Review Paper

# A review of heterogeneous calcium oxide based catalyst from waste for biodiesel synthesis

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**Abstract**
Biodiesel is one of the most promising method to replace the fossil fuels because it is more environmentally friendly. Nevertheless, biodiesel manufacturing costs are much higher compared to conventional fossil fuels. Thus, the biodiesel should be synthesizing from reusable wastes to minimize the production cost. Homogeneous catalyst is the most common catalyst employed in the commercial biodiesel field. However, there are some drawbacks in using homogeneous catalyst in the reaction such as the difficulties faced in separation process of the homogeneous catalyst from the mixture of product. The presence of promising current technology has proved that the utilization of heterogeneous catalyst can assist in overcoming the existing problem of homogeneous catalytic reaction, especially in wastewater generation. The heterogeneous catalysts are more environmentally friendly, easier to separate and its reusability property. Despite its low production cost and its beneficial use as an eco-friendly waste recycle method, waste materials may possess qualities and characteristics that differ from the conventional homogeneous catalyst prior to biodiesel production. This review paper focused in the recent discovery of the heterogeneous catalyst synthesized from natural bio-waste materials, especially CaO-based such as eggshells, seashells and bones for biodiesel production. Apart from that, gypsum, part of the construction waste is proposed as the newly found heterogeneous catalyst. Gypsum exists abundantly due to the rapid development of the economics where construction and demolition activities are happening daily. The utilization of these construction waste-based catalysts may able to provide a sustainable route for biodiesel production. This review will enhance the development and existing scientific data in the area of biodiesel production and the synthesis of CaO-based catalyst especially the synthesis of CaO-based catalysts from construction material.

**Keywords** Biodiesel · Biodiesel production · Heterogeneous catalyst · CaO catalyst · Gypsum

## 1 Introduction

The worldwide energy crisis is a result of the tremendous reduction of fossil fuels as the impact of the rapidly rising population especially in developing nations. In most of the developing countries are primary rely on the fossil fuels to act as the main source of energy. Fossil fuels are certainly able to fulfil the global energy mandate of exceeding 90% and act as the most reliable energy resources available [1]. Moreover, fossil fuels are considered to be eco-destructive because they are either indirectly or directly creating many problems such as global warming and climate change via discharging the greenhouse gases into the atmosphere. Presently, global climate change and depletion of natural resources are drawbacks to relying heavily on fossil fuels resources. Scientists have predicted that the fossil resources might run out in 2050 due to the high demand and consumption of fossil fuels at one hundred thousand

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times than the natural creation [2]. Besides, the global demand is projected beyond the double to ~ 30 Terawatt by the year of 2050 and triple to ~ 46 Terawatt at the end of the century [3]. Furthermore, the price of petroleum product is quite fluctuating. Thus, the energy alternative—biofuel derived energy is gradually gaining the global attention to cater the energy security problems. For instance, biodiesel has been touted to be a substitute to replace the existing traditional petroleum-based fuels. Apart from that, biodiesel is sustainable, biodegradable and eco-friendly [4]. Economical materials such as plant biomass with no greenhouse gases emission of biofuels cause it on stern renewable energy sources to overcome the energy and environment challenges [5]. Those renewable energy resources can be extracted from waste cooking oils, animal fats or vegetable oils which can produce biodiesel emitting less pollutants compared to fossil fuels [6].

The biodiesel is commonly synthesized through transesterification reaction with the help of homogeneous catalyst. There are several disadvantages can be found via this catalytic reaction such as being destructive to the environment by discharging the alkaline wastewater. There are also some limitations for the viable applications such as the complexity in the synthesis process and also complications in conducting the application of homogeneous catalyst on large-scale factories [7].

Therefore, several researches were suggested to use the heterogeneous catalyst instead of homogeneous catalyst in the transesterification reaction to obtain biodiesel. Heterogeneous catalyst does provide more advantages when compared to homogeneous catalyst. For example, it is environmentally friendly, non-corrosive and able to achieve higher yield of biodiesel [8].

The heterogeneous catalyst can be derived from various waste resources such as construction waste. Construction waste can be easily found in any countries, especially the developing countries whereas they built a lot of buildings to provide shelters to the citizens. The construction waste is a pertinent concern in protecting the environment. Taiwan has generated approximately 1.27 million tonnes of construction waste in 2009 [9]. The construction waste contains a wide-ranging proportion of municipal solid waste. The most common method to dispose construction waste is by landfilling. Hence, the potential to apply of using construction waste as the derivation for heterogeneous catalyst for the production of biodiesel can provide a cost-effective approach to recycle the waste and eventually can reduce the production cost for the biodiesel production [9].

## 2 Biodiesel

Biodiesel is an alternative fuel built up from monoalkyl esters through either catalyzed or non-catalyzed reaction of transesterification between triglycerides (oils or fats) and light alcohols (methanol or ethanol). The triglycerides sources can be virgin or used edible oils such as soybean oil, palm oil, vegetable oil or sunflower oil. These used vegetable oils can be exploited as the oil feedstocks even after been used in cooking. There are many advantages of biodiesel, for example, high lubricity, high flash point, low viscosity, biodegradable and environmentally friendly and low emission of greenhouse gases [1]. The physicochemical fuel properties of biodiesel will differ according to different oil feedstocks and alcohols used but biodiesel can be fully utilized as an alternative fuel for petroleum-based fuels to meet the growing energy demands safely and efficiently [10].

## 3 Biodiesel production

There are two typical types of catalyzed reaction: (1) homogeneous catalyst and (2) heterogeneous catalyst. The biodiesel production is commercially produced by homogeneous catalytic reaction with the availability of current implemented technology, i.e. transesterification [11]. The reaction with the occurrence of homogeneous catalyst is considered relatively fast and can achieve high conversion in short time. Nevertheless, it has severe hitches. The catalyst cannot be regenerated or recovered and must be neutralized then removed right after the completion of reaction. This will eventually generate a large amount of wastewater during the purification stage [9]. Furthermore, the separation process of homogeneous catalysts from products is challenging and tedious. Thus, this will involve more equipment and hence, cause higher capital cost [7].

A several promising technologies has implied that the use of heterogeneous catalyst in the transesterification process can help to cater the associated problems faced with homogeneous catalyst. The natural based heterogeneous catalysts are mostly non-corrosive and environmentally friendly. Separation the heterogeneous catalyst from the mixture is much easier than homogeneous catalysts [11]. Besides, the employment of heterogeneous based catalysts in the transesterification reaction can increase the yield and purity of the produced biodiesel [8].

Human generates wastes daily, for example construction activities create significant amount of waste which will impact the environmental either physically or chemically. These environmental impacts will lead to higher energy consumption, greater solid waste generation, more pollutions and more greenhouse gases emission. For example, in 2012, United Kingdom (UK) had produced 100 million tonnes of wastes from construction activities [12]. Despite that, Malaysia as one of the developing countries generates approximately twenty-six thousand tonnes of wastes daily from construction activities which means there are approximately nine million wasted produced annually [13]. The citizens' awareness of proper managing construction waste is considering as low in Malaysia and therefore, lead to an increase in illegal dumping sites. There are few types of construction waste, for example, wood, concrete, metals, bricks, drywall, roofing, plastics, cardboard and others. Some of these construction waste can convert into useful material or can be used as the raw material to produce heterogeneous CaO based catalyst. For instance, gypsum is a waste from false ceiling can be used as the raw material to synthesize the heterogeneous CaO based catalyst to produce biodiesel. Moreover, waste reuse and recycling allow to fully utilize these wastes and will eventually reduce the waste disposal.

## 4 Biodiesel catalyst

There are many types of technology that can be used to produce biodiesel. For instance, transesterification reaction between edible or non-edible oil with alcohols such as methanol with or without the presence of catalyst. There are also 2 main categories of catalysts which are the homogeneous catalyst and heterogeneous catalyst. Most of the biodiesel production are obtained with the assistance of homogeneous catalyst via present technologies, transesterification. However, there are many disadvantages using the homogeneous catalyst in the chemical reaction such as difficulties in separation of the biodiesel mixture and generate a large volume of wastewater [14]. Thus, scientists have further improved the existing technologies, the associated problems are able to be solved by using heterogeneous catalyst in the biodiesel production process. The heterogeneous catalyst can be synthesized from many resources such as biomass or non-biomass based, that is construction waste. The benefits and drawbacks of homogeneous and heterogeneous catalyst are shown in Table 1.

Presently, the research on the heterogeneous catalysts synthesized from biomass or non-biomass are explored extensively because of its potential to decrease the capital production cost. Through high temperature treatment, biomass will be chemically transformed into carbonaceous material and this carbon based material which contain the anticipated functional groups can be as a catalyst [20]. Besides that, the calcium carbonate content in some of the biomass waste can be converted to calcium oxide based catalyst to be applied in transesterification process [21].

Calcium oxide component (CaO) is non-corrosive, eco-friendly and abundantly. It is easily to handle due to the advantages of its low solubility, high alkalinity and most importantly, it can be reused after the reaction. Hence, it has been defined as one of the most frequent used heterogeneous catalyst in biodiesel production [22]. CaO can be derived from natural resources such as cement-based

**Table 1** Comparison of homogeneous and heterogeneous catalyst based on advantages and disadvantages

| Type of catalyst | Advantages | Hitches |
|---|---|---|
| Homogeneous base catalyst | There is no water formation in transesterification process [15]<br>Alkaline catalytic (2 steps) transesterification reaction from waste vegetable oil is economical in producing biodiesel [16]<br>Sodium hydroxide (NaOH) and potassium hydroxide (KOH) are easy obtainable and economically feasible [17]<br>The reaction occurred at trivial reaction which reduce the usage of energy [15]<br>The rate of reaction is roughly 4000 times faster than transesterification process using acidic catalyst [17] | There is occurrence of saponification when the free fatty acids composition in the oil is more than 2 wt% which will decrease the biodiesel yield and cause difficulties in purification process [15]<br>There will be more wastewater eliminated through purification stage [18]<br>The catalyst can only be used once<br>High sensitivity towards free fatty acids content [17] |
| Heterogeneous base catalyst | The catalyst is reusable and recyclable [18]<br>Separation process from product is easier [18]<br>The reaction rate is much faster than acid-catalysed reaction [17]<br>The life span of the catalyst is long [15]<br>The reaction happened at mild reaction which eventually minimize the energy utilization [17] | The catalyst is poisonous during exposure to ambient air [17]<br>The basicity property causes the sensitivity towards the free fatty acids content to be high [17]<br>There is limitation in diffusion [19]<br>The transesterification process will require more molar ratio of methanol to oil [19] |

wastes, seashells, eggshells or bones. The calcium oxide can be derived from calcium carbonate compound via high temperature calcination process ranging from 700 to 1000 °C [7]. The fully transform temperature for calcination process varies according to different calcium carbonate waste feedstocks. A summary table of comparison for CaO catalysts derived from several sources had been tabulated as shown in Table 2.

### 4.1 Heterogeneous catalyst derived from eggshell

The alkaline earth metal oxides such as calcium oxide which contains a high level of base is very suitable in synthesizing the heterogeneous catalyst. There are several researches have reported that the calcium oxide is the most reliable heterogeneous basic catalyst in biodiesel synthesis [38]. The calcium oxide is can be easily obtained in various of low-cost materials or wastes such as eggshell. This can be act as the heterogeneous catalyst and used in the biodiesel production with different types of oil feedstocks and alcohol [39].

Eggshell is plentifully available worldwide as the chicken egg is the most preferable food for breakfast and it is mostly daily consumed in most of the nations. Calcium carbonate is one of the major compounds in a chicken eggshell with 96% of oxides [40]. The utilization of eggshell as a catalyst can indirectly assist in reducing the landfill waste which can maintain the clean environment. Besides that, reuse of the waste eggshell can decrease the production cost for replacing or purchasing new catalyst in the biodiesel production.

Several researches showed the use of eggshell as a catalyst in the transesterification reaction for biodiesel production can increase the yield of biodiesel and can ease the separation process [8]. Atadashi et al. [15] have reported that converting eggshell into a heterogeneous catalyst for biodiesel production has simplified the purification stage and thus consumed less water and energy.

Usually, the obtained eggshell was washed with distilled water for dust and impurities removal [41]. The cleaned eggshell will be dried in an oven at 100 °C overnight and the particle size was further reduced using grinding mortar. Khemthong et al. [42] has stated that the high temperature calcination process took place within the temperature range between 500 and 1100 °C in air for 180 min in a muffle furnace. This calcination process

**Table 2** Catalytic performance for transesterification process over CaO catalysts derived from different source

| Source | Catalyst | | Type of oil | Operating conditions | | | | Yield (%) | References |
|---|---|---|---|---|---|---|---|---|---|
| | Type | Heat treatment | | Molar ratio of M:O | Catalyst loading (wt%) | Time (min) | Temperature (°C) | | |
| Chicken eggshells | CaO | 1000 °C, 2 h | Soybean | 9:1 | 3 | 180 | 65 | 95 | [23] |
| | CaO | 900 °C, 2 h | Karanja | 8:1 | 2.5 | 150 | 65 | 95 | [24] |
| Mud crab | CaO | 900 °C, 2 h | Palm olein | 0.5:1 | 5 | 150 | 65 | 98.8 | [25] |
| Cockle | CaO | 900 °C, 2 h | Palm olein | 0.54:1 | 4.9 | 180 | 65 | 99.4 | [26] |
| Mussel | CaO | 1050 °C, 2 h | Soybean | 24:1 | 12 | 480 | 60 | 94.1 | [27] |
| Clam | CaO | 900 °C, 3.5 h | Waste frying oil | 6.03:1 | – | 360 | 60 | > 89 | [28] |
| | CaO | 900 °C, 4 h | Palm olein | 9:1 | 1 | 120 | 65 | 98.0 | [29] |
| Turkey bones | CaO/biological tri-calcium phosphate (BTCP) | 909.4 °C, 4 h | Mustard | 9.9:1 | 4.97 | 180 | 70 | 91.22 | [30] |
| Bovine bones | CaO | 350 °C – 1000 °C, 6 h | Soybean | 6:1 | 8 | 180 | 65 | 97.0 | [31] |
| Pig bones | CaO/ hydroxyapatite (HAP) $K_2CO_3$ | 600 °C, 4 h | Palm | 9:1 | 8 | 90 | 65 | >90 | [32] |
| Sheep bones | CaO/ hydroxyapatite (HAP) | 600 °C, 8 h | Canola | 12:1 | 5 | 300 | 60 | 95.18 | [33] |
| Cement | CaO | 450 °C, 3 h | Soybean | 24:1 | 4 | 180 | 65 | 98.5 | [34] |
| Dolomite rock | CaO | 800 °C, 2 h | Palm kernel | 30:1 | 6 | 180 | 60 | 99.9 | [35] |
| Lime mud | CaO | 800 °C | Peanut | 15:1 | 6 | 180 | 64 | 94.4 | [36] |
| Red mud | CaO | 200 °C, 5 h | Soybean | 24:1 | 4 | 189 | 65 | 94.0 | [37] |

can fully transform the calcium carbonate to calcium oxide component.

### 4.2 Heterogeneous catalyst derived from seashell

There are different types of catalyst which can be derived from waste which can reduce the waste disposal and preserve the environment. Calcium oxide is considering as the high potential solid catalyst used for biodiesel production which can be derived from various sources such as seashells. These waste seashell sources are cheap and most importantly is they are eco-friendly [43]. The seashells of some certain species contain greater than 90% of calcium carbonate which can be used to convert into calcium oxide through calcination process at the temperature beyond 800 °C [44]. Nevertheless, there is one major concern when using calcium oxide as the heterogeneous catalyst for biodiesel production. The calcium oxide surface is easily contaminated by moisture attachment to form calcium hydroxide when exposed to ambient air because of the absorption of carbon dioxide and water [45]. The prevention of exposure of calcium oxide to atmosphere after the calcination process is considered to be not practical. Kouzu et al. [46] reported the heterogeneous catalytic activity of calcium oxide will be reduced once it exposes to the air as short as 3 min.

Also, there are a few studies showed the employment of seashells as a heterogeneous catalyst with doping of aluminum and zinc nitrate via dissolution–precipitation method. From the studies, the catalyst was calcined at 500 °C reported its surface contains a higher amount of calcium. The nanocrystallites of calcium oxide are well discrete and thus, it has the strongest ability to resist ambient air with relative humidity of 73% up to 72 h [47].

### 4.3 Heterogeneous catalyst derived from bones

There are quite a number of wastes can be re-calcined as a solid catalyst in biodiesel production such as animal bones. The reutilization of wasted animal bones can be a cost-effective method to synthesize the heterogeneous catalyst for biodiesel production [48]. The cost of catalyst is having a direct relationship with the overall production cost. Hence, the re-calcined of animal bones as a low-cost heterogeneous catalyst can help to reduce the production cost of biodiesel synthesis [49].

The animal bone is considered one of the best source of solid waste which can be used as catalyst due to the availability in the countries. The catalyst synthesized from animal bones has shown a good consistency and heterogeneous catalytic performance in the transesterification process but required higher catalyst loading with a longer reaction time. Hence, the usage of animal bones become impractical and non-economically feasible, in terms of operating cost. There are researches done to overcome the problems by impregnating the calcined animal bones in potassium hydroxide aqueous solution to improve the specific chemical properties of the bones which is much related to increase the heterogeneous catalytic activity [50].

The obtained animal bones were washed with hot water for ~ 240 min to remove the dust, impurities and cartilages on the bones. Then, it was dried at 110 °C for ~ 5 h. The animal bone particles were further crushed into powder form using a mechanical grinder. The powder was activated via calcination process within temperature of 500–1100 °C.

### 4.4 Heterogeneous catalyst derived from construction waste

Reusing and recycling is an effective method to reduce the landfill size and fully utilize them to produce and develop new products to reduce the dependency of these natural resources. Daily wastes are produced from either construction site, industries or households. The construction waste disposal is normally dumped to landfills or the irresponsible contractors will leave them there. There are many types of construction waste can be found, for example, concrete, woods, gypsum or bricks. These wastes can be fully utilized to reuse or regenerate as a new beneficial product, such as solid catalyst and so forth.

For example, the cement-based waste collected from construction site can be further processed into a useful heterogeneous base catalyst to be applied in the production of biodiesel. Most of the unwanted concretes are generally handled by burying them and this eventually will cause a severe environmental pollution. There are 180 million tonnes of shattered concrete generated from construction sites annually. Hence, these cement waste can be transformed into useful item instead of dumping them as a waste [9]. The cement-based waste is considered a promising calcium sulphate source due to its high conversion rate and the presence of metal oxides which further enhance the catalytic activity of the triglycerides alcoholysis in biodiesel production [2]. Quartz, portlandite, albite and calcite are the major mineral compounds can be found in concrete which have improve the overall heterogeneous catalytic activity.

There are some researches using cement-based waste as a solid basic catalyst to increase the transesterification reaction rate to produce more biodiesel with higher purity. The collected cement waste from the construction and demolition activities was grounded and then calcined at high temperature to be further applied into biodiesel as a solid catalyst. Gimbun et al. [4] has showed that the cement can be fully utilized as a heterogeneous catalyst and achieved a conversion of 95% in presence of excess oil at temperature of 60 °C and reaction time of 180 min. While, the 5 wt% catalyst loading has accomplished the highest conversion of 96.9% when the calcination temperature was set at 700 °C with a reaction temperature of 65 °C and reaction time of 480 min [4]. The physiochemical properties of the produced biodiesel are able to meet the range of specifications identified by the international biodiesel standard, ASTM D6751.

### 4.5 Heterogeneous catalyst derived from gypsum

Gypsum, a part of the construction waste is not fully exploit yet in the biodiesel field. Chemically, gypsum is a common natural sulphate mineral with a chemical formula of $CaSO_4{\cdot}2H_2O$ and this calcium sulphate component can be transformed into calcium oxide via the ordinary high temperature calcination process by eliminating sulphur dioxide gas. It can be found as a waste from construction site. For example, gypsum is appeared as a by-product when titanium (IV) oxide is extracted from ilmenite ores using digestion of sulphuric acid [51]. Subsequently, the unused acid will be treated by calcium carbonate to neutralize the mixture. Huntsman Tioxide is one of the most well-known company in manufacturing titanium dioxide ($TiO_2$) worldwide [52]. The annual capacity of this plant in Malaysia is approximately 56 thousand metric tonnes. Thus, plant is indirectly producing approximately 400 thousand tonnes of gypsum as by product per year [34]. Hence, it can be concluded gypsum is abundantly in Malaysia and suitable to act as an alternative to be used as a raw source to synthesize the calcium oxide based heterogeneous catalyst in biodiesel production. Apart from that, the gypsum waste can be found at construction site. Same procedure is proposed, i.e. crush and high temperature calcination are able to produce the efficient solid catalyst for transesterification reaction between the oil and alcohol in a batch of reactor system.

## 5 Conclusions

The review of this paper summarized the heterogeneous catalyst derived from different type of calcium rich wastes. This may benefit the petrochemical industries as the biodiesel production via reutilization of waste can decrease the production cost and is more environmentally friendly comparing to the common homogeneous catalyst (sodium hydroxide and potassium hydroxide). Besides that, recycling daily generated waste and optimized them as a solid catalyst in transesterification can reduce the waste disposal problems. The utilization of unwanted useful waste from landfills, especially construction site can further assist the nation to move from an oil-based economy towards a circular economy. A comprehensive analysis of the gypsum-based heterogeneous catalyst is crucial to be investigated to understand the heterogeneous catalytic activity and characterization before implementing this into biodiesel production. Nevertheless, there are limited researches investigated the feasibility of construction waste in biodiesel production. The physiochemical properties of the construction waste catalyst are believed to have an impact in the conversion of biodiesel by having a better particle size, porosity and effective functional group. Thus, it is significant to find the optimum calcination temperature and operating conditions to produce biodiesel with maximum yield. This review is hoped to able to give an insight in deriving the heterogeneous calcium oxide-based catalyst derived from different type of wastes such as gypsum which acts as the waste from construction site and they are able to act as efficient reusable basic catalyst to be employed in biodiesel production. To conclude, this review has successfully offered an insight to the various possibilities and opportunities of reusing different kind of wastes and huge possibility construction waste gypsum catalyst to be implemented in the near future. The future of biodiesel synthesis has also gained a potential method of advancement to be more economically affordable by classes of society alike as an attempt to reduce harmful emissions that are currently liberated through the use of fossil-based products.

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## Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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