

Synthesis and Characterization of Waste Eggshell-Based Montmorillonite Clay Catalyst for Biodiesel Production from Waste Cooking Oil

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Abstract. The depletion of resources and increase in demand for fossil fuel raise concerns as it is natural and non-renewable. Therefore, it will cause limitation on its availability and continuous reduction. This issue has led to the search for more economic, sustainable, and environmentally friendly alternatives which is biodiesel. The major drawback that reduces the possibility of biodiesel commercialization is the high cost of oil feedstock as it covers 75% of its total production cost. The waste cooking oil is used as feedstock in continuous transesterification as it is the primary option to lower the cost of biodiesel production. Biodiesel can be prepared using waste cooking oil and catalyst through transesterification reaction. In this research, the focus is on the utilization of chicken and quail waste eggshell to synthesize highly active Calcium Oxide (CaO)-based heterogenous catalyst with montmorillonite clay to catalyze efficient conversion of waste cooking oil to biodiesel. The formation of CaO/montmorillonite catalyst was confirmed based on the outputs from X-ray diffraction (XRD) and Scanning Electron Microscopy (SEM). The physio-chemically characteristics of catalysts exhibited a BET surface area from the ranging from 9.2-9.5 m²/g and presence of around 87% of elemental calcium as constituent through XRF characterization. In addition to this, high-performance liquid chromatography (HPLC) analysis is used to determine the conversion of biodiesel using conventional and microwave heating method which revealed a maximum biodiesel production yield of 98%. This optimum biodiesel yield was obtained at reaction temperature, molar ratio of waste cooking oil to methanol, and catalyst amount of 60 °C, 2 h, 1:12, 2.5 wt.% and 5 wt.% for both eggshells, respectively.

1. Introduction

Increase in demand for renewable resources is increasing every year in line with the raise in human population globally thus, developing environmental concerns resulting from continual dependence on fossil fuel sources [1-3]. The increase in number is a contribution from these sectors including transportation, power generation, industry, and households [4]. Currently, the fossil fuel crisis has globally floundered every region's economy post the decline in global stock of the oil consuming countries around the globe. In regard to that, the rapid depletion of fossil fuels and increase in the number of energy needs has encouraged countries to make savings and conduct studies to obtain renewable fuels [4,5]. Biodiesel is considered to act as a substitute or as an additive to diesel fuel which is nontoxic, renewable, and biodegradable [5-7].

Biodiesel fuel is considered to be high in cost in comparison to petroleum-based fuel, since 60-80% of the cost is affiliated with the feed-stock oil, due to the use of expensive, high quality virgin oils such as canola, soybean, sunflower, olive, palm, cottonseed, peanut, and linseed [8,9]. The increase in food commodities and improper waste disposal to the environment has made it necessary to use WCO as a biodiesel feedstock [10,11]. WCO helps in cost reduction and draws attention since they are renewable and readily available. Certain part of the waste cooking oil is utilized for soap preparation and fodder making, otherwise majority of the WCO are illegally dumped into landfills and other water bodies causing environmental pollution [11]. Typically, biodiesel can be produced using methanol and WCO through transesterification process in the presence of catalyst to produce fatty acid methyl ester (FAME) [4].

Transesterification process takes place using homogeneous acid/alkaline catalyst [11,12] that is

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usually referred as conventional process. Nevertheless, this process has side effects which includes formation of side products such as soap formation [13-15]. Free fatty acids (FFAs) are produced during the cooking of the oil causing the WCO inappropriate for traditional direct base-catalysed transesterification due to soap formation during biodiesel production [7]. Soaps are formed when basic catalyst reacts with FFAs with the feedstock. For alkaline-catalysed transesterification reactions, the oil should not contain more than 1% FFA. If the level of FFA reaches this volume, soap formation can impede the separation of the ester from the glycerine and also lower the rate of development of ester [14]. Soap production causes gel to form, increases viscosity, and significantly increases the cost of separating the product. A pre-treatment process is required to remove the FFAs to achieve high conversion with added cost of production [7]. One method to lower the cost is by using heterogeneous catalyst which helps to avoid the washing steps and further eliminates the wastewater generated.

This study focussed on the development of a solid CaO-based catalyst from two types of eggshells consisting of chicken and quail eggshells for the production of biodiesel using WCO [16]. The purpose of this study is to use CaO found in the eggshell as a solid catalyst rather than homogenous catalyst that will impact the decreasing cost of biodiesel production, therefore the biodiesel price can be used to compete with the price of diesel oil. Typical eggshell has a low porosity and high content of CaCO₃, which is important in the biodiesel industry [17]. The eggshell is combined with activated montmorillonite clay to produce CaO/montmorillonite catalyst. Calcium carbonate, magnesium carbonate, calcium phosphate, and other organic materials are components of the eggshells. Eggshell offers development opportunities as heterogenous catalysts based on the pore structure and content of CaCO₃ [18-20].

The use of this catalyst is expected to provide a solution for obtaining an effective and efficient biodiesel production process feasibility study that reduces environmental pollution. The study was conducted in the laboratory scale which involves several stages, namely catalyst preparation, catalyst characterization, and catalyst testing activity to determine the best operating conditions, including catalyst quantity and heating method. The aim of this research is to observe the comparison between the amount of fatty acid converted to biodiesel by using chicken/montmorillonite catalyst and quail/montmorillonite catalyst.

2 Materials and Methodology

2.1 Materials

Eggshells were collected from local eateries at Klang, Selangor, Malaysia. Waste cooking oil was procured from local cafeteria around Universiti Teknologi PETRONAS (UTP), Seri Iskandar, Perak. Montmorillonite clay was available at Block 5 laboratory, UTP. The transesterification reaction was

performed by using methanol as the alcohol and eggshell ash as heterogenous catalyst. All the solvents and chemicals were requested from UTP Block 4 Chemical Laboratory and directly used without any purification except for WCO. Toluene, potassium hydroxide (KOH), Isopropyl alcohol (2-propanol) and phenolphthalein indicator were used in the titration process to determine acid value. Lastly, n-hexane and isopropanol were used to dilute the biodiesel obtained for conversion and yield analysis.

2.2 Catalyst preparation

The chicken and quail eggshell were crushed into small chips separately using a cutting mill and washed thoroughly in tap water to remove any unwanted material adhered on its surface. Then, the crushed eggshells were soaked and rinsed several times with distilled warm water for the removal of excess impurities. The eggshell is then left in a hot air oven at 105 °C for 24 hrs. The dried eggshells were then reduced to small pieces and calcined in a muffle furnace at 900 °C for 4 hours under static air condition.

The activation of montmorillonite clay was done by chemical activation and kept under alkaline condition. The clay was calcined at 800 °C for 8 hours, then soaked in 0.4M potassium hydroxide (KOH) solution in 50 °C for 6 hours. The wet impregnation procedure is used to synthesize both the calcium oxide, CaO retrieved from the eggshells and clay as a solid catalyst. Therefore, the calcium oxide from both eggshells and the activated clay is mixed together. The catalyst is prepared by 15 mmol CaO for each 1-gram montmorillonite clay. This mixture was then soaked in KOH 0.02 M in 70 °C for 4 hours, kept for 24 hours, filtered, and dried at 100 °C for 20 hours, and then calcined for 2 hours at 900 °C to attain the CaO/Montmorillonite catalyst.

2.3 Catalyst characterization

The properties of the catalyst were assessed as follows: The surface characteristics of the catalyst was obtained by X-ray diffraction (XRD) technique to determine the diffraction lines located at different profile of catalyst. N₂ adsorption-desorption isotherms at -196 °C were measured by the static method in an adsorption analyzer (Model: X' Pert3 Powder & Empyrean, PANalytical). Scanning electron microscope was employed to determine the morphology structure of the element. X-ray fluorescence (XRF) (Model: Bruker; S8 Tiger) gave the elemental composition of the calcined catalyst (eggshell ash). Specific surface areas (S_{BET}) were determined by Brunauer-Emmett-Teller (BET) method using an ASAP 2020 surface area analyzer (Micromeritics). Fourier-transform infrared (FTIR) (Model: Perkin Elmer, Spectrum One) analysis to identify the crystallinity of sample and provide information on dimensions of unit cell.

2.4 Transesterification process

The WCO that have been collected, was filtered to remove food residues, and then heated at 100-105 °C for 30 minutes. Transesterification process is conducted in a batch type reactor which consists of three neck glass, equipped with reflux condenser and magnetic hot plate stirrer. Desired amount of catalyst prepared, methanol and WCO is introduced to the reactor and reaction will take place for 65 °C with range of 1 hour. The reaction mixture is required to be stirred thoroughly at constant speed using magnetic stirrer. Sample is collected after every 30 minutes after the start of the reaction in the batch type reactor for analysis. For the transesterification procedure, the variables that have been constant are the methanol to waste cooking oil ratio which was 12:1.

Besides, the catalyst loading is differed from 2% and 5% (wt.% to the waste cooking oil) for both eggshell catalysts. The similar procedure is repeated for the microwave irradiation process. The conversion of oil to fatty acid methyl esters is analyzed by using a High-Performance Liquid Chromatography (HPLC). The yield of biodiesel can be calculated by the equation defined below in Equation (1):

$$\text{Yield of Biodiesel (\%)} = (\text{weight of actual biodiesel} / \text{weight of oil used}) \times 100\% \quad (1)$$

The conversion of oil is determined using the following Equation (2):

$$\text{Conversion (\%)} = (\text{Weight of Triglycerol} - \text{Weight of biodiesel}) / (\text{Weight of biodiesel}) \times 100\% \quad (2)$$

2.5 Biodiesel Purification

The crude biodiesel produced through transesterification process was purified using distilled water. The purification was completed to meet the international standard specification provided by American Society for testing and materials [21]. The biodiesel produced during transesterification if of low value due to the contaminants such as residual methanol, glycerol, catalyst, glycerides and FFA is present [22]. Crude biodiesel was mixed with distilled water and centrifuged at 600 rpm for 5 minutes to avoid formation of emulsion, then slowly extracting the layer of biodiesel without water. The process was repeated twice to ensure colourless wash water is obtained, indicating complete removal of impurities.

3 Results and Discussion

3.1 Catalysts Characterization

3.1.1 Catalyst Composition

Table 1 depicts the chemical composition of chicken/montmorillonite catalyst and quail/montmorillonite catalyst from the X-ray fluorescence (XRF) analysis. 87.35 wt% of calcium in addition with phosphorus

amount was achieved by quail/clay catalyst since there is traces of phospholipid in the quail eggshell's membrane. Other elements are also traced as tabulated and all of these elements are impurities of the CaO catalysts. Increase in calcium content improves the quality of biodiesel because CaO exhibits higher active sites for the reaction to take place.

Table 1 Chemical composition of eggshell/clay catalysts.

Elements	Chicken/Clay	Quail/Clay
	Concentration (%)	
Ca	86.6	86.9
Si	6.86	7.02
Al	1.73	1.74
K	1.54	1.48
Fe	1.3	1.34
Mg	0.75	0.6
P	0.7	0.45

3.1.2 Physical Properties of Catalysts

Moderate pore diameter ranging from 17 nm to 20 nm and surface area within 9 m²/g is characterized for both the catalysts which allows reactants to diffuse into the interior of the catalyst as shown in Table 2 [23]. For better diffusion of reactant and product molecules, high pore diameter is desirable. The BET total surface area of CaO is low under the calcination condition of 800 °C which is similar to other studies observation at 9 m²/g [16]. There are possible differences in the surface area due to the initial particle size of the starting material. Trace pore volume of range 0.03 cm³/g to 0.04 cm³/g was obtained in agreement with previous study [24]. The total basic strength (pH 10.8) for quail/montmorillonite catalyst is recorded and higher than the weakest indicator (pH 9.8) that underwent a colour change. The basic site distribution of CaO (135.2 μmol/g) showed the presence of good basic strength. The active site of the oxide surface will interact with the methanol proton and contribute to the breakdown of the hydroxide bond, causing the transesterification reaction to initiate the formation of methoxide ion [25]. Such features indicate that the calcined eggshell ash collected have strong catalytic activity. Catalytic activity depends on the specific area of the substrate, base intensity and density of the base site.

Table 2. Physiochemical properties of eggshell catalysts using micromeritics physisorption and chemisorption analyzer.

Properties	Chicken/Montmorillonite catalyst	Quail/Montmorillonite catalyst
BET surface area (m ² /g)	9.259	9.549
Pore volume (cm ³ /g)	0.034	0.041
Pore diameter (nm)	17.879	20.454
Basicity (μmol/g)	130.2	135.2
Acidity (μmol/g)	87.4	95.03

Basic Strength (H ₊)	H_10.2	H_10.8
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3.1.3 Morphologies of Catalysts

The scanning electron microscopic image of activated montmorillonite clay and CaO catalyst is shown in Figure 1 above. For the catalyst, the morphology of CaO has irregular pores, shapes, and size, varying one surface to another (30µm of width). The shape of eggshell catalyst was formed by tiny crystals embedded on the large particles, probably due to the heterogenous distribution in the mechanical properties of the eggshell used which can be regarded as an attribute for high catalytic activity [10]. This characteristic heterogeneous morphology of CaO from calcined eggshell has been noted previously under similar conditions [8].

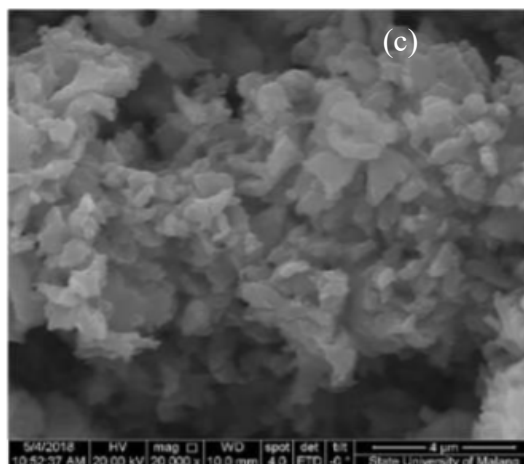
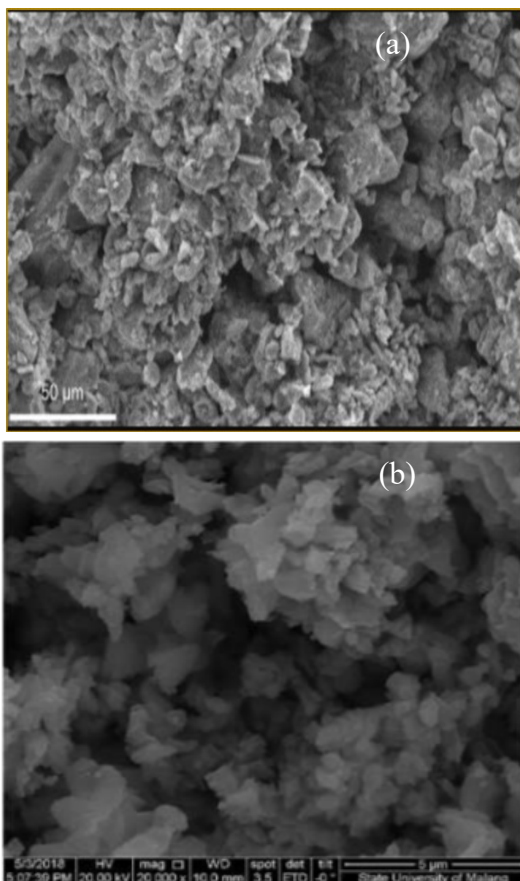


Fig. 1. (a) Morphological form of activated montmorillonite clay, (b) morphological form of chicken/montmorillonite catalyst, (c) morphological form of quail/montmorillonite catalyst.

The scanning electron microscopic image of activated montmorillonite clay and CaO catalyst is shown in Figure(s) 1,2 and 3 above. For the catalyst, the morphology of CaO has irregular pores, shapes, and size, varying one surface to another (30µm of width). The shape of eggshell catalyst was formed by tiny crystals embedded on the large particles, probably due to the heterogenous distribution in the mechanical properties of the eggshell used which can be regarded as an attribute for high catalytic activity [10]. This characteristic heterogeneous morphology of CaO from calcined eggshell has been noted previously under similar conditions [8].

3.1.4 Crystalline phases of Catalysts

Figure 2 shows the XRD patterns for activated montmorillonite clay and the eggshell catalysts. The peaks for activated montmorillonite clay denoted by the blue spectrum appeared at $2\theta = 7.88^\circ; 17.7^\circ; 19.8^\circ; 20.9^\circ$ and 27.6° . The peaks have been decreasing after the point 27.6° as there is a decrease in crystal size which might happened due to the exothermic conditions of the decarbonation process [26].

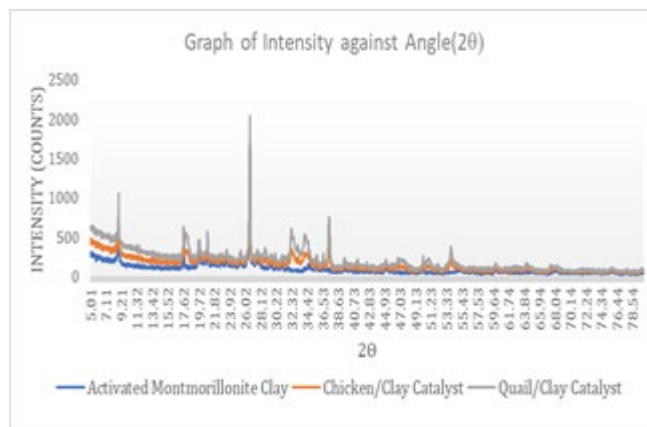


Fig. 2. XRD Spectrum of activated montmorillonite clay and catalyst.

The peaks in orange spectrum depicts the XRD patterns for chicken eggshell/montmorillonite catalyst which have been synthesized by mixing the CaO precursor and activated montmorillonite in KOH solution as explained in the experimental setup. Addition of activated clay into a CaO compound has the tendency to lower the existing high basicity of CaO [9]. The peaks appeared at $2\theta = 17.6^\circ; 26.5^\circ; 28.8^\circ; 33.9^\circ; 36.6^\circ; 37.5^\circ$ and 54° . The lower intensity for this catalyst could be due to the minimal production of crystallite size. The grey peaks above shows the XRD patterns for quail eggshell/montmorillonite catalyst and the diffraction lines are located at $2\theta = 17.8; 26.5^\circ; 29.8^\circ; 33.7^\circ; 35.6^\circ; 36.5^\circ; 46.7^\circ; 52^\circ$ and 54° . The peaks intensity is slightly higher in comparison with the chicken eggshell/montmorillonite catalyst due to higher crystallization.

3.1.5 Functional groups of Catalysts

Figure 3 shows the FTIR spectra for both chicken/montmorillonite clay catalysts. The spectrum of both the catalysts shows the presence of the out-of-plane bending, the asymmetric stretching and the in-plane bending modes of the carbonate groups located at $3642.35\text{ cm}^{-1}, 993.20\text{ cm}^{-1}$ and 894.76 cm^{-1} .

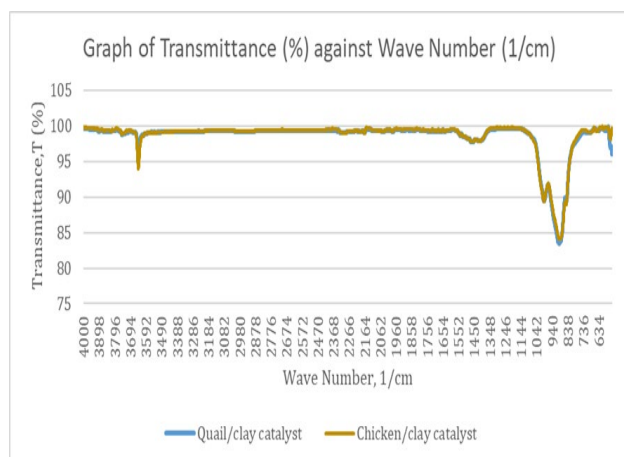


Fig. 3. FTIR spectra of eggshell/clay catalysts.

3.2 Catalytic Performance

Figure 4 depicts the conversion obtained for both the type of CaO/montmorillonite clay catalysts in biodiesel production. Conventional heating method have proven to achieve higher conversion compared to microwave heating in the duration of 2 hours for 2 different values of catalyst loading which is 2 wt.% and 5 wt.%. Quail/montmorillonite catalyst have proven to achieve higher conversion compared to chicken/montmorillonite catalyst in all heating methods and catalyst loading. As shown in Figure 6, lower yield for both conventional and microwave heating was obtained with low catalyst loading of 2 wt%, which was insufficient to catalyze the reaction to completion in the formation of methyl ester. Heterogenous catalyst in biodiesel production should range from 3-10 wt.% for

higher yield [6]. Higher biodiesel yield (99%) was observed at an optimum point of 5 wt.% catalyst loading as shown in Fig. 4. Reaction rates increases as reactants occupies more catalytic sites until saturation was reached. The impact of mass transfer limitation becomes more significant at higher catalyst loading, thus limiting reactants' accessibility to active sites [12]. The transesterification reaction is strongly dependent on the weight of catalyst which consequently affects the yield. An adequate increase in catalyst concentration results in an increase in the number of its active sites, thereby increasing the yield of methyl ester [16]. Excessive catalyst loading leads to high slurry viscosity and consequent poor reaction mixtures [2].

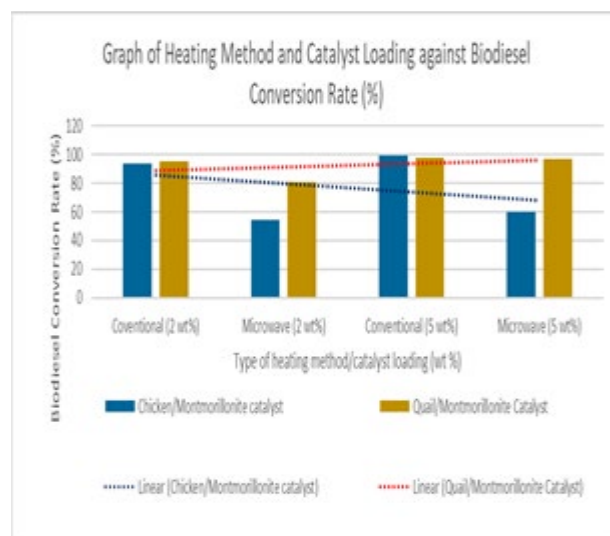


Fig. 4. Heating methods and catalysts activity for Biodiesel production

4 Conclusion

Calcium oxide (CaO) solid base catalyst has been successfully prepared from eggshell waste and activated montmorillonite clay. Three factors were investigated – oil-to-methanol ratio, catalyst loading and heating method – in order to optimize the process. Optimal reaction conditions obtained were methanol-to-oil molar ratio (12:1), catalyst loading of (5 wt.%) and temperature (60°C) using quail eggshell. It was assumed that the catalyst was used in sufficient amount with respect to oil in order to shift the reaction toward the formation of methyl ester. It is proved that the quail eggshell waste catalyst is capable to produce high biodiesel conversion via conventional heating method which is up to 98%. Thus, the study of developing and further engineering CaO solid base catalyst derived from shell waste still need to be enhanced to improve efficiency of biodiesel production from waste cooking oil.

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