Production of Bio hydrocarbons from Vegetable Oils and Animal Fats Using Magnesium Oxide as Catalyst

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Article Info

Abstract

Bio hydrocarbons are renewable fuels that can be produced through the catalytic deoxygenation of fatty acids, resulting in compositions like gasoline, kerosene, and diesel fractions derived from petroleum. The objective of this study is to generate gasoline, kerosene, and diesel from vegetable oils and animal fats using an MgO catalyst synthesized through the calcination of Mg-citrate. The characterization of the MgO catalyst, employing Fourier Transform Infrared (FTIR), X-Ray Diffraction (XRD) and Brunauer-Emmett-Teller (BET) surface adsorption method, revealed the presence of crystalline MgO and showed that mesoporous MgO with average pore size of 15.52 nm and exhibiting a surface area of 35.68 m^2 g^{-1}. The MgO catalyst was utilized in the deoxygenation reaction of palm oil, palm wax, and chicken fat, leading to the production of bio hydrocarbons with paraffin and olefin compositions like those found in gasoline, kerosene, and diesel derived from petroleum. Gas Chromatography-Mass Spectroscopy (GCMS) analysis of the liquid product demonstrated that the highest quantity of gasoline was derived from palm wax, followed by palm oil and chicken fat. Palm wax exhibits promising potential as a raw material for gasoline production through the deoxygenation reaction, specifically through decarboxylation and decarbonylation processes facilitated by the MgO catalyst.

Keywords: Bio hydrocarbons, chicken fat, palm oil, palm wax, deoxygenation, mesoporous MgO.

1. INTRODUCTION

In recent years, there has been a significant decline in the availability of petroleum-based fuels. This decrease has become a crucial factor in driving the exploration of alternative fuel sources. These alternative fuel sources can be produced from animal fats and vegetable oils. Animal fat and vegetable oil waste, widely used as feedstocks can be converted into alternative fuels, including biodiesel by means of transesterification reactions[1][2]. Animal fats and vegetable oils can also undergo deoxygenation and catalytic cracking reactions to produce green diesel and bio-hydrocarbons[3] as well as bio gasoline[4]. The triglyceride compounds found in animal fats and vegetable oils can be transformed into bio-hydrocarbon fuels like bio-gasoline, bio-kerosene, and bio-diesel through deoxygenation reactions[5][6], decarboxylation[7], and decarbonylation using catalysts[8].

Renewable energy sources comprise various materials such as palm oil, palm wax, used cooking oil, and animal fats like chicken fat, beef fat, fat derived from leather tanning waste, and fish oil. Notably, non-edible raw materials like animal fat waste have gained significant traction as a promising substitute for vegetable oil in the biofuel manufacturing sector[1].

Santos et al., (2017) used Na_2CO_3 as a catalyst to convert soap phase residue from the palm oil neutralization process into gasoline-like hydrocarbons through catalytic cracking[10]. Sembiring et al., (2018) used Pd/C to produce renewable gasoline from oleic acid through decarboxylation reaction[7]. J. Zhang et al., (2016)

used MgO as a basic catalyst for the catalytic deoxygenation of bio-oil[12].

The MgO catalyst can be used to convert beef fat into biofuel at a temperature of 300 °C, resulting in a liquid fraction containing alkanes, alkenes, alcohols, ketones, and cyclic compounds[13]. The mixed oxide CaO-MgO and Ni/γ-Al$_2$O$_3$ can be used as deoxygenation catalysts for the conversion of palm oil triglycerides, with optimal deoxygenation occurring at temperature of 375°C[14].

The MgO catalyst can be synthesized from magnesium oxalate, as demonstrated by Khromova et al., (2013) and used as a catalyst for the decarboxylation of pentanoic acid into dibutyl ketone[15]. The MgO catalyst can be synthesized from magnesium carbonate and used for catalytic conversion of beef tallow for biofuel production[13]. MgO can act as a catalyst due to its reactive surface, and its activity is attributed to the presence of defects in the crystal lattice, as well as Lewis and Brönsted acid-base sites[16]. MgO possesses thermal stability, the ability to capture and release carbon dioxide (CO_2)[17], as well as abundant and affordable availability, making it a cost-efficient choice as a catalyst in decarboxylation reactions.

In this study, palm oil, palm wax derived from palm fruit, and chicken fat are used as the raw materials. The catalyst magnesium oxide is synthesized through the calcination process of magnesium citrate which was obtained from the reaction between magnesium carbonate and citric acid. The raw materials used are non-toxic and environmentally friendly. The morphology of the synthesized catalyst will be characterized, and the liquid products will be analyzed to determine the percentages of bio gasoline, bio kerosene, and bio diesel.

1. RESEARCH METHODS
Materials and Tools

Commercial palm oil was obtained from PT. Bina Karya Prima, palm wax RC-166 was supplied from Raich Aromatic Supplies, commercial chicken fat was purchased from CV. Fosserca Indo Pangan, Food grade magnesium carbonate ($MgCO_3$) was obtained from Konoshima Chemical Co., Ltd, Food grade citric acid was obtained from RZBC Group Co., Ltd, stearic acid was obtained from Merck and the equipment for deoxygenation reaction used stainless steel reactor with a capacity of 150 cm^3.

Synthesis of Magnesium Oxide

Magnesium citrate was produced from the reaction between magnesium carbonate and citric

acid[18]. Magnesium citrate was formed as a result of a chemical reaction between magnesium carbonate and citric acid, the following reactions occur:

$$MgCO_{3(s)} + C_6H_8O_{7(aq)} \rightarrow MgC_6H_6O_{7(s)} + CO_{2(g)} + H_2O_{(l)}$$

Magnesium citrate was obtained through the reaction of 1 L citric acid 1 M solution with 0.5 mol (42 g) of solid $MgCO_3$ while being stirred using a magnetic stirrer and heated at a temperature of 60-70 °C for 3 hours. After this, the resulting mixture was dried by heating on a hotplate at a temperature of 90-100 °C until it formed a gel. The drying process was then continued using an oven at a temperature of 120 °C for 3 days, which resulted in the formation of magnesium citrate powder. Afterward, the magnesium citrate product was calcined at a temperature of 800 °C for 1.5 hours, yielding MgO catalyst powder.

The synthesized MgO was characterized using Fourier Transform Infrared Spectroscopy (FTIR) to analyze the relationship between infrared waves and chemical bonds in the MgO. X-ray Diffraction (XRD) was utilized to analyze the crystal structure of the MgO. The specific surface area and pore distribution of the MgO was determined by N_2 physical adsorption The Brunauer-Emmett-Teller (BET) measure.

Deoxygenation of palm oil, palm wax and chicken fat

The deoxygenation reaction was conducted using a 150 mL stainless steel reactor[19]. The raw materials that were used include palm oil, palm wax, chicken fat, and stearic acid as model compounds. The deoxygenation reaction was carried out by adding 1.5 g of MgO catalyst and 25 g of raw material into the reactor, after which it was heated to a temperature of 300 °C for 1.5 hours, the reactor was opened, allowing the hot vapor to flow through the distillation column and condense in the sample container. The resulting liquid product is collected, weighed, and analyzed using GC-MS.

2. RESULTS AND DISCUSSION

Synthesis and Characterization of Magnesium Oxide

The FTIR spectra of synthesized magnesium citrate can be seen in Fig.1 Figure 1(a) and 1(b) displayed the FTIR spectra of magnesium citrate before calcination and after calcination, respectively. The results of calcination indicated the formation of MgO. The peak on FTIR spectra corresponding to MgO was observed at a wavenumber of approximately 450 cm^{-1} [20].

(a)   (b)
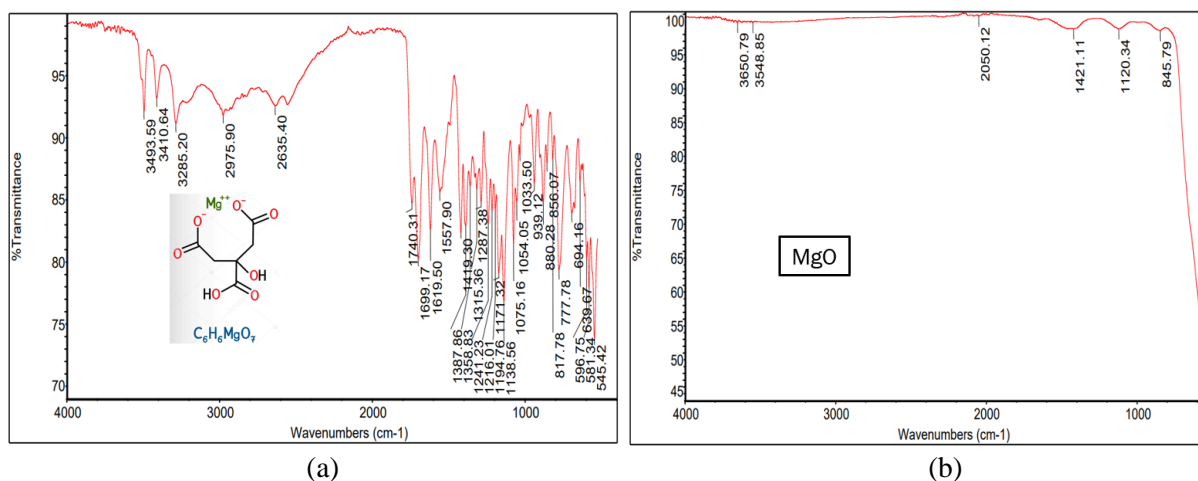
Figure 1. FTIR spectra of synthesized magnesium citrate: (a) before calcination (Magnesium Citrate) and (b) after calcination (MgO).
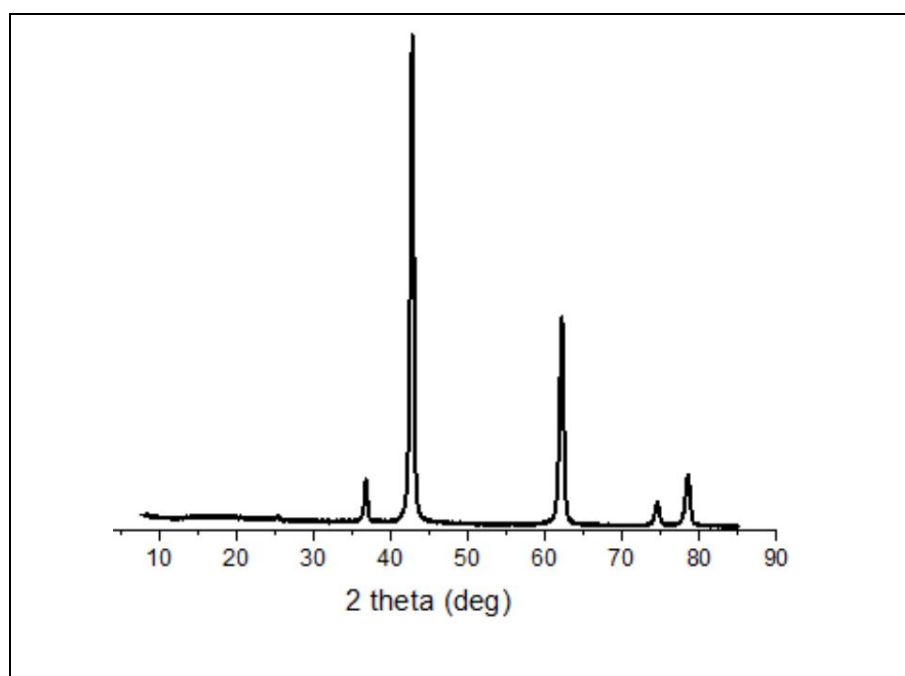

Figure 2. XRD patterns of synthesized MgO.

Figure 2 illustrates the XRD patterns of MgO. The XRD analysis results validate the successful production of the MgO catalyst, as all five characteristic MgO peaks are detected, aligning with the reference MgO (JCPDS No. 75-0447) [21]. These distinctive MgO peaks are observed at angles of 36.87°, 42.78°, 62.19°, 74.7°, and 78.42°. Furthermore, the synthesized MgO catalyst exhibits a notably high level of crystallinity, evidenced by the sharp peaks observed in the diffractogram. These findings are consistent with the research findings of Knoll et al., (2019) and Riyadhi et al., (2020). Additionally, the synthesized MgO catalyst possesses a surface area measuring 35.68 m^2g^{-1}, along with an average pore diameter of around 15.52 nm, classifying it as mesoporous.

The conversion reaction of stearic acid into bio hydrocarbons.

Stearic acid, also known as octadecanoic acid, is a saturated fatty acid that is solid at room temperature. The conversion of stearic acid into bio hydrocarbon was carried out using a stainless-steel batch reactor [23]. The liquid product obtained is 9.49 grams (37.96%), with the other products consisting of solid residue and gas. The GC-MS analysis results of the liquid product can be seen in Figure 3.
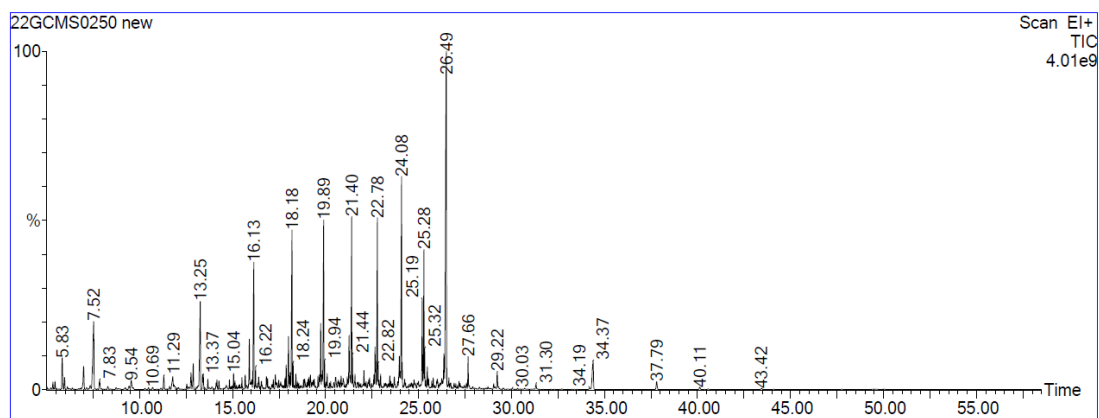
Figure 3. GC-MS analysis of the liquid product obtained from the deoxygenation of stearic acid into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

Table 1. Main components of the liquid product obtained from the conversion of stearic acid into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6 %.

No	Formula	Retention time	Compounds	Area (%)
1	$C_{17}H_{36}$	26.49	Heptadecane	18.87
2	$C_{15}H_{32}$	24.08	Pentadecane	7.34
3	$C_{12}H_{26}$	19.89	Dodecane	6.16
4	$C_{11}H_{24}$	18.18	Undecane	5.62
5	$C_{13}H_{28}$	21.40	Tridecane	5.05
6	$C_{14}H_{30}$	22.78	Tetradecane	4.83
7	$C_{10}H_{22}$	16.13	Decane	4.75
8	C_8H_{18}	7.52	Octane	4.69
9	C_9H_{20}	13.25	Nonane	4.33
10	$C_{16}H_{34}$	25.28	Hexadecane	3.56
11	$C_{16}H_{34}O$	25.19	2-Hexadecanol	2.25
12	$C_{12}H_{26}O$	34.38	2-Nonadecanone	2.18

Table 1 showed the main components of the liquid product obtained from the conversion of stearic acid into bio-hydrocarbon. The most abundant compound in the liquid product resulting from the conversion of stearic acid into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6% is heptadecane ($C_{17}H_{36}$) at 18.87%. Heptadecane was formed from the decarboxylation reaction of stearic acid [24]. In addition to the decarboxylation reaction, there was also a cracking reaction that forms shorter-chain compounds such as pentadecane ($C_{15}H_{32}$), dodecane ($C_{12}H_{26}$), undecane ($C_{11}H_{24}$), and other alkane compounds. Apart from alkane compounds, alcohol compounds such as 2-hexadecanol ($C_{16}H_{34}O$) and ketone compounds such as 2-nonadecanone ($C_{12}H_{26}O$) were also formed.

The liquid product is categorized into three categories based on the carbon chain length: gasoline (C_5-C_{11}), kerosene (C_{12}-C_{15}), and diesel (C_{16}-C_{22}). The highest composition of the liquid product is kerosene (35.43%), followed by gasoline (34.33%) and diesel (30.23%). The composition of gasoline, kerosene, and diesel products obtained from the conversion of stearic acid can be seen in Table 2.

Table 2. Composition of gasoline, kerosene, and diesel products from the liquid product obtained from the conversion of stearic acid into bio hydrocarbon.

Category	Compounds	Total Area (%)
Gasoline	C_5-C_{11} hydrocarbons	34.33
Kerosene	C_{12}-C_{15} hydrocarbons	35.43
Diesel	C_{16}-C_{22} hydrocarbons	30.23

The conversion reaction of palm oil into bio hydrocarbons

The GC-MS analysis results of the liquid product obtained can be seen in Figure 4.
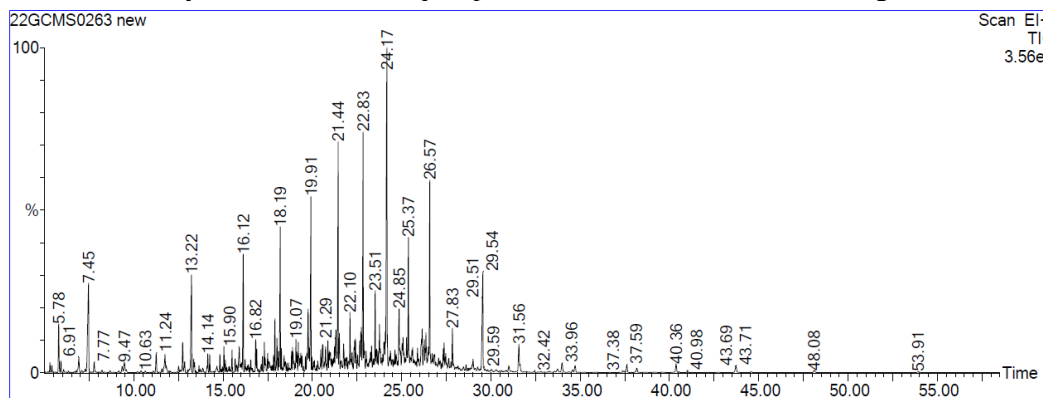
Figure 4. GC-MS analysis of the liquid product obtained from the deoxygenation of palm oil into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

Table 3. Main components of the liquid product obtained from the conversion of palm oil into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

No	Formula	Retention time	Compounds	Area (%)
1	$C_{15}H_{32}$	24.17	Pentadecane	13.18
2	$C_{14}H_{30}$	22.83	Tetradecane	5.86
3	$C_{13}H_{28}$	21.44	Tridecane	5.72
4	$C_{17}H_{36}$	26.57	Heptadecane	5.14
5	C_8H_{18}	7.45	Octane	5.07
6	$C_{17}H_{34}O$	29.54	2-Heptadecanone	4.75
7	$C_{12}H_{26}$	19.91	Dodecane	4.64
8	C_9H_{20}	13.22	Nonane	3.93
9	$C_{11}H_{24}$	18.19	Undecane	3.86
10	$C_{10}H_{22}$	16.12	Decane	3.42
11	$C_{16}H_{34}$	25.37	Hexadecane	3.19
12	$C_{15}H_{30}$	24.85	n-Nonylcyclohexane	2.33
13	$C_{17}H_{34}$	26.14	3-Heptadecene, (Z)-	2.02

In Table 3, it can be observed that the main compound present in the liquid product resulting from the conversion of palm oil into bio-hydrocarbon is pentadecane ($C_{15}H_{32}$). Pentadecane is formed through the decarboxylation reaction of palmitic acid ($C_{16}H_{32}O_2$) [25]. Palmitic acid ($C_{16}H_{32}O_2$) itself is a fatty acid commonly found in palm oil [26]. In addition to the decarboxylation reaction, there is also cracking, resulting in shorter hydrocarbon compounds. Apart from alkanes, small amounts of alkenes, cyclic compounds, and ketones are also found.

The conversion of palm oil into bio-hydrocarbon was carried out with a catalyst ratio of 6% MgO to palm oil at a temperature of 300°C for 90 minutes. The research results showed that 44.24% of the produced products were in liquid form. The liquid product consisted of kerosene, gasoline, and diesel, with the main compound found in the liquid product being pentadecane ($C_{15}H_{32}$). Based on Table 4, it can be observed that the largest proportion of the liquid product was kerosene (43.85%), followed by gasoline (34.08%), and diesel (22.07%).

Table 4. Composition of gasoline, kerosene, and diesel products from the liquid product obtained from the conversion of palm oil into bio hydrocarbon.

| Category	Compound	Total Area (%)
Gasoline	C_5-C_{11} hydrocarbons	34.08
Kerosene	C_{12}-C_{15} hydrocarbons	43.85
Diesel	C_{16}-C_{22} hydrocarbons	22.07

The conversion reaction of palm wax into bio hydrocarbon.

The GC-MS analysis results of the liquid product obtained can be seen in Figure 5 and Table 5. From Table 5, it is evident that the predominant compound present in the liquid product obtained from the conversion of palm wax is pentadecane ($C_{15}H_{32}$). Pentadecane is produced through the decarboxylation reaction of palmitic acid ($C_{16}H_{32}O_2$) [25]. Palmitic acid ($C_{16}H_{32}O_2$) is a fatty acid that is commonly found in wax or candles derived from palm oil. Apart from alkanes, small quantities of alkenes, cyclic compounds, and ketones are also generated.

In Table 6, it can be observed that the conversion of palm wax into bio-hydrocarbon results in a gasoline product of 58.25 %. Other products include kerosene at 32.34 % and diesel at 9.41 %. The research findings on the conversion of palm wax into bio-hydrocarbon using a 6 % MgO/palm wax catalyst ratio at a temperature of 300 °C for 90 minutes show a 38.16 % liquid product yield. The liquid product comprises gasoline, kerosene, and diesel, with the main compound being pentadecane ($C_{15}H_{32}$).
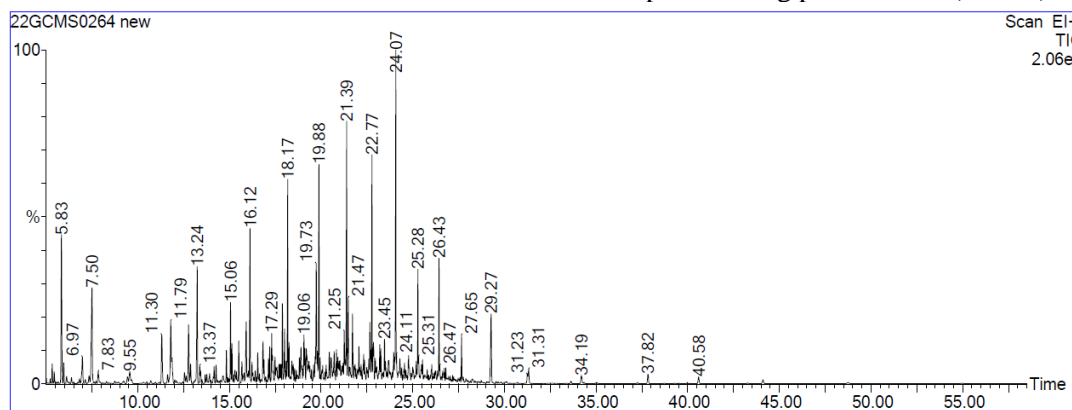
Figure 5. GC-MS analysis of the liquid product obtained from the deoxygenation of palm wax into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

Table 5. Main components of the liquid product obtained from the conversion of palm wax into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

No	Formula	Retention time	Compounds	Area %
1	$C_{15}H_{32}$	24.07	Pentadecane	7.92
2	$C_{13}H_{28}$	21.39	Tridecane	5.30
3	C_7H_8	5.83	Toluene	5.18
4	$C_{12}H_{26}$	19.88	Dodecane	4.87
5	C_8H_{18}	7.50	Octane	4.53
6	$C_{11}H_{24}$	18.17	Undecane	4.30
7	$C_{14}H_{30}$	22.77	Tetradecane	4.14
8	C_9H_{20}	13.24	Nonane	3.83
9	$C_{10}H_{22}$	16.12	Decane	3.79
10	$C_{10}H_8$	19.73	Naphthalene	3.33
11	$C_{17}H_{34}O$	29.27	2-Heptadecanone	3.11
12	C_8H_{10}	11.79	Benzene, 1,3-dimethyl-	2.43
13	$C_{17}H_{36}$	26.43	Heptadecane	2.31
14	$C_{11}H_{10}$	21.48	1H-Indene, 1-ethylidene-	2.24
15	C_8H_{10}	11.30	Ethylbenzene	2.16
16	C_9H_{12}	15.91	Benzene, 1,2,4-trimethyl-	2.09
17	$C_{10}H_{14}$	17.88	Benzene, 1-methyl-4-(1- methylethyl)-	2.09
18	C_8H_{10}	12.76	p-Xylene	2.01
19	$C_{16}H_34$	25.28	Hexadecane	2.00

Table 6. Composition of gasoline, kerosene, and diesel products from the liquid product obtained from the conversion of palm wax into bio hydrocarbon.

Category	Compound	Total Area (%)
Gasoline	C_5-C_{11} hydrocarbons	58.25
Kerosene	C_{12}-C_{15} hydrocarbons	32.34
Diesel	C_{16}-C_{22} hydrocarbons	9.41

The conversion reaction of chicken fat into bio hydrocarbon.

The GC-MS analysis of the liquid product obtained can be seen in Figure 6 and Table 7. In Table 7, it can be observed that the most abundant compound in the liquid product resulting from the conversion of chicken fat to hydrocarbons is pentadecane ($C_{15}H_{32}$). Pentadecane is formed through the decarboxylation reaction of palmitic acid ($C_{16}H_{32}O_2$) [25]. In addition to pentadecane, there are also significant amounts of other alkane compounds such as tetradecane ($C_{14}H_{30}$), tridecane ($C_{13}H_{28}$), octane (C_8H_{18}), and various other alkane compounds. Apart from alkane compounds, there are also small amounts of alkene, cyclic, and ketone compounds.

In Table 8, the liquid product resulting from the conversion of chicken fat contains a significant amount of kerosene, amounting to 43.59 %. This is followed by gasoline and diesel. The research findings on the conversion of chicken fat into bio hydrocarbon using a catalyst ratio of MgO/chicken fat of 6 % at a temperature of 300 °C for 90 minutes yielded a 35.12 % liquid product. The predominant liquid products obtained in sequence are kerosene, gasoline, and diesel, with the main compound in the liquid product being pentadecane ($C_{15}H_{32}$).
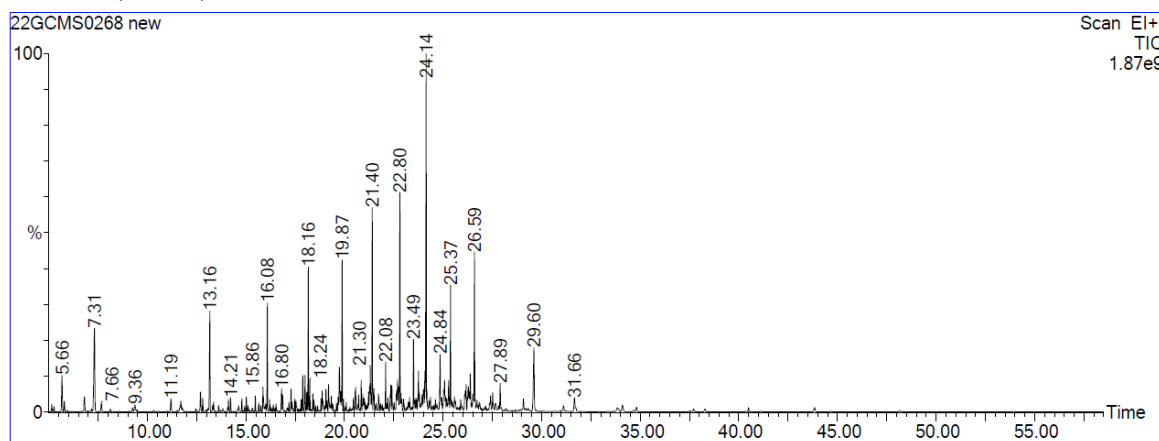
Figure 6. GC-MS analysis of the liquid product obtained from the deoxygenation of chicken fat into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

Table 7. Main components of the liquid product obtained from the conversion of chicken oil into bio hydrocarbon using MgO catalyst with a catalyst/feed ratio of 6%.

No	Formula	Retention time	Compounds	Area %
1	$C_{15}H_{32}$	24.14	Pentadecane	11.48
2	$C_{14}H_{30}$	22.80	Tetradecane	6.33
3	$C_{13}H_{28}$	21.40	Tridecane	5.92
4	C_8H_{18}	7.31	Octane	5.49
5	$C_{17}H_{36}$	26.59	Heptadecane	5.04
6	$C_{12}H_{26}$	19.87	Dodecane	4.35
7	C_9H_{20}	13.16	Nonane	4.28
8	$C_{11}H_{24}$	18.16	Undecane	3.97
9	$C_{17}H_{34}O$	29.60	2-Heptadecanone	3.91
10	$C_{16}H_{34}$	25.37	Hexadecane	3.52
11	$C_{10}H_{22}$	16.08	Decane	3.19
12	$C_{15}H_{30}$	24.84	n-Nonylcyclohexane	2.62
13	$C_{14}H_{28}$	23.49	Cyclopentane, nonyl-	2.10

Table 8. Composition of gasoline, kerosene, and diesel products from the liquid product obtained from the conversion of chicken fat into bio hydrocarbon.

Category	Compound	Total Area (%)
Gasoline	C_5-C_{11} hydrocarbons	32.09
Kerosene	C_{12}-C_{15} hydrocarbons	43.59
Diesel	C_{16}-C_{22} hydrocarbons	24.32

Table 9. Comparison of liquid product yields

Raw materials	Liquid product yields (%)
Palm oil	44.24
Palm wax	38.16
Stearic acid	37.96
Chicken fat	35.12

Comparison of the composition of products resulting from the deoxygenation of stearic acid, palm oil, palm wax, and chicken fat

The comparison of liquid product yields obtained from various raw materials can be seen in Table 9. The highest liquid product yield was consecutively obtained from the conversion of palm oil, followed by palm wax, stearic acid, and chicken fat.

In Table 10, it can be observed that the highest yield of gasoline is obtained from palm wax as the raw material, followed by stearic acid, palm oil, and chicken fat. The highest yield of kerosene is produced using palm oil as the raw material, followed by chicken fat, stearic acid, and palm wax. The highest yield of diesel is obtained using stearic acid as the raw material, followed by chicken fat, palm oil, and palm wax.

In Table 11, it can be observed that heptadecane is predominantly produced from stearic acid as the raw material, while pentadecane is produced in significant quantities from palm oil, palm wax, and chicken fat as the raw materials. The compositions of other products, such as octane, nonane, decane, undecane, dodecane, tridecane, and tetradecane, show minor differences, with insignificant variations in their amounts. However, the notable feature is the substantial production of pentadecane from palm oil and chicken fat, while heptadecane is predominantly produced from stearic acid. Similar with Simakova et al., (2009)[27], the catalytic deoxygenation of a mixture of palmitic and stearic acids using Pd catalysts at temperatures ranging from 260 to 300 °C resulted in the main products, specifically n-heptadecane and n-pentadecane.

Table 10. Comparison of the composition of gasoline, kerosene, and diesel products.

Product		Raw Material			
		Stearic acid	Palm oil	Palm wax	Chicken fat
Gasoline (C_5-C_{11})	(%)	34.33	34.08	58.25	32.09
Kerosene (C_{12}-C_{15})	(%)	35.43	43.85	32.34	43.59
Diesel (C_{16} -C_{22})	(%)	30.23	22.07	9.41	24.32

Table 11. Comparison of the composition of alkane compounds in liquid products

Formula	Compound		Raw Material			
			Stearic acid	Palm oil	Palm Wax	Chicken fat
C_8H_{18}	Octane	(%)	4.69	5.07	4.53	5.49
C_9H_{20}	Nonane	(%)	4.33	3.93	3.83	4.28
$C_{10}H_{22}$	Decane	(%)	4.75	3.42	3.79	3.19
$C_{11}H_{24}$	Undecane	(%)	5.62	3.86	4.30	3.97
$C_{12}H_{26}$	Dodecane	(%)	6.16	4.64	4.87	4.35
$C_{13}H_{28}$	Tridecane	(%)	5.05	5.72	5.30	5.92
$C_{14}H_{30}$	Tetradecane	(%)	4.83	5.86	4.14	6.33
$C_{15}H_{32}$	Pentadecane	(%)	7.34	13.18	7.92	11.48
$C_{16}H_{34}$	Hexadecane	(%)	3.56	3.19	2.00	3.52
$C_{17}H_{36}$	Heptadecane	(%)	18.87	5.14	2.31	5.04

3. CONCLUSIONS

The MgO catalyst was produced through the calcination process of magnesium citrate, which was formed from the reaction between magnesium carbonate and citric acid. This catalyst has a crystalline structure with mesoporous pores. Additionally, the catalyst has a surface area of $35.68 \ m^2g^{-1}$. In the context of gasoline production, the highest yield is obtained from palm wax as the raw material, followed by stearic acid, palm oil, and chicken fat. MgO is a basic compound that can act as a catalyst to remove carboxyl groups (-COOH) from fatty acid molecules. When fatty acids react with MgO under high-temperature conditions, a decarboxylation reaction occurs, transforming the fatty acids into lighter compounds. MgO has the ability to adsorb fatty acid molecules, enabling the decarboxylation reaction to take place on the surface of MgO. This adsorption aids in facilitating the interaction between fatty acids and MgO, thereby promoting a more efficient reaction.

ACKNOWLEDGMENTS

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