

Nickel-Lanthanum Impregnated into Natural Zeolite as a Catalyst for Biofuel Production from Sunflower Oil via Hydrocracking Process

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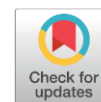
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Abstract

The increasing demand for crude oil or fossil fuel as a raw material for oil fuel has been steadily rising over time in line with the development that is taking place in Indonesia. However, biofuels are potential vegetable fuels that can be developed as alternative energy because they are renewable and can be renewed to overcome the energy crisis in the future. For this purpose, a double metal catalyst (impregnated with nickel and lanthanum), is used to make biofuels from sunflower seed oil. The effect of metal ratio on the yield of biofuel products is the concern in this study. The temperature of hydrocracking process was 250-330 °C with ratio of metal 5% and 10% (metal ratio 1:1 and 1:2). X-ray diffraction (XRD) shows that natural zeolite has a clinoptilolite phase, X-Ray Fluorescence (XRF) shows that acid and base activation increases the Si/Al ratio from 4.5 to 5, Scanning Electron Microscope – Energy Dispersive X-Ray (SEM-EDX) shows images of natural zeolite surfaces in the form of aggregate pieces, and Brunauer Emmett Teller (BET) shows that acid and base activation increases S_{BET} from 29.96 to 49.73 m²/g and forms a hierarchical natural zeolite. The impregnation of Ni-La/Zeolite catalyst has been successfully carried out using the incipient wetness impregnation method and the best catalyst results were obtained, namely Ni-La/Zeolite 10% (1:2) with a surface area of 15.33 m²/gram. The addition of Nickel and Lanthanum metals caused a decrease in the surface area and average pore diameter of the zeolite. The lowest surface area and average pore diameter were found in the variation of the Ni-La/Zeolite 10% (1:2) catalyst, namely 15.33 m²/gram and 13.99 nm. The highest hydrocarbon yield was found in the hydrocracking process with the Ni-La/Zeolite 10% (1:1) catalyst with gasoline, kerosene and gasoil fractions of 0.91; 0.39 and 8.32 (%wt), respectively. The hydrocarbon compound composition of the catalyst includes n-paraffin 4.43%, isoparaffin 0.21%, cycloparaffin 2.99% and olefin 2.71%.

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1. Introduction

As Indonesia continues to grow and develop, there is a corresponding rise in energy demand.

The transportation sector is among the highest consumers of energy across various industries. Fossil fuel have been used as a main source of energy for many years, but to fulfill the global energy demand, we have to get another energy source that can be better. The use of vegetable oil

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which is a renewable energy source expected to offer solutions to overcome the energy crisis [1]. The burning of the fossil fuels by the transportation of people can emit gaseous pollutants such as carbon dioxide, carbon monoxide, nitrogen oxide, sulfur oxide, volatile organic compounds, and particulate matter that can change the composition of the atmosphere [2]. A potential solution is to use renewable sources of energy, which have the ability to provide energy with reduced emission of air pollutants [3].

A biofuel is a fuel that is produced from biological processes or derived from the biomass of living organisms. Biofuels are arguably a potential renewable energy source in the transportation industry [4]. Biofuels contributed to the reduction of carbon emissions; it is because biofuels are carbon neutral. Besides that, biofuel can stabilize the global climate and improve energy security from use of conventional fossil fuels [5]. There are four generation categories of biofuel base on the type of feedstock used to produce biofuel. First of all is the biofuels that produced from edible feedstocks such as from corn, sugar cane and oil seed crops [6]. Second generation are produced from agricultural by-products or cellulosic materials such as wood, leaves and grass [7]. Third-generation are produced from aquatic cultivated stock such as algae. And last, the fourth generation are made from bioengineered microorganisms [8]. One of the vegetable oils that has a potential is sunflower oil. The sunflower is not only known as an ornamental plant but also known as a plant that produces oils [9]. The use of sunflower oil is still limited, especially in Indonesia. Sunflower oil mostly contains linoleic acid and oleic acid. The benefits of this oil are still not optimal. Sunflower oil consists of the same elements as other vegetable oils, such as carbon, hydrogen, and oxygen [10].

Zeolite is one of the non-metallic mineral commodities that have the potential to be catalysts in a cracking reaction. Zeolite is a metal support catalyst. The surface characteristics of zeolite can be changed by a modification with an acid process to clean the pores of adsorbed metals [11]. Lampung natural zeolite has good potential because it contains SiO_2 , Al_2O_3 , and several metals that are useful as catalysts [12,13]. With the addition of a metal component in the zeolite pore, it is expected to be able to make a high-quality hydrocarbon in biofuel. Nickel has been widely used as a catalyst before. It can give a positive contribution for cracking activity [14]. Several previous studies have conducted metal impregnation on natural zeolite. Such as the study by Kurniawan *et al.* [15] who conducted Ni and Mo impregnation on natural zeolite for the conversion of used cooking oil into biofuel. In

addition, there is a study conducted by Sihombing *et al.* [16], conducting Co and Mo impregnation on natural zeolite for the conversion of rubber seed oil into gasoline fractions. Several other studies have also conducted metal impregnation, such as Ni, Zn, Cu, and La on natural zeolite [17,18]. However, no one has combined Ni and La on natural zeolite for the hydrocracking reaction of sunflower oil. In the presence of a second metal, it is expected to be better. Therefore, nickel and lanthanum are combined for this study with zeolite [19].

Istadi *et al.* [20] reported that Co metal chemically doped on Ni/ZSM-5 catalyst for cracking palm oil has the highest conversion with 3Co-Ni/ZSM-5 to produce gasoline, kerosene, and diesel with yields of 2.61%, 4.38%, and 61.75%, respectively. Kurniawan *et al.* [15] also reported using NiMo/ γ - Al_2O_3 -zeolite catalyst with alumina-to-zeolite ratios of 75:25 and 25:75, effectively converting used cooking oil into bio-hydrocarbon products—green diesel and gasoline. Allwar *et al.* [21] also reported using NiO-CdO/biochar catalyst with hydrocracking of crude palm oil (CPO) has produced more liquid product than thermal cracking, at 56.55 wt%, 20.55 wt%, respectively.

Biofuel products from sunflower oil can be obtained through a process that cracks long-chain hydrocarbons into short-chain hydrocarbons. The method can be divided into three types: thermal cracking, catalytic cracking, and hydrocracking. Hydrocracking is a method that can convert vegetable oils to biofuel with the addition of a catalyst and hydrogen. Hydrocracking has several advantages, such as providing high conversions, high yields towards middle distillate, and producing alkanes with good quality [22]. Biofuel can be the energy that is environmentally friendly. To get the high yield of biofuel, it is usually influenced by several aspects, such as temperature reaction, time of cracking, weight of catalyst, and the type of catalyst [23]. Therefore, this study conducted acid and base pretreatment on natural zeolite. Then, Ni-La metal impregnation with varying metal ratios and metal loadings on natural zeolite was carried out, followed by sunflower oil hydrocracking reaction at varying reaction temperatures.

2. Materials and Methods

2.1 Materials

This study were used sunflower oil (from local market), natural zeolite from Lampung, nickel (II) nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), lanthanum(III) nitrate hexahydrate ($\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$) were purchased from Merck with purity $\geq 99\%$.

2.2 Preparation and Catalyst Characterization

Preparation of natural zeolite was made with acid activation and base activation or known as dealumination and desilication. Natural zeolite with a size of 200 mesh put into the muffle furnace at 500 °C for 3 h then base activation treatment with 500 mL NaOH 0.5 M at 75 °C for 1 h. After washing process with aquades until netral, then dry the zeolite in the oven at 110 °C for 24 h and activation with HCl 2 M for 2 h. Rewash the zeolite to pH of 5, dry in the oven at 110 °C for 24 h. Calcination was carried out at a temperature of 500 °C for 3 h in a free air atmosphere. Then impregnation the zeolite with nickel nitrate hexahydrate and lanthanum nitrate hexahydrate with metal ratio of 1:1 and 1:2, %loading of 5% and 10%. Impregnation is done by dissolving $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ dan $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ in methanol. Then, mixed with natural zeolite and stirred for 1h and left for 12 h. **Then, dried using an oven at a temperature of 110 °C for 12 h. 16. ????** The catalyst solid is then calcined using air at a temperature of 550 °C for 3 h and a catalyst solid is obtained that is ready to use.

The crystallinity of the catalysts was characterized by X-ray diffraction (XRD) using a XRD SmartLab Rigaku with Cu-K α radiation (1.541862 Å) in a 2 θ range from 5-100°. X-Ray Fluorescence (XRF) to determine the components in the catalyst. Scanning Electron Microscope – Energy Dispersive X-Ray (SEM-EDX) Analysis to analysis morphology and element composition. Brunauer Emmett Teller (BET) analysis produces surface area, pore size, and total pore volume.

2.3 Catalytic Tests

The catalytic tests were carried out using a batch reactor with four different temperatures (270, 290, 310, 330) °C (Figure 1). The 1 g of catalyst was placed in the reactor and 25 mL of sunflower oil was added. The condition reaction process at temperature of 270-330 °C with 20 bar

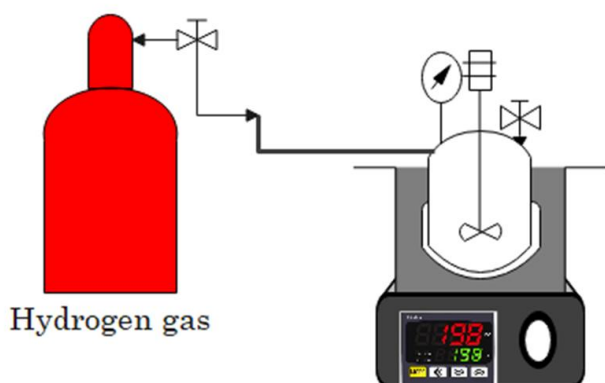


Figure 1. Schematic diagram of hydrocracking process.

H₂ gas cylinder pressure for 2 h. After reaching the variable temperature, the sample is left to cool to room temperature, then taken and placed in a sample bottle. The result of biofuel product was analyzed by GC-MS to determine hydrocarbon chain from the process. Analyses were performed using an Agilent gas chromatograph coupled to a mass selective detector (GC–MS). Separation was achieved on an Agilent HP-5 MS column (Catalog 19091S-433, 30 m × 0.25 mm × 0.25 μm, 5% phenyl-methylpolysiloxane). The results of yield and conversion were calculated based on the following equations below, where W of liquid product is the weight of the liquid product (g), W of sunflower oil is the weight of sunflower oil (g), W of remaining oil is the weight of the remaining oil (g), which was the following equation from Trisunaryanti *et al.* [24]:

$$\text{Liquid Product Yield (\%)} = \frac{\text{Weight of Liq.Product}}{\text{Weight of Sunflower Oil Feed}} \times 100\% \quad (1)$$

$$\text{Total Conversion (\%)} = \frac{\text{Weight of Sunflower Oil} - \text{Weight of remaining oil}}{\text{Weight of Sunflower Oil}} \times 100\% \quad (2)$$

3. Results and Discussion

3.1 Catalyst Characterization

The XRD patterns of Natural zeolite and modified zeolite shown in Figure 2. The main peaks of zeolite were observed at 2 θ of: 9,81°; 19,74°; 21,68°; 22,37°; 26,04°; 28,01°; 30,03° and 31,92°. Then the main peaks of modified zeolite were observed at 9,87°; 11,15°; 20,90°; 21,84°; 22,42°; 23,68°; 26,09°; 27,72°. These peaks indicate the clinoptilolite phase based on JCPDS card No. 25-1349 [12]. The activation process of zeolite with acids and bases results in a decrease in crystallinity as indicated by a decrease in the intensity of the typical clinoptilolite peaks [25].

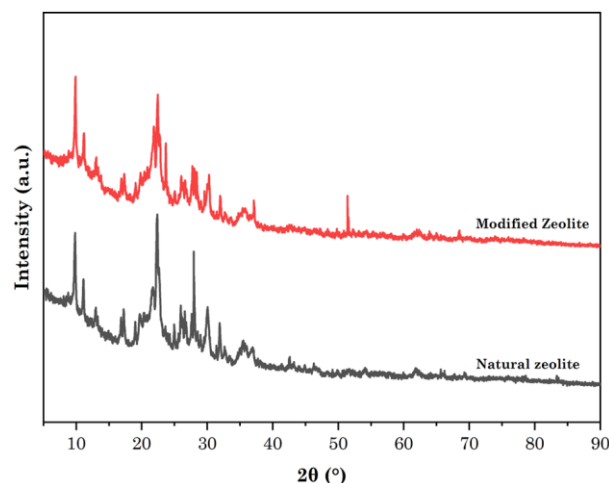


Figure 2. XRD pattern for zeolite and modified zeolite.

Figure 3 show the patterns of XRD for different ratio and metal loading amount from Ni-La/Zelite 5% (1:1) and Ni-La/Zelite 5% (1:2). The pattern with Ni-La/Zelite 5% (1:1) catalyst shown La diffraction peaks at $2\theta = 27,73^\circ$; NiO at $2\theta = 36,48^\circ$ and Ni at $2\theta = 50,09^\circ$ with low intensity. Then the XRD pattern for Ni-La/Zelite 5% (1:2) shown the diffraction peak of La at $2\theta = 27,70^\circ$; NiO at $2\theta = 42,56^\circ$ and Ni at $2\theta = 68,33^\circ$ with low intensity [14]. It shows that particle of La and Ni well dispersed on the zeolite [26].

Table 1 shows the result of XRF analysis in wt% units for zeolite and modified zeolite. It shows that there is an increase of SiO_2 on natural zeolite before the modification process from 72.50% to 75.90% after modification. Furthermore, there is a decrease of Al_2O_3 from 16.10% to 15.20%. Table 1 and Figure 4 show that the ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ before modification increased from 4.5 to 5.0 after modification. Increasing the ratio of Si/Al can increase the acidity or acid site on the zeolite, which will improve the activity of the zeolite. This is indicated a reaction of dealumination, with an aim

to optimize aluminum in zeolite to be stable at high temperatures [27]. Table 1 shows the result of XRF analysis in wt% units for zeolite that has impregnated with nickel and lanthanum. The ratio and % loading were 5% (1:1) and 5% (1:2). The characterization shows that there was an addition of nickel and lanthanum for all the ratios. On a Ni-La/Zelite 5% (1:1), the compositions of nickel and lanthanum are 2.60% and 2.00%. Then, for the catalyst Ni-La/Zelite 5% (1:2), the compositions of nickel and lanthanum are 3.90% and 2.70%. Impregnation was chosen as a dispersion method because it is an easy process, can be derived from its precursor salt solution, and is effective for dispersing active metal [28].

Figure 5 shows the SEM results for all catalysts. All catalysts have a similar morphology, resembling irregular aggregates. However, after modification, the material surface appears to be fragmented due to activation by acids and bases. Furthermore, after impregnation with Ni-La metal, the material has a more aggregated surface due to the presence of

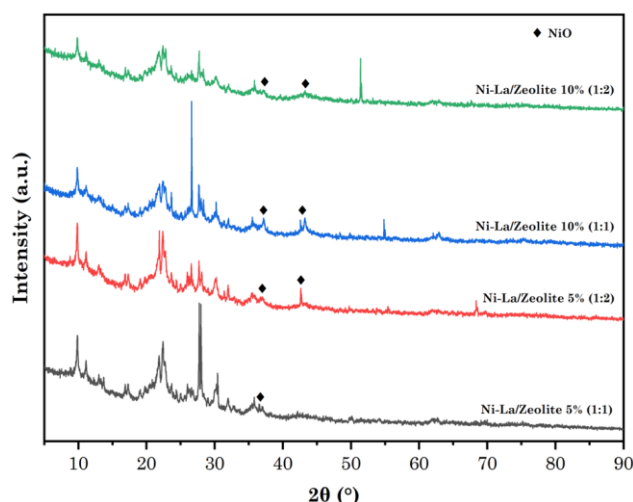


Figure 3. XRD pattern of Ni-La/Zelite.

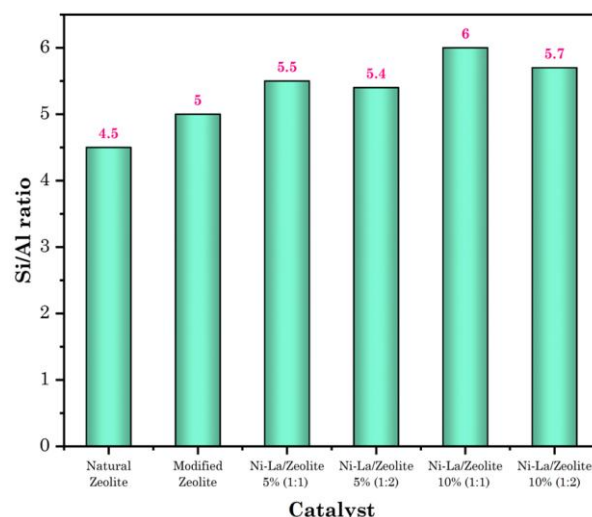


Figure 4. Si/Al ratio of the catalysts base on XRF results.

Table 1. XRF analysis for catalysts.

| Component | Catalyst composition (wt.%) | | | | | |
|-------------------------|-----------------------------|------------------|-----------------------|-----------------------|------------------------|------------------------|
| | Zeolite | Modified Zeolite | Ni-La/Zelite 5% (1:1) | Ni-La/Zelite 5% (1:2) | Ni-La/Zelite 10% (1:1) | Ni-La/Zelite 10% (1:2) |
| Al_2O_3 | 16.10 | 15.20 | 13.30 | 13.00 | 11.10 | 11.70 |
| CaO | 2.50 | 1.70 | 1.60 | 1.60 | 1.20 | 1.30 |
| Fe_2O_3 | 3.10 | 2.50 | 1.90 | 1.90 | 1.80 | 1.90 |
| K_2O | 2.30 | 0.90 | 1.20 | 1.20 | 0.90 | 0.90 |
| La_2O_3 | - | - | 2.30 | 3.20 | 5.70 | 8.30 |
| MgO | 2.00 | 0.80 | 1.60 | 1.50 | 1.40 | 1.40 |
| NiO | - | - | 3.20 | 4.90 | 9.70 | 6.90 |
| SiO_2 | 72.50 | 75.90 | 73.80 | 71.30 | 66.60 | 66.50 |

metal dispersed on the zeolite surface. This indicates that the metal has been successfully impregnated into the zeolite [29]. Figure 6 displays a mapping image of Ni and La metals in natural zeolite. The mapping results indicate that the metals are evenly dispersed on the zeolite surface, as indicated by the uniform distribution of colors [30]. Furthermore, EDX also indicates that Ni-La has been successfully impregnated, supporting the XRF analysis results.

Figure 7 displays the N₂ adsorption-desorption isotherms of natural zeolite, modified zeolite, and Ni-La impregnated zeolite. The isotherms show a hysteresis loop starting at P/P₀ = 0.4 to 1.0, which is a type IV isotherm. The presence of this hysteresis loop indicates the presence of mesopores (2-50 nm). The hysteresis loop formed at P/P₀ to 1.0 indicates the presence of intra- and interparticle mesopores [31]. While the isotherm at P/P₀ between 0-0.4 indicates

micropores in the material. Therefore, it can be concluded that based on the N₂ adsorption-desorption isotherms, all catalysts have a hierarchical pore type, namely a combination of micropores and mesopores [32].

Table 2 shows that natural zeolite after modification has a larger surface area value of 49.73 m²/g compared to natural zeolite which is 29.96 m²/g. Then after the impregnation process with nickel and lanthanum metals, the surface area and pore volume of the catalyst decreased. The surface area of the Ni-La/natural zeolite catalyst with 5% and 10% loading with a ratios of 1:1 and 1:2 were 23.69, 15.87, 19.35, and 15.33 m²/g, respectively. The decrease in the surface area of the catalyst was caused by the filling of the zeolite micropores-mesopores after the impregnation process with nickel and lanthanum metals. Furthermore, the reduced surface area of the catalyst is due to the metal particles spreading

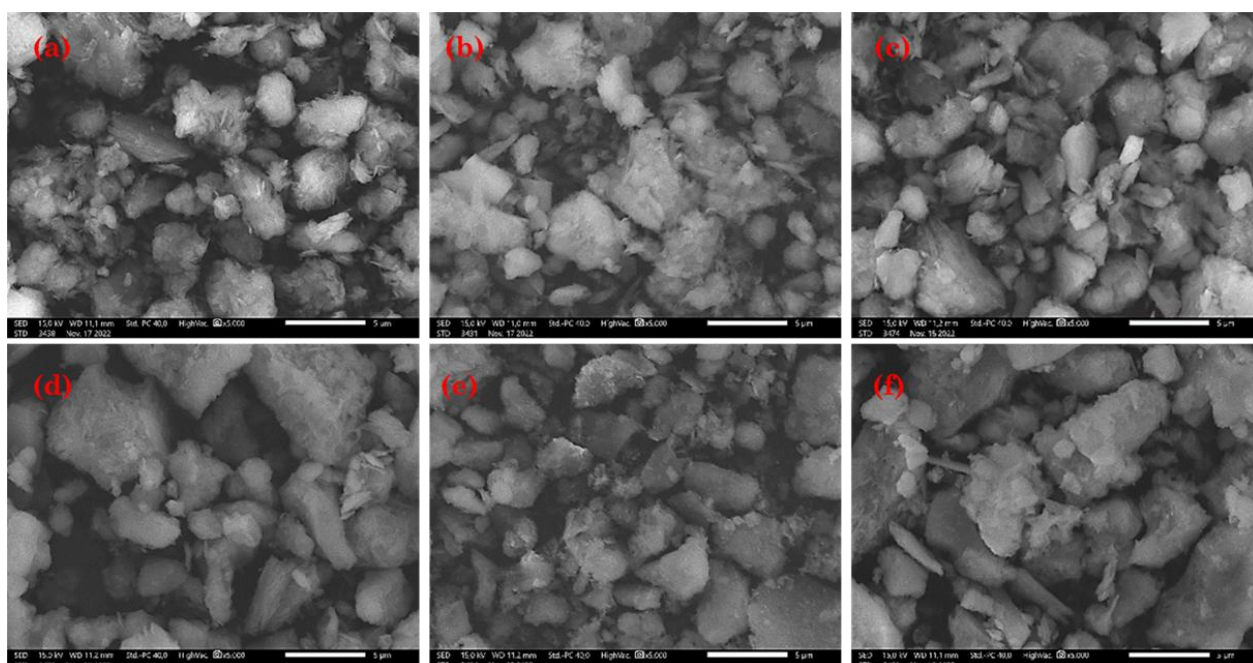


Figure 5. SEM image of Natural Zeolite (a), Modified zeolite (b), Ni-La/Zeolite 5% (1:1) (c), Ni-La/Zeolite 5% (1:2) (d), Ni-La/Zeolite 10% (1:1) (e), and Ni-La/Zeolite 10% (1:2) (f).

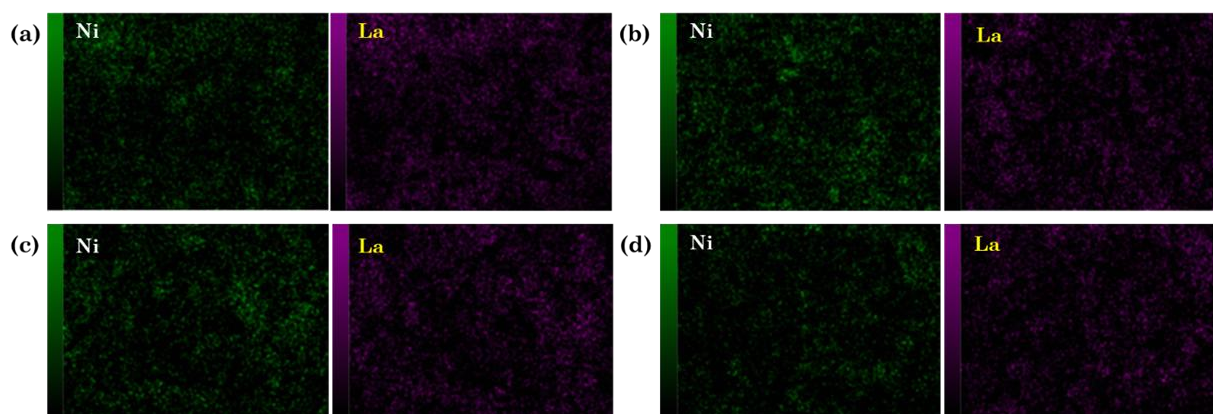


Figure 6. Metal mapping of the Ni-La/Zeolite 5% (1:1) (a), Ni-La/Zeolite 5% (1:2) (b), Ni-La/Zeolite 10% (1:1) (c), and Ni-La/Zeolite 10% (1:2) (d).

across the surface and entering the pores of the natural zeolite, thus reducing the pore volume of the catalyst [33]. This increases the active sites on the catalyst, which affects the reaction rate during the process without reducing the effectiveness and performance of the catalyst [34].

3.2 Catalytic Test

Figure 8 shows the conversion of sunflower oil in the hydrocracking reaction. The conversion rates are very high, almost all above 95%. This is because the reactor used is a batch reactor, resulting in a very large amount of converted feedstock. The unconverted residue is likely lost as gas during the reaction [35]. At a high temperature of 330 °C, conversion decreases because more gas is produced, thus reducing conversion [36]. A higher Si/Al ratio is known to generate stronger Brønsted acidity, which enhances cracking, decarboxylation, and deoxygenation reactions during hydrocracking.

This explains the higher formation of shorter-chain hydrocarbons and the broader distribution of gasoline–kerosene–gasoil obtained with the Ni-

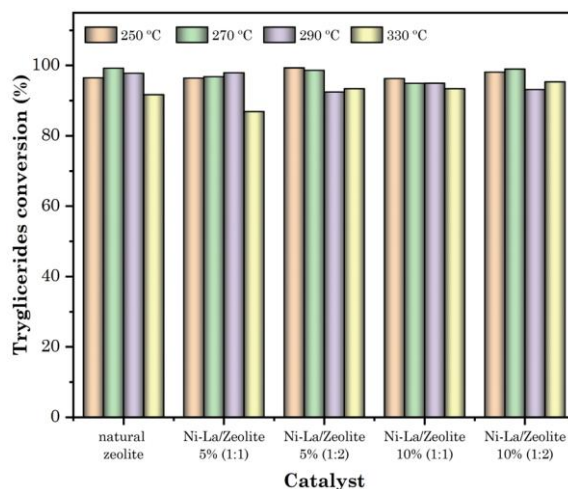


Figure 8. Tryglicerides conversion of the hydrocracking sunflower oil.

Table 2. BET analysis results of zeolite before and after modification and natural Ni-La/Zeolite catalyst. ^at-plot; ^bBJH

| Catalysts | S_{BET} (m ² /g) | $S_{\text{micro}}^{\text{a}}$ (m ² /g) | $S_{\text{ext}}^{\text{a}}$ (m ² /g) | Pore Volume ^b (cm ³ /g) | Average Pore Diameter (nm) |
|---------------------------------|---|---|---|---|----------------------------------|
| Natural zeolite | 29.96 | 1.27 | 28.69 | 0.08 | 11.50 |
| Modified natural zeolite | 49.73 | 11.96 | 37.76 | 0.11 | 8.49 |
| Ni-La/natural zeolite 5% (1:1) | 23.69 | 5.53 | 18.15 | 0.08 | 14.08 |
| Ni-La/natural zeolite 5% (1:2) | 15.87 | 1.09 | 14.77 | 0.07 | 18.48 |
| Ni-La/natural zeolite 10% (1:1) | 19.35 | 3.24 | 16.10 | 0.09 | 18.72 |
| Ni-La/natural zeolite 10% (1:2) | 15.33 | 2.72 | 12.61 | 0.05 | 13.99 |

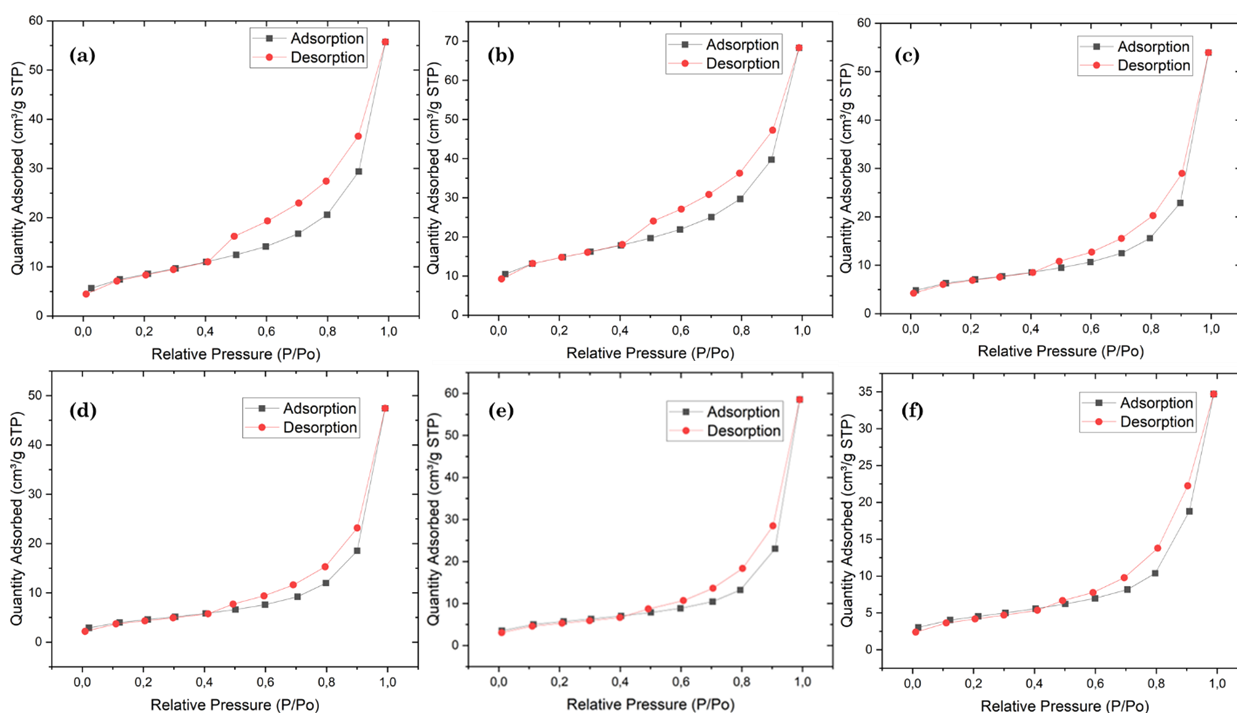


Figure 7. N₂ adsorption-desorption isotherm of catalysts.

La-modified catalysts, compared to natural zeolite which mainly produced carboxylic acids [37].

The process that has been typically used in the petroleum refinery plant for a long time to upgrade and improve product quality is hydroprocessing. It is divided into two main chemical reaction such as hydrotreating and hydrocracking. Hydrotreating is mainly used for eliminating heteroatom from crude oil in petroleum industry and hydrocracking is used to convert a larger molecule of hydrocarbon structure into hydrocarbon compounds of lower molecular weight [38]. Biofuel product from sunflower oil through hydrocracking with Ni-

La/Zeolite characterized by GC-MS. Hydrocarbon mixture produced by hydrocracking from vegetable oil is commonly called green diesel, it has a high cetane number mostly. The feedstock of hydrocracking reaction are one of the critical parameters for sustainable and feasible biofuel production [39].

The result is that biofuel can be classified into hydrocarbon compounds such as n-paraffin, isoparaffin, cycloparaffin, aromatic, olefin, alcohol, carboxylic acid, and polycyclic aromatic hydrocarbon. The main composition of the product still dominated by carboxylic acid. Figure 9 shows the product distribution from the hydrocracking

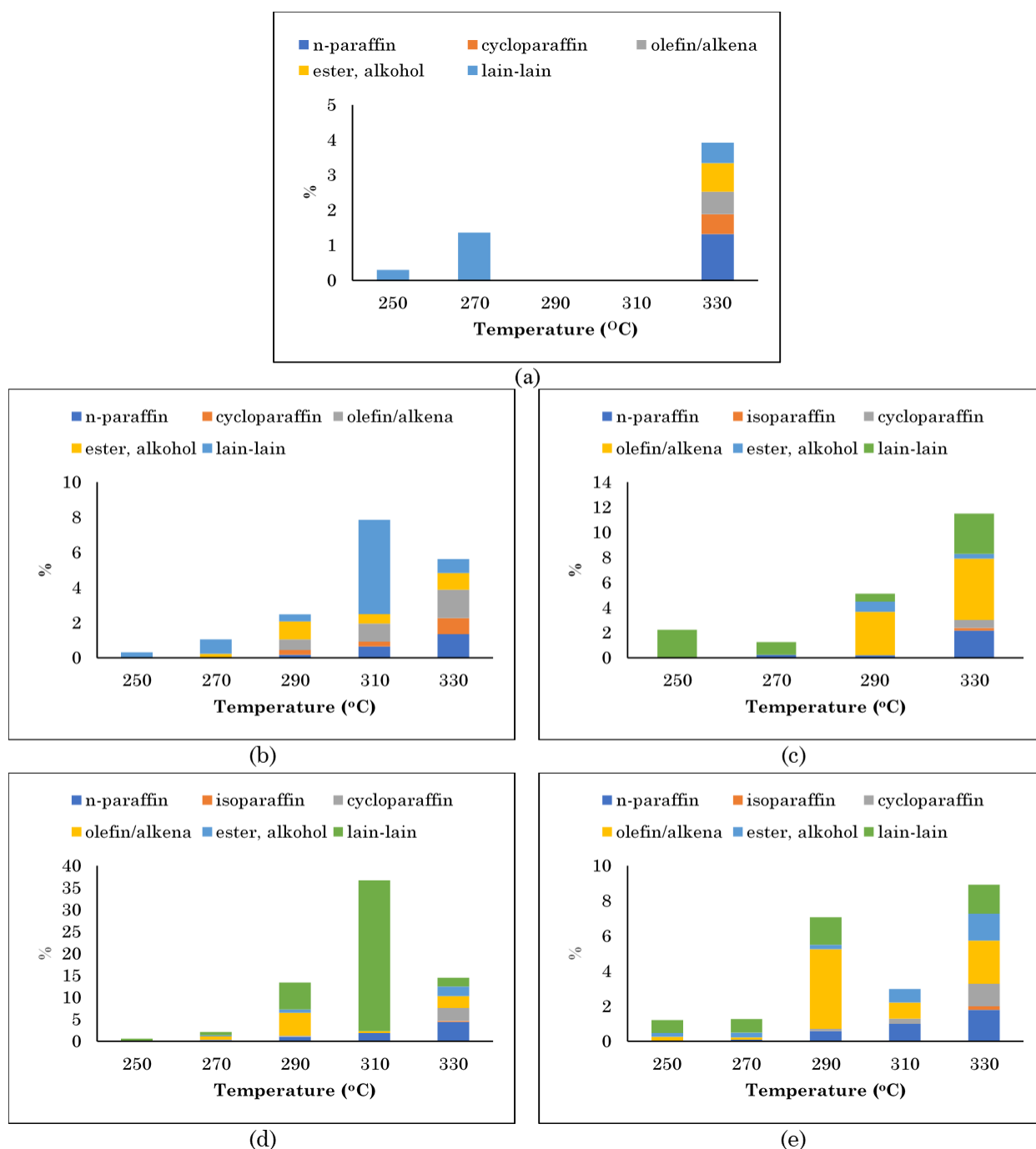


Figure 9. Composition for Natural zeolite (a), Ni-La/Zeolite 5% (1:1) (b), Ni-La/Zeolite 5% (1:2) (c), Ni-La/Zeolite 10% (1:1) (d), and Ni-La/Zeolite 10% (1:2) (e).

reaction of sunflower oil with a natural zeolite catalyst and Ni-La metal impregnation. A biofuel product from a natural zeolite catalyst contains only carboxylic acid. Then for the Ni-La Zeolite 5% (1:1) catalyst, there's a form of n-paraffin, cycloparaffin, and olefin. For Ni-La/Zeilite 5% (1:2) there's a component dominated with olefin and n-paraffin. GC-MS spectra of the biofuel show their own retention time for each component. The catalyst that is impregnated with nickel and lanthanum can form a variety of hydrocarbon components. Decreasing composition of n-paraffin with the addition of a catalyst ratio shows a cracking activation that can break the long-chain hydrocarbon into the short one. Hydrocarbon products from the reaction at 330 °C were greater than at lower temperatures. Furthermore, cyclic compounds also increased at 330 °C. This is because the cracking reaction has not yet occurred at lower temperatures but begins at higher temperatures [40]. It could also be due to a less active catalyst. Hydrocarbon biofuel mainly contains hydrocarbon and oxygenated compounds. Based on previous research, pentadecane and heptadecane were the most abundant hydrocarbon compounds in biofuel [41]. During the cracking process of sunflower oil, the active sites on the zeolite catalyst promoted the reaction such as cracking, decarboxylation, and decarbonylation [42]. Based on XRF and SEM-EDX, the uniform dispersion of Ni and La on the zeolite surface contributes to efficient hydrogenation-dehydrogenation functions, enabling the conversion of oxygenated intermediates into hydrocarbons. As a result, the Ni-La/zeolite catalysts produced n-paraffins, cycloparaffins, and olefins, whereas natural zeolite produced only carboxylic acids under the same conditions. La species, acting as promoters that enhance metal dispersion and stabilize acidic sites [41].

Figure 10 shows the results of hydrocarbon distribution based on carbon chain length. Biofuel types are categorized into 3, namely gasoline (C_5 - C_9), kerosene (C_{10} - C_{13}), and gasoil (C_{14} - C_{22}). Natural zeolite catalysts only produce gasoline and gasoil products at a temperature of 330 °C, while the zeolite catalyst impregnated with Ni-La produces gasoline, kerosene, and gasoil products at a temperature of 250-310 °C. This shows that Ni-La metal impregnation increases catalytic activity [43]. The catalyst with the best activity is Ni-La/Zeilite 10% (1:1) which shows the results of gasoline, kerosene, and gasoil at a temperature of 330°C with a large composition of more than 10%. This increase in catalytic activity can be associated with large pore size, large pore volume,

and a large meso/micropore ratio. The largest meso/micropore Ni-La/Zeilite 10% (1:1) ratio is 4.9, while natural zeolite without activation has a large ratio, but the catalyst is not yet active because activation has not been carried out. In addition, it can be caused by more active sites compared to other catalysts [44]. In addition, the Si/Al ratio also affects catalytic activity because it affects pore width and acid sites [45].

Textural analysis (Figure 7, Table 2) revealed that modified zeolite possessed a larger surface area (49.73 m²/g) and hierarchical micro-mesoporous structure, which facilitates the diffusion of bulky triglyceride molecules. Although impregnation decreased surface area due to partial pore filling, the presence of well-dispersed Ni and La species created more active sites. This is consistent with the observed higher catalytic activity, evident from (i) higher hydrocarbon production across C_5 - C_{22} , (ii) enhanced cracking at 330 °C, and (iii) the broader carbon chain distribution (Figure 9). The best performance of Ni-La/Zeilite 10% (1:1) correlates with its relatively larger mesoporous fraction, providing accessible sites for both metal-assisted hydrogenation and acid-catalyzed cracking pathways [46].

4. Conclusions

The impregnation of the Ni-La/Zeilite catalyst has been successfully carried out using the incipient wetness impregnation method, and the best catalyst results were obtained, namely Ni-La/Zeilite 10% (1:2) with a surface area of 15.33 m²/gr. The XRD diffractogram shows that natural zeolite has a clinoptilolite phase, the XRF result shows that acid and base activation increases the Si/Al ratio from 4.5 to 5, the SEM-EDX result shows images of natural zeolite surfaces in the form of aggregate chunks, and EDX shows Ni and La successfully impregnated into natural zeolite. The addition of nickel and lanthanum metals caused a decrease in the surface area and average pore diameter of the zeolite. The lowest surface area and average pore diameter were found in the variation of the Ni-La/Zeilite 10% (1:2) catalyst, namely 15.33 m²/gr and 13.99 nm. The highest hydrocarbon yield was found in the hydrocracking process with the Ni-La/Zeilite 10% (1:1) catalyst with gasoline, kerosene, and gasoil fractions of 0.91, 0.39, and 8.32 (% wt), respectively. The hydrocarbon compound composition of the catalyst includes n-paraffin (4.43%), isoparaffin (0.21%), cycloparaffin (2.99%), and olefin (2.71%).

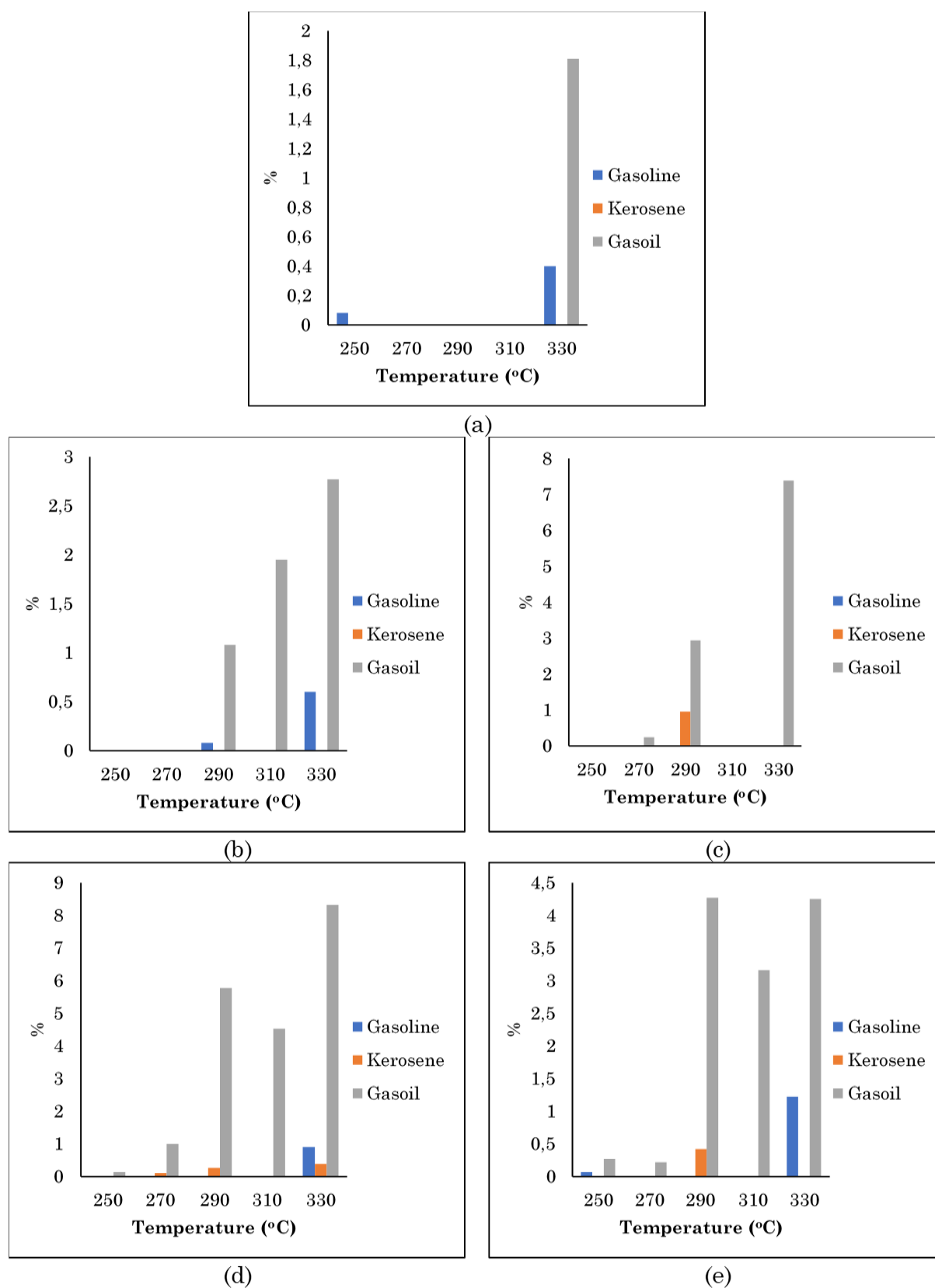


Figure 10. Distribution of hydrocarbon composition using Natural zeolite (a), Ni-La/Zeilite 5% (1:1) (b), Ni-La/Zeilite 5% (1:2) (c), Ni-La/Zeilite 10% (1:1) (d), and Ni-La/Zeilite 10% (1:2) (e).

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Credit Author Statement

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