



## MINI REVIEW

### Recent Accounts of Green Catalysts for Biodiesel Production

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Problems with solid waste management have become increasingly pressing in recent years. For some time now, it has been a major contributor to soil pollution, making the use of these materials in a variety of industrial applications an urgent necessity. Our search for replacement fossil fuels for transportation has been prompted by their diminishing availability. Biodiesel is one such alternative that doesn't require any adjustments to diesel vehicles. In this study, we studied the efficiency and reusability of several green catalysts used in the production of biodiesel.

**Keywords:** Biodiesel, Solid waste management, Alkalinity, Calcium, Potassium, Sodium, Catalyst.

#### INTRODUCTION

In present times, the aim of human society is to produce the energy from less carbon emitting sources and development of the eco-friendly, green fuel technology. There are many renewable energy sources like solar, wind, hydroelectric, nuclear energy and biofuels. Biofuels such as bioethanol, biodiesel are replaced as transportation fuels in place of fossil diesel fuels [1]. Biodiesel is an renewable fuel source which is free of sulphur, nitrogen, carbon and polycyclic aromatic compounds. This can be used in vehicle combustion engines without any alteration. Selection of feedstock is very important as it decreases 70% of biodiesel production cost. Initially, edible oils like sunflower, soyabean, palm, ground nut were used. Instead second generation feed stocks like animal fat, waste cooking oils, greases, algae oil are being used for biodiesel production. Nowadays, fifth generation biofuels are used for the production of biodiesel. Recently, linseed oil, mahua oil, jatropha oil, ponagamia oil and castor oil were discovered as an effective feed stocks for making of biodiesel. They are extracted using different extraction methods like organic solvent, supercritical fluid, ultrasound and biological methods [2]. Biodiesel is a fuel made by breakdown of triglycerides present in oil/free fatty acids to alkyl esters and this process is called transesterification [3-5].

Using chemical catalysts are very costly and require more time for the preparation as it involves multistep process [6-8]. So many researchers are working on new process technologies to develop methods, which improve the biodiesel yield and require less reaction time and low molar ratios. There are different production methods present in today's world to produce biodiesel from waste oils, plant based oils, algae oil and greases like microwave and non-catalytic methods. Following methods will be discussed in this review paper [9,10].

**Microwave method:** The microwave method for biodiesel production is a relatively new and innovative approach for producing biodiesel. It involves using microwave energy whose wavelengths ranges from 0.1 to 1 m and frequency range 0.3 to 300 GHz to facilitate the transesterification reaction that converts vegetable oil or animal fats into biodiesel. The process involves placing a mixture of the raw materials and a catalyst into a microwave reactor vessel and heating the mixture using microwave energy. Some of the advantages of the microwave method for biodiesel production include faster reaction times, higher yields of biodiesel and reduced energy consumption compared to traditional methods. However, the method requires specialized equipment and may have safety risks associated with the handling of flammable materials [11-13].

**Non-catalytic method:** Due to the high cost of catalysts on the market development of non-catalytic methods for producing biodiesel from oil includes the superficial non-catalytic method, which calls for the use of alcohol at extremely high temperatures (300-400 °C) and pressures (80 atm or 1200 psi). Triglycerides are simultaneously transesterified and fatty acids and lipids are methyl esterified. In terms of cost, this procedure is not more affordable than bio-catalytic transesterification. After reaching equilibrium, the reaction takes 3 to 5 min to complete and yields less biodiesel. High energy consumption necessitates the use of pricey high pressure vessels. Due to the limited miscibility of alcohol to oil, the rate of biodiesel production is quite low [14-16]. Catalysts are employed to solve this problem, thus various forms of catalysts are addressed below:

**Catalyst:** Catalysts are the substances that are known to speed up the reactions by providing them with an alternative path. Alike other reactions, there are two basic categories of catalysts when it comes to biodiesel production: Heterogeneous and homogeneous, which are further classified as alkalis, acidic and enzymatic. There is another classification for catalyst named as enzymatic that has recently attracted researchers for its huge advantages. Enzymatic catalyst are known to not form soap, this gives easier purification process. On the other hand, Leung *et al.* [17] explains the disadvantages of the same, stating that they are expensive and time taking processes, this limits their commercial application.

**Homogeneous acid catalyst:** The most often used acid catalysts are  $H_2SO_4$  and  $HCl$  [18]. Waste oils contain free unsaturated fats that can't be converted to biodiesel, which further encourages the formation of soap and prevents the ester, glycerine and wash water from separating when an alkali catalyst is used as a reaction catalyst [19]. Hence, homogeneous acid catalyzed biodiesel production is suggested as a solution to a number of issues caused by homogeneous alkali catalysts. The most prevalent acids used as catalysts in transesterification are sulphuric acid ( $H_2SO_4$ ), hydrochloric acid ( $HCl$ ), sulfuric acid ( $RS(=O)_2OH$ ) and ferric sulphate ( $Fe_2(SO_4)_3$ ) [20]. This reduces saponification since esterification and transesterification take place concurrently [17,21]. In this catalyst, the reaction rate is slower.

**Homogeneous alkali catalyst:** Due to the faster reaction than acid catalyst, homogeneous alkali catalysts are employed to produce biodiesel and the yield is greater than 96% [22-24]. Potassium hydroxide ( $KOH$ ), sodium hydroxide ( $NaOH$ ), sodium methoxide ( $NaOCH_3$ ) and sodium ethoxide are the most well-known homogeneous alkali catalysts ( $NaOCH_2CH_3$ ). Among these, sodium hydroxide and potassium hydroxide are frequently utilized due to their low cost and easily availability [21]. In order to produce biodiesel from vegetable oil, a non-ionic alkaline homogeneous catalyst's action and productivity have been described. Saponification will take place if the oil's free fatty acid level exceeds 2 wt.% [21,25] and as a result, a considerable amount of water will be wasted during the purifying process [17].

**Heterogeneous acid catalysts:** For the generation of biodiesel, it has been reported that commercial sulfonic ion-exchange resin is used. When catalysts are homogeneous, contamination

issues arise that necessitate labour-intensive product separation and purification procedures [26], which reduces the cost of the catalysts. Esterification and transesterification processes take place simultaneously despite sharing homogenous catalysts. Over homogeneous acid catalysts, heterogeneous acid catalysts have several advantages. Since the catalyst is in a distinct phase from the product, it is easier to separate the catalyst from the product, recover and reuse them. They also help in reducing corrosion issues because they are known to be insensitive to free fatty acid (FFA) content, which reduces soap production [21,27-30].

**Heterogeneous alkali catalyst:** To overcome the shortcomings of homogeneous catalysts in the production of biodiesel, many researchers are investigating many features of heterogeneous catalysts and their applications. Most heterogeneous catalysts used in the synthesis of biodiesel are either alkali earth metal composites or antacid oxide [27]. Compared to their homogeneous counterparts, heterogeneous base catalysts are shown to be more efficient than heterogeneous acid catalysts [31]. The catalyst cannot be recovered and must be reactivated at the end of the reaction, which limits its use in continuous processing methodologies [26]. This is necessary to prevent side reactions like hydrolysis and saponification or to avoid the need for additional response steps to initially switch over or remove the free fatty acids. By enhancing the catalyst's ability to be reused and provide opportunities for continuous operation, solid catalysts used in biodiesel amalgamation rather than homogeneous catalysts may result in lower production costs [32].

**Enzyme catalyst:** Enzyme catalyst has been the subject of a lot of research in recent years since it tends to reduce wastewater production and facilitate glycerol recovery, which ultimately helps make the process more efficient. Unfortunately, they are not economically viable and as a result, they are not commercially viable because the reaction time and yield are still not that favourable in comparison to the alkali catalyst. The two main kinds of enzymatic biocatalyst employed in the manufacture of biodiesel are intracellular lipases and extracellular. Research demonstrating the use of free lipases for transesterification have primarily concentrated on screening lipases [33,34] and further studying the factors that affect the response rate [35,36]. Solvent lipases offer the benefits of inexpensive preparation costs and a simple preparation process, but they may also typically be used immediately after inactivation [37,38].

**Copper impregnated  $TiO_2$  as heterogeneous nanocatalyst:** De & Boxi [39] used heterogeneous nanocatalysts to produce biodiesel. Catalyst was prepared using wet impregnation method, at a temperature of 45 °C, 3%  $TiO_2$  and 2% Cu produced the highest output of 90.93%. The molar ratio of methanol to oil was 20:1. The physical and chemical results of the biodiesel experiment showed that it was of a very high quality [40,41].

#### Bio-base catalyst used

**Shrimp shell:** A biodegradable shrimp shell catalyst for the synthesis of biodiesel was explored by Yang *et al.* [42].

Shrimp shells were heated to 450 °C without any air present, creating a solid porous substance that was subsequently loaded with 25 wt.% potassium fluoride and activated at 250 °C. Rapeseed oil and methanol were treated with the resulting catalyst to create FAME. At 65 °C, a combination of 9 parts methanol and 1 part oil was combined with a 2.5 wt.% shrimp shell catalyst, which produced an 89.1% conversion.

**Peanut shell:** The solid acid of amorphous carbon employed by Shang *et al.* [43] was made in a single step using a peanut shell as source material. For the synthesis of FAME, oleic acid and methanol were combined with a peanut shell catalyst. The ideal reactional circumstances led to a yield of 8%. Additionally, it was discovered that the catalyst could be used up to six times and produced biodiesel at a yield of 95% or higher, demonstrating the great operational stability of catalyst.

Using peanut shell as catalyst, Kim *et al.* [44] carried out research on the manufacture of biodiesel. The peanut waste (PW) was converted into peanut waste biochar (PWB) through pyrolysis. The PWB was used as catalyst to produce the best yield of biodiesel, which was 95.4%. The reaction was entirely saturable and the yield kept rising as the temperature was raised. Using KOH, a 90% yield of biodiesel was seen and a very long reaction time. Using peanut waste, three valorized goods were also produced (pyrolytic gases, oils, chars).

**Biont shell:** Biont shell was used to generate high performance heterogeneous catalyst as described by Xie *et al.* [45] in a three-step process using potassium fluoride. FAME was obtained through the reaction of rapeseed oil with methanol in the presence of biont shell catalyst resulting in the maximum yield of 97.5% biodiesel. Inadequate carbonization of biont shell resulted in active sites, these sites acted as a catalyst to create biodiesel. The biont shell catalyst matrix's weak polarity also contributed to poisoning the production of glycerol, which boosted the transesterification of rapeseed oil to biodiesel.

**Rice husk and eggshell:** In order to make biodiesel, Chen *et al.* [46] transesterified palm oil using a heterogeneous catalyst made of rice husk and eggshell that had been calcined. CaO was prepared through the calcination at 800 °C. Following incubation, the solution was dried for 24 h at 105 °C in a dry oven. The catalyst that had 30% RHA800-800 produced the highest output of 91.5% biodiesel, when it was manufactured under ideal circumstances. With a yield of more than 80%, catalyst demonstrates an outstanding reusability for more than 8 runs.

**Per pusilla torrey ash:** Chouhan & Sarma [47] employed L. Per Pusilla Torrey combustion-produced ashes. Ash was calcined at 550 °C to maximize its effectiveness as catalyst. The highest conversion of 89.43% of biodiesel was achieved when a 9:1 molar ratio of alcohol to oil was treated with 5% of ash as catalyst for 5 h at 65 °C. It was discovered that the catalyst may be used up to three times in a row with a yield of roughly 25%.

**De-oiled seed waste cake:** The utilization of catalysts produced from oil-cake waste (OCW) and carbonized de-oiled seed waste cake (DOWC) for the manufacture of FAME was described by Konwar *et al.* [48,49]. Sulfonated carbon materials were used as catalysts for the pre-treatment of acid oils. The

oil was first pre-treated with OCW catalyst. Moreover, the de-oiled seed waste cake catalyst shown outstanding activity in rupturing the cellulose structure to provide glucose yields of about 35-53% and up to 97%, when combined with fatty acid esterification.

De-oiled microalgal biomass (DMB) obtained from oleaginous microalgae (*Tetrademus obliquus*) was employed as catalyst by Roy & Mohanty [50]. The FAME yield was examined using different methanol and oil molar ratios (8:1 to 11:1) and catalyst concentrations (1-4 wt.%). With WCO, the catalyst had a greater catalytic activity and a maximum yield of 94.2% to 96.2%. Four successive repetitions of removing the catalyst from the process and reusing it resulted in a > 90% FAME yield.

**Eggshell using frying oil:** Quick lime (CaO), which was used by Niju *et al.* [51] to produce biodiesel using leftover cooking oil and a lab-scale continuous flow jacketed reactive distillation equipment, was obtained from eggshells. Methyl ester conversion was reported to be at its maximum (93.48%) at an inlet flow rate of 0.2 mL min<sup>-1</sup>, the catalyst bed height of 150 mm and a methanol to oil ratio of 4:1. Similarly, Dhawane *et al.* [52] carried out the research to produce biodiesel using used frying oil and used chicken egg shells. To determine the maximal production of biodiesel, 16 tests involving the transesterification of WFO under various processing conditions were conducted using the Toguchi method. The highest yield of biodiesel, in 4 wt.% catalyst was 93.27%.

**Turbonilla striatula waste:** *Turbonilla striatula* discarded shells were used by Boro *et al.* [53,54] to prepare a heterogeneous catalyst for the production of biodiesel from the mustard oil. The best results were observed when 3 wt.% of catalyst was handled at 65 °C with a 1:9 oil to methanol molar ratio; the yield was 93.3%. The calcined CaO was doped with Ba in varying amounts (0.5 to 1.5 wt.%) in an effort to boost production. Several of the active sites and the basicity for reuse were lost by catalyst. When 6:1 methanol to waste cooking oil was reacted for 3 h in the presence of 1.0 wt.% at the reaction temperature of 65 °C, the maximum conversion of less than 98% was noted.

**Malleus malleus shell:** Niju *et al.* [55] used Box-Behnken design (BBD) to optimize a study on malleus malleus shells (MMS) as a catalyst to create biodiesel. The wet impregnation method was used to prepare the catalyst. Highest biodiesel production of 93.81% was recorded using 7.5 wt.% catalyst, methanol to oil ratio of 11.85:1, reaction time of 86.25 min at 65 °C and BBD to optimize MMS catalyst for transesterification reaction.

**Capiz shell:** Capiz shell waste was also utilized for its potential as a biodiesel production catalyst as investigated by Suryaputra *et al.* [56]. The following conditions were used for transesterification, the molar ratio of palm oil to methanol was 1:8, the mixture was agitated at 700 rpm with a magnetic stirrer at 60 °C. The highest yield of biodiesel was produced at 3 wt.% of catalyst resulting in the conversion of 93.2%. Three further transesterification reaction cycles were conducted to assess the capacity of the catalyst for reuse.

**Mussel shell:** Hu *et al.* [57] conducted research on a catalyst made from freshwater mussel shell (FMS), which was produced

by a three-step. Chinese tallow oil was transesterified using the produced FMS catalyst. When an 1:12 oil to methanol combination was reacted at 70 °C for 1.5 h in the presence of 5% catalyst, an optimal yield of 90% and above was observed. Similarly, Rezaei *et al.* [58] also prepared the heterogeneous catalyst from waste mussel shell, which was calcined at 1050 °C. At a loading of 12 wt.%, the soybean oil to methanol ratio produced the highest output of 1:24 and a yield of more than 94% was attained with 100% purity. The catalyst's ability to be reused was tested in two separate ways: after each usage, it was washed and dried, then calcined at 1050 °C. After 5 runs, the yield for the repeatedly calcined catalyst dropped to more than 37% and the yield for the washed and dried catalyst dropped to 59.1%.

**Chicken bones using waste cooking oil:** Farooq *et al.* [59] investigated the use of leftover chicken bones as a catalyst for the generation of biodiesel from the waste cooking oil (WCO). Once bones were calcined at 900 °C, the byproduct was employed to transesterify the WCO and produced a maximum yield of 89.33% at 5.0 wt.% catalyst, 1:15 oil to methanol molar ratio and 65 °C reaction temperature. Further research showed that the catalyst provides a four-time reusability.

Tan *et al.* [60] conducted an investigation to determine whether or not bones from leftover fish and poultry might be used to produce a catalyst. Catalyst recovery after five consecutive runs was shown to be 50% effective. The waste bones were used as catalyst for 1.54 h at 65 °C, yielding a maximum of 89.5%. Since the catalyst has a greater surface area, a higher basicity value and more active sites, which improve the transesterification reaction, it was employed to create >90% biodiesel yield.

**Waste bones:** In order to create a low-cost heterogeneous catalyst for the manufacture of biodiesel from palm oil, Obadiah *et al.* [61] also employed leftover animal bones. When a 1:18 oil to methanol molar ratio was reacted for 4 h at 65 °C in the presence of 20 wt.% of catalyst with continuous stirring at 200 rpm, the best conditions for the generation of biodiesel were observed. Khan *et al.* [62] used Ostrich bones and waste cooking oil (WCO) for the creation of biodiesel. The highest production of 90.56% biodiesel was obtained after 4 h at 60 °C, 5 wt.% of catalyst and 15 wt.% of methanol. The catalyst was retrieved and four successive runs using it resulted in a yield of more than 80% biodiesel.

**Rohu fishbone using soybean oil:** In order to generate biodiesel from soybean oil, Chakraborty *et al.* [63] applied scrap Rohu fish (*Labeo rohita*) bone as raw material for the catalyst preparation. According to additional information, an optimal yield of 97.73% was obtained with a calcination temperature of 97.42 °C, oil to methanol molar ratio of 1:6.27 and a catalyst concentration of 1.01 wt.% oil. According to the findings, the synthesized catalyst has a reusability of up to six times and is a high-performance, inexpensive heterogeneous catalyst.

**Synthesis of beta-tricalcium phosphate from herring fishbone:** Using herring fish bone, Bitire *et al.* [64] produced a catalyst of tricalcium phosphate (TCP) (HFB). The process of Soxhlet extraction was used to prepare parsley seed oil. At

a catalyst concentration of 3 wt.%, a reaction period of 1.5 h, a temperature of 65 °C and a molar ratio of 10:1, biodiesel produced at the highest yield of 93%.

**Turritella terebra shell:** Mohiddin *et al.* [65] conducted research on the utilization of *Turritella terebra* shells (TTS) as catalyst in the synthesis of biodiesel, with chicken fat served as the feedstock. One step transesterification was carried out using 99% pure methanol. After producing biodiesel, catalyst was collected and used five times in a row to produce biodiesel with an 80% yield. *Turritella terebra* shells produced the maximum biodiesel output of 94.03%.

**Walnut shells:** Walnut shells have been used to develop a catalyst by Miladinovic *et al.* [66] by utilizing 5 wt.% catalyst and 12:1 molar ratio of methanol to oil at 60 °C, the maximum biodiesel yield of 98% was collected in just 10 min. Catalyst was recovered and utilized again for five more experiments in a row, producing a biodiesel yield of over 70%.

**Palm empty fruit bunch:** Yaakob *et al.* [67] conducted research on the transesterification of *Jatropha curcas* L. oil using palm empty fruit bunches (EFB). The best conditions were found to be an oil to methanol ratio of 1:15 while reacting at 65 °C for 90 min with 20 wt.% EFB ash catalyst. Moreover, for the KOH-EFB-catalyzed processes, an oil to methanol ratio of 1:15 combined with 45 min at 65 °C and 15 wt.% KOH doped ash produced a yield of greater than 98%. In the similar manner, Wong *et al.* [68] employed oil palm empty fruit to create a catalyst for the synthesis of glycerol-free biodiesel. Inter esterification was carried out to obtain glycerol free biodiesel. A 50:1 molar ratio with a 10 wt.% catalyst dosage at 100 °C for 8 h produced 50.5% biodiesel production.

**Musa balbisiana Colla underground stem:** Sarma *et al.* [69] investigated the use of an ash catalyst made from *Musa balbisiana* Colla underground stem (MBCUS) in a high temperature/high pressure (HT/HP) reactor for the production of biodiesel from *Jatropha curcas* L. oil (JCO) that contained a high concentration of free fatty acids. Under conditions of high temperature (275 °C) and internal pressure (4.2 MPa), the catalyst proved to be extremely useful and efficient and assisted in the production of 98% FAME from JCO. Also, it was found that the FAME produced had a significantly lower initial acidity (18.4 mg KOH/g) than JCO (3.4 mg KOH/g).

**Coconut husk:** Vadery *et al.* [70] conducted research on the husk of the coconut in order to produce a heterogeneous catalyst. The optimum calcination temperature was found to be 350 °C. A 30 min reaction at 45 °C with the addition of coconut husk resulted in a 97% biodiesel production.

A catalyst of the same type was produced by Thushari *et al.* [71] utilizing coconut coir husk (CCH), which contains waste palm oil (WPO) as the feed material. In this process, WPO didn't require to be pre-treated in order to produce biodiesel. It was found that reuse catalyst provided a decent FAME yield of 77% up to four cycles. In a 12:1 methanol to oil molar ratio, with 10% catalyst at 130 °C for 3 h, 89.8% FAME was produced from WPO using a CCH catalyst.

**Wood ash:** Sharma *et al.* [72] employed wood ash, a highly alkali substance, as a catalyst for the manufacture of biodiesel. To create activated wood ash catalysts, it was further

activated with  $K_2CO_3$  and  $CaCO_3$  via a double carbonate solid-state process. Wood ash was burned to high temperatures in the absence of oxygen to eliminate contaminants. When *Jatropha curcas* oil (JCO) was reacted with a 1:12 oil to methanol composition for 15 min at 40 °C in the presence of activated wood ash catalyst and calcined wood ash catalyst, over 99% of the biodiesel produced is produced.

**Egg shell, snail shell and wood ash mixed:** Adepoju *et al.* [73] developed a mixed catalyst for the manufacture of FAME out of three different waste materials. As a feedstock, palm kernel seed, ukpaka seed and dika nut were used to produce oil. The catalyst was recovered and used three times, improving the yield from 91.30% to 93.01%. Upon the 13th run, which had the following conditions: a molar ratio of 8:1, a reaction time of 64.71 min, a catalyst content of 4.5 wt.% and a reaction temperature of 61.61 °C, showed the highest FAME yield of 98%.

**Dolomite rocks:** The utilization of naturally existing dolomite rocks as a potential source of heterogeneous catalysts was examined by Ngamcharussrivichai *et al.* [74]. According to experiments, a 15:1 methanol to oil reaction carried out for 3 h in the presence of 10% calcined dolomite had 99.3% success rate. However, the catalyst did not demonstrate reusability over the long term and the yield declined to below 20% after the fourth repeated cycle. According to results from further process optimization, catalyst gave a yield of 98% and furthermore gave excellent reusability with 80% yield even after 10 cycles.

## Conclusion

Use of naturally occurring waste material derived catalysts for biodiesel production has gained increasing attention due to their potential for cost-effective and sustainable biodiesel production. Waste materials that have been investigated for use as catalysts include eggshells and animal bones, *etc.* In many cases, for example, eggshells have been found to be effective in the transesterification reaction of waste cooking oil, while animal bones have been used to prepare solid base catalysts for the transesterification of vegetable oils. The use of these materials as catalysts not only contributes to waste reduction and environmental protection but also provides a low-cost alternative to traditional catalysts. However, further research is needed to optimize the performance of these catalysts and to develop efficient and cost-effective production processes.

## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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