



Possibility of Using Commercial Hydrated Lime as a Catalyst for Rapeseed Oil Methanolysis

Azra Halilović^a, Sabina Begić^{b*}, Zoran Iličković^b, Amra Odošić^b,
Sanja Panić^c, Zorica Stojanović^d and Amir Fazlić^e

^a Faculty of Metallurgy and Technology, University of Zenica, Travnička Cesta 1, 72000 Zenica, Bosnia and Herzegovina.

^b Faculty of Technology, University of Tuzla, Urfeta Vejzagića 8, 75000 Tuzla, Bosnia and Herzegovina.

^c Faculty of Technology, University of Novi Sad, Bulevar cara Lazara 1, 21000 Novi Sad, Serbia.

^d Victoriaoil Ltd., Branka Erića br. 2, 22240 Šid, Serbia.

^e Faculty of Science, University of Sarajevo, Zmaja od Bosne 33-35, 75000 Sarajevo, Bosnia and Herzegovina.

Authors' contributions

This research was carried out in collaboration among all authors. Authors SB, ZI and AO have designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors AH and ZS performed experimental research and the laboratory analyses. Author SP performed the analysis and writing of the catalyst characterization results. Author AF performed the literature searches. All authors have read and approved the final manuscript.

Article Information

DOI: 10.9734/IRJPAC/2021/v22i1130442

Open Peer Review History:

This journal follows the Advanced Open Peer Review policy. Identity of the Reviewers, Editor(s) and additional Reviewers, peer review comments, different versions of the manuscript, comments of the editors, etc are available here: <https://www.sdiarticle5.com/review-history/83907>

Original Research Article

Received 20 October 2021
Accepted 26 December 2021
Published 28 December 2021

ABSTRACT

In this research, the possibility of using $\text{Ca}(\text{OH})_2$ in the form of commercial hydrated lime as a catalyst for the methanolysis of refined rapeseed oil was evaluated. Characterization of unused catalyst was performed by SEM-EDS, laser diffraction, XRF, BET, XRD and TG/DTG methods, and vegetable oil was analyzed for physicochemical characteristics. Within methanolysis, the effects of catalyst loading, reaction temperature and reaction time on methyl ester yield, density and viscosity of biodiesel, as well as the possibility of catalyst reuse were investigated. The obtained results showed that hydrated lime is an efficient catalyst for transesterification of vegetable oil, whereby the use of 3 wt% of catalyst relative to oil weight, at temperature 60°C and mixing rate 1000 rpm after 120 min of transesterification reaction achieved a yield of methyl esters 98.76%. With three reuses of hydrated lime, without intermediate washing and regeneration procedures, the yield of rapeseed oil methyl ester remained above 90%.

*Corresponding author: E-mail: sabina.begic77@gmail.com;

Keywords: Biodiesel; methanolysis; heterogeneous catalyst; hydrated lime.

1. INTRODUCTION

Depletion of fossil fuel stocks and environmental degradation related to their production and combustion have resulted in an exponential growth in interest in biofuels, among which biodiesel has attracted significant attention [1] due to favorable degradability and toxicity [2,3], engine performance and reductions in air pollutant emissions [4]. Biodiesel is a liquid biofuel, which consists of alkyl esters of fatty acids from lipid raw materials [5] such as vegetable oils, animal fats or used cooking oils [6]. Among the existing techniques of biodiesel production, transesterification of vegetable oils with alcohol in the presence of catalysts is conventionally used [7], which converts oil triglycerides into alkyl esters, resulting in biodiesel as the main product and crude glycerol as the main by-product [8]. This reaction is shown in Fig. 1. The process is also known as alcoholysis, ie methanolysis, since methanol is the most commonly used alcohol [9,10]. The use of catalysts increases the solubility of alcohol in oil and thus accelerates the transesterification reaction [11]. Important reaction conditions for efficient biodiesel production include: molar ratio of alcohol to oil, catalyst type and loading, temperature and reaction time [12,13]. Although significant advances have been made in biodiesel production technology, its commercialization continues to rely on the availability of suitable catalysts [14] for which, in addition to high catalytic performance, rapid separation and efficient post-reaction catalyst recycling are considered important requirements [15]. Commercial processes of biodiesel synthesis by transesterification are usually carried out with homogeneous alkaline catalysts, such as sodium hydroxide (NaOH) and potassium hydroxide (KOH), because they are low cost and allow production under low pressure and temperature conditions, with high conversion rate [16]. However, the total costs of biodiesel

production based on homogeneous catalysis are still not competitive enough compared to those of petroleum diesel production [17], because industrial processes face not only the challenges of using catalysts without regeneration, but also the difficulty of removing them from the media [18], resulting in significant consumption of washing water, as well as wastewater production [19]. High energy consumption during separation and purification processes is the main disadvantage of using a homogeneous catalytic system [20]. The use of heterogeneous catalysts in industrial biodiesel production could reduce its cost and make it competitive with fossil diesel, given the many advantages these catalysts offer, including higher reaction rates, easy separation from the product and reusability [21]. Therefore, research efforts have been made in recent decades to find efficient and affordable heterogeneous catalysts. Calcium oxide (CaO) has been used as a catalyst in a large number of published studies of heterogeneous base-catalyzed transesterification [22], due to its high catalytic activity under mild reaction conditions, wide availability and low cost [23,24]. Compared to CaO, there are significantly fewer published studies on the use of calcium hydroxide, Ca(OH)₂, for methanolysis of vegetable oils, and their findings are contradictory in terms of the achieved yield of methyl esters [25-28].

In this research, the possibility of using Ca(OH)₂ in the form of commercial hydrated lime as a catalyst for the methanolysis of rapeseed oil was evaluated. The effects of catalyst concentration, temperature and reaction time on the yield of fatty acid methyl esters (FAME), density and viscosity of biodiesel, as well as the possibility of catalyst reuse were investigated. After detailed physico-chemical characterization of the hydrated lime (HL), a correlation between its properties and achieved catalytic activity has been established.

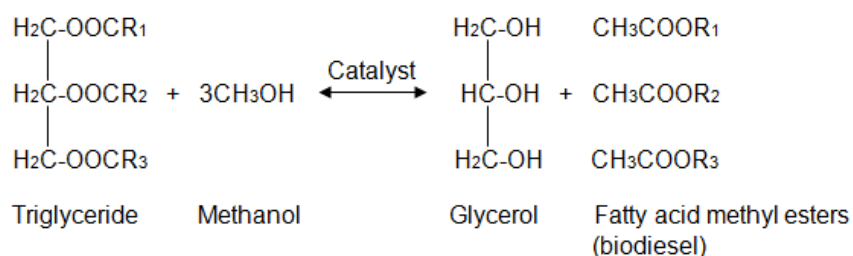


Fig. 1. General chemical reaction showing transesterification of triglycerides

2. MATERIALS AND METHODS

Refined rapeseed oil (*Brassica napus* L.) was procured from a domestic producer (Bimal Brčko, B&H). Commercial hydrated lime, used as a catalyst in this study, was also acquired from a domestic producer (Ingram Srebrenik, B&H). Methanol p.a. was purchased from Fluka. Oil, methanol and hydrated lime were used without further purification. All other analytical grade chemicals used were commercially obtained. The flowchart of the research methodology is shown in Fig. 2.

2.1 Rapeseed Oil Analysis

The methods of the International Organization for Standardization (ISO), listed in Table 1, were used to determine the physical and chemical properties of refined rapeseed oil. The following instruments and apparatus were used for oil analysis: Anton-Paar model DMA 35N digital densimeter for density determination, Ostwald-type viscometer for measuring kinematic viscosity, Abbe refractometer for determining refractive index, Agilent Technologies 7890A (G3440A) gas chromatograph with Agilent 7683 automatic liquid sampler for identification and quantification of fatty acids.

2.2 Characterization of Hydrated Lime

Elemental analysis of hydrated lime was performed by fluorescent X-ray analysis, using S8 TIGER Series 2 spectrometer, with characteristics: X-ray tube operating voltage of 40 kV, maximum current of 10 mA and maximum X-ray tube power of 400 W. The moisture content of the hydrated lime was determined by drying the sample in an oven for 24 h at a temperature of 105°C. Particle size analysis was performed by laser diffraction, using a Malvern Mastersizer 2000 analyzer. Brunauer-Emmet-Teller (BET) surface area, pore size and pore volume of the catalyst were measured from the multi-point nitrogen adsorption-desorption isotherm (at -196°C) using Micrometrics Gemini VII 2390. Determination of morphological properties of hydrated lime was performed by scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS), using JEOL JSM 6420 LV scanning electron microscope, with an acceleration voltage of 20 kV. The sample was coated with a 15 nm layer of gold, with a density of 19,32 g/cm³. The structural properties of the catalyst were analyzed by X-ray diffractometry. Diffractograms of the samples were recorded on

an automatic X-ray powder diffractometer, Philips PW1710. Diffractometer settings were 40 kV, 30 mA, a scan range of 2-80° (2θ) with a step size of 0.02° and a counting time of 1 s per step. Thermal analysis of hydrated lime was performed using a Q600 thermal analyzer (TA Instruments, New Castle, DE, USA). The sample was analyzed in a corundum pot under an inert atmosphere (nitrogen flow rate 100 cm³/min) in the temperature range 20 to 1000°C (heating rate 20°C/min). The weight of the sample was 3 mg.

2.3. Rapeseed Oil Methanolysis

Methanolysis experiments were conducted in a 500 cm³ three-neck round-bottomed glass flask, coupled to a water cooling recirculation system. Stirring and heating were performed using a mechanical stirrer and a hot plate. First, 62.5 cm³ of methanol and a certain amount of HL were added to the flask, after which 250 cm³ of refined rapeseed oil was added and the mixture was subjected to constant stirring (1000 rpm) at certain temperature and reaction time.

At the end of the reaction period, the catalyst was removed from the mixture by filtration using a vacuum pump and the liquid phase was transferred to a separation funnel where it was left for 24 h to allow biodiesel (methyl esters) and glycerol fractions to separate based on their different specific weights. After removal of the glycerol (lower) phase by discharge from the funnel, separation of fine catalyst particles and residual glycerol from the biodiesel fraction was performed by centrifugation (3000 rpm) using Eppendorf Centrifuge 5702, for 10 min at room temperature. The biodiesel fraction thus obtained was further analyzed for FAME content, density and viscosity.

Methanolysis experiments were carried out at different amounts of hydrated lime in relation to the mass of oil (2, 3, 4 and 5 wt%), at different temperatures (25, 45 and 60°C) and reaction times (60, 120 and 180 min). The methanolysis conditions that gave the highest yield of FAME were taken as optimal and applied in the catalyst reusability research, whereby previously used hydrated lime, after separation from the liquid phase and without subsequent regeneration, was added to the flask with fresh reactants.

Determination of fatty acid methyl ester content in samples obtained after methanolysis and separation from catalyst and glycerol was

performed by gas chromatography according to ISO 5508 method [39]. The density and viscosity

of the samples were additionally determined by the methods used for rapeseed oil.

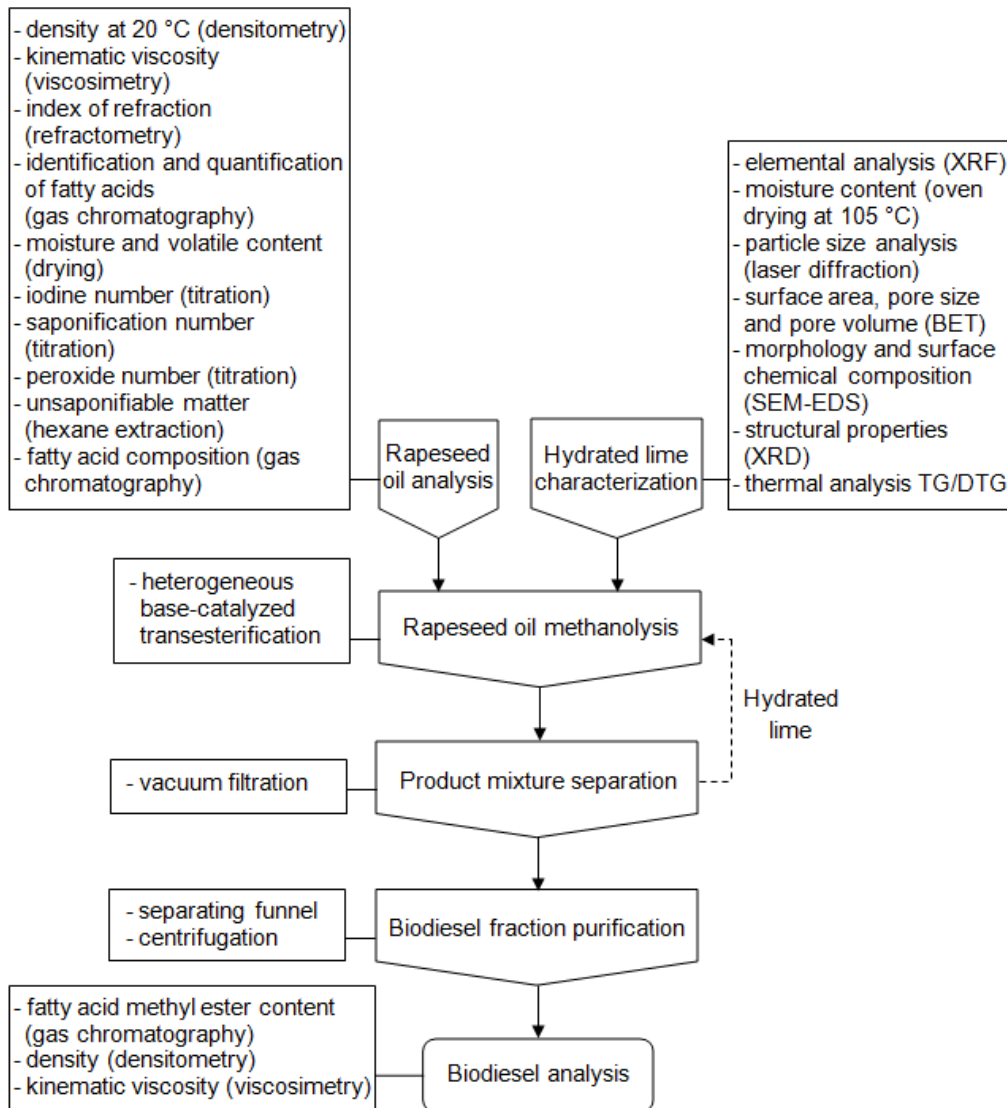


Fig. 2. Research methodology flowchart

Table 1. Methods of analysis of physical and chemical properties of rapeseed oil

Property	Method	Reference
Density	ISO 6883	[29]
Kinematic viscosity	ISO 3104	[30]
Index of refraction	ISO 6320	[31]
Free fatty acids content	ISO 660	[32]
Moisture and volatile content	ISO 662	[33]
Iodine number	ISO 3961	[34]
Saponification number	ISO 3657	[35]
Peroxide number	ISO 3960	[36]
Unsaponifiable matter	ISO 18609	[37]
Fatty acid composition	ISO 5509	[38]
	ISO 5508	[39]

3. RESULTS AND DISCUSSION

3.1 Rapeseed Oil Properties

The results of the refined rapeseed oil analysis are shown in Tables 2 and 3. The oil characteristics presented in Table 2 have values more or less similar to those published by other researchers [40-43]. While some properties, such as density, are naturally given and characterize rapeseed oil, others depend on the production process, from plant cultivation to oil extraction, cleaning and storage [44], which may explain the differences among analysis results published by researchers. The values of density, viscosity, iodine number, free fatty acids content and water content of the analyzed oil in this paper are within the limit values of the established properties for the use of rapeseed oil as a fuel for internal combustion engines compatible with vegetable oil [45]. However, high viscosity, which is generally 10-20 times higher in vegetable oils than in petroleum diesel [46], is a major obstacle to the direct application of this oil in unmodified diesel engines, as high viscosity affects fuel

dispersion, cold flow properties and causes the formation of carbon deposits on the internal components of engines [47].

Since biodiesel is a mixture of fatty acid esters, and taking the general assumption that the existing fatty acid profiles of the feedstock remain unchanged after transesterification [48], it follows that the properties of biodiesel largely depend on the fatty acid profile of starting material. The results of gas chromatography of rapeseed oil (Table 3) show the dominant content of four fatty acids (oleic, linoleic, linolenic and palmitic). From the aspect of desirable profile of fatty acids in raw materials for biodiesel production, given the influence of their methyl esters on fuel properties, high presence of monounsaturated fatty acids (such as oleic and palmitoleic acid), reduced presence of polyunsaturated, and controlled content of saturated fatty acids are recommended [49]. In this regard, the analyzed rapeseed oil has a favorable fatty acid composition, showing a total content (wt%) of monounsaturated acids 63.64, polyunsaturated 29.66 and saturated 6.7.

Table 2. Physical and chemical properties of refined rapeseed oil

Property	Unit	Value
Density (20°C)	kg/dm ³	0.917
Kinematic viscosity (40°C)	mm ² /s	30.81
Index of refraction	dimensionless	1.465
Free fatty acids content	wt%	0.02
Moisture and volatile content	%	0.03
Iodine number	dimensionless	110
Saponification number	mg KOH/g	186
Peroxide number	meq O ₂ /kg	0.89
Unsaponifiable matter	g/kg	9

Table 3. Rapeseed oil fatty acid profile

Fatty acid	Symbol	Content [wt%]
Myristic	C14:0	0.1
Palmitic	C16:0	4.12
Palmitoleic	C16:1	0.22
Stearic	C18:0	1.34
Oleic	C18:1	60.76
Linoleic	C18:2	20.9
Linolenic	C18:3	8.7
Arachidic	C20:0	0.68
Eicosenoic	C20:1	1.76
Eicosadienoic	C20:2	0.06
Behenic	C22:0	0.32
Erucic	C22:1	0.9
Lignoceric	C24:0	0.14

3.2 Catalyst Characterization

The determined moisture content of hydrated lime is 1.73 wt%. The surface morphology of commercial hydrated lime used as a catalyst is shown in Fig. 3. The present image reveals the nonuniform morphology in the form of aggregates (clusters) which consist of irregularly stacked angular crystals of $\text{Ca}(\text{OH})_2$. These aggregates are characterized by a great diversity in size and shapes as a consequence of unbalanced growth of hydroxide crystals from CaO during hydration. Additionally, due to random arrangement of the formed clusters, unevenly distributed porous regions (gaps and voids) have been created making the "open" structure of the hydrated lime [50]. The SEM analysis was done in combination with X-ray energy dispersion (EDS) analysis (Fig. 4) revealing the elemental composition (wt%) of the lime sample: 54.30% Ca, 39.05% O, 5.28% C, while the presence of Mg was detected in the amount less than 1%. The carbon registered in the sample might be an indication of the carbonate phase (CaCO_3) present in the sample. This will be confirmed by additional analysis below.

A wide range of particle/aggregate sizes, previously observed by SEM analysis, was verified by the particle size distribution (PSD) data shown on Fig. 5. Namely, the sizes of the particles of the hydrated lime are within a very broad range ($0.9 \mu\text{m} - 800 \mu\text{m}$) and can be characterized by monomodal volume distribution with maximum around $8 \mu\text{m}$. Only 10 vol% of the sample particles have diameter less than $2.8 \mu\text{m}$, while 50 vol% of them are smaller than $9.0 \mu\text{m}$. Additionally, since the 90 vol% of the particles are characterized by the diameter below $65.4 \mu\text{m}$, the average diameter is shifted towards lower value and amounts to $29.5 \mu\text{m}$.

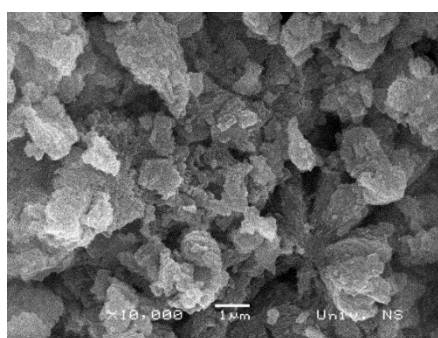


Fig. 3. SEM image of hydrated lime

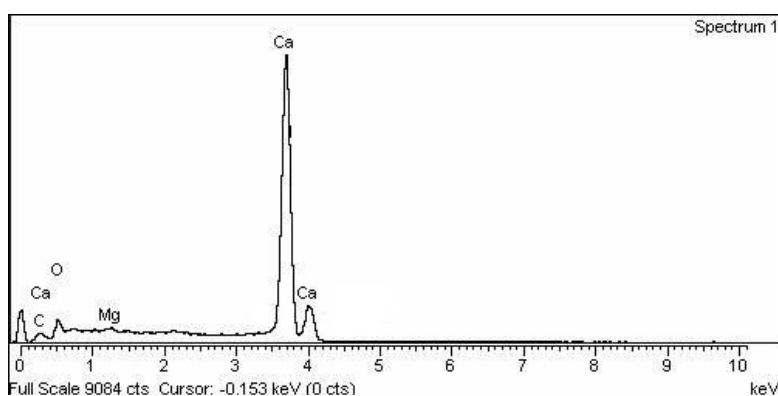


Fig. 4. EDS spectrum of hydrated lime

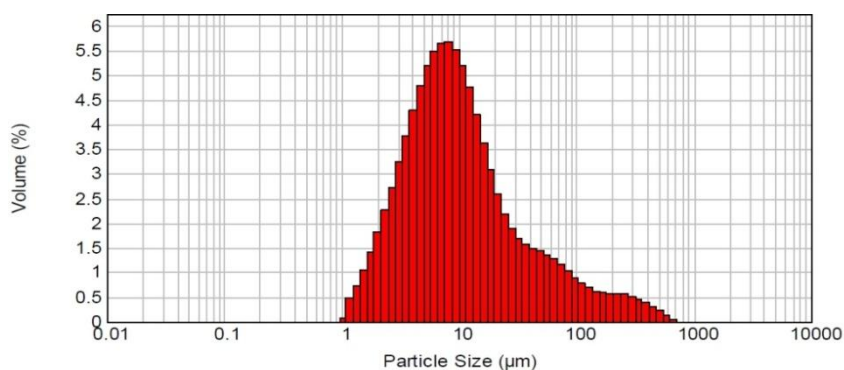


Fig. 5. Particle size distribution results of hydrated lime

Table 4. Chemical composition of hydrated lime obtained by XRF

Chemical composition	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	K ₂ O	TiO ₂	P ₂ O ₅	LOI*	Sum
wt%	0.53	0.35	0.21	68.92	1.02	0.29	0.04	0.002	0.02	26.68	98.06

*Loss on ignition

Chemical composition (in the form of oxides) of hydrated lime was determined by XRF analysis and the results were presented in Table 4. As can be seen, the sample consists mainly of CaO (68.92%), while MgO is detected in significantly lower concentration (1.02%). Other elements are present in very small amounts. The obtained LOI can be associated with the removed water and CO₂ from Ca(OH)₂ and CaCO₃ after sample exposure to high temperature.

The textural parameters of the hydrated lime are presented in Table 5. The sample has specific surface area of 25.4 m²/g and is characterized by relatively low total pore volume implying its undeveloped porous structure. This is also indicated by a high share of external surface area (21.8 m²/g) in BET area. According to the result of average pore diameter (2.1 nm), it can be suggested that the examined hydrated lime consists of certain portion of micropores along with the portion of pores in the meso-domain with diameter reaching the micro-range.

The XRD pattern of hydrated lime is given on Fig. 6. The diffractogram shows two components of the catalyst: portlandite – Ca(OH)₂ (COD database code: 1008781, Reference code 96-100-8782), with the most intensive peak around 2θ value of 18° and calcite – CaCO₃ (COD database code: 9000968, Reference code 96-900-0969), with the most intensive peak at 2θ

value between 29° and 30°. Since the calcite is identified by this method, the previous assumption from EDS results regarding its presence can be confirmed. According to the XRD results of quantitative analysis, the sample contains 96.16 wt% of portlandite, corresponding to 72.78 wt% of CaO. This coincided well with the XRF results.

In order to test the thermochemical changes, TG/DTG analysis of hydrated lime was performed, and the appropriate profile is shown on Fig. 7. The sample of hydrated lime is characterized by a three-step weight loss profile during heating in the flow of N₂ up to 800°C. The first step (up to 250 – 300°C) corresponds to the weight loss of 5.4% and can be ascribed to the loss of adsorbed water. With further heating up to 500°C, the mass of the sample reduces by additional 22.3%, which is due to the loss of structurally bound water (dehydroxylation of Ca(OH)₂) [51]. In the third step (in the temperature range 500°C - 770°C), the weight of the sample decreases for extra 5.2%, as a consequence of CaCO₃ decomposition into CaO and CO₂ [52,53]. The thermal changes at higher temperatures (over 800°C) indicate the impurities present in the sample along with the formed CaO. The results of TG analysis are consistent with other characterization results previously displayed.

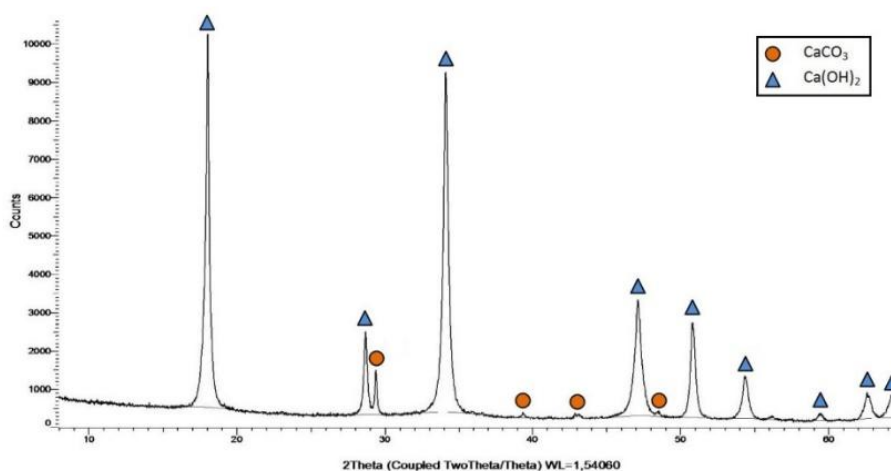
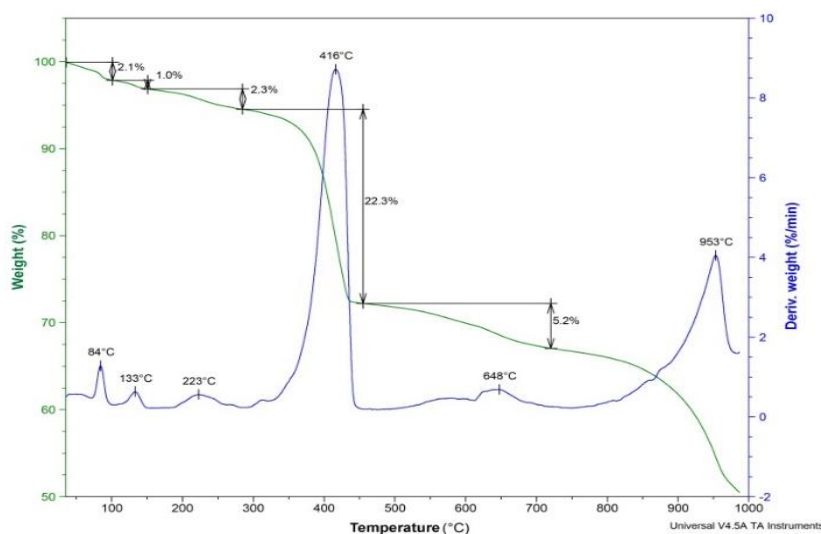
**Fig. 6. XRD pattern of hydrated lime**

Table 5. Textural parameters of hydrated lime

Parameter	BET (m ² /g)	Average pore diameter (nm)	Total pore volume (cm ³ /g)
Hydrated lime	25.4	2.1	0.016

**Fig. 7. TG/DTG profile of hydrated lime**

3.3 Influence of Methanolysis Parameters on FAME Yield and Properties

The effect of catalyst loading on FAME yield is shown in Fig. 8. Measured pH values of mixtures of methanol and catalyst (at catalyst concentrations of 2, 3, 4 and 5 wt%) immediately before the addition of oil were: 12.23, 12.26, 12.27 and 12.30, which indicates small differences. However, by increasing the hydrated lime loading (relative to oil weight) 2 to 3 wt% in oil methanolysis increased the yield of FAME 90.02 to 98.76%, which confirms the observed importance of catalyst concentration in transesterification yield optimization [54]. The lower yield, which was obtained at lower catalyst loading (2 wt%), can be explained by the insufficient number of active sites for the reaction of the reactants [18]. This is confirmed by the results of sunflower oil methanolysis [55], where 1 wt% Ca(OH)₂, after 120 min resulted in biodiesel yield <10%, while yield above 90% was obtained with 5 wt% of catalyst loading. Methanolysis of used soybean oil showed that the increase in conversion rate was proportional to the dose of hydrated lime [27]. In the study of methanolysis of refined sunflower oil [26], it was concluded that the applied concentration of Ca(OH)₂ (equivalent to 0.2% NaOH) was low, but even at higher catalyst concentrations (equivalent to 1% NaOH), oil conversions

remained modest (<70% after 24 h), which was explained in term of low specific surface area of the catalyst (0.9 m²/g). From that aspect, the higher yield of methyl ester obtained in this study can be associated with a significantly higher specific surface area of hydrated lime (25.4 m²/g). The maximum FAME yield of 98.76%, obtained using 3 wt% of hydrated lime, complies with the minimum requirements of the FAME content (96.5%) for biodiesel, according to the European standard EN 14214 [56]. At catalyst loadings 4 and 5 wt%, rapeseed oil methyl ester yields were 95.77 and 94.33%, respectively, indicating a decrease in reactant conversion. A declining trend in biodiesel yield after reaching a certain maximum value has also been observed in other studies of heterogeneous methanolysis [57-59]. Although the addition of a catalyst accelerates the transesterification reaction by increasing the contact surface between reactants [60], its excessive amount is related to mass transfer limitations caused by increased viscosity of the mixture [61] and adhesion of the product to the catalyst surface [62].

The effect of catalyst loading on density and viscosity of biodiesel from rapeseed oil is shown in Fig 9. HL loadings, applied in the range 2 - 5 wt%, had practically no effect on the density of the obtained biodiesel (0.882 - 0.883 g/cm³), because methanol, oil and esters have a very

similar density [63]. The determined FAME density values meet the European standard for biodiesel [56] which limits them to the range 860 - 900 kg/m³. Methanolysis of rapeseed oil with a kinematic viscosity of 30.81 mm²/s resulted in biodiesel viscosities in the range 3,215 - 2,640 mm²/s, thus confirming that after the transesterification process the viscosity values of vegetable oil methyl esters are significantly low [64].

In addition, an inverse correlation was observed between viscosity and FAME yield, i.e. higher yields resulted in lower viscosity. The determined viscosity values are slightly lower than those obtained by other researchers [40,41,65,66]. The

lowest determined viscosity value (2,640 mm²/s) meets the American standard for biodiesel D6751 [67], however, according to the European standard [56] it is lower than the minimum specified (3.5 mm²/s). Although low viscosity of biodiesel is generally desirable because it facilitates the handling of fuels at lower temperatures [68], too low values of viscosity are associated with power loss and excessive wear of fuel injection pumps [69]. The reason for such low viscosity of obtained FAME may be the insufficient removal of methanol after transesterification, because the presence of this alcohol in biodiesel can further reduce its viscosity [70].

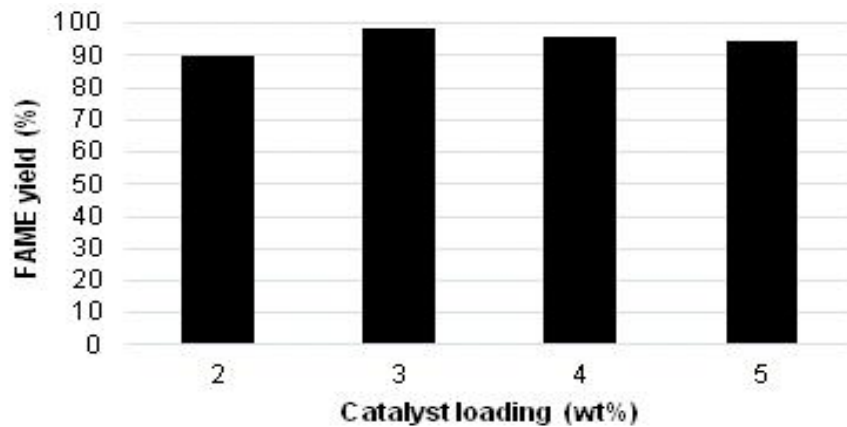


Fig. 8. Effect of HL loading on FAME yield

Process conditions: methanol:oil volume ratio 0.25, reaction temperature 60°C, reaction time 120 min

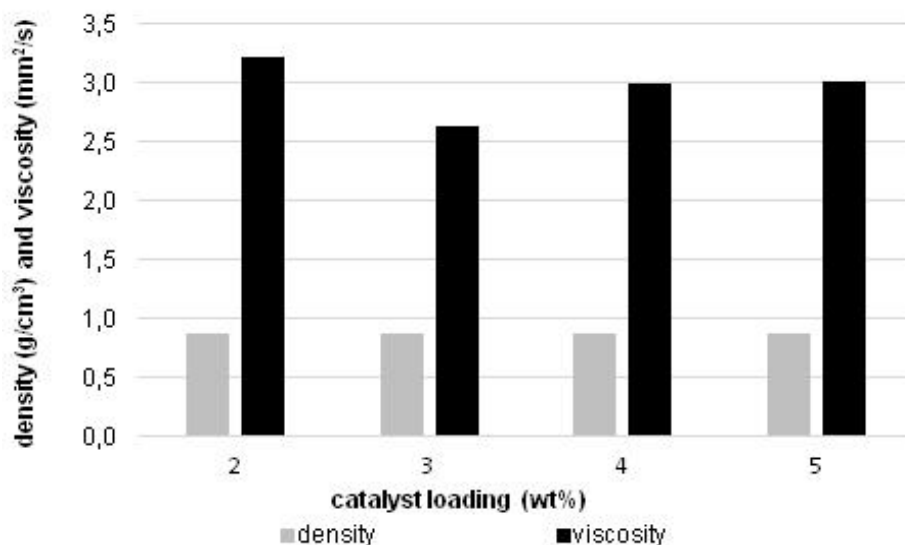


Fig. 9. Effect of HL loading on FAME yield on biodiesel density and viscosity

Process conditions: methanol:oil volume ratio 0.25, reaction temperature 60°C, reaction time 120 min

The effect of transesterification reaction time on FAME yield is shown in Fig. 10. During the first 60 minutes of methanolysis, a methyl ester yield of 68.04% was obtained, which increased to 98.76% during the additional 60 minutes, indicating the importance of ensuring sufficient time for the conversion of reactants into a product. The yield of FAME achieved after 120 min meets the yield requirement of the European standard for biodiesel [56], however, the yield obtained after 180 min is slightly lower (96.4%) than the minimum prescribed. This trend of increasing yield over time to a certain point, after which it decreases, was also observed in the methanolysis of sunflower oil [28] using hydrated lime acquired from the same producer. The decrease in yield that occurs with an excessive increase in reaction time is related to the reversibility of the transesterification reaction [71-73].

The effect of transesterification reaction time on density and viscosity of biodiesel from rapeseed oil is shown in Fig 11. The results of the densities of biodiesel obtained after 60, 120 and 180 min reaction time (0.898, 0.882 and 0.884 g/cm³) show that the density of vegetable oil is reduced by transesterification [74]. Since the viscosities of biodiesel after 60 and 180 min of methanolysis were higher (5.776 and 3.116 mm²/s) than after 120 min (2.64 mm²/s), and the FAME yields after 60 and 180 min were lower (68.04 and 96.4%) than those after 120 min (98.76%), an inverse correlation between viscosity and the achieved biodiesel yield at all given periods of methanolysis is observed. The viscosities of biodiesel obtained after all three examined transesterification periods meet the American standard for biodiesel [67], while their values are out of the range specified by the European standard [56].

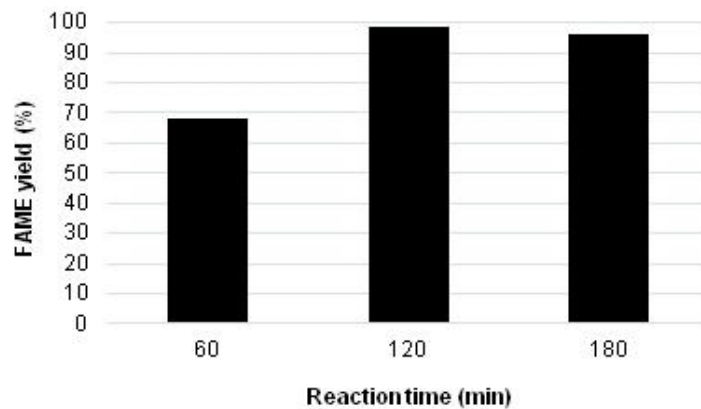


Fig. 10. Effect of reaction time on FAME yield

Process conditions: methanol:oil volume ratio 0.25, catalyst loading 3 wt%, reaction temperature 60°C

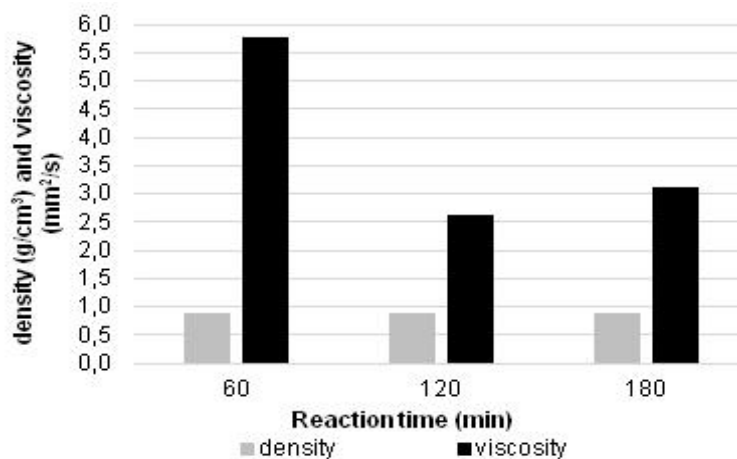


Fig. 11. Effect of reaction time on biodiesel density and viscosity

Process conditions: methanol:oil volume ratio 0.25, catalyst loading 3 wt%, reaction temperature 60°C

The effect of transesterification reaction temperature on FAME yield is shown in Fig. 12. Conducted methanolysis experiments at temperatures of 25, 45 and 60°C, in reaction time of 120 min resulted in the exponential growth of FAME yield, that is, an almost negligible yield (4.08%) was achieved at the lowest temperature, and increment the temperature to 45°C and 60°C, gave higher yields (25.12 and 98.76%). The increase in yield with reaction temperature is the result of a higher reaction rate and facilitated contact between the reactants, due to the decrease in oil viscosity [75]. The observed yield trend is similar to that of methanolysis of soybean oil using hydrated lime [27], where the authors reported that no conversion was achieved at a transesterification temperature of 25°C, while FAME yields were increasing (2, 19, 57 and 100%) with higher reaction temperatures (35, 45, 55 and 60°C). Although the higher temperature is necessary for increasing the reaction rate, conducting methanolysis at temperatures well above the boiling point of alcohol results in higher energy consumption, leads to alcohol loss by evaporation [76], and can accelerate triglyceride saponification [77], which will reduce the yield of biodiesel.

The effect of transesterification reaction temperature on density and viscosity of biodiesel from rapeseed oil is shown in Fig. 13. The densities of biodiesel obtained at transesterification temperatures 25°C (0.915 g/cm³) and 45°C (0.913 g/cm³), are slightly higher than that at 60°C (0.882 g/cm³), and there is an inverse correlation between densities and conversions of reactants at these temperatures. The inverse correlation between conversion of reactants and viscosity of biodiesel (24.97; 13.931; 2.640 mm²/s) is also observed at temperatures 25, 45, and 60°C.

3.4 Catalyst Reusability

After methanolysis and separation from the product mixture, hydrated lime was reused for transesterification of rapeseed oil, under conditions that were determined as optimal by previous experiments: catalyst concentration 3 wt%, methanol:oil volume ratio 0.25, reaction temperature 60°C and reaction time 120 min. If considering that the molecular weight of refined rapeseed oil is 869.16 g/mol [40], the molar ratio of methanol:oil in this study was 5.77:1. A total of three consecutive reuses (first, second and third reuse) were conducted, whose effects on FAME

yield and biodiesel density and viscosity compared to the effects of fresh HL (first use) are shown in Figs. 14 and 15. The reusability property is recognized as one of the advantages of heterogeneous catalysts over homogeneous ones, which is why reuse testing is an integral part of many studies of catalyst performance in oil transesterification [78-81].

After all three reuses of hydrated lime, FAME rapeseed oil yields were >91%, which is higher than those obtained by hydrated lime in the methanolysis of sunflower [28] and rapeseed [65] oil. However, the determined values of FAME yield after the first, second and third reuse of hydrated lime (98.42, 94.85 and 91.97%) are lower compared to the yield achieved with the use of fresh catalyst, which indicates reduced catalytic powers of HL. A correlation was observed between FAME yield and the number of repeated uses of hydrated lime, that is, a declining trend of yield with an increasing number of catalyst reuse. A similar trend has been observed with other heterogeneous catalysts [82-84]. This is due to the deactivation of the heterogeneous catalyst, for which the possible causes include: leaching of the active phase of the catalyst, clogging of pores or poisoning of surface active sites, and structural changes of the catalyst [85-88]. In the study of the efficiency of HL in the methanolysis of waste soybean oil [27], the reduced yield of FAME was primarily attributed to calcite, which was identified as a secondary phase in fresh hydrated lime, and after repeated catalyst use, as the main crystalline phase, which reduced catalytically active phase. This could also be true for HL in this study, since X-ray analysis of fresh hydrated lime (Fig. 3) identified calcite as the secondary crystalline phase. In numerous research papers, the characterization of biodiesel is limited to reporting methyl ester content, which is considered to be its most important property, followed by properties of density and viscosity [17]. Although according to the European specification [56] only biodiesel obtained by the first reuse of catalysts meets the requirements for FAME content, and samples obtained by further successive use of catalysts have a slightly lower methyl ester content than the specified limit, their values are in the upper part of the mentioned range of 79 - 99% which is usually obtained by using heterogeneous catalysts [17]. Considering the high value of FAME yield achieved after the third reuse of HL, it can generally be assumed that this catalyst would give satisfactory FAME yields even with multiple

reuses. The measured FAME densities after each reuse of HL had negligible changes in values (0.882 - 0.883 g/cm³), while the FAME viscosities after the first, second and third reuse were 2.738, 3.128 and 3.248 mm²/s indicating an inverse correlation between viscosity and yield of rapeseed oil methyl ester. Biodiesel obtained by the first reuse of the catalyst, according to EN 14214 standard [56], has an acceptable density value, while its viscosity is slightly lower than the

minimum required. Other biodiesel samples, obtained by the second and third reuse of HL, have densities that also meet the European standard, but their viscosities are outside the required range. However, after each of the three consecutive uses of hydrated lime as a catalyst, the obtained biodiesel met the specifications of the American standard D6751 [67], in terms of the analyzed properties.

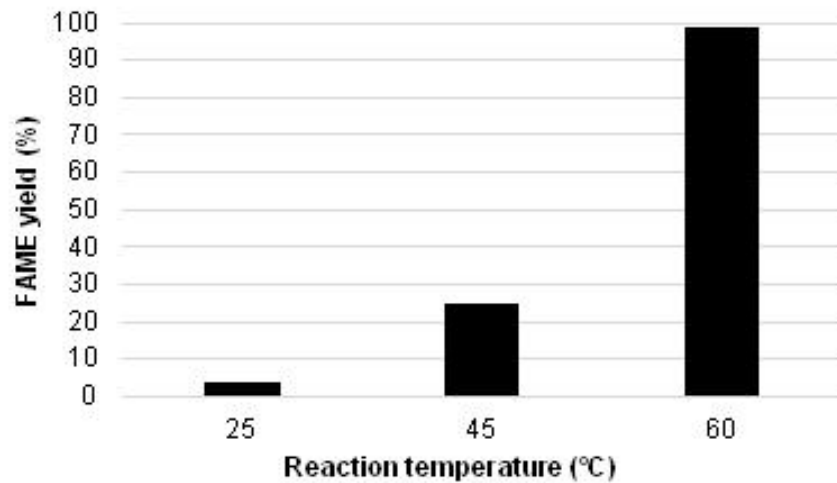


Fig. 12. Effect of reaction temperature on FAME yield

Process conditions: methanol:oil volume ratio 0.25, catalyst loading 3 wt%, reaction time 120 min

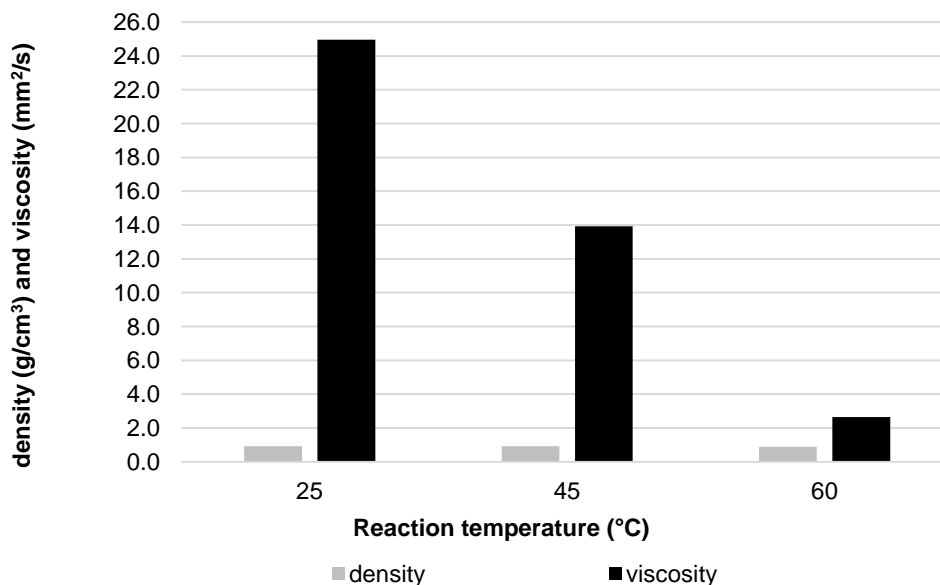


Fig. 13. Effect of reaction temperature on biodiesel density and viscosity

Process conditions: methanol:oil volume ratio 0.25, catalyst loading 3 wt%, reaction time 120 min

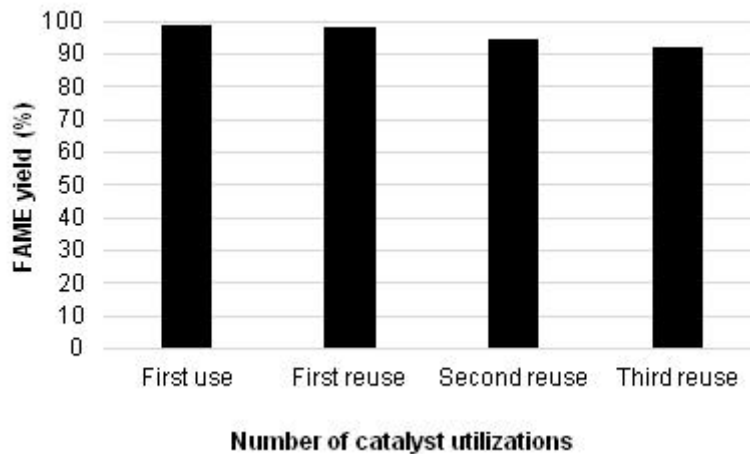


Fig. 14. Effect of HL reuse on FAME yield

Process conditions: methanol:oil volume ratio 0.25, catalyst loading 3 wt%, reaction temperature 60°C, reaction time 120 min

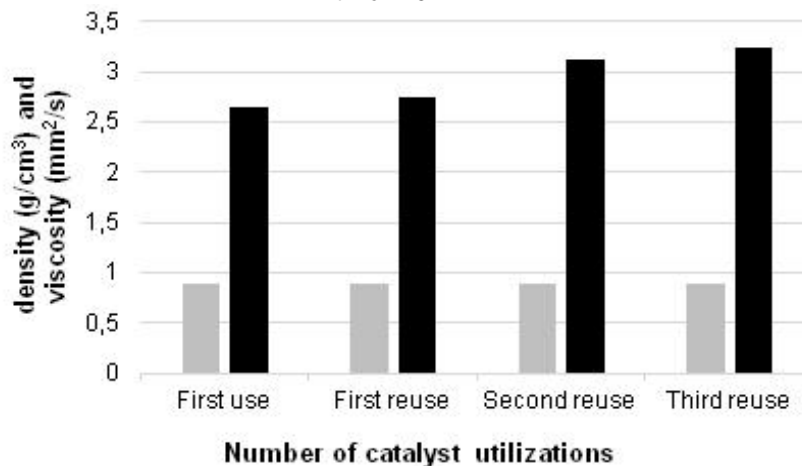


Fig. 15. Effect of HL reuse on biodiesel density and viscosity

Process conditions: methanol:oil volume ratio 0.25, catalyst loading 3 wt%, reaction temperature 60°C, reaction time 120 min

4. CONCLUSION

The results of the research on the methanolysis of rapeseed oil showed the possibility of using hydrated lime as a catalyst for the transesterification of vegetable oil. The determined optimal conditions of the investigated methanolysis were catalyst loading 3 wt%, methanol to oil volume ratio of 0.25, reaction time 120 min and temperature 60°C, resulting in maximum biodiesel yield of 98.76%. Among these conditions, a temperature value of 60°C was determined as the minimum required to obtain a high yield of methyl ester (>90%). In this regard, a comparison of energy costs versus biodiesel yield increase may be useful when considering the application of higher

methanolysis temperatures in industrial production. Although the increase in FAME yield is a function of the increase in catalyst concentration and reaction time, it is necessary to determine the optimal parameters experimentally, since their excessive values can result in reduced yields. Three reuses of hydrated lime, without any intermediate washing and activation procedures, resulted in yields of rapeseed oil methyl ester above 90%, indicating the possibility of multiple reuses of the catalyst, which is particularly advantageous in terms of process costs. In addition, biodiesel samples, obtained with the first use and the first reuse of hydrated lime, met the requirements of the European standard EN 14538, in terms of FAME content and density of biodiesel, while their

viscosities are below the specified limit. Despite numerous studies focused on the synthesis of new transesterification catalysts, larger production capacities require readily available catalysts with reusability properties without demanding intermediate separation and reactivation steps. From that aspect, the distribution of raw materials and well-established processes of hydrated lime production, as well as its experimentally proven efficiency as a heterogeneous catalyst with properties of reusability without prior reactivation, are certainly attributes that make it worth considering for use in industrial biodiesel production practice. Given the relatively high catalytic activity shown during successive uses in the transesterification reaction, the application of hydrated lime would be feasible in a continuous biodiesel production process, which would be interesting to investigate experimentally beforehand. The overall results of this research can serve as a basis for further research of catalysts of this type, purchased from various manufacturers, and linking their properties with catalytic activity in the methanolysis of various vegetable oils.

DISCLAIMER

Hydrated lime and rapeseed oil were procured directly from domestic producers with their consent for use in this study. There is absolutely no conflict of interest between the authors and producers of the mentioned materials, because we do not intend to use these products as an avenue for any litigation but for the advancement of knowledge. Also, the research was not funded by the producing company rather it was funded by personal efforts of the authors.

ACKNOWLEDGEMENTS

The authors acknowledge Bimal, Brčko (Bosnia and Herzegovina) and Victoriaoil, Šid (Serbia), for their support through the provision of necessary raw materials and chemicals, and for kindly allowing the use of laboratory equipment for research purposes.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Nasreen S, Nafees M, Qureshi LA, Asad MS, Sadiq A, Ali SD. Review of Catalytic Transesterification Methods for Biodiesel

- Production. In: Biernat K, editor. *Biofuels - State of Development*. IntechOpen; 2018.
DOI: 10.5772/intechopen.75534
2. Vicente G, Coteron A, Martinez M, Aracil J. Application of the factorial design of experiments and response surface methodology to optimize biodiesel production. *Ind. Crop. Prod.* 1998;8:29-35.
DOI: 10.1016/S0926-6690(97)10003-6
3. Al-Zuhair S. Production of biodiesel: Possibilities and challenges. *Biofpr.* 2007;1(1):57-66.
DOI: 10.1002/bbb.2
4. Wu G, Ge JC, Choi NJ. A comprehensive review of the application characteristics of biodiesel blends in Diesel Engines. *Appl. Sci.* 2020;10(22):8015.
DOI: 10.3390/app10228015
5. Makareviciene V, Sendzikiene E. Non-catalytic biodiesel synthesis under supercritical conditions. *Processes.* 2021;9(1): 138.
DOI: 10.3390/pr9010138
6. Demirbas A, Bafail A, Ahmad W, Sheikh M. Biodiesel production from non-edible plant oils. *Energy Explor. Exploit.* 2016;34(2):290–318.
DOI:10.1177%2F0144598716630166
7. Deshpande SR, Sunol AK, Philippidis G. Status and prospects of supercritical alcohol transesterification for biodiesel production, *WIREs Energy Environ.* 2017; 6(5):252.
DOI: 10.1002/wene.252
8. Hájek M, Vávra A, de Paz Carmona H, Kocík J. The catalysed transformation of vegetable oils or animal fats to biofuels and bio-lubricants: A review. *Catalysts.* 2021;11(9):1118.
DOI: 10.3390/catal 11091118
9. Xiao M, Mathew S, Obbard JP. Biodiesel fuel production via transesterification of oils using lipase biocatalyst, *Glob Change Biol Bioenergy.* 2009;1(2):115-125.
DOI: 10.1111/j.1757-1707.2009.01009.x
10. Musa IA. The effects of alcohol to oil molar ratios and the type of alcohol on biodiesel production using transesterification process. *Egypt. J. Pet.* 2016;25(1):21-21.
DOI: 10.1016/j.ejpe.2015.06.007
11. Mainali K. Base catalytic transesterification of vegetable oil. *Sci. Prog.* 2012;95(1):50–72.
DOI:10.3184/003685011X1320510399987

12. Gebremariam SN, Marchetti JM. Biodiesel production technologies: Review. *AIMS Energy*. 2017;5(3):425-457. DOI: 10.3934/energy.2017.3.425
13. Dash SK, Lingfa PA. Review on production of biodiesel using catalyzed transesterification. In: *AIP Conf Proc*. 2017;1859(1):020100. DOI: 10.1063/1.4990253
14. Yusuff AS, Adeniyi OD, Olutoye MA, Akpan UG. A review on application of heterogeneous catalyst in the production of biodiesel from vegetable oils. *J. Appl. Sci. Process Eng*. 2017;4(2):142-157. DOI: 10.33736/jaspe.432.2017
15. Miceli M, Frontera P, Macario A, Malara A. Recovery/reuse of heterogeneous supported spent catalysts. *Catalysts*. 2021;11(5):591. DOI: 10.3390/catal11050591
16. Mazaheri H, Ong HC, Amini Z, Masjuki HH, Mofijur M, Su CH, Anjum Badruddin I, Khan TMY. An overview of biodiesel production via calcium oxide based catalysts: Current state and perspective. *Energies*. 2021;14(13):3950. DOI: 10.3390/en14133950
17. Romero R, Martínez SL, Natividad R. Biodiesel production by using heterogeneous catalysts. In: Manzanera M, editor. *Alternative Fuel*. IntechOpen; 2011. DOI: 10.5772/23908
18. Chua SY, Periasamy LAP, Goh CMH, Tan YH, Mubarak NM, Kandedo J, Khalid M, Walvekar R, Abdullah EC. Biodiesel synthesis using natural solid catalyst derived from biomass waste — A review. *J. Ind. Eng. Chem*. 2020;81:41–60. DOI: 10.1016/j.jiec.2019.09.022
19. De Lima AL, Ronconi CM, Mota CJA. Heterogeneous basic catalysts for biodiesel production. *Catal. Sci. Technol*. 2016;6(9):2877–2891. DOI: 10.1039/C5CY01989C
20. Bhuiya MMK, Rasul MG, Khan MMK, Ashwath N, Azad AK, Hazrat MA. Second generation biodiesel: Potential alternative to-edible oil-derived Biodiesel. *Energy Procedia*. 2014;61:1969-1972. DOI: 10.1016/j.egypro.2014.12.054
21. Faruque MO, Razzak SA, Hossain MM. Application of heterogeneous catalysts for biodiesel production from microalgal oil - A review. *Catalysts*. 2020;10(9):1025. DOI:10.3390/catal10091025
22. Kesić Ž, Lukić I, Zdujić M, Mojović Lj, Skala D. Calcium oxide based catalysts for biodiesel production: A review. *Chem. Ind. Chem. Eng. Q*. 2016;22(4):391–408. DOI: 10.2298/CICEQ160203010K
23. Ortega MB, Hurtado AC, Duarte SAG. Triglyceride transesterification in heterogeneous reaction system with calcium oxide as catalyst. *Rev. Fac. Ing. Univ. Antioquia*. 2011;57:7-13. Available: <http://www.scielo.org.co/pdf/rfiua/n57/n57a01.pdf>
24. Urasaki K, Takagi S, Mukoyama T, Christopher J, Urasaki K, Kato S, Yamasaki A, Kojima T, Satokawa S. Effect of the kinds of alcohols on the structure and stability of calcium oxide catalyst in triolein transesterification reaction, *APPL CATAL A-GEN*. 2012;411–412:44–50. DOI:10.1016/j.apcata.2011.10.019
25. Gryglewicz S. Rapeseed oil methyl esters preparation using heterogeneous catalysts. *Biores. Technol*. 1999;70(3):249-253. DOI: 10.1016/S0960-8524(99)00042-5
26. Arzamendi G, Arguinarena E, Campo I, Zabala S, Gandia LM. Alkaline and alkaline-earth metals compounds as catalysts for the methanolysis of sunflower oil. *Catal. Today*. 2008;133–135:305–313. DOI:10.1016/j.cattod.2007.11.029
27. Sanchez-Cantu M, Perez-Diaz LM, Rosales R, Ramirez E, Apreza-Sies A, Pala-Rosas I, Rubio-Rosas E, Aguilar-Franco M, Valente JS. Commercial hydrated lime as a cost-effective solid base for the transesterification of wasted soybean oil with methanol for biodiesel production. *Energy Fuels*. 2011;25(7):3275–3282. DOI: 10.1021/ef200555r
28. Iličković Z, Andrejaš F, Subašić E, Stuhli V. Mogućnost primjene hidratiziranog vapna kao katalizatora u procesu metanolize suncokretova ulja. *Kemijska Industrija*. 2018;67(9-10):377–383. DOI: 10.15255/KUI.2017.049
29. International Organization for Standardization (ISO). ISO 6883: Animal and vegetable fats and oils — Determination of conventional mass per volume (litre weight in air). Geneva, Switzerland: International Organization for Standardization; 2017.
30. International Organization for Standardization (ISO). ISO 3104: Petroleum products — Transparent and opaque liquids — Determination of

- kinematic viscosity and calculation of dynamic viscosity. Geneva, Switzerland: International Organization for Standardization; 2020.
31. International Organization for Standardization (ISO). ISO 6320: Animal and vegetable fats and oils — Determination of refractive index. Geneva, Switzerland: International Organization for Standardization; 2020.
 32. International Organization for Standardization (ISO). ISO 660: Animal and vegetable fats and oils — Determination of acid value and acidity. Geneva, Switzerland: International Organization for Standardization; 2009.
 33. International Organization for Standardization (ISO). ISO 662: Animal and vegetable fats and oils — Determination of moisture and volatile matter content. Geneva, Switzerland: International Organization for Standardization; 2016.
 34. International Organization for Standardization (ISO). ISO 3961: Animal and vegetable fats and oils — Determination of iodine value. Geneva, Switzerland: International Organization for Standardization; 2018.
 35. International Organization for Standardization (ISO). ISO 3657: Animal and vegetable fats and oils — Determination of saponification value. Geneva, Switzerland: International Organization for Standardization; 2020.
 36. International Organization for Standardization (ISO). ISO 3960: Animal and vegetable fats and oils — Determination of peroxide value - Iodometric (visual) endpoint determination. Geneva, Switzerland: International Organization for Standardization; 2017.
 37. International Organization for Standardization (ISO). ISO 18609: Animal and vegetable fats and oils — Determination of unsaponifiable matter - Method using hexane extraction. Geneva, Switzerland: International Organization for Standardization; 2000.
 38. International Organization for Standardization (ISO). ISO 5509: Animal and vegetable fats and oils — Preparation of methyl esters of fatty acids. Geneva, Switzerland: International Organization for Standardization; 2000.
 39. International Organization for Standardization (ISO). ISO 5508: Animal and vegetable fats and oils — Analysis by gas chromatography of methyl esters of fatty acids. Geneva, Switzerland: International Organization for Standardization; 1990.
 40. Rezki B, Essamlali Y, Aadil M, Semlal N, Zahouily M. Biodiesel production from rapeseed oil and low free fatty acid waste cooking oil using a cesium modified natural phosphate catalyst. *RSC Adv.* 2020;10:41065–41077. DOI: 10.1039/d0ra07711A
 41. Encinar JM, Pardal A, Sánchez N, Nogales S. Biodiesel by transesterification of rapeseed oil using ultrasound: A kinetic study of base-catalysed reactions. *Energies.* 2018;11(9):2229. DOI:10.3390/en11092229
 42. Neagu A-A, Nița I, Botez E, Geaca S. A physico-chemical study for some edible oils properties. *Ovidius University Annals of Chemistry.* 2014;24(2):121-126. DOI:10.2478/auoc-2013-0020
 43. Demirbas A. Biodiesel fuels from vegetable oils via catalytic and non-catalytic supercritical alcohol transesterifications and other methods: A survey. *Energy Conversion and Management.* 2003;44:2093–2109. DOI:10.1016/S0196-8904(02)00234-0
 44. Thuneke K, Remmele E, Widmann B, Wilharm T. Standardisation of rapeseed oil as a fuel. In: *Proceedings of the 1st World Conference on Biomass for Energy and Industry, Sevilla, Spain, 2000*;5–9. Available:https://www.tfz.bayern.de/mam/cms08/en/dateien/standardisationofrapeseedoil_biomassevilla2000_th.pdf
 45. Deutsches Institut für Normung (DIN). DIN 51605: Kraftstoffe für pflanzenöläugliche Motoren - Rapsölkraftstoff - Anforderungen und Prüfverfahren; 2020. DOI: 10.31030/3178560
 46. Balat M. Modeling vegetable oil viscosity. *Energy sources a: Recovery util. Environ. Eff.* 2008;30(20):1856-1869. DOI: 10.1080/15567030701457392
 47. Bello EI, Mogaji TS, Agge M. The effects of transesterification on selected fuel properties of three vegetable oils. *J. Mech. Eng. Res.* 2011;3(7):218-225. Available:<http://www.academicjournals.org/jmer>
 48. Hoekman SK, Broch A, Robbins C, Cenicerros E, Natarajan M. Review of biodiesel composition, properties, and

- specifications, *Renew. Sust. Energ. Rev.* 2012;16(1):143-169.
DOI: 10.1016/j.rser.2011.07.143
49. Pinzi S, Garcia IL, Lopez-Gimenez FJ, Castro MDL, Dorado G, Dorado MP. The ideal vegetable oil-based biodiesel composition: A review of social, economical and technical implications. *Energy Fuels.* 2009;23(5):2325–2341.
DOI: 10.1021/ef801098a
 50. Cardoso FA, Fernandes HC, Pileggi RG, Cincotto MA, Vanderley JM. Carbide lime and industrial hydrated lime characterization. *Powder Technol.* 2009;195(2):143-149.
DOI: 10.1016/j.powtec.2009.05.017
 51. Khachani M, El Hamidi A, Halim M, Arsalane S. Non-isothermal kinetic and thermodynamic studies of the dehydroxylation process of synthetic calcium hydroxide Ca(OH)₂. *J. Mater. Environ. Sci.* 2014;5(2):615-624.
Available:https://www.jmaterenvironsci.com/Document/vol5/vol5_N2/75-JMES-684-2014-Khachani.pdf
 52. Kim T, Olek J. Effects of sample preparation and interpretation of thermogravimetric curves on calcium hydroxide in hydrated pastes and mortars. *Transp. Res. Rec.* 2012;2290(1):10-18.
DOI: 10.3141/2290-02
 53. Rodriguez-Navarro C, Ruiz-Agudo E, Burgos-Cara A, Elert K, Hansen EF. Crystallization and colloidal stabilization of Ca(OH)₂ in the presence of nopal juice (*Opuntia ficus indica*): Implications in architectural heritage conservation. *Langmuir.* 2017;33(41):10936-10950.
DOI: 10.1021/acs.langmuir.7b02423
 54. Ismail S, Ahmed AS, Anr R, Hamdan S. Biodiesel production from castor oil by using calcium oxide derived from mud clam shell. *Journal of Renewable Energy.* 2016;5274917:8.
DOI: 10.1155/2016/5274917
 55. Stamenkovic OS, Veljkovic VB, Todorovic ZB, Lazic ML, Bankovic-Ilic MB, Skala DU. Modeling the kinetics of calcium hydroxide catalyzed methanolysis of sunflower oil. *Bioresour. Technol.* 2010;101(12):4423–4430.
DOI: 10.1016/j.biortech.2010.01.109
 56. Institut za Standardizaciju Srbije (ISS). SRPS EN 14214: Tečni naftni proizvodi – Metilestri masnih kiselina (MEMK) za primenu u dizel-motorima i za potrebe zagrevanja – Zahtevi i metode ispitivanja. Beograd, Srbija: Institut za Standardizaciju Srbije; 2019.
 57. Li Y, Qiu F, Yang D, Li X, Sun P. Preparation, characterization and application of heterogeneous solid base catalyst for biodiesel production from soybean oil, *Biomass Bioenergy.* 2011;35(7):2787-2795.
DOI: 10.1016/j.biombioe.2011.03.009
 58. Deshmane VG, Adewuyi YG. Synthesis and kinetics of biodiesel formation via calcium methoxide base catalyzed transesterification reaction in the absence and presence of ultrasound. *Fuel.* 2013;107:474–482.
DOI: 10.1016/j.fuel.2012.12.080
 59. Al-Muhtaseb AH, Jamil F, Al-Haj L, Zar Myint, MT, Mahmoud E, Ahmad M, Hasan AO, Rafiq S. Biodiesel production over a catalyst prepared from biomass-derived waste date pits. *Biotechnol Rep (Amst).* 2018;20:e00284.
DOI: 10.1016/j.btre.2018.e00284
 60. Mandari V, Devarai SK. Biodiesel production using homogeneous, heterogeneous, and enzyme catalysts via transesterification and esterification reactions: A critical review. *Bioenerg. Res.* 2021.
DOI: 10.1007/s12155-021-10333-w
 61. Xie W, Zhao L. Heterogeneous CaO-MoO₃-SBA-15 catalysts for biodiesel production from soybean oil. *Energy Convers. Manag.* 2014;79:34-42.
DOI: 10.1016/j.enconman.2013.11.041
 62. Lani NS, Ngadi N, Taib MR. Parametric study on the transesterification reaction by using cao/silica catalyst. *Chem. Eng. Trans.* 2017;56:601-606.
DOI:10.3303/CET1756101
 63. Encinar JM, González JF, Pardal H, Martínez G. Transesterification of rapeseed oil with methanol in the presence of various co-solvents. In: *Proceedings Venice 2010: Third International Symposium on Energy from Biomass and Waste.* 2010;1–17.
Available:<http://hdl.handle.net/10400.26/1297>
 64. Acaroglu M, Demirbas A. Relationships between viscosity and density measurements of biodiesel fuels. *Energy Sources A: Recovery Util. Environ. Eff.* 2007;29(8):705-712.
DOI: 10.1080/00908310500280827
 65. Kouzu M, Hidaka J, Komichi Y, Nakano H, Yamamoto M. A process to transesterify

- vegetable oil with methanol in the presence of quick lime bit functioning as solid base catalyst. *Fuel*. 2009;88:1983–1990.
DOI: 10.1016/j.fuel.2009.03.013.
66. Baczewski K, Szczawiński S. Investigation properties of rapeseed oil methyl esters/aviation turbine fuel Jet A-1 blends. *Journal of KONES Powertrain and Transport*. 2011;18(1). Available:<http://yadda.icm.edu.pl/yadda/element/bwmeta1.element.baztech-article-BUJ8-0008-0012>
 67. ASTM (American Society for Testing and Materials) International. Standard Specification for Biodiesel Fuel Blend Stock (B100) for Middle Distillate Fuels (D6751 – 20a); 2020.
DOI: 10.1520/D6751-20A
 68. Knothe G. Biodiesel: Current trends and properties. *Top Catal*. 2010;53:714–720.
DOI: 10.1007/s11244-010-9457-0
 69. Ayetor GK, Sunnu A, Parbey J. Effect of biodiesel production parameters on viscosity and yield of methyl esters: *Jatropha curcas*, *Elaeis guineensis* and *Cocos nucifera*. *Alex. Eng. J*. 2015;54:1285-1290.
DOI: 10.1016/j.aej.2015.09.011
 70. Berrios M, Skelton RL. Comparison of purification methods for biodiesel. *Chem. Eng. J*. 2008;144:459–465.
DOI:10.1016/j.cej.2008.07.019
 71. Buasri A, Chaiyut N, Loryuenyong V, Worawanitchaphong P, Trongyong S. Calcium oxide derived from waste shells of mussel, cockle, and scallop as the heterogeneous catalyst for biodiesel production. *Sci. World J*. 2013;460923:7.
DOI: 10.1155/2013/460923
 72. Salamatinia B, Hashemizadeh I, Ahmad Zuhairi A. Alkaline earth metal oxide catalysts for biodiesel production from palm oil: Elucidation of process behaviors and modeling using response surface methodology. *IJCCE*. 2013;32(1):113-126.
DOI: 10.30492/ijcce.2013.5911
 73. Colombo K, Ender L, Barros AAC. The study of biodiesel production using CaO as a heterogeneous catalytic reaction. *Egypt. J. Pet*. 2017;26:341–349.
DOI: 10.1016/j.ejpe.2016.05.006
 74. Bano S, Ganie AS, Sultana S, Sabir S, Khan MZ. Fabrication and Optimization of Nanocatalyst for Biodiesel Production: An Overview. *Front. Energy Res*. 2020;8:579014.
DOI: 10.3389/fenrg.2020.579014
 75. Chozhavendhan S, Singh MVP, Fransila B, Kumar RP, Devi GK. A review on influencing parameters of biodiesel production and purification processes. *Curr. Opin. Green Sustain. Chem*. 2020(1-2):1–6.
DOI: 10.1016/j.crgsc.2020.04.002
 76. Gaide I, Makareviciene V, Sendzikiene E, Kazancev K. Natural rocks–Heterogeneous Catalysts for Oil Transesterification in Biodiesel Synthesis. *Catalysts*. 2021;11(3):384.
DOI: 10.3390/catal11030384
 77. Mathiyazhagan M, Ganapathi A. Factors affecting biodiesel production. *Res. Plant Biol*. 2011;1(2):1–5.
Available:<https://updatepublishing.com/journal/index.php/ripb/article/view/2566/2544>
 78. Istadi I, Mabruo U, Kalimantini BA, Buchori L, Anggoro DD. Reusability and Stability Tests of Calcium Oxide Based Catalyst (K₂O/CaO-ZnO) for Transesterification of Soybean Oil to Biodiesel. *Bull. Chem. React. Eng. Catal*. 2016;11(1):34-39.
DOI:10.9767/bcrec.11.1.413.34-39
 79. Chen C, Cai L, Shangguan X, Li L, Hong Y, Wu G. Heterogeneous and efficient transesterification of *Jatropha curcas* L. seed oil to produce biodiesel catalysed by nano-sized SO₄ 22/TiO₂. *R. Soc. Open Sci*. 2018;5: 181331.
DOI: 10.1098/rsos.181331
 80. Go YW, Yeom SH. Fabrication of a solid catalyst using coal fly ash and its utilization for producing biodiesel. *Environ. Eng. Res*. 2019;24(2):324-330.
DOI: 10.4491/eer.2018.029
 81. Dantas J, Leal E, Cornejo DR, Kiminami RHGA, Costa ACFM. Biodiesel production evaluating the use and reuse of magnetic nanocatalysts Ni_{0.5}Zn_{0.5}Fe₂O₄ synthesized in pilot-scale. *Arab. J. Chem*. 2020;13:3026-3042.
DOI: 10.1016/j.arabjc.2018.08.012
 82. Maleki H, Kazemeini M, Bastan F. Transesterification of canola oil to biodiesel using CaO/Talc nanopowder as a mixed oxide catalyst. *Chem. Eng. Technol*. 2017;40(10):1923-1930.
DOI:10.1002/CEAT.201600579
 83. Fereidooni L, Enayati M, Abbaspourrad A. Purification technology for renewable production of fuel from methanolysis of waste sunflower oil in the presence of high

- silica zeolite beta. Green Chem Lett Rev. 2021;14(1):2-14.
DOI: 10.1080/17518253.2020.1856426
84. Mansir N, Teo SH, Mijan N-A, Taufiq-Yap YH. Efficient reaction for biodiesel manufacturing using bi-functional oxide catalyst. Catal. Commun. 2021;149:106201.
DOI: 10.1016/J.CATCOM.2020.106201
85. Tesser R, Vitiello R, Carotenuto G, Garcia Sancho C, Vergara A, Maireles Torres PJ, Li C, Di Serio M. Niobia supported on silica as a catalyst for Biodiesel production from waste oil. Catal. Sustain. Energy. 2015;2:33-42.
DOI: 10.1515/cse-2015-0002
86. Ajala OE, Aberuagba F, Odetoye TE, Ajala AM. Biodiesel: Sustainable energy replacement to petroleum-based diesel fuel—A review. Chem Bio Eng Reviews. 2015;2(3):145-156.
DOI:10.1002/cben.201400024
87. Atadashi IM, Aroua MK, Abdul Aziz AR, Sulaiman NMN. The effects of catalysts in biodiesel production: A review. J Ind Eng Chem. 2013;19(1):14-26.
DOI: 10.1016/j.jiec.2012.07.009
88. Islam A, Taufiq-Yap YH, Chan E-S, Moniruzzaman M, Islam S, Nabi MN. Advances in solid-catalytic and non-catalytic technologies for biodiesel production. Energy Convers. Manag. 2014; 88:1200-1218.
DOI: 10.1016/j.enconman.2014.04.037

© 2021 Halilović et al.; This is an Open Access article distributed under the terms of the Creative Commons Attribution License (<http://creativecommons.org/licenses/by/4.0>), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Peer-review history:
The peer review history for this paper can be accessed here:
<https://www.sdiarticle5.com/review-history/83907>