Effect of nanocatalysts on the transesterification reaction of first, second and third generation biodiesel sources- A mini-review

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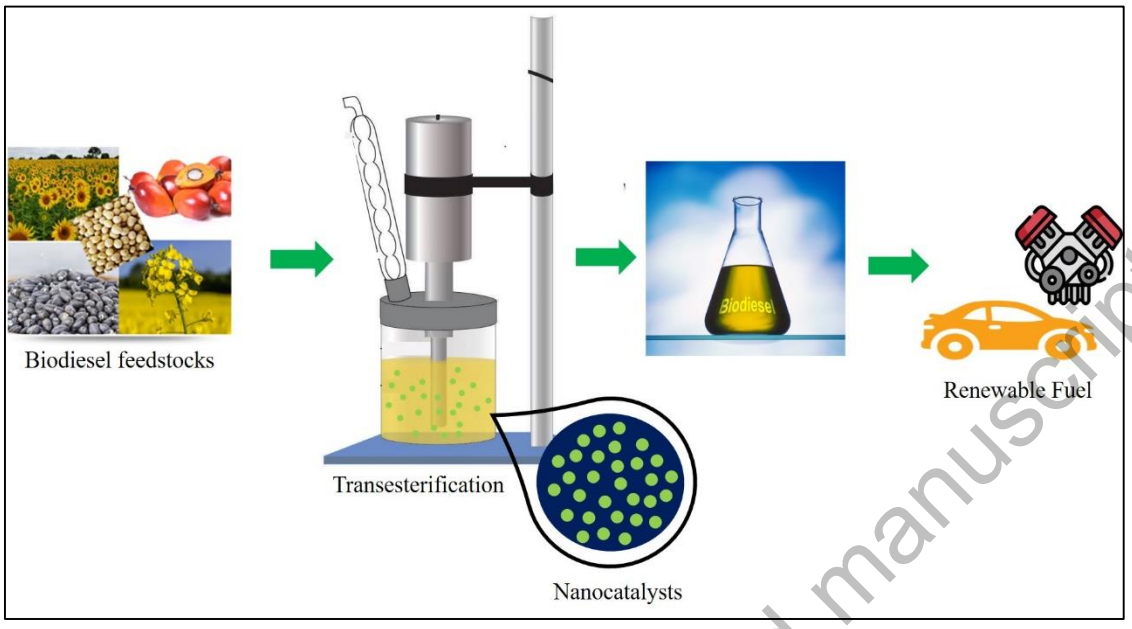
Abstract

Biodiesel is a fuel that has numerous benefits over traditional petrodiesel. The transesterification process is the most popular method for biodiesel production from various sources, categorised as first, second and third generation biodiesel depending on the source. The transesterification process is subject to a variety of factors that can be taken into account to improve biodiesel yield. One of the factors is catalyst type and concentration, which plays a significant role in the transesterification of biodiesel sources. At present, chemical and biological catalysts are being investigated and each catalyst has its advantages and disadvantages. Recently, nanocatalysts have drawn researchers' attention to the efficient production of biodiesel. This article discusses recent work on the role of several nanocatalysts in the transesterification reaction of various sources in the development of biodiesel. A large number of literature from highly rated journals in scientific indexes is reviewed, including the most recent publications. Most of the authors reported that nanocatalysts show an important influence regarding activity and selectivity. This study highlights that in contrast to conventional catalysts, the highly variable surface area of nanostructure materials favours interaction between catalysts and substrates that efficiently boost the performance of products. Finally, this analysis provides useful information to researchers in developing and processing cost-effective biodiesel.

Keywords: Biofuel feedstock; biodiesel production technologies; alternative fuel; clean energy; nanocatalysts development; environmental sustainability.

40 **Graphical abstract**



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List of abbreviation

FFA	Free Fatty Acids
FAME	Fatty Acid Methyl Ester
NP	Nanoparticles
MNPs	Magnetic nanoparticles

1. Introduction

Biodiesel has been known as one of the most promising renewable fuels because of its biodegradability, sustainability, and role in the reduction of pollutant emissions in recent years (Naylor and Higgins 2017, Muhammad et al. 2021). Many countries around the world are producing biodiesel from different sources (Figure 1). In addition, biodiesel has become increasingly more affordable and is commonly used in many parts of the world because of the introduction of subsidies and tax exemptions. Biodiesel is the ester of a long chain (C14–C24), and is synthesized from several lipid content sources including vegetable oils, animal fats and waste oil (Khoobbakht et al. 2016, Mukhopadhyay et al. 2017). Glycerol is a by-product of the biodiesel production process and is estimated to enhance the financial benefits of the biodiesel industry further. It has been reported that about 10 wt.% glycerol can be obtained from the total production volume and it can be used as a combustion improver of diesel/biodiesel (Damanik et al. 2018). Biodiesel shows similar characteristics to the diesel fuel in terms of beneficial physical and chemical properties, including viscosity, flash point and cetane number (Fattah et al. 2014, Arbab et al. 2015, Ong et al. 2019).

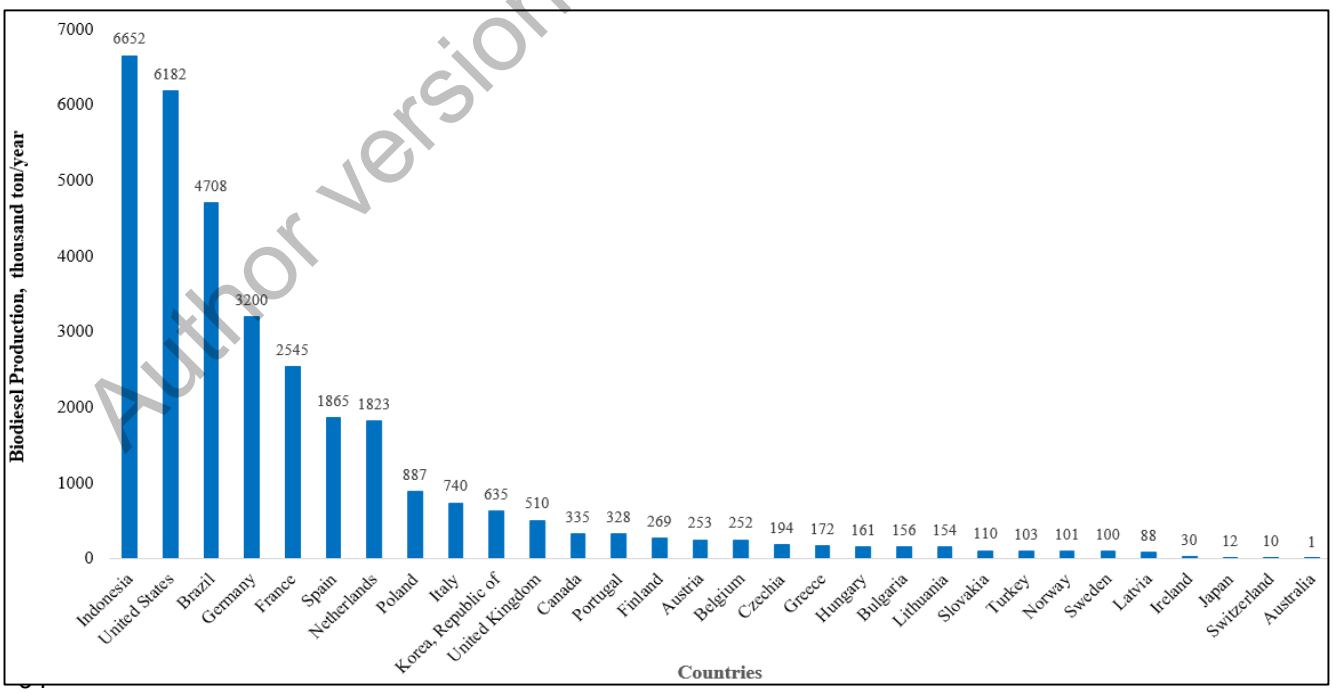

Figure 1: Global biodiesel production by country in 2018 (UNdata 2018).

Over 350 oil-bearing plants worldwide have been listed as possible sources of biodiesel, which can generally be graded into first, second, and third generation biodiesel (Ahmad et al. 2011). Figure 2 shows the main feedstocks of biodiesel in different countries.

Figure 2. Source of biodiesel in different countries, adopted from (Gardy et al. 2019) with permission.

Biodiesel is usually categorized as first, second and third generation based on its source (Ong et al. 2014, Coh et al. 2019, Silitonga et al. 2019, Lau et al. 2020, Silitonga et al. 2020). First generation biodiesel feedstocks are derived from food and edible oils. Commonly used feedstocks for first generation biodiesel include soybean, sunflower, oil palm, rapeseed, canola and cottonseed (Samani et al. 2021). However, it has been argued that the use of edible food crops for the production of first generation biofuels effectively reduces the amount of edible food for human consumption, thus increasing food prices in the global food market (Bhuiya et al. 2020). Although first generation biofuels help satisfy the human need for fuel, at the same time it depletes some resources intended for the even more important human need for nourishment. This provides an incentive for researchers to explore other sources of biofuels that do not disrupt the human food supply.

Second generation biodiesel is obtained from feedstocks from non-edible sources, e.g. crops, non-edible oil and other non-edible sources such as wood, husk, etc., which are then processed to produce biodiesel (Rahman et al. 2016, Rahman et al. 2017). These sources practically eliminate our dependency on edible food crops for the production of fuel, which sparked the "food vs fuel" debate in the first place. Feedstocks used for the production of second generation biodiesel include *jatropha*, mahua, jojoba oil, tobacco seed, *Calophyllum*, and sea mango (Ong et al. 2014, Lee et al. 2020). Commercial and residential waste is also included in this category. The use of these feedstocks to produce second generation biodiesel has been proven to be more efficient and more environmentally friendly compared to the feedstocks used for first generation biodiesel (Pinzi 2009). However, some problems remain. By its very nature, crops require fertile land to grow, and the cultivation of non-edible crops for second generation biodiesel requires an extensive amount of fertile land, which competes with land used for the cultivation of edible food crops. Third-generation biodiesel reduces both the food and land problems related to first and second generation biodiesel. Algae, specifically microalgae, are used as feedstocks for the production of third generation biodiesel (Chia et al. 2018, Mofijur et al. 2019). The use of microalgae for biodiesel production is considered a more feasible alternative compared to feedstocks used for first and second generation biodiesel (Saladini et al. 2016, Leong et al. 2018, Hossain et al. 2020), with microalgae having the potential to produce a yield of 15–300 times more than the yield from a traditional crop in relation to plantation area (Hossain et al. 2019, Hossain et al. 2019). Table 1 shows the advantages and challenges of first, second and third generation biodiesel sources.

Table 1. Advantages and challenges of first, second and third generation biodiesel sources. (Mofijur et al. 2013, Mofijur et al. 2013, Leong et al. 2018).

Biodiesel types	**Sources**	**Advantages**	**Challenges**
First generation	Edible oil feedstock	• Renewable source • Environment-friendly • Easy conversion into biofuel	• Competes with food crops (food-energy conflict)

			• Rising cost of food due to food competition • Land scarcity
Second generation	Non-edible oil feedstock	• Renewable source • Environment-friendly • Does not compete with food crops • Effective land utilization (non-arable lands)	• Land and water use competition • Requires sophisticated downstream processing technologies • High production cost • Uncertain long-term supply of oil yield
Third generation	Oleaginous microbes	• Renewable source • Environment-friendly • No conflict with food or land usage • Higher growth rate tendencies • High cell lipid accumulation	• Insufficient biomass production for commercialization • High initial production and setup costs for economic viability (Large scale).

Oils and fats can be used in different ways, including direct use, blending, micro-emulsions, pyrolysis and transesterification (Mofijur et al. 2012). Among these methods, transesterification is the most common method of converting oils and fats (Mofijur et al. 2016). As mentioned, one of the critical factors that affect the transesterification process is the type and concentration of catalysts (Mofijur et al. 2017). The use of catalysts in the transesterification process speeds up the reaction rate, thus increasing the biodiesel yield. Besides, the use of a catalyst in the production process contributes to a tangible response to the production rate. Different types of catalysts are used to produce biodiesel through the transesterification process from different sources. Nevertheless, these catalysts can be categorized into four major groups, i.e., homogenous catalysts, heterogeneous catalysts, biocatalysts, and nanocatalysts (Ruhul et al. 2015, Akubude et al. 2019). A number of recent advances in catalytic converting of oils and fats to biodiesel has been observed. Among them, the development of biodiesel using nanocatalysts offers some advantages over traditional acid/base catalysts. Nanocatalysts typically enhance reaction kinetics by enabling a reaction to occur at a lower temperature, reducing side reactions and increasing recycling levels and energy recovery (Ghanbari et al. 2017). The highly variable surface area and superficial energy of nanoscale catalysts contribute to high catalytic activity. Nanocatalysts offer promising

alternatives for efficient biodiesel production from oil and fat as their higher surface areas and catalytic activity mitigate the particular problem related to traditional catalysts (Hoseini et al. 2018). Whilst there is more and more literature on the effects of nanocatalysts on biodiesel transesterification in recent decades, fewer researchers have reviewed and analysed them. There are limited reviews in scientific databases on the impact of nanocatalysts on first, second and third generation biodiesel production processes. This paper therefore critically analyses in detail the influence of various nanocatalysts on first, second, and third generation biodiesel production processes, which is very important for ongoing research into the development and processing of cost-effective biodiesel production.

2. Biodiesel production technologies

As mentioned before, biodiesel can be produced using two different approaches, the physical approach and the chemical approach. Physical approaches include direct use, blending and microemulsion, which allows the oils to be used directly in their neat form. Chemical approaches include the pyrolysis and transesterification process, which result in a chemically modified form of natural oils (Shahabuddin et al. 2013, Uddin et al. 2018). Table 2 shows the benefits and drawbacks of various biodiesel production technologies. The thermal degradation of biodiversity with the help of a catalyst with no oxygen present is known as pyrolysis (Ong et al. 2019). Vegetable oil, animal fats, and natural fatty acids are examples of pyrolyzed materials. Many investigators have studied the pyrolysis of triglycerides to obtain suitable fuels for the diesel engine (Ashok et al. 2019). Transesterification is the popular chemical method for transforming natural oils and fats into biodiesel fuel using a process where three moles of alcohol such as methanol stoichiometrically react with one mole of triglyceride (Fattah et al. 2013, Fattah et al. 2014, Rashed et al. 2016). In general, the transesterification process takes place at 60–70 ºC with a catalyst resulting in a mono-alkyl ester (biodiesel) as the main product and glycerol as a co-product. The conversion of triglycerides into monoglyceride occurs in

three successive reversible reactions, as shown in Figure 3 (Mofijur et al. 2013). Firstly, methanol reacts with triglyceride, producing diglycerides. Then diglycerides react with methanol producing monoglyceride. Finally, monoglyceride reacts with methanol that results in glycerol (Atabani et al. 2014).

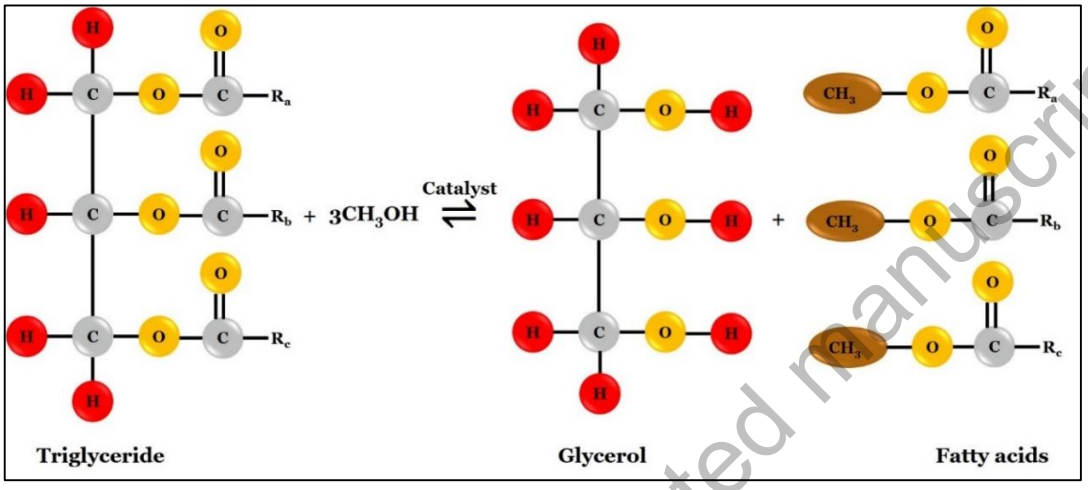

Figure 3. The chemical reaction of the biodiesel production process.

Different parameters influence the transesterification process, which relies on the reaction conditions (Mofijur et al. 2014, Anwar et al. 2018). If the conditions are not optimized, the process is either ineffective or the performance significantly reduced. Thus, every parameter is equally critical to accomplishing a high level of efficiency in producing biodiesel that complies with regulatory requirements. The most significant factors that influence the transesterification reaction are free fatty acids, water content, types of alcohol and molar ratio used, catalytic types and concentrations, reactivity temperature and duration, stirring rate and method, final product purification, mixing speed, organic co-solvent effect and specific gravity (Tan et al. 2019).

Table 2. Benefits and drawbacks of various biodiesel production technologies, reprinted with permission from (Tabatabaei et al. 2019).

Method	Advantage	Disadvantage
Direct use and blending	- Low capital and production costs - Simple production	- Solidification of blend at cold temperatures - Impractical and inappropriate for direct use in diesel engines - High viscosity - Gum formation

		- Lubricating oil thickening - Incomplete fuel combustion - Oil deterioration - High level of free fatty acid - Low volatility - Unsaturated hydrocarbon chains reactivity - Injector nozzles clogging - Poor atomization - Engine durability reduction, - Higher air pollution emission - Higher engine maintenance costs - Higher engine wear
Microemulsion	- Biodiesel formation with lower viscosity and higher liquidity - Lower nitrogen oxide emissions - No by-product or waste formation - Clear, single phase, and thermodynamically stable colloidal equilibrium dispersion of biodiesel fluid	- Heavy deposition of carbon residue - Inadequate combustion - Lubricating oil thickening - Random injector needle sticking
Pyrolysis	- Suitable for areas with well-established hydro-processing industry - Generation of value-added by-products like syngas - Biofuel with satisfactory physical and chemical properties	- High production cost - Complex equipment requirement - Biofuel has no oxygenated value - Producing short chain molecules with more similarities to gasoline than diesel fuel
Transesterification	- The most common method for production of biodiesel - Unreacted feedstock can be recycled - The by-product *(i.e.*, glycerol) can be converted into value-added products	- Dry alcohol and oil must be used to increase biodiesel yield by avoiding saponification - Glycerol must be efficiently separated to avoid generation of hazardous gases (*i.e.*, acetaldehyde, formaldehyde) - Expertise requirement - Complex equipment requirement

3. Nanocatalysts for the biodiesel production process

Catalytic technologies are essential in the production of different petrochemical products. A state-of-the-art catalyst relies on producing fewer investment products. In terms of energy, environment, and nanomedication, nanocatalysts play a crucial role as the use of nanoparticles has become evident in many chemical and electrochemical reactions as efficient catalysts. Recently, due to their specific benefits, nanocatalysts have received significant attention for the development of biodiesel.

3.1 Types and characteristics of nanocatalysts

Nanocatalysts are mesoporous, magnetic, carbon-based or metal oxide-based. Carbon-based nanocatalysts include graphite, carbon black, buckyball, fullerene, and inorganic nanotubes while metal oxides nanocatalysts include aluminium, iron, silver, titanium oxide, cobalt, iron oxide, cerium oxide, calcium oxide and zinc oxide (Thangaraj et al. 2019). Other nanocatalysts also exist, including clays and quantum dots. Among various types of nanocatalysts, metal oxide nanocatalysts are regarded as the most promising and have been therefore widely studied for the development of biodiesels from a range of feedstocks. The use of carbon-based materials as solid acid catalysts for biodiesel production using various feedstocks has increased. Nevertheless, this form of catalyst has disadvantages such as, primarily, deactivation, thermal instability, higher methanol-to-oils requirements, side reactions and higher reaction times. In transesterification of different biodiesel sources, mesoporous nanocatalysts are attracted by high surfaces, broad pore widths, and better accessibility, which promote the diffusion of reactants into the active sites of the catalyst's acid. In the early 1990s, after the discovery of porous materials, different approaches have been used to develop and design materials with enhanced structural properties, such as pores, strength and active sites, to enhance their efficiency in different reactions. The size of the pore may vary between 15 and 300 Å depending upon the method of development and the interaction between precursor and template particles (Melero et al. 2006). However, catalyst designs based on magnet nanoparticles have gained significant attention and become a key feature in the development of biodiesel from low-cost sources. This can be explained by the magnetic properties that allow the isolation of the nanocatalyst from the reacting medium, which can eliminate centrifugal and ultrafiltration techniques in industrial applications. A wide variety of magnetic nanocatalysts have been synthesized recently and used to produce biodiesel from low-cost feedstock. The characteristics of different types of nanocatalysts include a high surface area and catalytic activity, being

adsorbent, being prone to agglomeration, having a range of possible chemistries (natural and synthetic) and useful in a wide range of applications (Rao 2010).

3.2 Advantages and disadvantages of nanocatalysts in the transesterification process

There are some advantages of using nanocatalysts in the transesterification process, including high catalytic efficiency compared to other catalysts (Rao 2010, Fattah et al. 2020). The surface areas of nanocatalysts are small, resulting in increased activity over conventional catalysts. Further, they are highly stable, possess superior saponification resistance, and have an effective surface/volume ratio and high reusability (Rahmani Vahid et al. 2017). Nanocatalysts can be synthesized by a range of techniques, including high-temperature, microwave burning, traditional hydrothermal, hydrothermal, solvothermal, and solo-gel techniques, co-precipitation, impregnation, condensation, chemical vapour and electrochemical techniques, vacuum coating, vapour, etc. (Quirino et al. 2016, Ambat et al. 2018). The disadvantages of nanocatalysts are that their synthesis comes at a high cost and more alcohol is needed for the efficient transesterification process. Table 3 summarizes the comparative advantages and disadvantages of different catalysts.

Table 3. Comparative advantages and disadvantages of different types of catalysts.

Catalyst types	Advantages	Disadvantages	Reference
Homogeneous acid catalyst	• High biodiesel yield • Suitable for low-quality feedstock, hence insensitive to FFA content • Simultaneous occurrence of esterification and transesterification • Less energy consumption than homogeneous base catalysis	• Chances of damaging equipment due to acid corrosiveness • Higher yield of free glycerol • Higher temperature requirement but less than that of supercritical method • Separation of catalyst is difficult from product. • Takes longer time to complete than base catalysed reaction	(Mahanta and Shrivastava 2004, Marchetti et al. 2008, Guan et al. 2009, Lam et al. 2010, Farag et al. 2011, Gebremariam and Marchetti 2017)
Homogeneous base catalyst	• Reaches completion faster than acid catalysed reaction • Mild reaction condition and less energy intensive • Catalysts are cheap and widely available • Less corrosive than acid catalysts	• Production depends on FFA content in the oil • Low quality feedstock poses the issue of saponification of oil • Glycerol recovery is difficult • Wastewater generated in washing steps is alkaline and requires post-treatment	(Dias et al. 2008, Marchetti et al. 2008, Demirbas 2009, Lam et al. 2010, Leung et al. 2010, Parawira 2010, Gebremariam and Marchetti 2017)

Heterogeneous base catalysis	• Superior selectivity • Catalysts are easy to separate from the reaction mixture • Reduced wastes • Catalysts can be regenerated and reused • Mild reaction conditions and less energy intensive	• Poisoning of the catalyst occurs when exposed to ambient air • Production depends on FFA content in the oil • High FFA content in oil results in saponification which in turn reduces yield and complicates purification • Leaching of active sites in the catalyst may result in product contamination	(Furuta et al. 2006, Lam et al. 2010, Parawira 2010, Borges and Díaz 2012, Jagadale and Jugulkar 2012, Gebremariam and Marchetti 2017)
Heterogeneous acid catalysis	• Catalysts can be separated from reaction mixture easily • Reduces the process stages and waste • Insensitive to FFA content in oil • Preferred for transesterification of low- grade oil • Catalyst can be easily removed and recycled	• Complicated catalyst synthesis procedures lead to higher cost • Requires high reaction temperature, high molar ratio of alcohol to oil and long reaction time. • Relatively energy intensive	(Peng et al. 2008, Melero et al. 2009, Lam et al. 2010, Borges and Díaz 2012, Gebremariam and Marchetti 2017)
Lipase catalysts	• Suitable for low quality feedstock as the process is insensitive to FFA and water content in the oil • Generally carried out at low reaction temperature • Easy separation of glycerol and other by-products simplifying the purification step • Yields high purity product (esters) • Immobilized enzymes can be reused	• Enzymes are expensive • Yield is relatively low • Very long reaction time • Lipase deactivation caused by methanol and glycerol	(Mahanta and Shrivastava 2004, Devanesan et al. 2007, Marchetti et al. 2007, Ranganathan et al. 2008, Bajaj et al. 2010, Lam et al. 2010, Leung et al. 2010, Amini et al. 2017, Gebremariam and Marchetti 2017)
Nanocatalysts	• Relatively short reaction time • High specific surface area of catalyst requiring less amount of catalyst • Catalyst can be reused many times • Wide range of catalyst choice	• More alcohol is required than other processes for effective yield • Preparation of appropriate catalysts costs more in some cases	(Wen et al. 2010, Chaturvedi et al. 2012, Sivakumar et al. 2013, Rengasamy et al. 2014, Rengasamy et al. 2014, Sharma et al. 2015, Hashmi et al. 2016, Gebremariam and Marchetti 2017)

Ionic liquid catalysts	• Ease of separation of final products due to the formation of biphasic mixture • Process is time efficient • Tailor made catalysts to suit a particular need • Ease of separation of catalyst and can be reused many times • High catalytic activity with excellent stability	• Ionic liquid production is expensive • More alcohol is required than other processes for an effective yield	(Gamba et al. 2008, Dupont et al. 2009, Earle et al. 2009, Andreani and Rocha 2012, Guo et al. 2013, Ren et al. 2014, Gebremariam and Marchetti 2017)
Supercritical transesterification	• Faster completion • Insensitive to the water content of the feedstocks • No catalyst is used hence no washing is required • Easier to design as a continuous process	• Higher temperature and pressure required • High operating cost due to high pressures and temperatures • Very high methanol consumption	(Kusdiana and Saka 2001, Bunyakiat et al. 2006, Marchetti et al. 2008, Song et al. 2008, Shahid and Jamal 2011, Santana et al. 2012, Kiss et al. 2014, Micic et al. 2014, Gebremariam and Marchetti 2017)

3.3. Application of nanocatalysts in first, second and third generation biodiesel production processes

The utilization of nanocatalysts in the production process for different types of biodiesel has been investigated by many researchers around the world, e.g., for first generation, second generation and third generation biodiesel production. Figure 6 shows the mechanism of the transesterification reaction using nanocatalysts. Nanocatalysts can accomplish high production yield with very moderate reaction conditions and lower reaction periods. The reusability of these catalysts is also superb because they maintain good performance even after 11 cycles.

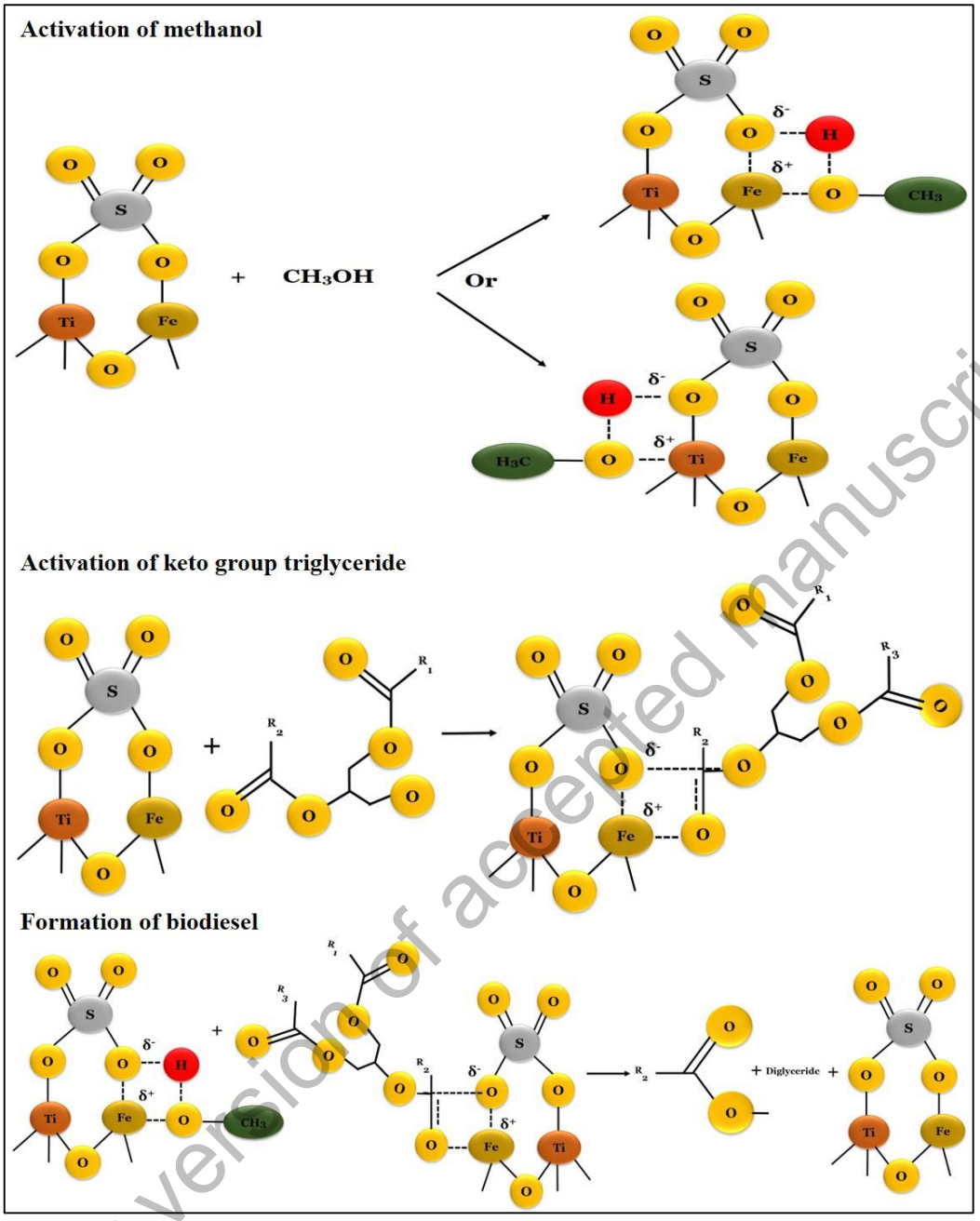

Figure 4. Mechanism of transesterification reaction using nanocatalysts (Carlucci et al. 2019).

As discussed previously, nanocatalysts have attained special attention because of their high specific surface area and high catalytic efficiency. These characteristics can solve the issues associated with heterogeneous catalysts such as resistance to mass transfer, longer reaction time, and fast deactivation (Ambat et al. 2018, Fattah et al. 2020). Nanocatalysts can be used either in the supported form with the help of solids such as zeolites, carbon, and oxides, or without any support (Akia et al. 2014). Numerous nanocatalysts have been used for the

transesterification of first, second, and third generation feedstocks. Depending on the nanocatalysts used, the reaction time and yield were found to vary for all three generations of biodiesel. For example, canola oil (a first generation biodiesel feedstock) was used in various studies to produce biodiesel using different nanocatalyst combinations, which showed a wide variation in biodiesel yield. Kazemifard et al. (2018) studied potassium hydroxide reinforced $Fe_3O_4@Al_2O_3$ core-shell nanocatalysts in the production of biodiesel from canola oil. They found that 25 wt.% of Fe_3O_4 to Al_2O_3 (K/FeAl-0.25) showed appropriate magnetic properties and catalytic activity to use as a suitable nanocatalyst for biodiesel production. They were able to convert 98.8% of canola oil to biodiesel under 12:1 methanol/oil, 4 wt.% of catalyst in 6 h. On the other hand, Alsharifi et al. (2017) produced nanocatalyst by implanting lithium onto TiO_2 to enhance the surface properties TiO_2 with 30 wt.% of Li which showed the highest activity for FAME formation. The obtained catalyst exhibited 98% FAME conversion under the optimum conditions of 24:1 M methanol to oil ratio, 5 wt.% catalyst dosage in just 3h. However, when the same nanocatalyst was used for transesterification of waste cooking oil (a second generation feedstock), a 91.73% FAME conversion was observed for the same reaction conditions. Another prominent first generation biodiesel feedstock, palm oil, was used by (Zhang et al. 2020) to produce biodiesel using nanocatalysts. They focused on optimizing the transesterification reaction parameters using response surface methodology when mesoporous $NaAlO_2/\gamma\text{-}Al_2O_3$ with a 30% mass ratio of $NaAlO_2$ to $\gamma\text{-}Al_2O_3$ nanocatalyst was used to catalyse the reaction. The maximum FAME yield of 97.65% was obtained at the optimum reaction conditions of 20.79:1 M methanol to oil, 10.89 wt.% catalyst, and a 64.72 °C reaction temperature. The efficacy study of SiO_2/ZrO_2 nanocatalyst in the transesterification process of soybean oil by (Faria et al. 2009) indicated that nanocatalyst offers an improved biodiesel conversion yield (96.2 ± 1.4%) after 3 h of reaction. They also identified that after recovery, the catalyst can be reused for at least six more cycles with a small penalty (12% less) in its

catalytic efficiency. Qiu et al. (2011) studied the performance of $C_4H_4O_6HK$ nanocatalyst in the transesterification of soybean oil and revealed that the use of 6% nanocatalyst at a molar ratio of 16:1 and temperature of 60°C offers a maximum biodiesel yield of 98.03%. Saeedi et al. (2016) reported that the use of KNa/ZIF-8 nanocatalyst in the transesterification of soybean oil also enhances the catalytic performance. The maximum conversion efficiency (>98%) was found at the molar ratio of 10:1 after 3.5 h of reaction time. Nevertheless, they also reported that this nanocatalyst could be recycled and reused for at least three additional cycles.

The use of second generation biodiesel feedstock, such as waste cooking oil, for biodiesel production using nanocatalysts has been extensively studied (Milano et al. 2018). For example, Borah et al. (2018) studied biodiesel production from waste cooking oil using in-situ TiO_2/RGO nanocomposite as a nanocatalyst. Results show that 98% biodiesel conversion was achieved with an optimized oil to methanol molar ratio of 1:12 at 65 °C with 1.5 wt.% catalyst loading and reaction time of 3 h. On the other hand, Kaur et al. (2018) studied the production of biodiesel from waste cooking oil using 20 wt.% tungsten (W) supported TiO_2/SiO_2 nanocatalyst. The complete transesterification of waste cooking oil was observed with an optimum reaction condition of 1:30 M oil to methanol ratio, at 65 °C in 4 h. Manivannan and Karthikeyan (2013) studied the efficacy of Mg-Al nanohydrotalcite in the transesterification of neem oil and reported that the reaction temperature played an important role in the improvement of biodiesel yield. They found that the Mg-Al nanohydrotalcite offers the highest yield of 84% at 65 °C and a further increase in temperature declined the yield of FAME. Wang et al. (2015) reported that Fe/Fe_3O_4 nanocatalyst is an efficient recoverable catalyst and it offers superb performance during the biodiesel production from waste cooking oil. The catalysts were tested in the transesterification of glyceryl trioleate and in the esterification of oleic acid in methanol. While the sulfonic acid functionalized MNPs showed low reusability, with a conversion drop to 62% at the fifth run, sulfamic acid functionalized MNPs maintained 95%

conversion throughout five reaction cycles. Amalia et al. (2019) observed a promising transesterification using 70% KOH/zeolite catalyst for the transesterification of castor oil at 55 °C and 7 h of reaction time. Venkat Reddy et al. (2006) found that the use of a CaO-based nanocatalyst in the transesterification of poultry fat at room temperature and 10:3 molar ratio offers100% biodiesel yield.

Minimal research has been carried out on the production of third generation biodiesel utilizing nanocatalysts. For example, Teo et al. (2016) studied biodiesel production from algae (*Nannochloropsis* sp.) using nano $Ca(OCH_3)_2$ (calcium methoxide) as a catalyst. They obtained a maximum FAME yield of 99% for 30:1 M methanol to oil, over 3 wt.% of catalyst loading at 80 °C in 3 h.

Based on different studies of nanocatalyst applications for biodiesel production, it is evident that the utilization of different generation feedstock oils for transesterification reactions using nanocatalysts shows the important influence of nanocatalysts on activity and selectivity. The presented results reveal that the high specific surface area of nanostructure materials in comparison with bulk catalysts is favourable for the contact between catalyst and substrates, which effectively improves product yield. A summary of the research findings on the application of nanocatalysts in first, second, and third generation biodiesel production is presented in Table 4.

Table 4. Summary of the research findings on the application of nanocatalysts in first, second and third generation biodiesel production.

Feedstock (Type)	Catalyst type	Concentration (wt. %)	Oil to alcohol ratio	Temperature (°C)	Time (min)	Yield (%)	Reference
Canola oil (First generation)	$KOH/Ca_{12}Al_{14}O_{33}$	3.5	1:12	65	240	96.70	(Nayebzadeh et al. 2016)
	$KOH/Fe_3O_4@Al_2O_3$	4	1:12	65	360	88.40	(Kazemifard et al. 2018)
	Li/TiO_2	5	1:24	65	180	98	(Alsharifi et al. 2017)
	Calcined dolomite	5.3	1:7.6	60	150	96.60	(Korkut and Bayramoglu 2018)
Castor oil (Second generation)	Si-MMT- pH-SO_3H	5	1:12	60	300	89.80	(Negm et al. 2017)
	Ni doped ZnO nanocatalysts	11	1:8	55	60	95.20	(Baskar et al. 2018)
	TiO_2/RGO	1.5	1:12	65	180	98	(Baskar et al. 2018)
Corn oil (First generation)	$Ca/\gamma\text{-}Al_2O_3$	6	1:12	65	300	34.64	(Moradi et al. 2015)
Cottonseed oil (Second generation)	Ti/SiO_2	5	1:30	65	204	>98	(Kaur et al. 2018)
	CeO_2/Li/SBA-15	10	1:40	65	240	>98	(Malhotra and Ali 2018)
Date seed oil (Second generation)	Eggshell derived catalyst	5	1:12	65	90	93.50	(Farooq et al. 2018)
Euphorbia lathyris oil (Second generation)	Acid-based HPA catalyst $(C_6H_{15}O_2N_2)_2$ $HPW_{12}O_{40}$ (ly_2HPW)	9	374.4 mmol	65	720	91.20	(Zhang et al. 2018)
Jatropha oil (Second generation)	CaO	0.02:1	1:5.15	65	133.1	95.80	(ANR et al. 2016)
Karanja oils (Second- generation)	Li–CaO	5	1:12	65	120	>99	(Kaur and Ali 2011)
Madhuca indica oil (Second generation)	Heteropoly acid (HPW)–coated ZnO	2 g	30 ml	55	300	98	(Thangaraj and Piraman 2016)
Mahua oil (Second generation)	Mn-doped ZnO	8%	1.7% (v/v)	50	50	97	(Baskar et al. 2017)
Microalgae (Third generation)	$Ca(OCH_3)_2$	3	1:30	80	180	99	(Teo et al. 2016)
	CaO	0.5-2	1:9	55	-	96.3	(Siva and Marimuthu 2015)
	CaO	1.5	1:9	60	-	96.3	(Manikandan and Rajasekaran 2013)
Olive oil (First generation)	Zinc dodecatungstophosphate ($Zn_{1.2}H_{0.6}PW_{12}O_{40}$; ZnPW)	2.3	1:28	65	720	97.2	(Woodford et al. 2014)
Oleic acid (Second generation)	$25\%MoO_3$/B-ZSM-5	3	1:20	160	360	93	(Mohebbi et al. 2020)
Soybean oil (First generation)	Cs-Na_2ZrO_3 Basic heterogeneous	1	1:30	65	15	98.8	(Torres-Rodríguez et al. 2016)

	Fe_3O_4@SiO_2@CPTMS@amine	6	1:36	160	180	96	(Farzaneh et al. 2018)
	Calcined marble slurry and hydroxyapatite	6	1:9	65	180	94	(Gupta et al. 2018)
	CaO-K_2O	15	1:4.6	70	240	99	(Fernandes et al. 2016)
	Calcinated form of waste tucuma peels	1	1:15	80	240	97.30	(Mendonça et al. 2019)
	Magnetic $LiFe_5O_8$-$LiFeO_2$	8	1:36	65	120	96.50	(Dai et al. 2018)
	CaO (Nanocrystalline-1)	1 mmol;	10 ml	25	360	100	(Venkat Reddy et al. 2006)
	CaO/CaN, CaO/ss	8	1:12	65	360	93 (CaO/CaN) 96 (CaO/ss)	(Gupta and Agarwal 2016)
	ZrO_2–$C_4H_4O_6HK$(Zirconia-loaded potassium bitartrate)	6	16:1	60	120	98.03	(Qiu et al. 2011)
	Mixed iron/tin oxide (ISnO)	1 g	6 g	200	180	90	(Alves et al. 2014)
Sunflower oil (First generation)	MgO/$MgAl_2O_4$	3	1:12	110	180	91.10	(Rahmani Vahid et al. 2017)
	CaO-based/Au nanoparticles	3	1:9	65	180	94–97	(Bet-Moushoul et al. 2016)
	CaO/Fe_3O_4@SiO_2	6	1:15	65	300	97	(Feyzi and Norouzi 2016)
	Aluminum dodecatungstophosphate ($Al_{0.9}H_{0.3}PW_{12}O_{40}$) (AIPW)	3	1:34	65	840	96	(Vahid and Haghighi 2017)
	Cs–MgO	2.8	1:30	90	1440	93	(Alaei et al. 2018)
	Ca(30%)/Al-MCM-41	10	1:12	70	480	84.20	(Vardast et al. 2019)
	MgO/$MgFe_2O_4$	4	1:12	110	240	82.40	(Alaei et al. 2018)
	Cs/Al/Fe_3O_4	6	1:14	58	120	88	(Feyzi et al. 2013)
Palm oil (First generation)	γ-Al_2O_3/KI	4	1:14	60	240	79	(Islam et al. 2015)
	CaO functionalized with strontium	5	1:9	65	30	98.31	(Li et al. 2016)
	CaO	9	1:12	60	120	90	(Uprety et al. 2016)
	30Ca/APB-700	5	1:8	65	150	93.40	(Wang et al. 2019)
	Strontium and Nickel	2	1:9	65	300	97	(Abreu et al. 2017)
	$NaAlO_2$/γ-Al_2O_3	10.89	1:20.79	64.72	180	93.29	(Zhang et al. 2020)
Rapeseed oil (First generation)	K_2O/γ–Al_2O_3)	3	12:1	70	180	94	(Han and Guan 2009)
Rubber seed oil (Second generation)	Sodium metasilicate	9	1:9	65	40	97	(Roschat et al. 2017)
Waste kernel oil (Second generation)	Mn@MgO-ZrO_2	3	1:15	90	240	96.40	(Jamil et al. 2018)
Waste cooking oil (Second generation)	Cr/Ca/γ-Al_2O_3	6	1:18	65	180	78.29	(Sulaiman et al. 2017)
	SO_4/Fe-Al-TiO_2 solid acid	3	1:10	90	150	96	(Gardy et al. 2018)
	Zinc-doped CaO	5	1:12	65	132	>98	(Kataria et al. 2017)
	$FeCl_3$ -modified resin	8	1:10	90	120	92	(Guldhe et al. 2017)
	Tungsten supported TiO_2/SiO_2	5	1:30	65	240	>98	(Kaur et al. 2018)

	CaO–MgO	1.2 g (CaO, 0.7 g; MgO, 0.5 g)	1:7	75	360	98.95	(Tahvildari et al. 2015)
	KF/CaO	4	1:12	65	150	96.80	(Zheng et al. 2006)
	MgO	300 mg	1:4	70	40	99	(Li et al. 2009)
	Iron (II)-doped ZnO	14	1:12	55	50	91	(Wang et al. 2009)
	Sulfonated graphene	10	1:20	100	840	98	(Borah et al. 2018)

4. Challenges in using nanocatalysts in biodiesel production

There are some issues with catalytic biodiesel production, such as excessive processing time, delay in reaction time, and the need to isolate the catalyst and saponified contaminants from the biodiesel (Mahlia et al. 2020). These issues are not present in the non-catalytic process of transesterification. For example, the supercritical process uses less energy and is completed in a very short period (2-4 minutes). Moreover, since no catalyst is required, biodiesel filtration and glycerol recovery are much simpler, hassle-free and are less harmful to the environment. However, reactor and maintenance costs are high and methanol consumption is high (Atabani et al. 2012).

The utilization of nanocatalysts in the catalytic transesterification process offers some advantages over other catalysts (Wen et al. 2010, Chaturvedi et al. 2012, Sivakumar et al. 2013, Rengasamy et al. 2014, Rengasamy et al. 2014, Sharma et al. 2015, Hashmi et al. 2016). Despite many advantages, nanocatalysts have some issues in responding to the transesterification process (Ajala et al. 2020). Nanoparticles sintering is the principal downside of nanocatalysts. Metal atoms are unstable at high temperatures in the reactive atmosphere in various catalytic processes, which leads to major changes in the metal nanoparticles' size and shape. Such structural changes lead to unwanted effects such as non-uniformity, selectivity loss or reversal, and catalytic discontinuation (Zuliani et al. 2018). Thus, sintering in nanocatalysts may also restrict their use to low temperature and short-term applications, unless preventive measures are taken. The best approach to avoid agglomeration in nanoparticles is by using ligand or coating materials such as carbon and inorganic components. Beside the above challenge, some metal-based nanocatalysts display some challenges during the recovery stage. In fact, lattice oxygen species form hydrogen bonds to methanol and glycerine in the transesterification reaction, increase the viscosity of glycerine, and form solids in a suspended form with some nanocatalyst types, which is then difficult to recover.

5. Conclusions

While biodiesel offers a competitive alternate to diesel fuel in different areas, efficient output is often jeopardized due to high feedstock costs and the absence of sustainable technology. Various investigators have suggested different biodiesel production techniques, which are typically based on feedstock properties. Transesterification, which relies mostly on the catalytic mechanism, is among the methods used to burn fat and oil into biodiesel. Many technologies are used for the transesterification of biodiesel, each of which requires a different raw material property and ideal operating conditions for efficient processing. From this study, it can be summarised that nanocatalysts can be used in lower temperature approaches and their utilization in transesterification reactions speeds up the reaction process. Further, nanocatalysts are not affected by the free fatty acids and water content of feedstocks. Nanocatalysts are also reusable, which offers cost advantages compared to other catalysts. Nevertheless, more alcohol is required for a successful yield and it can be costly to prepare suitable catalysts. Developing efficient and economic catalysts in an environmentally sustainable approach is critical to solving current challenges. Therefore, a catalyst with these characteristics developed for successful transesterification would represent a landmark in the fuel industry. Furthermore, to address the existing challenges of the energy-efficient production of biodiesel, efforts should concentrate on gaining a thorough understanding of surface catalytic reaction mechanisms, which is crucial for developing rational ideas for advanced catalysts with predetermined improved catalytic efficiency for target reactions.

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