Science and Technology Indonesia
e-ISSN:2580-4391 *p*-ISSN:2580-4405
Vol. 8, No. 3, July 2023

Research Paper

Catalytic Oxidative Desulfurization of 4-Methyldibenzothiophene by Ni/Al Modified Titanium Dioxide and Zinc Oxide

Risfidian Mohadi[1,2]*, Neza Rahayu Palapa[1], Sahrul Wibiyan[1], Mardiyanto[3], Rohmatullaili[1], Erni Salasia Fitri[1], Aldes Lesbani[1,2]

[1]*Research Center of Inorganic Materials and Coordination Complexes, Universitas Sriwijaya, Palembang, 30662, Indonesia*
[2]*Master of Materials Science, Graduate Program, Universitas Sriwijaya, Palembang, 30662, Indonesia*
[3]*Department of Pharmacy, Faculty of Mathematics and Natural Sciences Universitas Sriwijaya, Ogan Ilir, 30662, Indonesia*
***Corresponding author**: risfidian.mohadi@unsri.ac.id

Abstract
In this study, composite layer double hydroxide-metal oxide catalysts (Ni/Al-TiO_2 and Ni/Al-ZnO) were successfully prepared and utilized for the oxidative desulfurization of 4-methyldibenzothiophene. The catalysts were characterized using XRD, FTIR, and SEM-EDS analysis. The diffraction patterns confirmed the successful synthesis of the composite materials, while the FTIR spectra exhibited absorption bands at specific wavenumbers 3480 cm^{-1}, 1630 cm^{-1}, 1376 cm^{-1}, 830 cm^{-1}, 775 cm^{-1}, and 683 cm^{-1}. The surface morphology of the catalysts showed variations, with Ni/Al-LDH having a smooth particle surface and long particle shape, while Ni/Al-ZnO and Ni/Al-TiO_2 formed grain-like or aggregated structures. EDS analysis confirmed the presence of Ni/Al, Ti, and Zn elements in the composites, with Ti and Zn accounting for 29.3% and 10.7% of the composite mass, respectively. The optimized reaction conditions included a reaction time of 40 minutes, catalyst dosage of 0.25 g, temperature of 50°C, and the use of n-hexane as the solvent. The catalysts demonstrated a heterogeneous nature and proved to be reusable, as the conversion of 4-methyldibenzothiophene remained above 90% even after three catalytic reactions, with Ni/Al-TiO_2 and Ni/Al-ZnO composites maintaining conversion rates of 99.36% and 99.32%, respectively.

Keywords
Desulfurization, 4-Methyldibenzothiophene, Layered Double Hydroxide, Composite

Received: 10 March 2023, Accepted: 15 June 2023
https://doi.org/10.26554/sti.2023.8.3.414-421

1. INTRODUCTION

Vehicles play a crucial role in our daily lives by providing a convenient mode of transportation. However, the use of vehicles can lead to energy depletion and result in air pollution (Fayad et al., 2022). Air pollution arises from the vehicle fuels combustion, including sulfur dioxide (SO_2) (Mahmoudi et al., 2021). Consequently, there has been significant global interest in the process of deep desulfurization of fuels. The key objective is to produce fuels with extremely low levels of sulfur by eliminating sulfur-containing compounds such as benzothiophene (BT), dibenzothiophene (DBT), and their derivatives such as 4-methyldibenzothiophene (Mjalli et al., 2014). In order to comply with strict environmental regulations In Europe, in the year 2005, the permissible sulfur content in fuel oil was 10 parts per million (ppm) it is imperative to decrease the sulfur content of fuels to levels below 10 parts per million (ppm) (Ren et al., 2016; Wang et al., 2013; Abro et al., 2016). Figure 1 depicts the Oxidative Desulfurization of 4-methyldibenzothiophene.

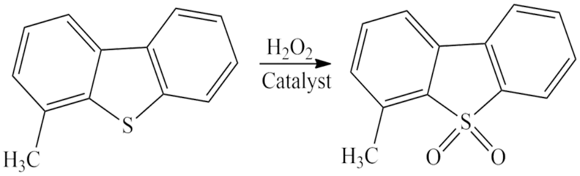

Figure 1. Oxidative Desulfurization of 4-methyldibenzothiophene

Adsorptive desulfurization, extractive desulfurization, and oxidative desulfurization are among the alternative processes used for deep desulfurization of liquid fuels (Malani et al., 2021). Adsorptive desulfurization, which aims to reduce the sulfur compound content, faces challenges in terms of sulfur selectivity. On the other hand, hydrodesulfurization, another method for desulfurization, necessitates high temperatures and pressures (Abedini et al., 2021). Oxidative desulfurization is highly effective in converting 4-methyldibenzothiophene without the need for high temperatures (Ahmad et al., 2023).

Oxidative desulfurization (ODS) has been extensively researched as a prominent approach for decreasing sulfur compounds (Raeisi et al., 2022). ODS offers several advantages, including its ability to operate under mild conditions and its efficiency in converting sulfur compounds. In the process of ODS, the catalyst plays a crucial role in activating oxidants, thereby significantly influencing the overall outcome (Deng et al., 2022). Therefore, the need for a highly efficient catalyst arises (Xun et al., 2023). To enhance the reaction rate in the oxidative desulfurization process, the presence of a catalyst is necessary (Luna et al., 2022). Research on the desulfurization process is ongoing as scientists continue to explore effective materials for converting sulfur through chemical methods, particularly by employing catalytic oxidation reactions with synthesized catalysts. Several catalysts have been documented for desulfurization, including polyoxometalate (Ahmad et al., 2022a), red mud (Liu et al., 2023), pentaethylenehexamine-phosphotungstic acid (Wu et al., 2023) and layered double hydroxide (Song et al., 2021). An ideal catalyst is characterized by its ability to be rapidly synthesized, produced at a low cost, and demonstrate high efficiency (Ahmad et al., 2022b).

Layered double hydroxides (LDHs) typically exhibit a layered structure consisting of brucite-like layers. These layers contain both divalent metal ions and trivalent cations, which are uniformly dispersed at the atomic level. The substitution of divalent cations with trivalent cations in the LDH structure results in the formation of positively charged layers (Wu et al., 2018). The type of catalyst is greatly influenced by how it can be separated and regenerated. One promising candidate is the layered double hydroxide (LDH) catalyst. LDH offers several advantages as it can be produced quickly, at a low cost, and with high efficiency (Palapa et al., 2021). One drawback of LDH is its susceptibility to easy exfoliation, which affects its reusability and reduces its attractiveness as a catalyst. To overcome this limitation, LDH is often combined with metal oxides. By subjecting LDH to high-temperature calcination, organic pollutants can be removed, facilitating the). With these advantages and disadvantages, the modification of Ni/Al-LDH with zinc oxide and titanium oxide was carried out.

During the experimental phase, catalysts such as Ni/Al-TiO_2 and Ni/Al-ZnO were synthesized, with 4-methyldibenzothiophene selected as the sulfur compound. To ensure the successful preparation of the catalysts, XRD, FTIR, and SEM-EDS analyses were conducted for characterization purposes. The optimization process of oxidative desulfurization of 4-methyldibenzothiophene involved exploring various parameters, including time, catalyst dosage, temperature, solvent selection (n-pentane, n-hexane, and n-heptane), conducting heterogeneous tests, and evaluating catalyst reusability.

2. EXPERIMENTAL SECTION

2.1 Chemical and Instrumental

Sigma-Aldrich provided 4-methyldibenzothiophene, which was used as received without any additional treatment. Additional chemicals, such as hydrogen peroxide, acetonitrile, pyridine, n-pentane, n-hexane, n-heptane, nickel (II) nitrate hexahydrate, aluminum nitrate nonahydrate, sodium carbonate, sodium hydroxide, titanium(IV) oxide, and zinc(II) oxide, were also utilized directly without the need for further purification.

The materials were characterized using several instruments, including the Rigaku Miniflex-6000 X-ray diffractometer, EMC-18PC-UV UV-Vis spectrophotometer, Shimadzu Prestige-21 Fourier transfer infrared spectrometer, and Quanta 650 scanning electron microscope. These instruments were employed to analyze and investigate the properties and structure of the materials in question.

2.2 Preparation of Ni/Al-Layered Double Hydroxide

To synthesize Ni/Al-LDH, the following procedure was followed: Initially, a solution was created by dissolving 0.75 M Nickel (II) nitrate hexahydrate and 0.25 M Aluminum nitrate in 100 mL of distilled water. The solution was then stirred for a period of two hours. Next, a mixture of Sodium Hydroxide and Sodium Carbonate in a 2:1 ratio was slowly added to the solution until the pH reached 10. After stirring the mixture for 17 hours at a temperature of 70°C, it was filtered and subsequently dried (Lesbani et al., 2021).

2.3 Preparation of Ni/Al Composite

The Ni/Al composite preparation was conducted as follows: 0.75 M zinc(II) nitrate hexahydrate and 0.25 M aluminum nitrate nonahydrate were dissolved in 100 mL of pure water, and the mixture was stirred for 10 minutes. Then, 2 M sodium hydroxide was slowly added to adjust the pH to 8. The mixture was stirred for 4 hours at a temperature of 70°C. Next, 3.7 g of TiO_2/ZnO (with a Ni/Al and metal oxide ratio of 1:1) was added, and the mixture was shaken for 3 hours. After that, 150 mL of 0.37 M sodium hydroxide was added, and the mixture was shaken for an additional 4 hours at 70°C. The mixture was then filtered, dried, and calcinated at a temperature of 300°C (Yuliasari et al., 2022).

2.4 Oxidative Desulfurization Method

4-methyldibenzothiophene with a concentration of 500 ppm was prepared in n-hexane and transferred to a two-pronged catalytic reaction flask. The flask was connected to a condenser to prevent the evaporation of n-hexane. Next, 0.25 g of Ni/Al-Oxide catalysts was added, followed by the addition of 1 mL of 30% hydrogen peroxide. The reaction progress was observed at 10-minute intervals by extracting samples with acetonitrile and measuring them using a UV-Visible spectrophotometer at 235 nm. The percentage conversion of 4-methyldibenzothiophene was determined using the following Equation 1.

$$\%\text{Conversion of 4-Methyl(DBT)} = \frac{(C_0 - C_f)}{C_0} \times 100\% \quad (1)$$

In the context of the desulfurization process of 4-methyldibenzothiophene, C_0 represents the initial concentration of the compound, while C_f represents the final concentration of 4-methyldibenzothiophene after the desulfurization process has taken place. These concentration values are used to calculate the extent of sulfur conversion or the percentage of desulfurization achieved. The optimization process for the oxidative desulfurization of 4-methyldibenzothiophene involved varying several parameters. These parameters included time (ranging from 10 to 60 minutes), catalyst dosage (ranging from 0.05 to 1 gram), temperature (ranging from 30 to 50 degrees Celsius), solvent selection (n-pentane, n-hexane, and n-heptane), conducting heterogeneous tests, and assessing catalyst reusability. To evaluate the reusability of the catalyst, the reaction mixture was centrifuged after 1 hour to recover the Ni/Al-oxide catalyst. The catalyst was then washed with n-hexane multiple times, dried, and subsequently reused in the desulfurization of 4-methyldibenzothiophene.

2.5 Heterogeneity Test

During the heterogeneous test, the desulfurization process was performed on a 500 ppm solution of 4-methyldibenzothiophene in 30 mL of n-hexane. The reaction took place at a constant temperature of 323 K for a duration of 10 minutes. After the reaction, the catalyst and the 4-methyldibenzothiophene solution were separated. Following the initial reaction, the desulfurization process of the 4-methyldibenzothiophene solution continued for an additional 20 to 30 minutes, maintaining the same temperature of 323 K. Subsequent analyses were conducted to determine the extent of sulfur conversion of the 4-methyldibenzothiophene compound.

3. RESULT AND DISCUSSION

The surface morphology and EDS characterization shown in Figure 2. Surface morphology Ni/Al-LDH has a smooth particle surface and a long particle shape, while Ni/Al-ZnO and Ni/Al-TiO_2 have a surface morphology that forms grains or aggregates. EDS serves as a tool employed for identifying the constituent elements within a given sample. The analysis conducted using EDS on the Ni/Al-ZnO composite revealed the presence of various elements, such as Ni, Al, and Zn. Similarly, the EDS analysis performed on the Ni/Al-TiO_2 composite detected the presence of several elements, like Ni, Al, and TiO where the element Ti with 29,3 mass percentage appears after the composite of Ni/Al-LDH with TiO_2 and Zn with 10,7 mass percentage appears after the composite of Ni/Al-LDH with ZnO. This substantiates the successful creation of the composite material through the research conducted.

The FTIR spectrums of Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, Ni/Al-ZnO are shown in Figure 3. The peak at wave number 3480 cm^{-1} in Ni/Al-LDH(a), Ni/Al-TiO_2(b), and Ni/Al-ZnO(c) indicates the presence of O-H stretching vibrations in the hydroxyl layer (Priatna et al., 2023). In the Ni/Al-TiO_2 and NiAl-ZnO composites, the peak is lower because the O-H groups evaporate due to calcination at high temperatures. The peaks observed at wave numbers approximately 1630 cm^{-1} and 1376 cm^{-1} were determined to correspond to H-O-H bonding and NO_3^- originating from Ni/Al-LDH (Lv et al., 2022). The peaks observed at wave numbers around 830 cm^{-1}, 775 cm^{-1}, and 683 cm^{-1} were identified as indicative of metal oxide presence in Ni/Al-LDH, TiO_2, and ZnO, respectively (Intachai et al., 2021).

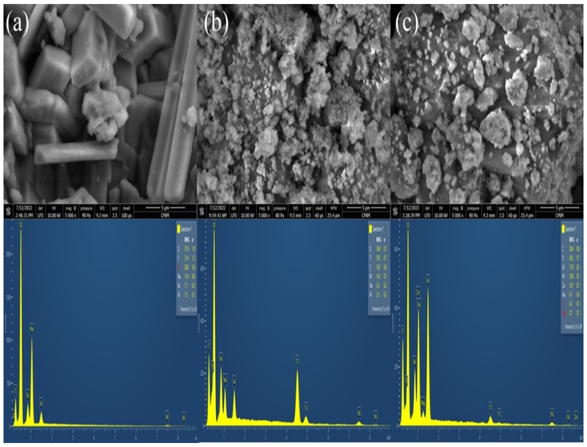

Figure 2. Surface Morphology and EDS Characterization of Ni/Al-LDH(a), Ni/Al-TiO_2(b), and Ni/Al-ZnO(c)

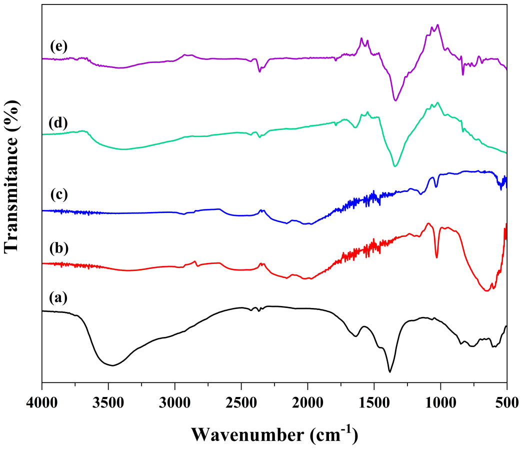

Figure 3. FTIR Spectrum of Ni/Al-LDH(a), TiO_2(b), ZnO(c), Ni/Al-TiO_2(d) and Ni/Al-ZnO(e)

XRD diffraction of Ni/Al-LDH, TiO_2 , ZnO, Ni/Al-TiO_2, Ni/Al-ZnO shown in Figure 4. The XRD patterns observed at angles 2θ = 11.52°, 22.96°, 35.22°, 39.34°, 45.99°, 60.98°, and 62.34° in Figure 4(a) show diffraction patterns that match the diffraction patterns reported by Madram and Atashbar, for Ni/Al-LDH (Madram and Zandi Atashbar, 2015). In Figure 4(b), the diffraction pattern of TiO_2 was observed at 2θ angles of 25.59° (101), 37.09° (004), 48.16° (200), 54.03° (211), 55.26° (105), and 62.29° (204). Additionally, in Figure 4(c), the diffraction pattern of ZnO was shown at 2θ angles of 31.75° (100), 34.41° (002), 36.24° (101), 47.52° (002), 56.56° (110),

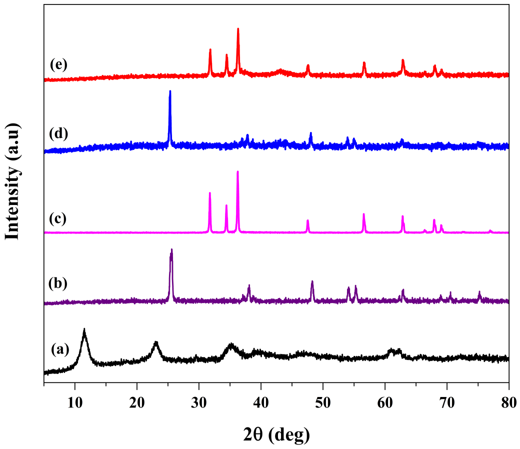

Figure 4. XRD Diffraction of Ni/Al-LDH(a), TiO_2(b), ZnO(c), Ni/Al-TiO_2(d) and Ni/Al-ZnO(e)

and 62.84° (103). These diffraction patterns of TiO_2 and ZnO corresponded to the JCPDS numbers 73-1764 and 36-1451, respectively (Basnet et al., 2019). The diffraction pattern of the base material and precursor material is observable in the synthesized composite, as evident from the comparison of the diffraction patterns in Figure 4(b) with Figure 4(d) and Figure 4(c) with Figure 4(e). This consistent diffraction pattern, without any noticeable changes, indicates the successful synthesis of the composite material.

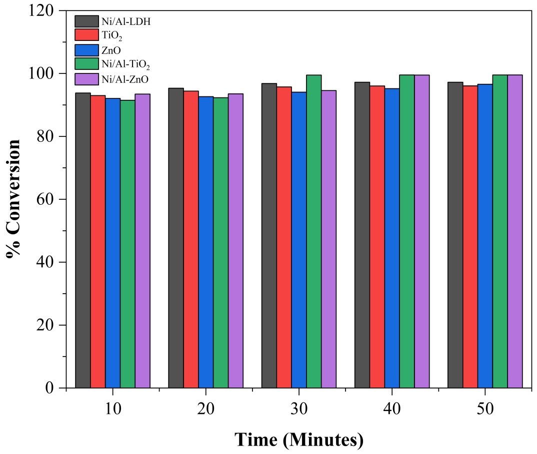

Figure 5. Effect of Reaction Time on Oxidative Desulfurization of 4-methyldibenzothiophene

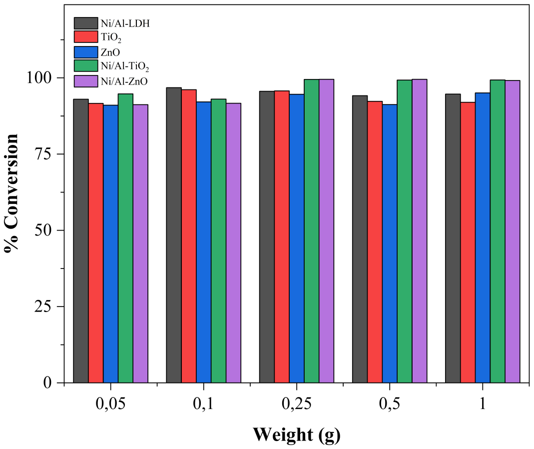

Figure 6. Effect of Catalyst Dosage on Oxidative Desulfurization of 4-methyldibenzothiophene

Figure 5. shows the effect of reaction time on the effectiveness of the catalyst to remove 4-methyldibenzothiophene. The effect of reaction time is one of the important factors. The more the reaction time increases, the greater the %conversion obtained, and when it reaches the optimum condition, the %conversion becomes linear (Zhou et al., 2021). The optimum response time was at 40 minutes, where at the next time the change no longer changed drastically and became linear. The optimum conversions at Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO were 97.22%, 96.03%, 95.19%, 99.53%, and 99.50%, respectively. Therefore, increasing the reaction time can reduce the %conversion of 4-methyldibenzothiophene. The order of the optimum conditions of %conversion obtained from the largest to the smallest is Ni/Al-ZnO>Ni/Al-TiO_2>Ni-/Al-LDH>ZnO>TiO_2.

The effect of catalyst dosage on the %conversion of 4-methyldibenzothiophen can be seen in Figure 6. Higher catalyst dosage offers increased opportunities for interaction between the catalyst active site and the adsorbate in this study is 4-methyldibenzothiophene (Kumar et al., 2012). This study showed that at a dose of 0.1 g Ni,Al-LDH and TiO_2 resulted in the highest %conversion with percentages of 96.77% and 96.1%. In ZnO, Ni/Al-TiO_2 and Ni/Al-ZnO, the best dose was 0.25 g with %conversion obtained by 94.61%, 99.48% and 99.53% . This difference indicates that higher catalyst doses can cause more intensive competition between the active sites of the catalyst and oxidant molecules (Subhan et al., 2019).

A study was carried out to assess the impact of various solvents on the selection of the optimal solvent for the oxidative desulfurization of 4-methyldibenzothiophene can be seen in Figure 7. In particular, n-pentane, n-hexane, and n-heptane were used as solvents for the experiment. The analysis of solvent effects revealed that n-hexane demonstrates superior performance when compared to both n-pentane and n-heptane for the oxidative desulfurization of 4-methyldibenzothiophene In terms of percentage conversion, the results from highest to lowest are as follows: Ni/Al-ZnO (99.50%), Ni/Al-TiO_2 (99.47%), Ni/Al-LDH (96.84%), TiO_2 (95.74%), and ZnO (93.9%).

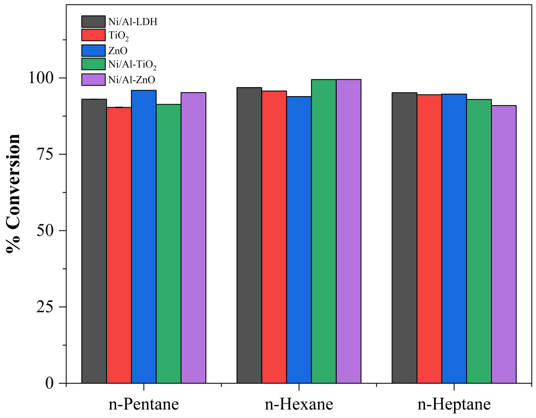

Figure 7. Effect of Solvent on Oxidative Desulfurization of 4-methyldibenzothiophene

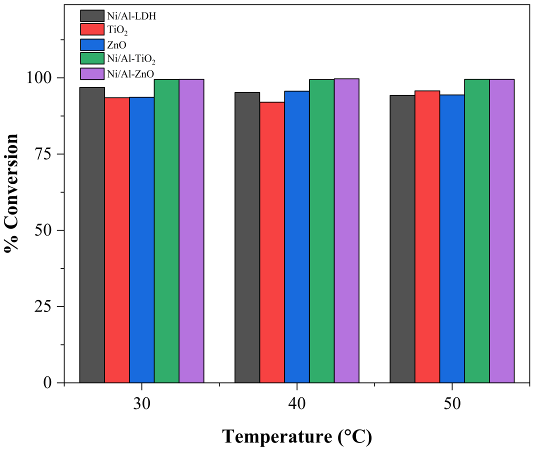

Figure 8. Effect of Temperature on Oxidative Desulfurization of 4-methyldibenzothiophene

Based on the data presented in Figure 8, the optimal temperature for the desulfurization of 4-methyldibenzothiophene is determined to be 50°C. The oxidative desulfurization process was conducted within a temperature range of 30°C to 50°C. It was observed that higher temperatures promote a more rapid rate of molecular diffusion and enhance the interaction between 4-methyldibenzothiophene and catalysts, resulting in a more efficient desulfurization process (Cao et al., 2020). The catalysts, including Ni/Al-TiO_2, Ni/Al-ZnO, TiO_2, ZnO, and Ni/Al-LDH, exhibited varying levels of conversion rates. Among them, Ni/Al-TiO_2 and Ni/Al-ZnO demonstrated the highest conversion rates, reaching percentages of 99.51% and 99.50% respectively. TiO_2 and ZnO followed with conversion rates of 95.70% and 94.41% respectively, while Ni/Al-LDH exhibited a slightly lower conversion rate of 94.27%.

The heterogeneity test is used to differentiate between homogeneous and heterogeneous catalysts. Homogeneous catalysts dissolve in the reactants or products of the reaction, while heterogeneous catalysts remain insoluble. In this study, the heterogeneity test was performed at a temperature of 50°C for 10 minutes. After the test, the catalyst and 4-methyldibenzothiophene solution were separated. The reaction process was then allowed to continue for an additional 20-30 minutes without the presence of any catalyst. The reaction exhibited a significant acceleration before the catalyst and 4-methyldibenzothiophene were separated at the 10-minute mark. Subsequently, the reaction proceeded in a linear fashion without any further changes. Indeed, the presence of an unchanged 4-methyldibenzothiophene concentration confirms that the system is heterogeneous. Figure 9 showcases the performance of catalysts Ni/Al-LDH, TiO_2, ZnO, Ni/Al-TiO_2, and Ni/Al-ZnO. Heterogeneous catalysts provide several advantages. One advantage is the ease of separating the catalyst from the product, as mentioned in reference (Houda et al., 2018). This characteristic simplifies the purification process and aids in the recovery of the desired product. Another advantage is the potential for reusability of heterogeneous catalysts, as discussed in reference (Polikarpova et al., 2020). This property allows the catalyst to be utilized multiple times, resulting in cost savings and environmental benefits.

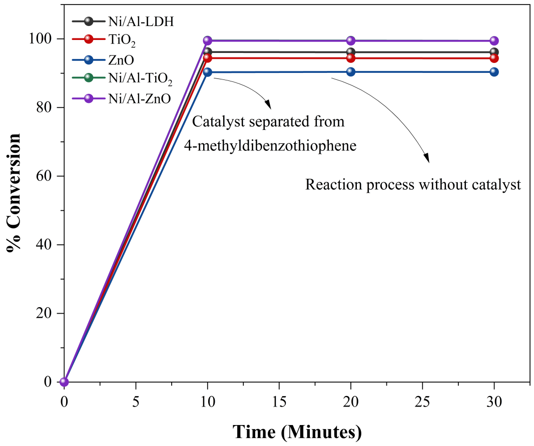

Figure 9. Heterogeneity Test of Catalyst

The reusability of catalysts is of great importance in the industry, as it leads to cost savings (Jiang et al., 2017). To assess the reusability of the catalysts in this study, n-hexane was used as the solvent. After each cycle, the catalyst was recovered, and 4-methyldibenzothiophene was desorbed by subjecting it to ultrasonic treatment with n-hexane as the solvent. The catalyst was subsequently dried and reused in the oxidative desulfurization process (Ribeiro et al., 2019). Based on the data presented

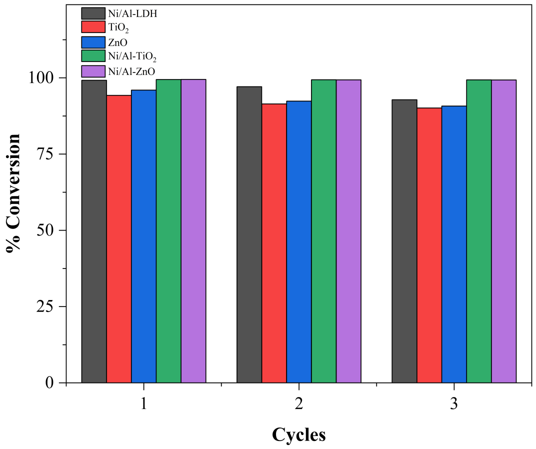

Figure 10. Reusability of Catalyst on Oxidative Desulfurization of 4-methyldibenzothiophene

in Figure 10, it can be observed that the repeated use of the catalyst remains effective, yielding favorable percentage conversion values. In the first stage of catalyst reuse, Ni/Al-LDH achieved a conversion value of 99.20%, TiO_2 achieved 94.27%, ZnO achieved 95.96%, Ni/Al-TiO_2 achieved 99.46%, and Ni/Al-ZnO achieved 99.47%. As the catalysts were reused in stages 2 and 3, there was a slight decrease in the percentage values, but they still remained within the range of 90%, indicating that the repeated use of the catalysts up to three times remains highly effective.

4. CONCLUSION

This study successfully synthesized Ni/Al-LDH layered double hydroxides and modified them with titanium dioxide (TiO_2) to create Ni/Al-TiO_2, and with zinc oxide (ZnO) to form Ni/Al-ZnO. The synthesis of these catalysts was confirmed through various analyses, including XRD pattern, FTIR spectra, SEM images, and EDS analysis. The composite catalyst demonstrated superior performance compared to both the precursor and base material in terms of desulfurization of 4-methyldibenzothiophene. This conclusion is based on the significantly higher percentage conversion values obtained across all tested reaction factors. The improved desulfurization efficiency of the composite catalyst highlights its enhanced ability to facilitate the desired chemical reaction compared to the individual precursor and base materials. The optimal conditions for the oxidative desulfurization process were determined to be a reaction time of 40 minutes, a catalyst dosage of 0.25 grams, a temperature of 50°C, and the use of n-hexane as the solvent. The study also confirmed that the catalyst utilized in this research is heterogeneous in nature and can be reused multiple times. In summary, this research showcases the successful synthesis of Ni/Al-TiO_2 and Ni/Al-ZnO catalysts and their effectiveness in the oxidative desulfurization of 4-methyldibenzothiophene. The study identifies the optimal reaction conditions and establishes the heterogeneous nature and reusability of the catalyst.

5. ACKNOWLEDGMENT

Our research were supported from Reseach Center of Inorganic Materials and Complexes Universitas Sriwijaya and Hibah Profesi 2023 as additional research output Contract Number SP DIPA-023.17.2.677515/2023 accordance with SK Rector No. 0187/UN9.3.1/SK/2023.

REFERENCES

Abedini, F., S. Allahyari, and N. Rahemi (2021). Oxidative Desulfurization of Dibenzothiophene and Simultaneous Adsorption of Products on Biobr-C_3N_4/MCM-41 Visible-Light-Driven Core–Shell Nano Photocatalyst. *Applied Surface Science*, **569**; 151086

Abro, R., S. Gao, X. Chen, G. Yu, A. A. Abdeltawab, and S. S. Al-Deyab (2016). Oxidative Desulfurization of Gasoline by Ionic Liquids Coupled with Extraction by Organic Solvents. *Journal of the Brazilian Chemical Society*, **27**(6); 998–1006

Ahmad, N., M. Badaruddin, N. Yuliasari, F. S. Arsyad, and A. Lesbani (2022a). Highly Efficient Catalytic Oxidative Desulfurization of Dibenzothiophene using Layered Double Hydroxide Modified Polyoxometalate Catalyst. *Bulletin of Chemical Reaction Engineering & Catalysis*, **17**(4); 821–830

Ahmad, N., E. S. Fitri, A. Wijaya, A. Amri, M. Mardiyanto, I. Royani, and A. Lesbani (2022b). Catalytic Oxidative Desulfurization of Dibenzothiophene Utilizing Composite Based Zn/Al Layered Double Hydroxide. *Bulletin of Chemical Reaction Engineering & Catalysis*, **17**(4); 733–742

Ahmad, N., N. Yuliasari, F. S. Arsyad, I. Royani, R. Mohadi, and A. Lesbani (2023). Catalytic Oxidative Desulfurization of Dibenzothiophene by Heterogeneous M^{2+}/Al-Layered Double Hydroxide (M^{2+}= Zn, Mg, Ni) Modified Zinc Oxide. *Iranian Journal of Catalysis*, **13**(1); 35–45

Basnet, P., D. Samanta, T. Inakhunbi Chanu, J. Mukherjee, and S. Chatterjee (2019). Assessment of Synthesis Approaches for Tuning the Photocatalytic Property of ZnO Nanoparticles. *SN Applied Sciences*, **1**(6); 1–13

Cao, Y., H. Wang, R. Ding, L. Wang, Z. Liu, and B. Lv (2020). Highly Efficient Oxidative Desulfurization of Dibenzothiophene using Ni Modified MoO_3 Catalyst. *Applied Catalysis A: General*, **589**; 117308

Deng, C., H. Zhu, Y. Huang, H. Liu, P. Liu, P. Cui, Y. Chao, J. Liu, R. Wang, and P. Wu (2022). High Temperature Oxidizing-Resistant Magnetic High Entropy Catalyst for Efficient Oxidative Desulfurization. *Catalysis Today*, **405**; 66–74

Fayad, M. A., M. T. Chaichan, H. A. Dhahad, A. A. Al-Amiery, and W. N. R. Wan Isahak (2022). Reducing the Effect of High Sulfur Content in Diesel Fuel on NO_x Emissions and PM Characteristics Using a PPCI Mode Engine and Gasoline–Diesel Blends. *ACS Omega*, **7**(42); 37328–37339

Houda, S., C. Lancelot, P. Blanchard, L. Poinel, and C. Lamonier (2018). Oxidative Desulfurization of Heavy Oils with High Sulfur Content: A Review. *Catalysts*, **8**(9); 344

Intachai, S., T. Nakato, and N. Khaorapapong (2021). ZnO Decorated on Low Carbonate NiAl-Layered Double Hydroxide as Efficient Photocatalyst for Methyl Orange Degradation. *Applied Clay Science*, **201**; 105927

Jiang, W., D. Zheng, S. Xun, Y. Qin, Q. Lu, W. Zhu, and H. Li (2017). Polyoxometalate-Based Ionic Liquid Supported on Graphite Carbon Induced Solvent-Free Ultra-Deep Oxidative Desulfurization of Model Fuels. *Fuel*, **190**; 1–9

Kumar, S., V. C. Srivastava, and R. Badoni (2012). Oxidative Desulfurization by Chromium Promoted Sulfated Zirconia. *Fuel Processing Pechnology*, **93**(1); 18–25

Lesbani, A., N. R. Palapa, R. J. Sayeri, T. Taher, and N. Hidayati (2021). High Reusability of NiAl LDH/Biochar Composite in the Removal Methylene Blue from Aqueous Solution. *Indonesian Journal of Chemistry*, **21**(2); 421–434

Liu, S., Z. Liu, H. Zhu, Z. Wang, J. Guo, X. Zhang, H. Yu, X. Yue, P. Ning, and B. Li (2023). The Roles of Red Mud as Desulfurization and Denitrification in Flue Gas: A Review. *Journal of Environmental Chemical Engineering*, **11**(3); 109770

Luna, M. L., M. A. Taboada-Ortega, M. A. Alvarez-Amparán, and L. Cedeño-Caero (2022). Effect of Iron Incorporation on W Based Catalysts for Oxidative Desulfurization of Dibenzothiophene Compounds. *Catalysis Today*, **394**; 336–347

Lv, H., H. Rao, Z. Liu, Z. Zhou, Y. Zhao, H. Wei, and Z. Chen (2022). NiAl Layered Double Hydroxides with Enhanced Interlayer Spacing via Ion-Exchange as Ultra-High Performance Supercapacitors Electrode Materials. *Journal of Energy Storage*, **52**; 104940

Madram, A. R. and N. Zandi Atashbar (2015). Application of Pd-Substituted Ni-Al Layered Double Hydroxides for the Hydrogen Evolution Reaction. *Iranian Journal of Hydrogen & Fuel Cell*, **2**(4); 253–261

Mahmoudi, V., A. M. Kermani, M. Ghahramaninezhad, and A. Ahmadpour (2021). Oxidative Desulfurization of Dibenzothiophene by Magnetically Recoverable Polyoxometalate-Based Nanocatalyst: Optimization by Response Surface Methodology. *Molecular Catalysis*, **509**; 111611

Malani, R. S., A. H. Batghare, J. B. Bhasarkar, and V. S. Moholkar (2021). Kinetic Modelling and Process Engineering Aspects of Biodesulfurization of Liquid Fuels: Review and Analysis. *Bioresource Technology Reports*, **14**; 100668

Mjalli, F. S., O. U. Ahmed, T. Al-Wahaibi, Y. Al-Wahaibi, and I. M. AlNashef (2014). Deep Oxidative Desulfurization of Liquid Fuels. *Reviews in Chemical Engineering*, **30**(4); 337–378

Palapa, N. R., T. Taher, A. Wijaya, and A. Lesbani (2021). Modification of Cu/Cr Layered Double Hydroxide by Keggin Type Polyoxometalate as Adsorbent of Malachite Green from Aqueous Solution. *Science and Technology Indonesia*, **6**(3); 209–217

Polikarpova, P., A. Akopyan, A. Shlenova, and A. Anisimov (2020). New Mesoporous Catalysts with Brønsted Acid Sites for Deep Oxidative Desulfurization of Model Fuels. *Catalysis Communications*, **146**; 106123

Priatna, S. J., Y. M. Hakim, S. Wibyan, S. Sailah, and R. Mohadi (2023). Interlayer Modification of West Java Natural Bentonite as Hazardous Dye Rhodamine B Adsorption. *Science and Technology Indonesia*, **8**(2); 160–169

Raeisi, A., A. N. Chermahini, and M. M. Momeni (2022). A Novel Photocatalytic and Photoelectrocatalytic System for Oxidative Desulfurization of Model Fuel using $BiVO_4$@HKUST-1 Composite in Powder and Deposited on Fluorine-Doped Tin Oxide. *Journal of Photochemistry and Photobiology A: Chemistry*, **433**; 114190

Ren, X., G. Miao, Z. Xiao, F. Ye, Z. Li, H. Wang, and J. Xiao (2016). Catalytic Adsorptive Desulfurization of Model Diesel Fuel using TiO_2/SBA-15 under Mild Conditions. *Fuel*, **174**; 118–125

Ribeiro, S. O., C. M. Granadeiro, P. L. Almeida, J. Pires, M. C. Capel-Sanchez, J. M. Campos-Martin, S. Gago, B. De Castro, and S. S. Balula (2019). Oxidative Desulfurization Strategies using Keggin-Type Polyoxometalate Catalysts: Biphasic Versus Solvent-Free Systems. *Catalysis Today*, **333**; 226–236

Song, Y., J. Bai, S. Jiang, H. Yang, L. Yang, D. Wei, L. Bai, W. Wang, Y. Liang, and H. Chen (2021). Co-Fe-Mo Mixed Metal Oxides Derived from Layered Double Hydroxides for Deep Aerobic Oxidative Desulfurization. *Fuel*, **306**; 121751

Subhan, S., A. U. Rahman, M. Yaseen, H. U. Rashid, M. Ishaq, M. Sahibzada, and Z. Tong (2019). Ultra-Fast and Highly Efficient Catalytic Oxidative Desulfurization of Dibenzothiophene at Ambient Temperature Over Low Mn Loaded Co-Mo/Al_2O_3 and Ni-Mo/Al_2O_3 Catalysts using NaClO as Oxidant. *Fuel*, **237**; 793–805

Wang, L., Y. Chen, L. Du, S. Li, H. Cai, and W. Liu (2013). Nickel-Heteropolyacids Supported on Silica Gel for Ultra-Deep Desulfurization Assisted by Ultrasound and Ultraviolet. *Fuel*, **105**; 353–357

Wu, C., C. Chen, Z. Qi, J. Chen, Q. Wang, C. Ye, and T. Qiu (2023). Facile Synthesis of Efficient Pentaethylenehexamine-Phosphotungstic Acid Heterogeneous Catalysts for Oxidative Desulfurization. *Chinese Journal of Chemical Engineering*

Wu, M., B. Chang, T.-T. Lim, W.-D. Oh, J. Lei, and J. Mi (2018). High-Sulfur Capacity and Regenerable Zn-Based Sorbents Derived from Layered Double Hydroxide for Hot Coal Gas Desulfurization. *Journal of Hazardous Materials*, **360**; 391–401

Xun, S., C. Wu, L. Tang, M. Yuan, H. Chen, M. He, W. Zhu, and H. Li (2023). One-Pot $In-Situ$ Synthesis of Coralloid Supported VO_2 Catalyst for Intensified Aerobic Oxidative Desulfurization. *Chinese Journal of Chemical Engineering*, **56**(43); 136–140

Yuliasari, N., A. Wijaya, R. Mohadi, E. Elfita, and A. Lesbani (2022). Photocatalytic Degradation of Malachite Green by Layered Double Hydroxide Based Composites. *Bulletin of Chemical Reaction Engineering & Catalysis*, **17**(2); 240–249

Zhou, X., X. Pei, R. Wang, and T. Zhao (2021). Catalytic Conversion for Hydrogen Sulfide. *Aerosol and Air Quality Research*, **21**(9); 210065