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RESEARCH ARTICLE

Using chicken-eggshell-derived calcium oxide as an ideal catalyst towards esterification of high-included free fatty acid waste cooking oil; Application of Taguchi method

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ABSTRACT

In this study, we used chicken eggshells as a low-cost catalyst for the esterification process of oil with a high fatty acid (FFA) content. We used the Taguchi method to optimize both esterification and transesterification steps. The waste chicken eggshell was calcinated at a temperature of 700 °C for 4 hours to synthesize a calcium oxide (CaO) active catalyst. The newly obtained catalyst was characterized by BET, TEM, and SEM. Then, this catalyst was employed for the transesterification of waste cooking oil. This work aimed to optimize critical parameters in the preparing of biodiesel for the production of waste cooking oil to maximize efficiency. Hence, the effects of reaction temperature, reaction time, catalyst amount, and methanol/ oil molar ratio on biodiesel yield were investigated and optimized through the Taguchi method. The maximum biodiesel efficiency is calculated to be 84% through transesterification at the following optimum conditions: the reaction temperature of 80 °C, the reaction time of 1.5 h, the ratio of methanol to oil of 10:1, and the catalyst amount of 2% w/w.

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INTRODUCTION

Recently, renewable energy sources such as biodiesel and biomass from environmental wastes have received significant attention. These new biofuels reduce any dependence on fossil fuels. They can be used instead of fossil fuels which are responsible for environmental pollution. For this reason, today, biofuels are known as alternative fuels [1]. Biodiesel (frequently defined as an alkyl monoester) is the best alternative for petroleum-derived diesel fuel [2, 3]. Owing to their energy content similar to diesel, they can be used commonly in fuel blends [4, 5], resulting in a significant reduction in emissions of gaseous pollutants such as CO, SOx, and organic compounds [6]. Transesterification is the simplest and most cost-effective way to produce biodiesel from oil [4].

Transesterification, called alcoholysis, is the reaction of a fat or oil with an alcohol to form esters and glycerol. In that reaction, one mole of triglyceride reacts with three moles of alcohol (normally methanol) in the presence of catalysts. Usually, transesterification of vegetable oil to biodiesel (fatty acid methyl ester, FAME) can be catalyzed by either acids or bases [7, 8]. High values of FFA cause the production of biodiesel to be incompatible with essential catalysts. Therefore, there are two alternative methods for solving this problem. The first method is a two-step process that needs primary acid-catalyzed esterification of the FFA, followed by a basic catalyzed transesterification. A single process uses an acidic catalyst that simultaneously catalyzes esterification and transesterification reactions in the second method.

Heterogeneous catalysts provide easy and cost-effective separations in which the regeneration of the catalyst is possible [9]. Heterogeneous essential catalysts include alkaline earth metal oxides such as calcium oxide (CaO), magnesium oxide (MgO), and hydrotalcite ($Mg_6Al_2CO_3(OH)_{16}{\cdot}4(H_2O)$) [10, 11].

Eggshells are a network of protein fibers associated with crystals of calcium carbonate ($CaCO_3$), magnesium carbonate ($MgCO_3$), calcium phosphate ($Ca_3(PO_4)_2$), and also some organic substances and water. $CaCO_3$, the significant component of eggshells (96%), is an amorphous crystal that can be crystallized to calcite (hexagonal crystal) [12, 13].

$CaCO_3$ particles of eggshell can be used as raw materials to form CaO heterogeneous catalysts; not only will the waste materials be eliminated (environmentally favorable), but the cost of biodiesel processing will be decreased (cost-effectiveness). Many experts have conducted numerous studies using eggshell waste as a renewable catalyst. [14, 15, 16]. Recently, natural calcium sources from waste materials have been considered a new trend for biodiesel production [17].

Wei et al. [16] used eggshells as a catalyst in the transesterification reaction of soybean oil with methanol. They reported that the modification of eggshells at a temperature of seven hundred ^oC could be active in their particles as catalysts for the transesterification process .

Niju et al. [18] used successive steps (calcination-hydration-dehydration) to activate eggshell-derived CaO catalysts. The calcination process at a temperature of nine hundred degrees Celsius was applied in their work. However, the only problem in their work is the cost of the operation, which is necessary for the modification of the resulting catalyst. Moreover, they did not use their resulting catalyze to optimize conditions for biodiesel production.

We first calcinated the chicken eggshell (as a cost-effective source) to create a CaO active catalyst in the present study. Then, this catalyst was utilized for the transesterification of waste cooking oil. In this study, Taguchi methodology was used to optimize different biodiesel production variables, including temperature, time, methanol/oil ratio, and the amount of catalyst, to get the maximum efficiency.

EXPERIMENTAL

Materials

Frying oil waste was collected from a local restaurant in Babol, Iran. This oil includes solid impurities such as food residues and water. For this reason, before any use, we filtered this crude oil to make it ready to use for biodiesel production. The main chemical substances used in our study are listed: methanol (Merck), potassium hydroxide (Merck), ethanol (Flucka), hydrochloric acid (Fluka), and phenolphthalein (Sigma-Aldrich).

Eggshells were collected from a pastry shop in northern Iran. Before any use, they were washed carefully in tap water to eliminate any surplus material adhering to their surface and washed two times with distilled water. The rinsed eggshells were dried for 24

hours and then crushed into powder (see Fig. 1).

Catalyst characterization

Scanning Electron Microscopy (SEM), Model: F E I Quanta FEG 200 and Transmission Electron Microscope (TEM), Hitachi High-Technologies Europe GmbH, Krefeld, Germany, model HF2000 analyses were used to confirm the morphology of the calcined catalyst. The surface areas of calcined and non-calcined CaO were determined by BET analysis using an ASAP 2020 surface area analyzer (Micromeritics).

Taguchi method and design of experiment

The Taguchi optimization method and experiment design apply fractional factorial test designs that reduce the number of experiments. The Taguchi method and investigation technique can evaluate several process variables that disturb the performance characteristic while minimizing the number of test runs [9]. As shown in Tables 1 and 2, we considered four factors: time (minutes), temperature (°C), methanol/oil ratio, and catalyst amount at four different levels. Using Taguchi methodology, the average impact of each variable on the acidity (in esterification) and the biodiesel production efficiency (in transesterification) were investigated. As a result, the best possible experimental outcomes are determined and examined, which is the main advantage of the present study compared to other biodiesel production methods.

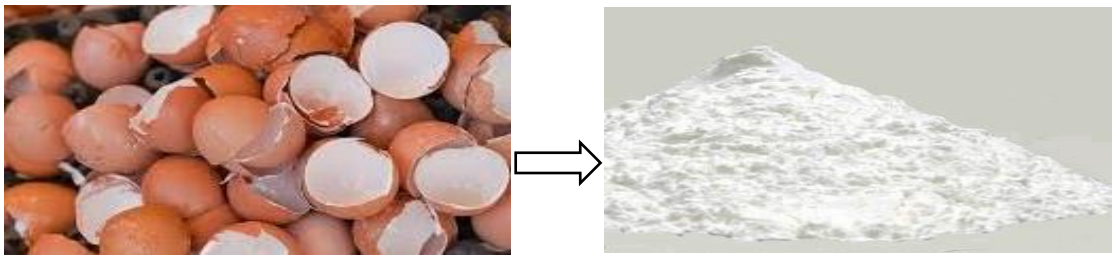


Figure 1. Preparation of CaO catalyst derived from eggshell waste

Table 1.The values of different levels for esterification

level	Factors			
	HCl(gr)	Methanol/Oil (molar ratio)	Temperature (•c)	Time (min)
1	0.5	3:1	60	30
2	1	6:1	80	60
3	1.5	9:1	100	90
4	2	12:1	120	120

Table 2.The values of different levels for transesterification

level	Factors			
	eggshell-derived CaO catalyst (gr)	Methanol/Oil (molar ratio)	Temperature (•c)	Time (hr)
1	1	6:1	60	1.5
2	2	8:1	80	2.5
3	3	10:1	100	3.5
4	4	12:1	120	4.5

RESULTS AND DISCUSSION

The morphology of eggshell-derived CaO

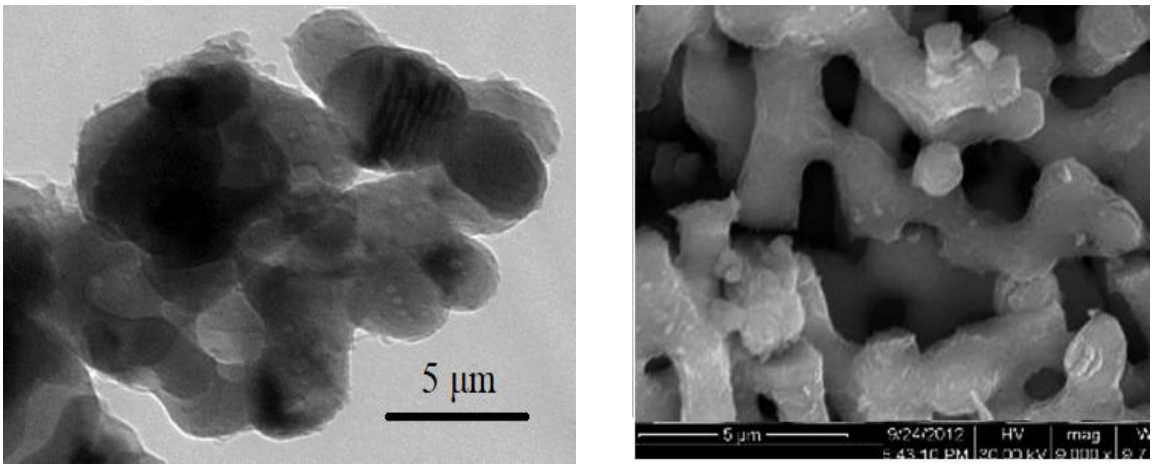
The dried eggshell was shaped into small pieces using crushing and then calcined in a muffle furnace under static air conditions at 700 °C for four hours to produce active CaO particles.

The TEM and SEM images of the eggshell-derived CaO catalyst are given in Fig. 2. Both TEM and SEM analyses confirm that the typical morphology of particles is a micrometer in width (1.7 to 2.4 µm). Moreover, the BET analysis was used to conclude the specific surface area of the particles. According to the BET analysis, the mean surface area of the eggshell-derived CaO was calculated to be 3.17 m^2/gr. This high surface area points towards the suitability of the achieved CaO particles as an ideal catalyst for the transesterification process.

Esterification

The measured free fatty acid (FFA) content for the crude oil was 6.8%, which means it is not suitable to be used directly for transesterification due to the saponification. Therefore, it requires a pretreatment step (esterification) to reduce the FFA content of fat to less than 1% [7]. Literature reviews show that heterogeneous catalysts for the esterification process have not yielded satisfactory results [7, 8, 9]. Another drawback of heterogeneous catalysis is the necessity for harsh reaction conditions [9]. Solve the HCl homogenous catalyst to solve these problems as a cost-effective material.

Tables 1 and 2 depict the runs for the esterification and transesterification processes, respectively. For each run, the FFA content value was considered. All deals for 16 Taguchi suggested runs and FFA percentage results are listed in Table 3.



(a) **(b)**

Figure 2. The TEM (a) and SEM (b) pictures of eggshell derived CaO catalyst

Table 3.The suggested runs by Taguchi methodology in the esterification stage along with the results of % FFA (HCl: gr, Methanol/oil: mol/mol, time: min, temperature: °C)

No.	Factor	Quantity	level	FFA	No.	Factor	Quantity	Level	FFA
1	**HCl**	1	1	%0.95	9	**HCl**	3	3	1.68%
	Methanol/Oil	3	1			**Methanol/Oil**	3	1	
	Time	30	1			**Time**	120	4	
	Temperature	60	1			**Temperature**	100	3	
2	**HCl**	1	1	0.95	10	**HCl**	3	3	%0.72

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	Methanol/Oil	6	2		**Methanol/Oil**	6	2	
	Time	60	2		**Time**	90	3	
	Temperature	80	2		**Temperature**	120	4	
3	**HCl**	1	1	1.68%	**11**	**HCl**	3	3	0.67%
	Methanol/Oil	9	3			**Methanol/Oil**	9	3	
	Time	90	3			**Time**	60	2	
	Temperature	100	3			**Temperature**	60	1	
4	**HCl**	1	1	0.67%	**12**	**HCl**	3	3	0.67%
	Methanol/Oil	12	4			**Methanol/Oil**	12	4	
	Time	120	4			**Time**	30	1	
	Temperature	120	4			**Temperature**	80	2	
5	**HCl**	2	2	%0.89	**13**	**HCl**	4	4	%1.06
	Methanol/Oil	3	1			**Methanol/Oil**	3	1	
	Time	90	3			**Time**	60	2	
	Temperature	80	2			**Temperature**	120	4	
6	**HCl**	2	2	%0.72	**14**	**HCl**	4	4	%0.89
	Methanol/Oil	6	2			**Methanol/Oil**	6	2	
	Time	120	4			**Time**	30	1	
	Temperature	60	1			**Temperature**	100	3	
7	**HCl**	2	2	%0.72	**15**	**HCl**	4	4	%0.95
	Methanol/Oil	20.59	3			**Methanol/Oil**	9	3	
	Time	30	1			**Time**	120	4	
	Temperature	120	4			**Temperature**	80	2	
8	**HCl**	2	2	%0.78	**16**	**HCl**	4	4	0.67%
	Methanol/Oil	27.45	4			**Methanol/Oil**	12	4	
	Time	60	2			**Time**	90	3	
	Temperature	100	3			**Temperature**	60	1	

Then the obtained FFA values in all experiments were entered into the Taguchi algorithm program, and the mean graph of each variable on the amount of %FFA was achieved. The average impact of HCl amount (Fig. 3), the molar ratio of methanol/oil (Fig. 4), time (Fig. 5), and temperature (Fig. 6) are respectively indicated in the amount of %FFA. The minimum amount of the average impact of the factor in each chart is considered the best level.

In each graph, the horizontal axis indicates the variable levels. According to the minimum levels in the specified levels for the variables, the highest effect of decreasing the %FFA is determined. The optimal values of the variables are achieved with the amount of the 2% w/w HCl catalyst (1 gr catalyst with 50 gr oil), the molar ratio of alcohol to oil (12:1), the time of 30 minutes, and the temperature of 60°C. FFA content was reduced from 6.8% to 0.67 by applying these situations to the esterification of crude oil.

Transesterification

Reducing the crude oil's FFA content to 0.7% makes it possible for transesterification afterward. To optimize the parameters of transesterification, we again used the Taguchi algorithm. The values of the levels for transesterification are listed in Table 2. On the other hand, Table 4 represents the transesterification parameters of 16 Taguchi suggested runs and their efficiency in biodiesel production.

In each run, the specified amount of eggshell-derived CaO catalyst, the molar ratio of methanol/oil, temperature, and time have been used to investigate their effect on biodiesel production. The biodiesel efficiency is calculated according to the following relationship and entered into the Taguchi algorithm. Finally, the mean effect of each variable is achieved.

$$\text{Biodiesel efficienc} = \left(\frac{\text{methyl ester mass}}{\text{oil mass}}\right) \times 100 \quad \text{(eq. 1)}$$

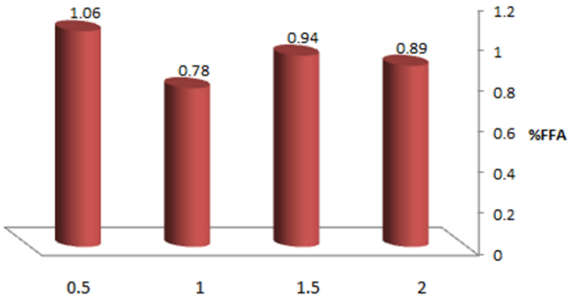

Figure 3. Average effects of HCl concentration (left to right: 0.5, 1, 1.5, and 2 gr) on % FFA

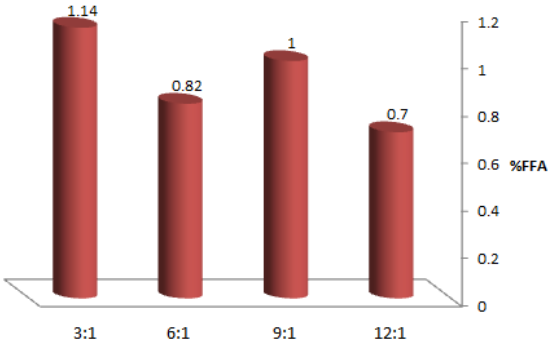

Figure 4. Average effects of Methanol/Oil (left to right: 3:1, 6:1, 9:1, and 12:1) on % FFA

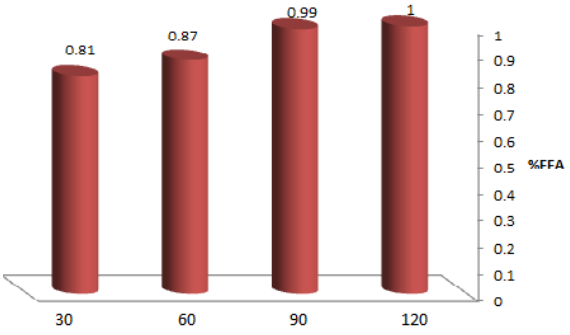

Figure 5. Average effects of time (left to right: 30, 60, 90, and 120 min) on % FFA

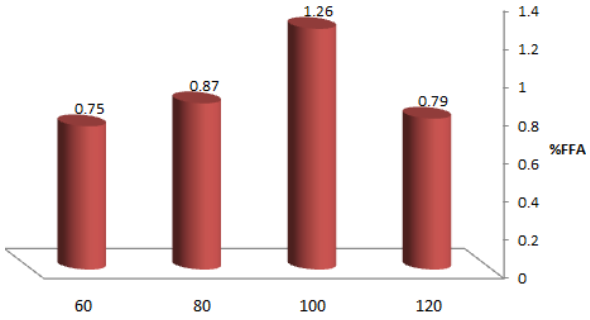

Figure 6. Average effects of temperature (left to right: 60, 80, 100, and 120 °C) on %FFA

Table 4.The suggested runs by Taguchi methodology in the transesterification stage along with the results of biodiesel yield (% Yield)

No	Factor	Quantity	Level	%Yield	No	Factor	Quantity	Level	%Yield
1	**CaO %Wt**	2	1	76	**9**	**CaO %Wt**	2	1	81
	Methanol/Oil	6	1			**Methanol/Oil**	10	3	
	Time	1.5	1			**Time**	3.5	3	
	Temperature	60	1			**Temperature**	120	4	
2	**CaO %Wt**	4	2	90	**10**	**CaO %Wt**	4	2	70
	Methanol/Oil	8	2			**Methanol/Oil**	12	4	
	Time	1.5	1			**Time**	3.5	3	
	Temperature	80	2			**Temperature**	100	3	
3	**CaO %Wt**	6	3	80	**11**	**CaO %Wt**	6	3	85
	Methanol/Oil	10	3			**Methanol/Oil**	6	1	

	Time	1.5	1			**Time**	3.5	3	
	Temperature	100	3			**Temperature**	80	2	
4	**CaO %Wt**	8	4	83	**12**	**CaO %Wt**	8		75
	Methanol/Oil	12	4			**Methanol/Oil**	8		
	Time	1.5	1			**Time**	3.5	3	
	Temperature	120	4			**Temperature**	60	1	
5	**CaO %Wt**	2	1	86	**13**	**CaO %Wt**	2	1	93
	Methanol/Oil	8	2			**Methanol/Oil**	12	4	
	Time	2.5	2			**Time**	4.5	4	
	Temperature	100	3			**Temperature**	80	2	
6	**CaO %Wt**	4	2	88	**14**	**CaO %Wt**	4	2	87
	Methanol/Oil	6	1			**Methanol/Oil**	10	3	
	Time	2.5	2			**Time**	4.5	4	
	Temperature	120	4			**Temperature**	60	1	
7	**CaO %Wt**	6	3	50	**15**	**CaO %Wt**	6	3	75
	Methanol/Oil	12	4			**Methanol/Oil**	8	2	
	Time	2.5	2			**Time**	4.5	4	
	Temperature	60	1			**Temperature**	120	4	
8	**CaO %Wt**	8	4	85	**16**	**CaO %Wt**	8	4	
	Methanol/Oil	10	3			**Methanol/Oil**	6	1	
	Time	2.5	2			**Time**	4.5	4	5
	Temperature	80	2			**Temperature**	100	3	

In the following, the average effects of eggshell-derived CaO catalyst (see Fig. 7), molar ratios of methanol/oil (see Fig. 8), times (see Fig. 9), and temperatures (see Fig. 10) on biodiesel efficiency have been indicated. The maximum amount of the average impact of the factor in each figure is considered the best level. In each graph, the horizontal axis indicates the level of variables .Considering the maximum biodiesel yield at each level, the highest effects on the efficiency of biodiesel are determined. According to Figs. 7-10, the optimal values of the variables are found in the following situations: methanol/oil molar ratio 10:1, catalyst amount: 2% w/w (1 gr catalyst in 50 gr oil), reaction time: 90 min, and reaction temperature: 80 °C, where the highest efficiency for biodiesel is indicated to be 84%.

As shown in Tables 3 and 4, the optimal alcohol/oil molar ratio values in the esterification and transesterification stages have been reported as 12:1 and 10:1, respectively. A proper amount of alcohol/oil molar ratio is required to break chains of glycerol fatty acids, but its amount should not exceed a specific limit. In addition to the presence of excess alcohol, the purification process becomes more difficult and expensive. It should be mentioned that the excessive use of methanol has no adverse effect on the result and only leads to increasing the separation cost [19, 20].

In most cases, the reaction temperature is close to the boiling point of alcohol in the atmosphere. The boiling point of methanol is between 60 and 70 ° C at 1 atm. However, the rising temperature can generally affect the efficiency of both esterification and transesterification processes. This study reported the optimal temperature for esterification and transesterification at 60 °C and 80 °C, respectively. In addition, the effects of different reaction times on biodiesel efficiency have been studied. We found 30 min and 90 min as the optimal reaction times for

esterification and transesterification processes. The amount of eggshell-derived CaO catalyst is one factor that has the highest effect on biodiesel efficiency. Compared to the critical values for the catalyst concentration reported between 0.25 and 3% oil weight (w/w) in biodiesel production [21, 22], we obtained the optimal values of the eggshell-derived CaO catalyst to be 2% w/w for the transesterification process.

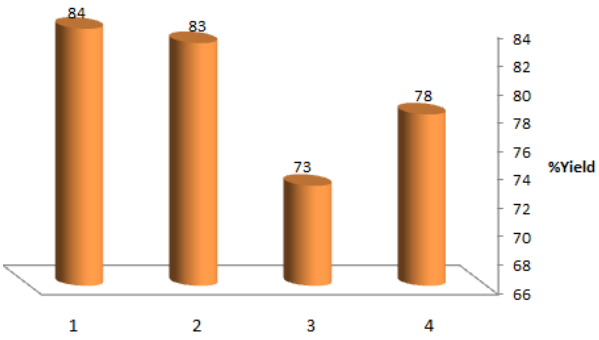

Figure 7. Average effects of the amounts of CaO catalyst (left to right: 1, 2, 3, and 4 gr) on %Yield

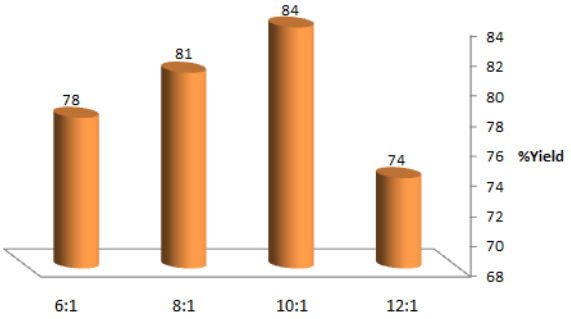

Figure 8. Average effects of methanol/oil ratio (left to right: 6:1, 8:1, 10:1, and 12:1) on % Yield

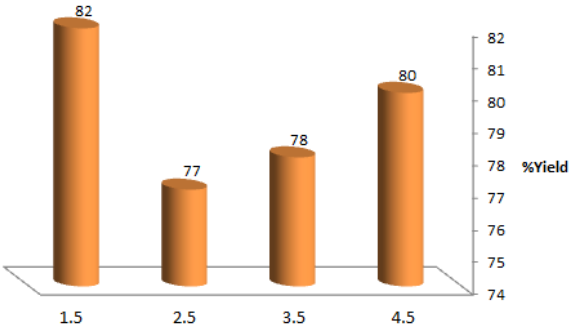

Figure 9. Average effects of different times of reaction (left to right: 1.5, 2.5, 3.5, and 4.5 hr) on % Yield

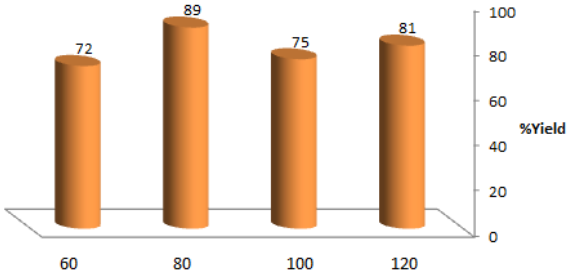

Figure 10. Average effects of the different temperatures of the reaction mixture (left to right: 60, 80,100, and 120 °C) on %Yield

CONCLUSION

In this study, a two-step process was developed to convert waste cooking oil to methyl ester. We synthesized and characterized eggshell-derived CaO as an inexpensive catalyst for the transesterification of waste cooking oil. The first step was acid treatment, which reduced the oil's FFA content to less than 1% using an acid-catalyzed reaction with a ratio of methanol/oil of 12:1 at 60˚C and 30 min reaction time. The effect of molar ratio, amount of catalyst, reaction temperature, and reaction time were also analyzed in the transesterification process. After acid treatment, an alkaline transesterification reaction was carried out at a 1 gr eggshell-derived CaO catalyst with a ratio of methanol/oil of 10:1 at 80˚C for 90 min. The maximum yield of biodiesel is calculated to be 84% using this catalyst.

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