

Biodiesel Production from Waste Cooking Oil Using Catalyst Calcium Oxide Derived of Limestone Lintau Buo

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Abstract

Biodiesel is an alternative to fossil fuels. It is produced by transesterification of triglycerides with short-chain alcohols in the presence of a catalyst. Sources of triglycerides include vegetable oils and animal fats. Among alkali metal oxide catalysts, calcium oxide (CaO) is widely used in the transesterification reaction. CaO is strongly alkaline and is cheap and environmentally friendly. It has low solubility in methanol. CaO can be sourced from natural limestone deposits. In this study, a solid CaO catalyst was obtained from Lintau Buo limestone and calcined at 900°C for 5 h. The results showed that this catalyst had a significant effect on biodiesel production.

Keywords: CaO, Heterogeneous catalyst, Transesterification, Waste Cooking Oil, Biodiesel

INTRODUCTION

Renewable and environmentally friendly fuels are being developed as a consequence of the scarcity of fossil fuels, global warming, and environmental problems [1-3]. Biodiesel, fatty acid methyl ester, or FAME, is a monoalkyl ester of fatty acids from vegetable oils or animal fats are an alternative solution [4, 5]. Its biodegradability, renewability, and physical similarity to diesel petroleum but with less toxic emissions make it a feasible sustainable energy source [6, 7].

FAME is produced by the transesterification of vegetable oils or animal fats with short-chain alcohol in the attendance of a catalyst [8]. The use of land to produce vegetable oils for FAME reduces that available for food production which can cause social problems. To minimize the effect on food production, waste cooking oil can be used as an alternative raw material making the process efficient and effective in minimizing environmental impact [9].

Homogeneous catalysts like NaOH, KOH, and H₂SO₄ are effective for transesterification [10, 11], producing a high yield in short reaction time. Nevertheless, homogeneous catalysts can result in problems such as saponification during the reaction, production of large amounts of wastewater, corrosion due to emulsification, difficulty in separation, and side reactions such as decomposition and polymerization [12]. The employment of heterogeneous catalysts can reduce some of these problems.

Heterogeneous catalysts are easily separated. They are recyclable, environmentally friendly, and do not produce

large amounts of wastewater [13]. Heterogeneous transesterification catalysts that have been studied include alkaline earth metal oxides, alkali metal compounds supported on alumina, zeolites, solid acids, and enzymes. Previous studies have reported that alkaline earth oxides which have high alkalinity can be used to produce FAME [14]. The most frequently used heterogeneous base catalyst is calcium oxide (CaO) [15]. CaO is low-cost, long-lived, moisture resistant, and has low solubility in a liquid mixture. Also, it produces a high yield of FAME [14].

CaO is widely used as a heterogeneous catalyst because there are many natural sources of calcium available. Natural calcium carbonate from eggshells, shells, limestone, dolomite, cuttlebone, or other sourced can be converted to CaO via calcination [16].

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The presence of high amounts of calcium in lime makes it a potential source of CO for a heterogeneous catalyst. However, little research has been conducted on the potential of heterogeneous base catalysts derived from natural limestone sources. Hence, low-cost heterogeneous base catalysts synthesized from local limestone is the focus of this study.

The source of limestone used is the Lintau Bou area of West Sumatra. This limestone contains Ca (97%) along with Mg (0.5%), Al (0.4%), (0.3%), Fe (0.1%), Ag (1.1%). Morphological and elemental properties of the calcinated limestone were characterized by SEM and XRF. Structural properties were identified through XRD analysis. Catalytic activity was evaluated through a transesterification reaction with waste cooking oil (WCO).

MATERIAL AND METHODS

Material

Limestone was collected from Lintau Buo Regency; West Sumatra. Waste cooking oil (used three times for frying) was collected from fried food vendors in Jati Padang, West Sumatra, and then analyzed for chemical composition using GCMS. Methanol, ethanol, and n-hexane were purchased from Merck Limited, Padang, Indonesia.

Catalyst Preparation

Limestone was rinsed using distilled water to remove surface impurities and dried at 105°C. It was then crushed using a mortar and calcined at 900°C for 5 h in a furnace to produce CaO which was mashed using a mortar and placed in an airtight container to avoid contact with CO₂ and H₂O.

Characterization of waste cooking oil and catalyst

WCO was filtered with Whatman filter paper no. 42 to remove sludge. After that, the WCO was dried at 100°C to

constant weight. The fatty acid composition of WCO was analyzed by GCMS. The crystal structure of the catalyst before and after transesterification was determined using X-ray diffraction (XRD) by PanAnalytical Expert Pro which uses Cu K α radiation. Elemental analysis was performed by X-ray fluorescence (XRF) analysis using the PANalytical Epsilon 3 analysis model. Morphological characteristics were analyzed using scanning electron microscopy (SEM) and X-ray spectroscopy (EDX) dispersive energy (JX-J), JSM-6290LV.

Transesterification

The transesterification was conducted in a 500 ml three-neck flask equipped with a reflux condenser and a magnetic stirrer. First, methanol and catalyst were added to the flask and heated to the reaction temperature. Then WCO was added and heated at 50°C while stirring at 850 rpm. Amount of catalyst was varied (1, 3, 5, 7 wt%) as was the molar ratio of methanol: oil (3:1, 6:1, 9:1, 12:1), reaction temperature (55, 60, 65, 70°C) and reaction time (2, 4, 6, 8 h). After the reaction was completed, the solid catalyst was filtered out using a funnel complete with filter paper. The filtrate was transferred to a separating funnel and left overnight. The top layer was collected and washed with hot distilled water until clear. The FAME was dried at 110°C to remove the remaining washing water, and the FAME content was analyzed by gas chromatography and yield calculated using the following equation:

$$\text{Methyl ester \%} = \frac{\% \text{FAME Area From GC} \times \text{Weight of product}}{\text{weight of feedstock used}} \times 100\% \quad [17]$$

RESULTS AND DISCUSSION

Determination of the content of free fatty acids

Based on (figure1) linoleic fatty acid content was 49.56% (Table 1). Mahreni (2010), found a similar sample contained 55% linoleic acid [18, 19].

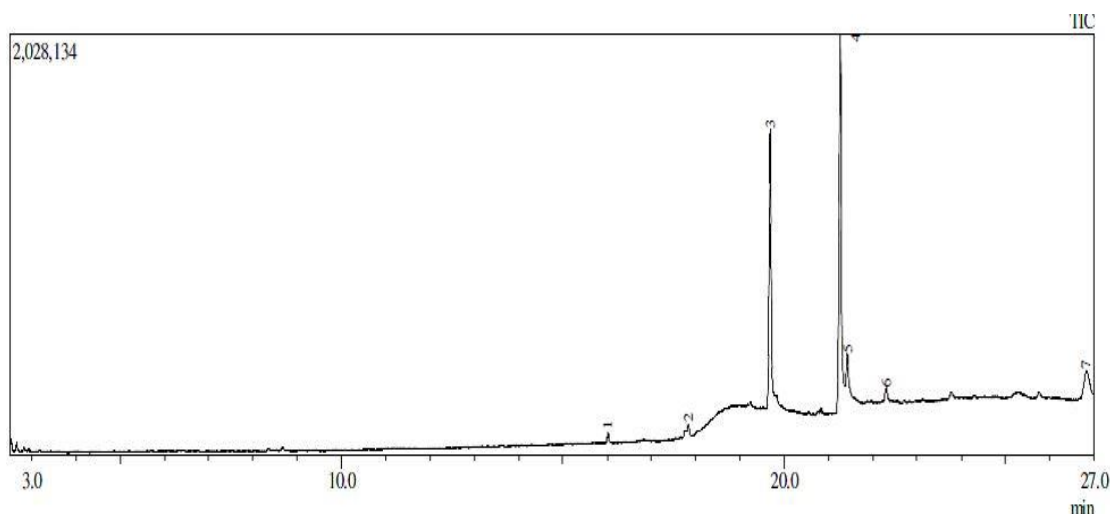


Figure 1. Chromatogram of waste cooking oil from Gas Chromatography

Peak#	R.Time	Area%	Height	
1	16.017	0.86	46322	Palmitate
2	17.834	1.09	53076	Oleate
3	19.688	31.73	1339759	Palmitin
4	21.270	49.56	1809337	Linoleic
5	21.430	7.49	234934	Stearin
6	22.310	1.54	61366	Hexadecanoic
7	26.836	7.72	124911	Borane

X-ray diffraction (XRD) analysis

The XRD pattern of the limestone (Figure 2a) shows the presence of CaCO₃ peaks at 2θ: 23°, 29°, 35°, 39°, 43°, 47°, and 48° (ICDD Reference No: 01-078-4614). It can be seen that the main component of limestone is calcium carbonate. These results are consistent with XRF analysis, which also indicates that calcium (Ca) is the main element. Peaks corresponding to CaO and Ca (OH)₂. CaO at 2θ: 32°, 37°, 53°, 64°, 67°, 91° (ICDD Reference Number: 01-077-2010) and Ca (OH)₂ at 2θ: 17°, 28°, 34°, 46°, 50°, 54°, 62° (ICDD

Reference Number: 01-084-1265) can also be observed (Figure 2b). This indicates that after calcination this limestone should produce CaO with high crystallinity. Above 700°C, CaCO₃ turns into CaO and 900°C is the optimal temperature for calcination [20]. The appearance of the Calcium diglyceroxide at 2θ: 20° (ICDD Reference No: 00-021-1544) peak after CaO has been used in transesterification (figure 2c) corresponds to its formation in the process of combining calcium oxide with glycerol [21]. Calcium diglyceroxide is an active species that can function as a catalyst in the transesterification reaction [22].

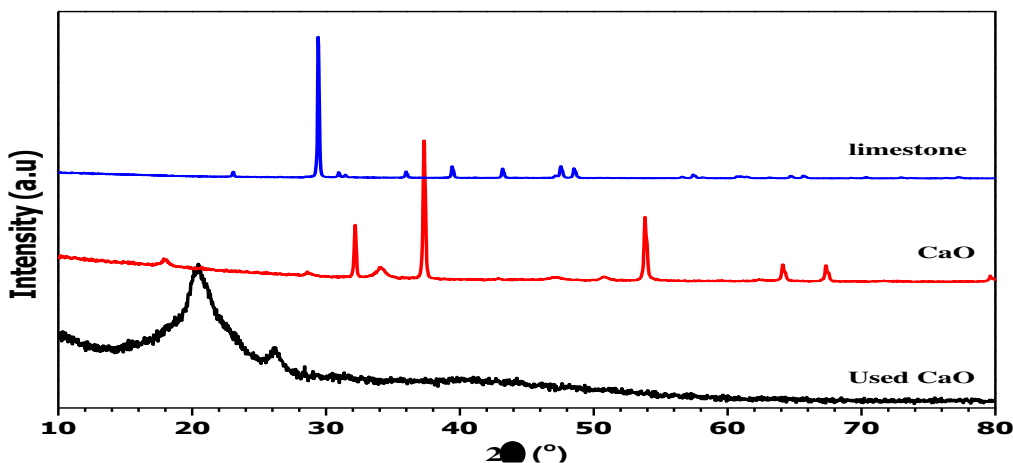


Figure 2. X-ray diffraction analysis (a) Limestone (b) CaO (c) Used CaO

X-ray fluorescence (XRF) analysis

The composition of the limestone and CaO were analyzed by XRF (Table 2). Ca content in limestone is 97%, while Ca content is 98% in CaO after calcination. Ca content decreased

to 93% after the first transesterification reaction cycle. A small amount of the CaO catalyst reacted with methanol to form Ca (C₃H₇O₃)₂ and Ca (OH)₂, but most was able to be recovered as CaO(20).

Limestone		CaO		Used CaO	
Compound	Conc	Compound	Conc	Compound	Conc
Mg	0,5%	Mg	0,3%	Mg	1,7%
Al	0,4%	Al	0,4%	Al	0,7%
Si	0,3%	Si	0,4%	Si	0,8%
Ca	97%	Ca	98%	P	1,7%
Fe	0,1%	Fe	0,1%	Ca	93%
Ag	1,1%	Ag	0,9%	Fe	0,4%
				Ag	1,4%

SEM analysis

(Figure 3) shows an SEM of the CaO catalyst before and after the transesterification reaction showing morphological changes after the reaction. Fresh catalyst has a uniform and irregular distribution of particles, whereas used catalyst show particles agglomerated with other reactant molecules. Morphological changes occur due to the blockage of active

sites with by-products such as diglycerides, monoglycerides, glycerol, and FAME and contamination by moisture and CO₂ in the air during the catalyst filtration process [23]. These physical changes also contribute to a decrease in catalytic performance. The morphological changes to a catalyst after the first reaction is known to result in a new active surface which has a significant impact on catalytic activity [24].

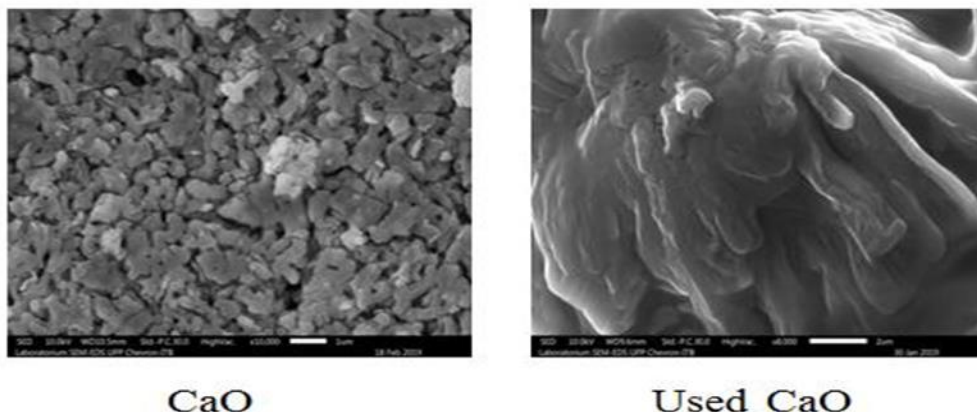


Figure 3. SEM Analysis (a) CaO (b) Used CaO

Amount of catalyst

The effect of the amount catalyst on transesterification was significant (Figure 4). When other conditions were constant (molar ratio alcohol/oil 6:1, 65°C, 4 h) a small amount of catalyst produced a low yield of FAME. Optimal results were

obtained at 5wt% CaO which produced a 86% yield. Higher amounts of CaO than this reduced the yield. A critical mass of catalyst is needed to drive a reaction [25, 26]. On the other hand, too much catalyst makes the reactant mixture too thick, which causes problems with mixing and separation [27].

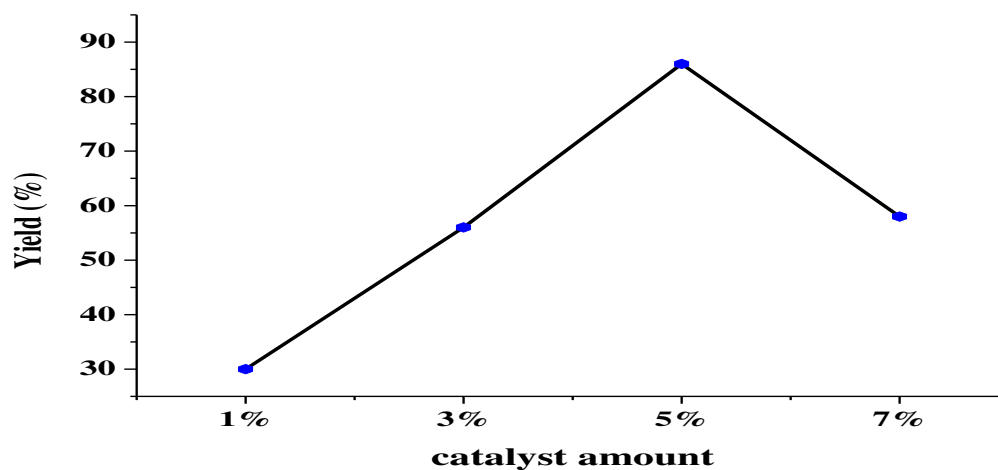


Figure 4. Yield for different amounts of catalyst

Methanol to oil ratio

The molar ratio of alcohol/oil in the transesterification stage under constant conditions (5wt% catalyst, 65°C, 4h) (Figure 5), had a significant effect on yield which increased with increasing methanol/oil ratios up to 6: 1 (yield of 86%). Exceeding this optimum value reduced yield. Stoichiometry of the transesterification reaction indicates that one mole of oil (or triglyceride) needs three moles of alcohol to produce three moles of FAME and one mole of glycerol. Because

transesterification is reversible, excess methanol is required to move the equilibrium towards the formation of methyl esters [28]. Large amounts of methanol increase the methoxy ions on the surface of the catalyst [29]. Increased numbers of methoxy ions push the equilibrium toward FAME production [30]. A further increase in the methanol/oil ratio will dissolve most of the glycerol, which inhibits the reaction between methanol and the other reactants and the catalyst, thereby interfering with glycerin separation and product reduction [31].

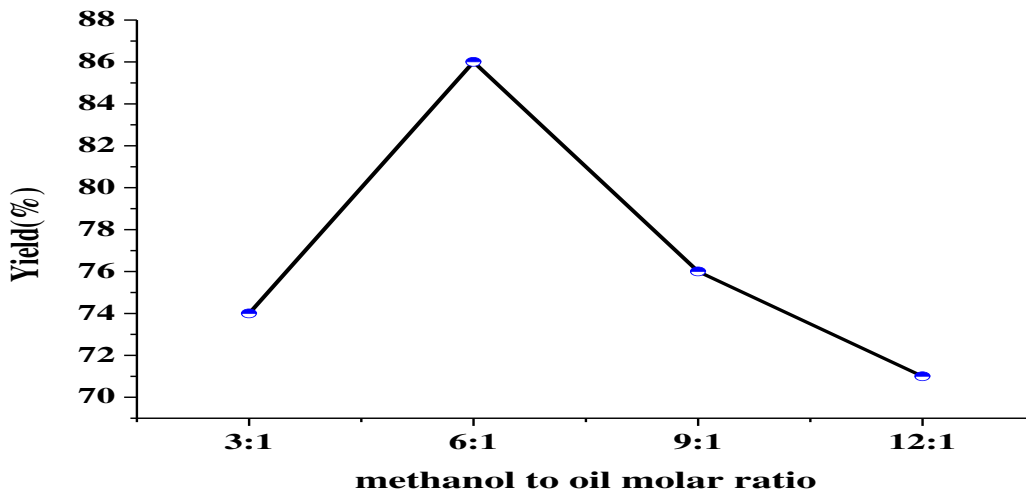


Figure 5. Effect of the methanol to oil molar ratio

Reaction time

The reaction time also significantly influences the transesterification yield when carried out under constant conditions (5 wt% catalyst, alcohol/oil molar ratio of 6: 1, 65°C). Figure 6 shows that yield increases with reaction time up until 4 hours with the optimal yield being 86%. Longer

reaction times reduce yield. Optimal reaction time is reached when there is time for sufficient reactant interactions to produce the final product [32]. A shorter time means fewer reactant interactions and lower yield [25]. Conversely, a time longer than the optimal point causes a back reaction to occur and results in the formation of soap/saponification, which can reduce the amount of product [28].

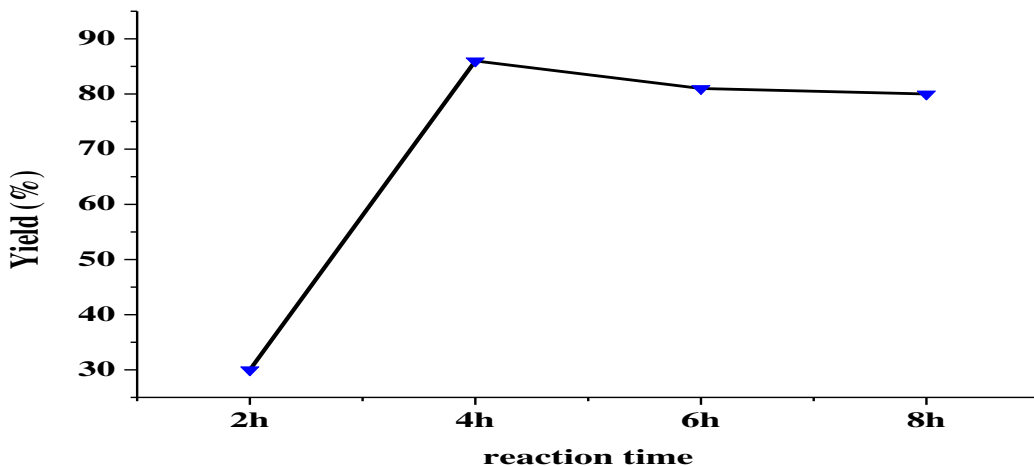


Figure 6. Effect of reaction time on the yield of FAME

Temperature

The effect of temperature on the transesterification process under fixed conditions (5wt% catalyst, the molar ratio of alcohol: oil 6: 1, 4h) is shown in Figure 7. Increasing the temperature increased yield up until 65°C with the optimal yield being 86%. Higher temperatures reduce yield. FAME production is an endothermic reaction that requires high

temperatures [33]. The reaction rate is slow at low temperatures due to diffusion resistance; Heterogeneous catalysts form three phases producing an oil-methanol catalyst system [26]. Increasing the reaction temperature beyond the optimal conditions will cause the evaporation of methanol, which encourages saponification so that FAME production is reduced [34].

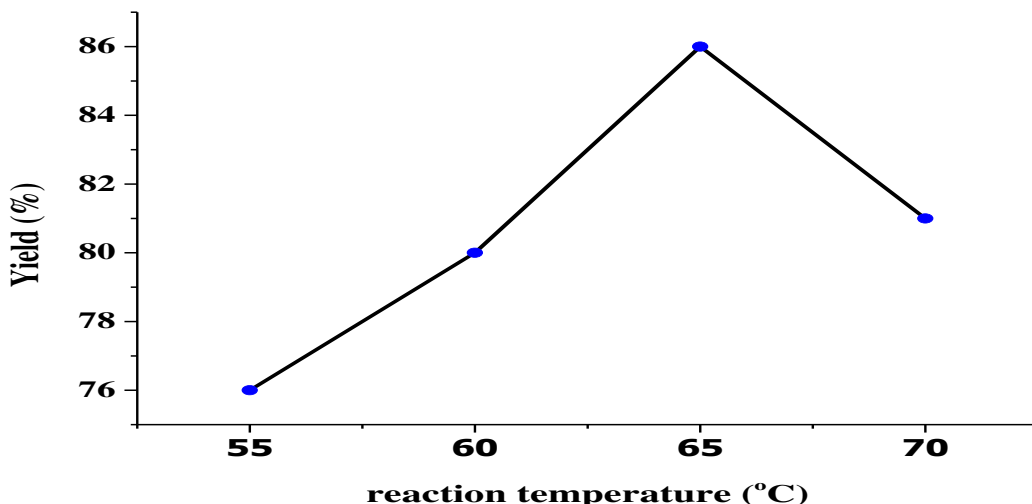


Figure 7. Effect of Reaction Temperature on yield

Reusability

After the reaction for 4h, the catalyst was separated from the mixture by filtration and washed using methanol and n-hexane [29] and dried at 110°C for reuse without prior recalcination. The FAME yield obtained from the reused catalyst for each reaction cycle shown in (Figure 8). The catalyst's effectiveness gradually decreases with repeated use.

Catalytic activity is reduced due to the washing process [28]. Besides, the formation of Calcium diglyceroxide $\text{Ca}(\text{C}_3\text{H}_7\text{O}_3)_2$ as indicated by the XRD pattern will result in some saponification [23] which causes difficulties in the separation process, reducing catalyst recovery [35]. According to the SEM image illustrates the agglomeration of particles with other reactant molecules or catalyst leaching. This causes the loss of active sites, resulting in a reduced product yield [36].

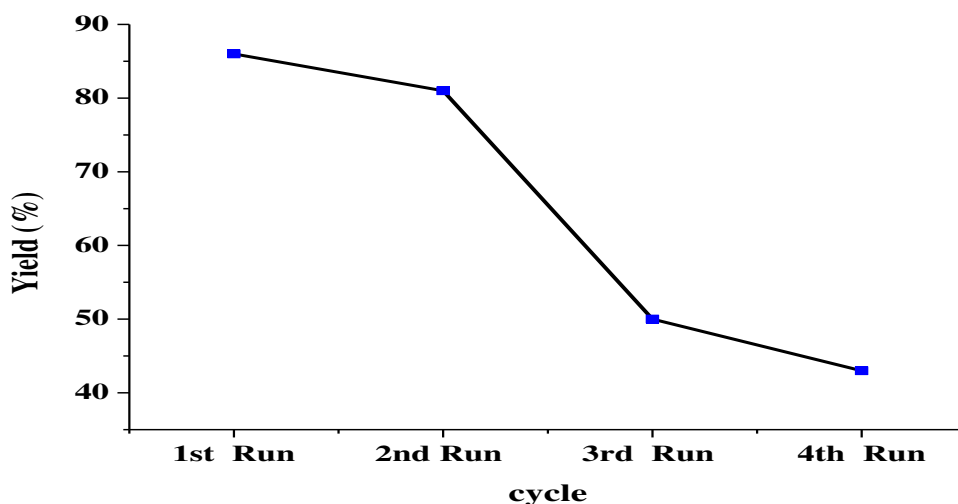


Figure 8. Reusability of the CaO catalyst

CONCLUSION

Lintau Bou limestone has high calcium content and can be used to produce a low-cost heterogeneous base CaO catalyst. Calcination (900°C for 5 hours) produces CaO particles which can be used in transesterification. Optimization of parameters indicated that the maximum yield of FAME obtained is 86%, which can be achieved using 5% by weight catalyst, 6: 1 ratio of methanol to oil, 4 hour reaction time, and a temperature of 65°C. The CaO catalyst could be reused three times indicating good reusability.

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