the principle of catalytic oxidation continue to be developed until now. Several desulfurization technologies for dibenzothiophene compounds have been carried out such as photocatalytic desulfurization (Mgidlana et al., 2021), adsorptive desulfurization (Subhan et al., 2019), and oxidative desulfurization (ODS) (Li et al., 2020). In addition, in petroleum which is explored together with gas containing sulfur, the desulfurization process is carried out using a separator column and the extractive desulfurization process (EDS) (Rezaee et al., 2021). Until now, the desulfurization process is still being researched to find effective materials to convert sulfur by chemical processes, especially with the principle of catalytic oxidation reactions using synthesized catalysts. Catalysts have been reported for desulfurization of DBT, including montmorillonite (Kang et al., 2018), Fe promoted Ni/Co-Mo/Al_2O_3 (Muhammad et al., 2018), silica (Teimouri et al., 2018), and layered double hydroxide (Masoumi and Hosseini, 2020; Wu et al., 2018).

The potential catalyst to be used is Layered Double Hydroxide (LDH). LDH can be made easily, with low cost, and has a high-efficiency level (Taher et al., 2021). LDH is derived from the mineral clay brucite, whose general formula is $Mg(OH)_2$. In the catalytic process, LDH has been used for water remediation (Karim et al., 2022), n-heptane hydroconversion (Zhu et al., 2019), and biodiesel production (Gabriel et al., 2021). LDH is interested in catalysis due to its large surface area and homogeneous distribution of various essential components (Zhu et al., 2019). The disadvantage of LDH is easily exfoliated, so the reusability of LDH is less attractive. Therefore, LDH is composited with metal oxide. LDH is easily made into composites with metal oxides by calcining at high temperatures to remove organic pollutants (Dang et al., 2021).

In the experiment part, Ni/Al-TiO_2 and Ni/Al-ZnO were prepared as catalysts and DBT was the sulfur compound. Characterization of catalysts used XRD, FTIR, and SEM-EDS to know the successful preparation of the catalysts. The process of optimizing the oxidative desulfurization of DBT was carried out with variations in time, UV-Vis spectrum, the dosage of catalyst, temperature, solvent (n-pentane, n-hexane, and n-heptane), acidity test, heterogeneous test, and reusability.

2. EXPERIMENTAL SECTION

2.1 Chemicals and Instrumentation

Dibenzothiophene (DBT) was obtained from Sigma-Aldrich and directly used as received. Other chemicals such as hydrogen peroxide (H_2O_2), acetonitrile (CH_3CN), pyridine (C_5H_5N), n-pentane (C_5H_{12}), n-hexane (C_6H_{14}), n-heptane (C_7H_{16}), nickel (II) nitrate hexahydrate ($Ni(NO_3)_2.6H_2O$), aluminum nitrate nonahydrate ($Al(NO_3)_2.9H_2O$), sodium carbonate (Na_2CO_3), and sodium hydroxide (NaOH), titanium(IV) oxide (TiO_2), and zinc(II) oxide (ZnO) were also directly used without further purification. Water was supplied from the Research Center of Inorganic Materials and Complexes FMIPA Universitas Sriwijaya after several cycles of water purification using Purite®. Instrumentation such as X-Ray Diffractometer (XRD) type Rigaku Miniflex-6000, EMC-18PC-UV Spectrophotometer, Fourier Transfer Infra-Red (FTIR) type Shimadzu Prestige-21, and Scanning Electron Microscope Energy Dispersive Spectrometer (SEM-EDS) Quanta 650.

2.2 Synthesis and Preparation of Catalyst and Characterization

Synthesis of Ni/Al-LDH was conducted according to Lesbani et al. (2021) as follows: 0.75 M ($Ni(NO_3)_2.6H_2O$) and 0.25 M ($Al(NO_3)_2.9H_2O$) dissolved in 100 mL of distilled water, stirred for 2 h. Then slowly added the mixture of NaOH and Na_2CO_3 (ratio 2:1) to pH 10. The mixture was stirred for 17 h at 70°C, then filtered and dried.

Preparation Ni/Al-Oxide was conducted 0.75 M ($Ni(NO_3)_2.6H_2O$) and 0.25 M ($Al(NO_3)_2.9H_2O$) dissolved in 100 mL distilled water, stirred for 2 h. Then slowly added the mixture of NaOH and Na_2CO_3 (ratio 2:1) to pH 10. The mixture was stirred for 17 h at 70°C then added TiO_2/ZnO (ratio 1:1), shaken for 3 h. The mixture was added 150 mL 0.37 M NaOH, shaken for 17 h at 70°C, filtered, dried, and then calcinated at 300°C for 7 h.

2.3 Oxidative Desulfurization of Dibenzothiophene

Dibenzothiophene with the concentration of 500 ppm was prepared in n-hexane and transferred to a two-pronged catalytic reaction flask. The flask is connected to a condenser to prevent evaporation of n-hexane. 0.25 g Catalysts (Ni/Al-Oxide) followed by adding 1 mL of 30% hydrogen peroxide. The reaction was observed per 10 minutes time interval through extraction with acetonitrile followed by measuring using a UV-Visible spectrophotometer at 235 nm. The percentage conversion of DBT followed the equation:

$$\%\text{conversion of DBT} = \frac{(C_{0,\text{DBT}} - C_{\text{f,DBT}})}{C_{0,\text{DBT}}} \times 100$$

Where, $C_{0,DBT}$ and $C_{f,DBT}$ are the initial and final concentrations of DBT, respectively.

The process of optimizing the oxidative desulfurization of DBT was carried out with variations in time (10-60 min), UV-Vis spectrum (220-250 nm), the dosage of catalyst (0.05-1 g), temperature (30-50°C), solvent (n-pentane, n-hexane, and n-heptane), acidity test, heterogeneous test, and reusability. Reusability of catalyst is carried out by centrifugation of the reaction mixture after 1 h to recover the Ni/Al-oxide catalyst. The catalyst was washed with n-hexane several times, dried, and reused in the desulfurization of DBT.

3. RESULTS AND DISCUSSION

XRD powder diffraction of Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO is shown in Figure 2. XRD peaks of Ni/Al-LDH were analyzed from the JCPDS No. 15-0087 (Chen et al., 2022). Ni/Al-LDH peaks were detected at 2θ= 11.48°(003), 23.30°(002), 35.03°(311), and 61.40°(013) (see

Figure 2a). The diffraction peaks at 2θ= 11.48°(003) and 61.40°(013) indicate crystal planes of Ni/Al-LDH (Xie et al., 2021). Figure 2b shown diffraction of TiO_2 at 2θ= 25.59°(101), 37.09°(004), 48.16°(200), 54.03°(211), 55.26°(105), and 62.29° (204). Figure 2c shown diffraction of ZnO at 2θ= 31.75°(100), 34.41°(002), 36.24°(101), 47.52°(002), 56.56°(110), and 62.84° (103). TiO_2 and ZnO followed JCPDS No. 73-1764 and 36-1451, respectively (Basnet et al., 2019). The structure of Ni/Al-LDH, TiO_2, and ZnO in composite Ni/Al-TiO_2 and Ni/Al-ZnO was consistent, which also indicated that the preparation of composite did not change the form of precursors (see Figures 2d and 2e).

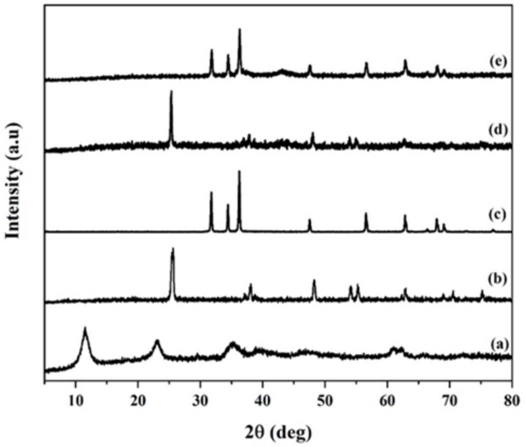

Figure 2. XRD Powder Diffraction of Ni/Al-LDH (a), TiO_2 (b), ZnO (c), Ni/Al-TiO_2 (d), and Ni/Al-ZnO (e)

FTIR spectra of Ni/Al-TiO_2 and Ni/Al-ZnO absorption band at 3398, 1639, 1339, 832, 731, and 682 cm^{-1} (see Figures 3d and 3e). An absorption band at 3398 cm^{-1} was the O-H stretching vibrations in the hydroxyl layer (Normah et al., 2021; Palapa et al., 2021). 1639 and 1339 cm^{-1} as H-O-H and NO_3^- stretching from Ni/Al-LDH (Lv et al., 2022). The peaks at 832, 731, and 682 cm^{-1} can be assigned metal oxide in Ni/Al-LDH, TiO_2, and ZnO (Intachai et al., 2021).

Table 1. EDS of Catalysts

Element	Ni/Al-LDH (%wt)	Ni/Al-TiO_2 (%wt)	Ni/Al-ZnO (%wt)
Ni	33.9	20.7	15.6
Al	5.2	3.1	13.9
Ti	-	29.3	-
Zn	-	-	18.2
O	43.2	34.8	27.2

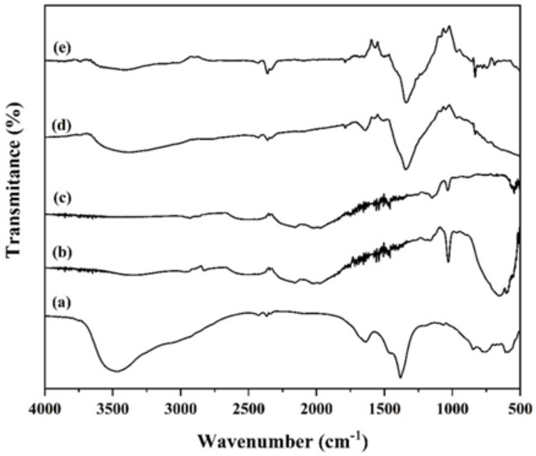

Figure 3. FTIR spectrum of Ni/Al LDH (a), TiO_2 (b), ZnO (c), Ni/Al-TiO_2 (d), and Ni/Al-ZnO (e)

Figure 4 shows the SEM Image and EDS of Ni/Al-LDH, Ni/Al-TiO_2, and Ni/Al-ZnO. SEM image investigated the morphology of catalysts at 2500 times magnification. The catalysts have an irregular structure, TiO_2 and ZnO adhere to the surface of Ni/Al-LDH. EDS analysis in Table 1 shows the Ni, Al, Ti, Zn, and O atom percentages. Ti and Zn atoms appear after composited into Ni/Al-TiO_2 and Ni/Al-ZnO. The percent mass of Ti and Zn at 29.3% and 18.2%, respectively. Thus, preparation of Ni/Al-TiO_2 and Ni/Al-ZnO has been a success.

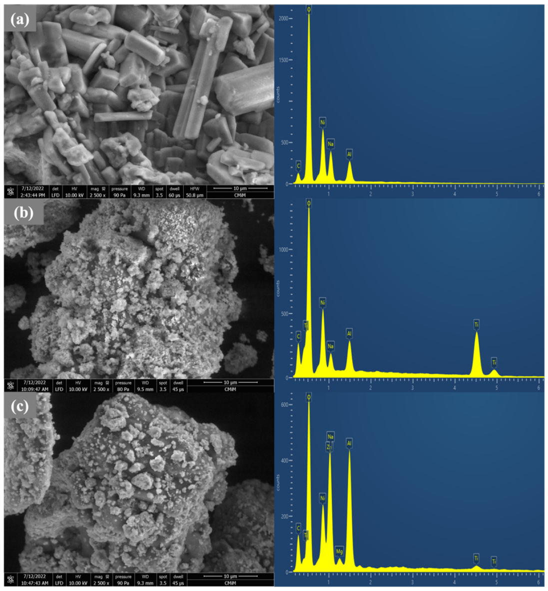

Figure 4. SEM Image and EDS of Ni/Al-LDH (a), Ni/Al-TiO_2 (b), and Ni/Al-ZnO (c)

The acidity test was carried out using the gravimetric method with pyridine as the adsorbate base. Pyridine has a large size causing bonding to occur only on the surface. The results of the determination of the acid site for each catalyst are shown in Table 2. Ni/Al-LDH increased after being composited with TiO_2 and ZnO. the acidity of Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO were 0.148, 0.298, 0.782, 0.714, 0.184

mmol/gram, respectively. The increase in the acidity of Ni/Al-LDH, because it has been reduced, will lack electrons so that it has a higher ability to absorb pyridine. Acid sites of catalysts are polyacid to convert DBT into DBT-sulfone (Trisunaryanti et al., 2021).

Table 2. Acidity of Catalyst

Catalyst	Acidity (mmol/g)
Ni/Al-LDH	0.148
TiO_2	0.298
ZnO	0.782
Ni/Al-TiO_2	0.714
Ni/Al-ZnO	0.184

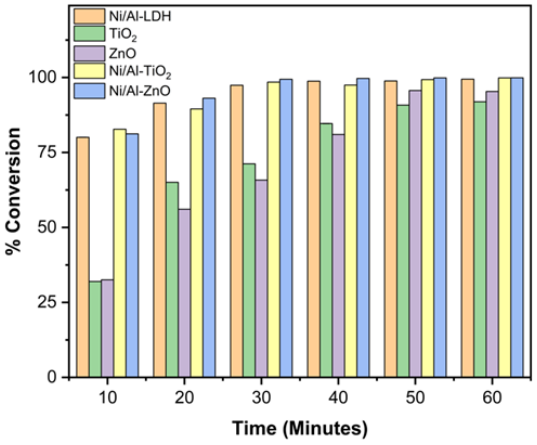

Figure 5. Profile of Desulfurization by Time Over Composite Catalysts

The catalytic oxidative desulfurization of DBT is strongly influenced by time. The effect of the desulfurization time of DBT is displayed in Figure 5. The catalytic data showed a long reaction time was directly proportional to the high %conversion of DBT (Muhammad et al., 2018). The optimum reaction time was 30 min and the percentage conversion of DBT on Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO were 99.44%, 91.92%, 95.36%, 99.88%, and 99.90%, respectively. The composite increases the ability of the catalyst in the desulfurization of DBT. In this study, Ni/Al-ZnO is better than Ni/Al-TiO_2 in conversion of DBT. The DBT that appeared in the acetonitrile phase was rapidly oxidized and converted to DBT-sulfone after extraction.

UV-Vis spectrum of oxidative desulfurization dibenzothiophene by composite catalysts is shown in Figure 6. UV-vis spectrum used wavelength 220-250 nm. The Absorbance of DBT at 235 nm decreased with increasing desulfurization time. The decrease in absorbance gradually indicates the concentration of DBT is decreasing. At 30 min, %conversion of DBT >90% with the most significant reduction indicated by Ni/Al-ZnO catalyst.

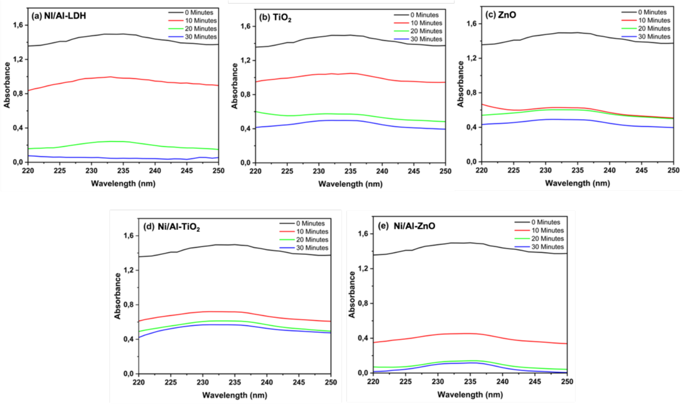

Figure 6. UV-Vis Spectrum of Oxidative Desulfurization Dibenzothiophene by Composite Catalysts

The effect of the dosage of catalyst is presented in Figure 7. Generally, a higher catalyst dosage will provide more opportunities for interaction between the active site of the catalyst and the DBT (Ye et al., 2020). However, in contrast to this study, the optimum dosage for all catalysts was 0.25 g. A higher catalyst dosage will increase the active site of the catalyst while competing with oxidant molecules (Subhan et al., 2019). Therefore, increasing the catalyst dosage can reduce the %conversion of DBT. Percentage of Desulfurization order of DBT: Ni/Al-ZnO > Ni/Al-TiO_2 > Ni/Al-LDH > ZnO > TiO_2.

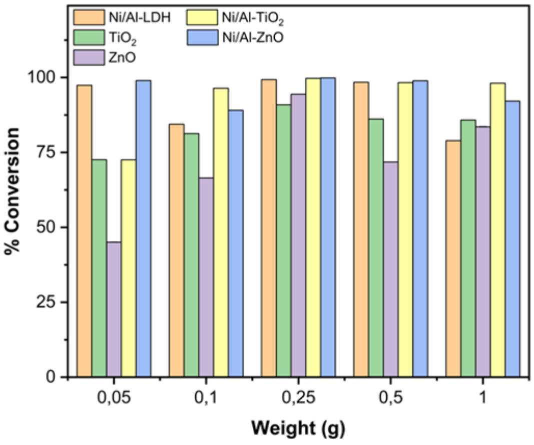

Figure 7. Effect of Catalyst Dosage on Desulfurization of Dibenzothiophene

Figure 8 shows the effect of temperature catalytic oxidative desulfurization of DBT. The catalytic data shows that high temperature is directly proportional to the high %conversion of DBT. Desulfurization of DBT using H_2O_2 is better at high temperatures by converting DBT to DBT-sulfone (Fraile et al., 2016; Lesbani et al., 2015). Desulfurization of DBT at 50°C is better than at 30°C and 40°C. Percentage of Desulfurization order of DBT: Ni/Al-ZnO > Ni/Al-TiO_2 > Ni/Al-LDH > ZnO > TiO_2. The fuel oil is operated at 60-70°C to overcome different oxidant dissociation energies. Therefore, this study is superior because the temperature is lower with the same efficiency.

The effect of solvent was carried out to determine the best solvent in the oxidative desulfurization of DBT. n-pentane, n-hexane, and n-heptane were used as DBT solvents. The solvent effect shows that n-hexane is better at desulfurizing DBT than n-pentane and n-heptane. The results are compiled in Figure 9 showing the desulfurization of DBT on Ni/Al-ZnO > Ni/Al-TiO_2 > Ni/Al-LDH > ZnO > TiO_2.

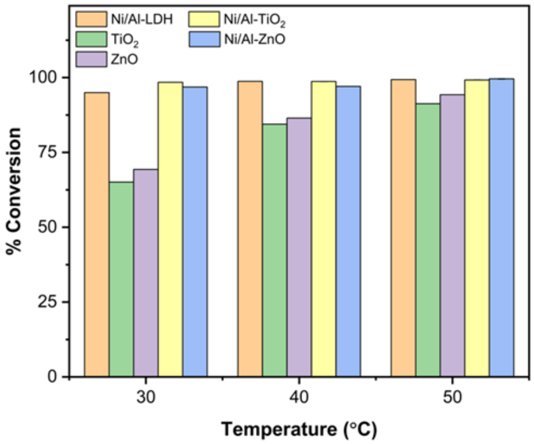

Figure 8. Effect of Various Temperatures on Desulfurization of Dibenzothiophene Using Composite Catalysts

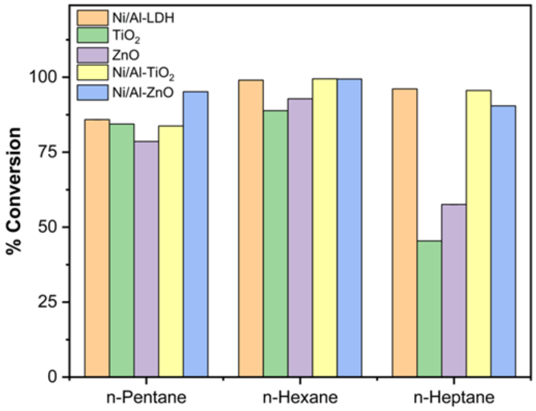

Figure 9. Effect of Solvent on Desulfurization of Dibenzothiophene by Composite Catalysts

Figure 10 shows the heterogeneous test of catalyst. Heterogeneous test the catalyst to determine whether the catalyst is homogeneous or heterogeneous. Homogeneous catalysts are soluble in the reactants/products of the reaction, while heterogeneous catalysts are insoluble. The Heterogeneous test was carried out by desulfurization of DBT at 50°C for 10 min, the catalyst and DBT solution were separated. DBT solution continued with desulfurization process for 20-30 min. The unchanged DBT concentration indicates that Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO are truly heterogeneous system. The advantage of heterogeneous catalysts is that can be used for reusability (Vallés-García et al., 2020).

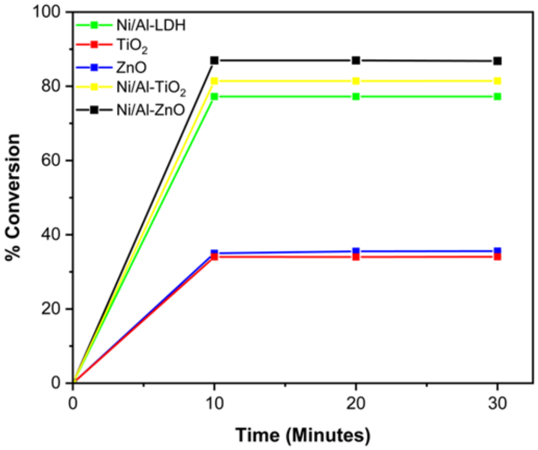

Figure 10. Heterogeneous Test of Catalyst

Reusability of catalysts is very influential for the industry to save operating costs (Song et al., 2021). Reusability is suitable for Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO catalysts because they are heterogeneous. Figure 11 shows the good reusability of catalyst. After 3 times catalytic reactions, the conversion of DBT still retains more than 80%, even Ni/Al-TiO_2 and Ni/Al-ZnO composites still 97.79% and 98.99%, respectively. FTIR analysis was carried out to investigate changes in the structure of the catalyst after the reusability process. The new peak appearing at 1200 cm^{-1} is sulfone. Figure 12 shows that the FTIR Ni/Al-TiO_2 has undergone structural changes, while Ni/Al-ZnO is stable in reusability process.

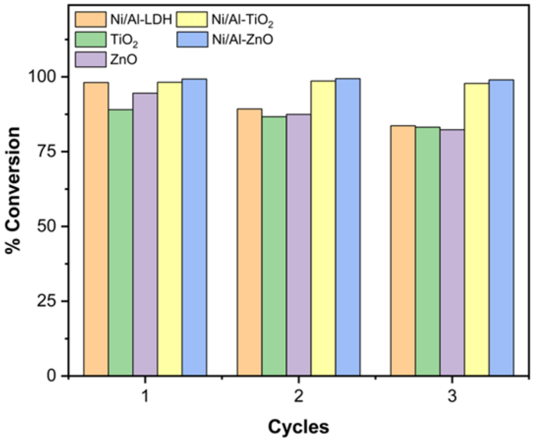

Figure 11. Reusability of Catalyst

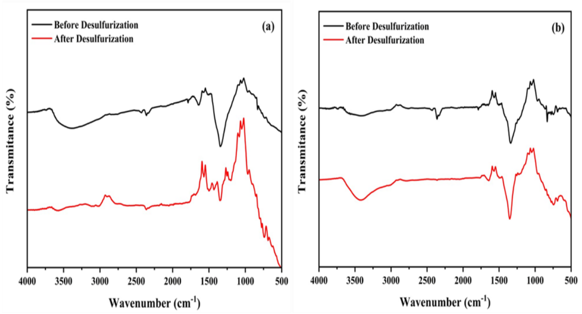

Figure 12. FTIR Spectrum of Composites Ni/Al-TiO_2 (a), Ni/Al-ZnO (b) Before and After Desulfurization of Third Cycle

4. CONCLUSIONS

In the present study, composite layer double hydroxide-metal oxide (Ni/Al-TiO_2 and Ni/Al-ZnO) was successfully prepared and used as catalyst of oxidative desulfurization of dibenzothiophene. The acidity of Ni/Al-LDH increased after being composited with TiO_2 and ZnO. The optimum reaction time, dosage catalyst, and temperature were 30 min, 0.25 g, and 50°C, respectively, and n-hexane as a solvent. The percentage conversion of dibenzothiophene on Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO were 99.44%, 91.92%, 95.36%, 99.88%, and 99.90%, respectively. The catalysts are heterogeneous system and the advantage is that can be used for reusability. After 3 times catalytic reactions, the conversion of dibenzothiophene still retains more than 80%, even Ni/Al-TiO_2 and Ni/Al-ZnO composites still 97.79% and 98.99%, respectively.

5. ACKNOWLEDGEMENT

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