A Short Review on Catalyst, Feedstock, Modernised Process, Current State and Challenges on Biodiesel Production

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Abstract: Biodiesel, comprising mono alkyl fatty acid esters or methyl ethyl esters, is an encouraging option to fossil fuels or diesel produced from petroleum; it has comparable characteristics and its use has the potential to diminish carbon dioxide production and greenhouse gas emissions. Manufactured from recyclable and sustainable feedstocks, e.g., oils originating from vegetation, biodiesel has biodegradable properties and has no toxic impact on ecosystems. The evolution of biodiesel has been precipitated by the continuing environmental damage created by the deployment of fossil fuels. Biodiesel is predominantly synthesised via transesterification and esterification procedures. These involve a number of key constituents, i.e., the feedstock and catalytic agent, the proportion of alcohol to oil, the circumstances of the reaction and the product segregation and purification processes. Elements that influence the yield and standard of the obtained biodiesel encompass the form and quantity of the feedstock and reaction catalyst, the proportion of alcohol to feedstock, the temperature of the reaction, and its duration. Contemporary research has evaluated the output of biodiesel reactors in terms of energy production and timely biodiesel manufacture. In order to synthesise biodiesel for industrial use efficaciously, it is essential to acknowledge the technological advances that have significant potential in this sector. The current paper therefore offers a review of contemporary progress, feedstock categorisation, and catalytic agents for the manufacture of biodiesel and production reactors, together with modernised processing techniques. The production reactor, form of catalyst, methods of synthesis, and feedstock standards are additionally subjects of discourse so as to detail a comprehensive setting pertaining to the chemical process. Numerous studies are ongoing in order to develop increasingly efficacious techniques for biodiesel manufacture; these acknowledge the use of solid catalytic agents and non-catalytic supercritical events. This review appraises the contemporary situation with respect to biodiesel production in a range of contexts. The spectrum of techniques for the efficacious manufacture of biodiesel encompasses production catalysed by homogeneous or heterogeneous enzymes or promoted by microwave or ultrasonic technologies. A description of the difficulties to be surmounted going forward in the sector is presented.
Keywords: biodiesel; feedstock; heterogeneous catalyst; esterification/transesterification; FAME

1. Introduction

The current population growth and development in the world are some of the reasons that could contribute to the possible depletion of fossil fuel and increased in global energy demand for human activities such as production of food, goods, and services, [1,2]. Environmental problems such as global warming that has direct effect on our planet and the ecosystem at large are largely associated with fossil based fuel, hence the search for alternative energy source, that can comfortably replace the conventional fossil fuel [3]. The current global economy is largely dependent on the transportation of goods and services through road, air, and sea routes, thereby exhausting more than 60% of the produced fossil fuel worldwide [4,5]. Biofuels are generally found to be the best replacement for conventional fossil fuel as renewable sources of energy, as a result of their biodegradability, nontoxicity, availability of feedstock, and low emission of poisonous gases [6,7]. Biofuels are purely renewable and therefore considered to be the safest fuel to be used on our planet for the production of goods and services. Biofuels are composed of ethanol, which is predominantly being produced and utilized in Brazil [8,9], biodiesel that is being produced from many edible and non-edible oils in Europe, Asia, and other parts of the world [10]. There is also green diesel produced through cracking, pyrolysis, hydrodeoxygenation, and deoxygenation process of oxygenates compounds from vegetable oils [11]. All the aforementioned biofuels are much safer and healthier to be used as fuels when compared to petroleum-based fuel [12].

Upon all the aforementioned biofuels, biodiesel is considered to be one of the most viable renewable fuel alternative to petroleum-derived fuel [13]. This is due to the availability of its wide range of feedstock such as soybean oil [14], sunflower oil [15], cotton seed oil [16], and rapeseed oil [17]. The feedstock also includes non-edible oils such as Jatropha Curcas oil [18], palm fatty acid distillate (PFAD) [19] and waste cooking oil [20]. Some of these feedstocks, despite being renewable, are also cheap [21] and available [22]. Besides, biodiesel physical and chemical properties such as flash point, cetane number, cloud point, and pour point are similar to that of conventional fuel [23].

Biodiesel is also referred to as fatty acid methyl esters (FAME), and is usually produced by esterification or transesterification process of animal fats or vegetable oils, respectively with methanol or ethanol as a solvent in the presence of a catalyst [24,25]. During a transesterification reaction, a vegetable oil such as palm oil is transformed using base catalyst in the presence of methanol and temperature over the period of time, producing FAME and glycerol as the byproduct [26]. The catalyst could be homogeneous, such as NaOH or KOH, or heterogeneous, such as CaO, MgO, or dolomite [27]. While in the esterification reaction process, a high free fatty acid (FFA) feedstock such as PFAD, animal’s fat, or waste cooking oil is used over either homogeneous acid catalyst such as HCl, H2SO4, or HNO3 [28] or heterogeneous acid catalyst such as Zeolites, Heteropolyacids, Sulphonic acid group, and mixed metal oxides [29] to produce FAME and water as byproduct [30]. Besides the environmental and availability of feedstock advantages, biodiesel also offers the potential advantages various socioeconomic aspect such as employment for the unemployed youth in the rural area and additional source of income for the farmers [31]. This review will focus on the detailed process of biodiesel production using catalytic and non-catalytic routes. The chemical composition of various feedstock and the produced biodiesel will also be discussed. The current and future prospects and challenges of biodiesel production are also discussed.

2. Feedstock for Biodiesel Production

Generally, biodiesel can be produced from renewable sources of feedstock such as vegetable oils and animal fats [25]. These feedstock consist of more than 95% edible oils
from different plants [32]. Different countries utilized different vegetable based oils as feedstock; this includes sunflower oil in United States, soybean oil in Europe, and palm oil in Malaysia and Indonesia [33,34]. Other vegetable oils used for biodiesel production include soybean oil [35], peanut oil, and rapeseed oil [36]. However, utilizing edible vegetable oils for the production of biodiesel is posing the biggest threat to the food industry around the world [37]. To overcome the increased demand of edible oil in food industries with high cost of feedstock [38], the selection and studies of the potentials different of non-edible oil feedstock for low costly biodiesel production is highly required. Non-edible feedstock that are currently utilized for the effective low-cost biodiesel production are Jatropha curcas oil, waste cooking oil, grease oil, and palm fatty acid distillate (PFAD) [39]. Many palm oil factories produced waste from crude palm oil refining in Malaysia, which reaches up to 17.4 million metric tons generated from the palm oil industries [40]. The utilization of the PFAD feedstock and waste cooking oil for biodiesel production led to reduced production cost and improved waste management in the most of palm factories [41]. Other non-edible oils commonly used for biodiesel production such as jatropha oil [10,42–44], Karanja (Pongamia pinnata) [45], microalgae [46], microbial oils [47], Pine oil [48], neem oil [49], and rubber seed oil [50] were reported. Moreover, animal fat (chicken fat, cow fat, etc.) were reported to be used as an alternative feedstock for effective, low-cost biodiesel production [51,52].

2.1. Edible Oil as Biodiesel Feedstock

Vegetable oils have always been used for both edible purposes and for a wide range of industrial applications, such as biodiesel, illumination oil, soaps, cosmetics, pharmaceuticals, emulsifiers, lubricants and greases, drying and semi-drying oils in paint, etc. [53,54]. Since vegetable oils are renewable, biodegradable, and environmentally friendly, they have become important feedstock for producing biodiesel [55–57]. Due to increasing demand of edible oils for both food and fuel purposes, non-edible oils became more attractive feedstock for biodiesel production. The physiochemical properties of some edible oils used for biodiesel production is presented in Table 1. Vegetable oils in general have high viscosity, which is a major cause of poor fuel atomization and operational problems such as engine deposits. However, the engine deposits can be reduced by blending the biodiesel derived from edible feedstock with Petro diesel through micro-emulsification.

Table 1. Physiochemical Properties and Yield of Biodiesel from Different Feedstocks.

| Type of Oil        | Kinematic Viscosity at 40 °C (cSt) | Density (g/cm³) | Saponification Number | Iodine Value | Acid Value (mg KOH/g) | Cetane Number | Heating Value (MJ/kg) | Yield % | References |
|--------------------|-----------------------------------|-----------------|------------------------|--------------|-----------------------|---------------|-----------------------|---------|------------|
| Soybean            | 4.08                              | 0.885           | 201                    | 138.7        | 0.15                  | 52            | 40                    | >95     | [58–62]    |
| Rapeseed           | 4.3–5.83                          | 0.88            | Nd                     | Nd           | 0.25–0.45             | 49–50         | 45                    | 95–96   | [63–65]    |
| Sunflower          | 4.9                               | 0.88            | 200                    | 142.7        | 0.24                  | 49            | 45.3                  | 97.1    | [61,64,66] |
| Palm               | 4.42                              | 0.86–0.9        | 207                    | 60.07        | 0.08                  | 62            | 34                    | 89.23   | [67–69]    |
| Peanut             | 4.42                              | 0.883           | 200                    | 67.45        | Nd                    | 62            | 40.1                  | 89      | [70–72]    |
| Corn               | 3.39                              | 0.88–0.89       | 202                    | 120.3        | Nd                    | 58–59         | 45                    | 85–96   | [66,73,74] |
| Cotton             | 4.07                              | 0.875           | 204                    | 104.7        | 0.16                  | 54            | 45                    | 96.9    | [75–77]    |
| Jatropha curcas    | 4.78                              | 0.8636          | 202                    | 108.4        | 0.496                 | 61–63         | 40–42                 | 98      | [78–82]    |
| Pongamia pinnata   | 4.8                               | 0.883           | Nd                     | Nd           | 0.62                  | 60–61         | 42                    | 97–98   | [53,83–85] |

2.2. Non-Edible Oils as Feedstock for Biodiesel Production

Globally, there is a lot of interest in non-edible oils as these are ubiquitously accessible. Furthermore, utilizing these products can eradicate any conflict between their use for sustenance or energy. This feedstock category has also been deemed more efficacious and less harmful to the environment. The by-products obtained are also valuable; utilisation of non-edible oils is additionally more cost-effective [86]. Considered to be the second-generation of feedstock for biodiesel [29], non-edible oils include oils such as castor oil (Ricinus communis L.), Pongamia pinnata (Karanja), Neem (Azadirachta indica), yellow oleander (Thevetia peruviana Schum.), Cerbera odollam (sea mango), Jatropha curcas L., Hevea brasiliensis (rubber), Moringa oleifera, Sapindol sebiferum, Milo (Thespesia populnea L.), Calophyllum
inophyllum, Croton megalocarpus, Madhuca indica (*Mahua*), Madhuca longifolia, silk cotton tree (*Ceiba pentandra*), and Eruca Sativa Gars. A relatively large proportion of free fatty acids (FFA) is found in the majority of these non-edible oils. A notable quantity of soap can be harvested when high FFA feedstock undergoes transesterification catalysed by an alkali-based agent. Soaps act as emulsifiers, which make glycerol and ester phase segregation more challenging. Moreover, any catalyst that has been transformed into soap loses its ability to speed up the process of biodiesel synthesis; thus, significant levels of catalyst loading are necessary [87]. This issue can be resolved by the use of esterification using an acid-based catalyst, although this has a slow reaction speed, mandating long reaction times.

Studies have generally indicated that the optimum strategy for biodiesel synthesis from non-edible oils with elevated FFA content is a two-step process, i.e., acid esterification with subsequent acid-based transesterification [88].

Despite the fact that globally, over 350 plant species that produce oil are possible resources for use in biodiesel manufacture, the overwhelming majority of biodiesel to date has been derived from edible oils, e.g., rapeseed, sunflower, palm oil, soybean, and coconut oils [88]. Indeed, the use of non-edible feedstock still hard to commercialize due to its low availability.

2.3. Composition of Biodiesel Feedstock

Various vegetable-based oils are composed of different chemical compositions, which helps in achieving the targeted yield at the end of the reaction process. Vegetable oils are formed of triglyceride molecules, which contain three types of fatty acids: saturated (Cn:0), monounsaturated with one double bond (Cn:1), and polyunsaturated with two or three double bonds (Cn:2,3) [89]. It should be noted that the majority of biodiesel’s important fuel qualities are highly influenced by the fatty acid level of the feedstock [64]. Hence, fatty acid compositions of feedstock particularly will initially be investigated prior utilizing it for the effective production of biodiesel. The fatty acid profiles of the various vegetable oils used for biodiesel production are summarized in Table 2.
Table 2. Chemical composition of biodiesel feedstock.

| Fatty Acid | Lauric | Behenic | Palmitic | Stearic | Palmioleic | Linolenic | Oleic | Gadoleic | Linoleic | Arachidic | Myristic | Erucic |
|------------|--------|---------|----------|---------|------------|-----------|-------|----------|----------|-----------|----------|-------|
| C12H24O2  | C16H32O2 | C18H36O2 | C16H30O2 | C18H30O2 | C18H34O2 | C20H38O2 | C18H32O2 | C20H40O2 | C14H28O2 | C22H42O2 |
| C12:0      | C16:0   | C18:0   | C16:1    | C18:3   | C18:1     | C20:1    | C18:2   | C20:0    | C14:0    | C22:1     |

| Source      | Nd     | Nd     | Nd       | Nd      | Nd         | Nd        | Nd     | Nd       | Nd       | Nd         | Nd       |
|-------------|--------|--------|----------|---------|------------|-----------|--------|----------|----------|------------|----------|
| Algae       | 0.33   | 15.64  | 2.10     | 0.32    | 4.88       | 54.89     | Nd     | 19.56    | 2.24     | Nd         | Nd       |
| Soybean     | 0.57   | 11.43  | 4.03     | 0.07    | 3.34       | 24.85     | Nd     | 55.33    | 0.25     | 0.07       | Nd       |
| Sunflower   | 0.46   | 5.93   | 3.44     | 0.14    | 0.38       | 36.22     | Nd     | 52.95    | 0.23     | 0.08       | Nd       |
| Corn        | 0.13   | 12.23  | 2.62     | 0.13    | 0.85       | 31.40     | Nd     | 51.21    | 0.32     | 0.02       | Nd       |
| Cottonseed  | 0.14   | 21.47  | 2.61     | 0.56    | 0.15       | 18.21     | Nd     | 55.45    | 0.06     | 0.69       | Nd       |
| Canola      | 0.07   | 6.23   | 2.49     | 0.34    | 5.11       | 61.46     | Nd     | 22.12    | 1.43     | 0.05       | Nd       |
| Olive       | 0.24   | 13.27  | 3.69     | 0.86    | 0.76       | 68.00     | Nd     | 12.48    | 0.48     | Nd         | Nd       |
| Safflower   | Nd     | Nd     | 6.70     | 2.40    | 0.08       | 0.15      | 11.50  | Nd       | 79.00    | Nd         | 0.10     |
| Hazelnut    | Nd     | Nd     | 5.82     | 2.74    | 0.29       | 0.46      | 79.30  | Nd       | 10.39    | 0.16       | 0.13     |
| Rapeseed    | Nd     | Nd     | 3.49     | 0.85    | Nd         | 8.23      | 64.40  | Nd       | 22.30    | Nd         | Nd       |
| Palm oil    | 0.1    | 0.1    | 36.7     | 6.6     | 0.1        | 0.3       | 46.1   | 0.2      | 8.6      | 0.4        | 0.7      |
| Jatropha    | Nd     | Nd     | 14.2     | 7.0     | 0.7        | 0.2       | 44.7   | Nd       | 32.8     | 0.2        | 0.1      |
| Palm kernel | 47.8   | 8.4    | 2.4      | Nd      | Nd         | 15.4      | Nd     | 2.4      | 0.1      | 16.3       | Nd       |
| Animal fats | Nd     | 0.01   | 28.4     | 15.7    | Nd         | 0.6       | 42.2   | 0.86     | 9.4      | 0.16       | 2.52     |
| WCO         | Nd     | 0.03   | 20.4     | 4.8     | 4.6        | 0.8       | 52.9   | Nd       | 13.5     | 0.12       | 0.9      |

Nd: Not provided.
3. Biodiesel Production Processes

The production of biodiesel involves the utilization of vegetable-based oil or animal fats on homogeneous or heterogeneous catalyst system [117]. The conversion of triglyceride in vegetable oils to biodiesel and glycerol using a homogeneous or heterogeneous base catalyst system is referred as transesterification reaction process [118]. Moreover, utilizing high FFA feedstock such as waste cooking oil, PFAD, and animal fat over a homogeneous or heterogeneous acid catalyst to produce FAME and water as by-product is termed esterification reaction process. Each of the two production processes could involve high or low temperature depending on the catalyst system adopted. A homogeneous base catalyst system reaction usually proceeds under relatively lower reaction temperature when compared to a heterogeneous catalyst system. Given the availability of feedstock, the transesterification process is one of the most cost-effective and appealing methods for producing significant amounts of biodiesel, making it the most versatile commercial biodiesel production technology [119–121].

3.1. Transesterification and Esterification of Vegetable Oil Using Homogeneous Acid and Base Catalysts

As discussed in the above section, esterification and transesterification of triglycerides or fatty acids to FAME can be performed with the help of a base or acid catalyst in both homogeneous and heterogeneous phases. Homogeneous catalysis generally employs acid and alkali catalysts in liquid phases. Acid catalysts favor FFA esterification, while base catalysts favor triglycerides transesterification reaction. The esterification reaction is represented by the general equation shown in Figure 1a, and transesterification is shown in Figure 1b. Both of reactions consists in three equivalent, consecutive, and reversible reactions. In esterification, the FFA reacts with an alcohol (typically methanol or ethanol) in the presence of an acid catalyst, forming FAMEs and an alcohol. In the case of transesterification, the triglyceride is converted stepwise to diglyceride, monoglyceride, and finally, glycerol. At each reaction step, one molecule of FAME is produced. The detailed mechanism of both of reactions is displayed in Figure 2a,b. In acid catalysis, both of the Brønsted-type acid sites and Lewis-type acid sites could catalyse the FFA esterification. The mechanism of Brønsted-type acid sites catalysed esterification as shown in Figure 2a, in which protonation of the acid group gives an oxonium ion that is readily attacked by an alcohol through an exchange reaction accordingly generates the corresponding ester after losing a proton [29]. In base catalysis, the protonation of the carbonyl group of the triglycerides occurs followed by an attack of the protonated carbon by the alcohol to form a tetrahedral intermediate (Figure 2b). The most important aspect of a homogeneous-base catalysed reaction is the production of a nucleophilic alkoxide from the alcohol, which is used to attack the electrophilic portion of the triglyceride’s carbonyl group [122]. There are three stages to breaking down a triglyceride. In the first stage, an unstable intermediate tetrahedral (triglyceride) is produced. In the second stage, this tetrahedral is further broken down into a diglyceride ion and fatty acid ester. In the third and final stage the catalyst is recovered through proton transfer. The process is then repeated twice more resulting in three fatty acid esters and a glycerol [123].

The main process used in biodiesel production employs homogeneous catalysts. However, although this is a proven technique, the use of heterogeneous catalysts in the process has been the subject of recent research as it has the potential to increase the productivity and sustainability through the utilization of non-edible oils as feedstock.
**Figure 1.** Overview of the chemical reaction (a) acid-catalyzed esterification and (b) base-catalyzed transesterification reaction pathways [124].

**Figure 2.** Chemical reaction (a) acid-catalyzed transesterification reaction pathways and (b) base-catalyzed transesterification [125,126].
3.1.1. Homogeneous Base Catalysts for Biodiesel Production

Biodiesel is typically manufactured using a batch process, which employs a homogeneous catalyst system. The most common catalysts employed in the production of biodiesel at the industrial scale are soluble alkaline catalysts, typically NaOH and KOH as they have demonstrated high catalytic activities [127], resulting in a less energy-intensive and faster process. For example, a reaction time of 1 to 2 h can achieve a high yield of more than 90% with sufficient qualitative feedstock and appropriate reaction conditions [128–130]. There is additional benefit of less methanol-to-oil ratio utilization during the reaction, which will make the entire reaction process low cost. It has been shown that with a methanol:oil ratio of 5–15:1 and temperature of 60–75 °C, a high yield of FAME can be achieved in a short period when homogeneous base catalysts are used [131,132].

Alkaline metal alkoxides are potential homogeneous base catalysts for the transesterification of triglycerides; these include CH$_3$ONa and CH$_3$CH$_2$ONa [133]. Although they are reported to have high activities, and thereby the potential to have a high biodiesel yield of 98% in only 30 min even at low molar concentrations (as low as 0.5 mol%), they require the absence of water in the feedstock, which makes them difficult to be used at the industrial scale, as most commercial-grade feedstock contain some level of water [134]. Moreover, they are significantly more expensive than alkaline metal hydroxides. As a result, the preferred method for production of biodiesel still incorporates the use of alkaline metal hydroxides.

One of the main drawbacks of the homogeneous base catalyst system is that they cause corrosion within the reactors and pipelines, which means that biodiesel production in a continuous flow reactor is not sustainable [135]. Also, at the end of the process, the catalyst needs to be separated from the product. In the case of homogeneous base catalysts in biodiesel, this is a difficult process, which is highly energy intensive as it requires several stages including aqueous quench and neutralisation [136]. The by-products of these stages include stable emulsions, soap, and a significant quantity of sewage. Although the by-product glycerol could theoretically be sold for further use, practically, resale is precluded as it is highly contaminated due to the aqueous waste salt streams [99]. The use of heterogeneous base catalysts for triglyceride transesterification could address some of these issues [137].

3.1.2. Homogeneous Acid Catalysts for Biodiesel Production

The homogeneous acid catalysts have also been employed for the synthesis of biodiesel from renewable-based feedstock such as animal fat, PFAD, grease, and waste cooking oil [138]. The common homogeneous acid catalysts used for this reaction process include H$_2$SO$_4$, HCl, BF$_3$, and H$_3$PO$_4$ [139]. The process for synthesizing biodiesel with acid catalysts has many downsides, including slow reaction times, and it requires increased quantities of methanol. For example, it takes over 50 h to achieve a 100% conversion of soybean oil at 65 °C using H$_2$SO$_4$ and a methanol:oil ratio of 30:1 [140]. Scientists normally increase the catalyst concentration, methanol:oil ratio (20–300:1), and reaction temperatures (150–250 °C) to increase the reaction rate for research purposes [141]. In addition to slow reaction times, homogeneous acid catalysts are corrosive, lead to excessive amounts of waste water, and complex procedures are required to separate the catalyst from the reaction products for reuse [142]. Finally, the biodiesel produced is substandard as the residual acid has an adverse effect on the sulphur content [143]. This has led to the development of heterogeneous acid catalysts for this process.

3.2. Transesterification and Esterification of Vegetable Oil Using Heterogenous Acid and Base Catalysts

Due to the rise of concern regarding separation, waste, and corrosion problems, the use of a heterogenous catalyst such as solid base and solid acid catalyst are frequently used in the biodiesel production process. Indeed, the use of solid catalyst eliminates the separation problem; hence, this process is more environmentally friendly than the
conventional homogeneously catalysed process. Since the heterogenous acid and base catalyst can be easily removed from the product via filtration, this process is also very economical. Although the use of heterogenous solid catalyst is environmentally and economically effective, the superior reaction activity occurs as higher reaction temperature (between 100 and 250 °C) and uses a higher methanol:oil ratio (10–25:1) and a higher concentration of catalyst (3–10 wt%). Indeed, the reaction times are much slower in a heterogeneous process when compared with homogenous reaction process [144].

3.2.1. Heterogeneous Base Catalysts for Biodiesel Production

The use of heterogeneous solid base catalysts (single metal oxides [145], mixed metal oxides [146], supported alkali metal/metal ion [147], clay mineral (hydrotalcites) [148] and organic solid bases [149,150] in the synthesis of biodiesel from vegetable oil-based feedstock has received a lot of attention. These included the metal oxides La$_2$O$_3$, MgO, ZnO, and CaO. Based on their individual performances towards biodiesel synthesis from vegetable oil, it was found that reaction rates of these oxides were related to their basicity, and was particularly related to their strong basic sites. The order of their activity towards biodiesel synthesis was therefore found to be La$_2$O$_3$ > CaO > MgO > ZnO. Among these oxides, CaO is one of the most attractive catalysts for industrial processes as it is readily available, relatively cheap, and has a low toxicity [151,152]. However, leaching of metal ions from the surface of the CaO into the biodiesel was observed [153,154].

The second class of base catalysts, e.g., mixed metal oxide catalysts, have also received some attention for this application as they offer improved basicity and mechanical strength compared to single metal oxides [155]. Biodiesel yields greater than 80% were achieved at temperatures of 60–64.5 °C when mixed metal oxides or bimetallic oxide catalysts such as Al$_2$O$_3$-SnO, Al$_2$O$_3$-ZnO [156], CaO-CeO$_2$, CaZrO$_3$ [157], and CaO-MgO [158] were used. However, it was observed that the activity of the CaO-MgO catalyst on biodiesel production using vegetable-oil-based feedstock reduced significantly after four successive trials [25]. This was likely caused by the blockage of the catalyst active sites arising from adsorption of either the intermediates or the products, such as glycerine, monoglycerides, and diglycerides or contamination during the filtering process arising from oxygen, water, and carbon dioxide in the air [159]. However, it proved possible to regain the original activity level by high-temperature treatment when extra calcium precursor was used [159,160]. Mg-La oxides have also been shown to give excellent results for the conversion of sunflower oil with yields as high as 100% at room temperature [161,162].

The third category of base catalysts are supported alkali metal/metal ion, have also been considered for triglyceride transesterification [163]. Metal-supported catalysts demonstrated superior basicity and catalytic activity to single metal oxides [164,165]. Support materials (e.g., alumina, zirconia, activated carbon, etc.) are modified by the addition of basic oxides (e.g., CaO, ZnO, BaO, and MgO) to alter their chemical properties to produce this type of solid base catalyst [137,166–168]. It is also possible to incorporate metals such as Na, K, Li, Ba, and Mg either on their own or as ions in their halide, carbonate, hydroxide, or nitrate forms [169]. Conversion rates greater than 90% have been achieved with NaOX/Na-X Titanasilicate at the temperature of range of 100–120 °C (ETS-10) [170]. Albuquerque et al. (2008) reported similar results of conversion using KOH/Na-X, CaO/SBA-15 catalysts, with 85% conversion at the reaction temperature of 60 °C and 95% conversion at reaction temperature of 65 °C, respectively. However, similar to the CaO discussed earlier, a reduction in activity was observed over time due to leaching of the active species and/or blockage of the active site arising from adsorption of organic molecules [171,172].

The other category of base catalysts used for biodiesel production are the anionic clays such as hydrotalcite [173] or layered double hydroxide (LDH) [174]. In particular, Mg-Al LDHs have received attention for application in biodiesel synthesis [175]. The main advantage of Mg-Al LDHs is that, although they do not have high activity levels, their catalytic activity is retained when exposed to water or high FFA [176]. Activity levels for these catalysts can be altered by changing the Mg/Al ratio and calcination
The highest activity levels were achieved with a combination of an Mg/Al at a molar ratio of approximately 3 and a calcination temperature of 500 °C [178,179]. In a study carried out by Zhang et al., (2008), biodiesel conversion of 99% was achieved for acidic cottonseed oil (FFA 9.5 wt%). These experiments used 1 wt% Mg-Al LDHs with a methanol:oil ratio of 6:1, reaction time of 3 h, and reaction temperature of 200 °C. The FFA content was significantly reduced down to 1 wt%, at the end of the process. Mg-Al LDHs have been modified by some researchers in order to increase its reactivity without affecting the retention of activity upon exposure to FFA and water. For example, Bo et al., (2007) have looked at Mg-O rich Mg-Al LDHs and KF/Mg-Al LDHs and MacAla et al., (2008) examined Fe-doped Mg-Al LDHs. It was found that Mg-Al LDHs did experience a reduction in activity similar to the other classes of catalyst due to adsorption of hydrocarbons on the active sites [180]. This can easily be rectified through the process of air calcination [181].

The last category of base catalysts that are potentially suitable for the synthesis of biodiesel are the organic solid bases, which include alkyl guanidine [122], Zn-arginate [182], and anion exchange polymer resins [183,184]. The two main benefits of these catalysts are (1) that they work with unrefined acidic oil and (2) separation of the biodiesel and glycerol is easier because they do not produce soap or emulsion by-products, as the organic base-FFA complex is soluble in the reaction mixture. In order to achieve maximum performance of these catalysts, there are a number of areas that require further study, including heterogenization of guanidine, optimization of the catalysts, and reaction conditions, and prevention of the adsorption of organics on the active sites, which leads to the reduction of activity.

The main issue with all of the heterogeneous base catalysts discussed above is the observed reduction in activity over time. Depending on the catalyst, this can arise from leaching of the active metal ions, exposure to H₂O or CO₂ or the adsorption of organics. This issue affects both the reuse of the catalyst and the purity of the biodiesel and glycerol at the end of the process [141]. These particular issues make these categories of catalysts an important topic for future research. In addition, ways of reducing reaction temperatures, methanol requirements, and the concentration of the catalyst need to be examined.

It is conclusively proven that, in general, base catalysts outperform acid catalysts in the transesterification of triglycerides as shorter reaction times can be achieved with very high conversion.

### 3.2.2. Heterogeneous Acid Catalysts for Biodiesel Production

As discussed earlier, the heterogeneous catalysts are generally more suitable for continuous biodiesel reaction processes. Separation from the reaction products and subsequent purification is relatively simple, which improves recyclability of the catalysts [185]. The catalysts are safer and easier for industrial-scale processes as a result of less waste water production at the end of the reaction. However, heterogeneous solid catalysts have received little attention to date for the production of biodiesel via direct esterification of oils despite the fact that solid acid products are readily available [186]. The common solid acid catalysts, which are used for biodiesel production, include sulphated metal oxides [187], mixed metal oxides [188], sulphonated solids [189], zeolite [190], resins [191], and heteropolyacids [191]. Sulphated zirconia has seen some interest for this process as a result of its strong acidity and the fact that it can be regenerated via liquid-phase chemistry [192]. During studies of the stability and activity of this catalyst it was found that the biodiesel conversion of 84% was achieved in 2 h using a commercial-grade sulphated zirconia for tricaprylin transesterification [193]. The reaction was carried out at the reaction temperature of 120 °C and required elevated pressure of 6.8 atm [194]. However, a decrease in catalyst activity level was observed during the ensuing cycles due to leaching of the sulphate ions into the product.

In order to increase the performance of sulphated zirconia catalyst, the methods to improve the acidity and stability have been developed. It was found that WO₃/ZrO₂ catalyst sample displayed improved stability in comparison with the SO₄²⁻/ZrO₂, sam-
ple as loss of the acid sites into the solution was significantly reduced [195]. However, it was found that WO$_3$ was susceptible to a reduction in activity arising from oxidation caused by exposure to the reducing agent FFA; WO$_3$ is an active metal oxide for biodiesel production from high FFA feedstock as a result of its bi-functional catalytic activity and reduced susceptibility to leaching. Excellent biodiesel yield from high FFA waste cooking oil were recorded using WO$_3$-supported catalysts as reported by [196]. Activity levels could be recovered easily through air recalcination. In order to improve the performance of this catalyst, more work needs to be done to develop both the processes and the catalyst itself including an understanding of the oxidation state of WO$_3$. Another sulphated metal oxide catalyst that has been studied for this process is SO$_4^{2-}$/TiO$_2$. Chen et al. demonstrated that a yield of 90% can be achieved for the transesterification and esterification of cotton seed oil with this catalyst, although the reaction required elevated temperatures of 230 °C. The stability of this catalyst is yet to be studied.

3.3. Bio-Catalyst for Biodiesel Production

Recently, biocatalyst have been used as catalysts for biodiesel production process. Biocatalysts consist of free lipase, traditionally immobilized lipase (lipase immobilized on nonmagnetic material) and lipase immobilized on Magnetic Nanoparticles (MNPs). Notably, enzymes are better than homogeneous catalysts because of their biocompatibility, biodegradability, and environmental acceptability [197]. Lipases have excellent catalytic activity and stability in nonaqueous media. This catalyst has been proven to be effective in the esterification and transesterification of carboxylic esters during biodiesel production, as well as in aminolysis [161]. Intracellular lipase from microbial cells is thought to be affordable and effective as a biocatalyst. However, whole cell-based catalysts are used in transesterification, which exhibited poor activity on an oily substrate and thus yielded limited biodiesel percentage [198]. It is worth noting that the biodiesel reaction activity can be improved (biodiesel conversion: 95–96%), but only for a longer period of time. The usage of an isolated enzyme can theoretically boost catalytic activity, due to the high cost of the enzyme, difficulty in enzyme recovery from the products, non-reusability, and exhaustion of enzyme activity limiting its application in industries [199].

The use of immobilization technique was found be effective in enhancing the enzyme properties including stability, activity, specificity, and selectivity coupled with the reduction of inhibition that may positively influence the operating conditions [200,201]. Physical adsorption, covalent bonding, entrapment, encapsulation, and cross-linking are the methods described [202]. Indeed, the nonmagnetic materials are frequently used as carriers for lipase immobilization [180,181]. These studies revealed that the enzyme’s activity is primarily influenced by two factors: pH and temperature. Notably, immobilised enzymes have better stability than free-enzyme catalyst. Immobilised lipase was found to be effective at pHs up to 8.0, whilst free lipase was found to be active at pHs up to 7.5 [183]. Many researchers have investigated the impact of temperature on lipase immobilisation. Similarly, the immobilised lipase remains stable when the reaction temperature increases to a higher temperature. This is supported by Zheng and his co-worker, who found that at 50 and 60 °C, the relative activity of free lipase was 88 and 75%, respectively, while immobilised lipase was 96 and 81% [203]. It can be suggested that the immobilized lipase is well protected due to its rigid conformation. Similarly with above discussion, large amount of methanol positively influenced biodiesel conversion. High enzyme dosage is not favourable for the reaction. Water content has been shown to disrupt the action of an enzyme in a nonaqueous media [204]. Water has a significant impact on lipase’s catalytic activity and stability. Water expands the interfacial area, which contributes to the preservation of lipase activity. Because lipase is more flexible in the aqueous phase, an excess of water promotes hydrolysis. Lipase has a high level of stability at optimal water content [205,206]. Based on former finding [207] in transesterification of propyl oleate over free lipase (P. fluorescens) and immobilized lipase, the result showed highest biodiesel conversion at at 60 °C, whereas the activity of free lipase is low at 70 °C. In the same study,
the reaction rate of immobilized lipase increased at 70 °C, which revealed the stability of immobilized lipase. It is worth noting that the majority of the immobilised lipase had a high recycle ability (>10 cycles) [208,209].

MNPs are commonly used to immobilise proteins, peptides, medicines, and enzymes. MNPs offer some advantages including higher specific surface area/volume ratio allowing more enzyme binding, low mass-transfer resistance, and less fouling for effective separation catalyst and product recovery by external magnetic field. Indeed, nanoparticles result in high thermal stability of catalyst. Fe₃O₄-coated Si (Fe₃O₄@SiO₂) is a common MNP used in enzyme immobilisation. Based on former findings from studies on the transesterification of olive oil over Burkholderia lipase immobilized on an alkyl-functionalized Fe₃O₄@SiO₂ catalyst [210,211], high conversion of biodiesel (90%) is achieved using 11 wt.% catalyst for 30 h reaction time. Similarly, P. cepacia lipase immobilised on MNPs achieved a high conversion (79%) of biodiesel [212], whereas Thermomyces lanuginosus lipase immobilised on Fe₃O₄ achieved a high conversion (97%) of palm oil transesterification. Similarly with immobilised lipase, the immobilised MNPs catalyst showed high operational stability.

3.4. Recent Trend of Biomass-Based Catalyst

The application of innovative heterogeneous catalysts derived from agricultural waste has been prioritized. The utilization of biomass is still attractive and highly recommended for producing highly efficient catalyst for producing high-quality, cost-effective, and long-lasting biodiesel. Carbon-based acid catalysts have several distinct advantages, including lower cost, due to the fact that they may be made from low-value biomass waste and have a large surface area and porosity [213]. This was in agreement with Table 3 findings, whereby the majority of the biomass-based catalysts exhibited high surface area (~13.30 to 1411 m²/g) and large pore size [167,168,172,213–215]. Indeed, most of these studies still used the method of chemical activation method using H₂SO₄, H₃PO₄, and KOH for improving the acidity and textural properties of the activated carbon catalyst, hence promoting the catalytic esterification and transesterification reaction (FAME yield: 44–97%). Tang et al. used direct sulfonation with concentrated H₂SO₄ to synthesize sugarcane bagasse and oil-palm-trunk-derived catalyst to catalyse the esterification process between waste oil and methanol. FAME yields were reported to be higher than 93.49% and 72.09%, respectively [216]. Notably, instead of using chemical activation, recent findings also tried to improve the textural properties of carbon catalyst using water steam approach [217–219]. Using steam approach can improve the surface area by 90% enlarged from its parent carbon. Despite the fact that this process is regarded a green way for synthesizing an effective activated carbon catalyst, it has not been extensively researched in the biodiesel generation reaction.

### Table 3. Different type of biomass-based catalyst.

| Biochar Sources      | Activation Conditions               | Surface Area (m²/g) | Pore Volume (cm³/g) | Pore Size (nm) | Type of Reaction | FAME Yield a | References |
|----------------------|------------------------------------|---------------------|---------------------|----------------|------------------|--------------|------------|
| Woody biomass        | Sulfonation, Activation temperature: 875 °C | 1411                | 0.71                | 2.20           | Transesterification | 44.20        | [220]      |
| CCAC900              | Sulfonation at 900 °C              | 972.66              | 0.11                | 2.43           | Esterification     | 93.49        | [216]      |
| Peanut hull          | Sulfonation, Activation temperature: 100 °C | 242                 | 0.13                | 1.05           | Esterification     | 97.00        | [221]      |
| Sugarcane bagasse    | Sulfonation, Activation temperature: 150 °C Charring temperature: 470 °C | 54.74               | -                   | 2.7            | Esterification     | 80.00        | [222]      |
| Mixture of dried leaves | Charring temperature: 470 °C        | 18.767              | 0.019               | 2.785          | Transesterification | 85.00        | [223]      |
| Irul wood biomass    | Sulfonation                        | 13.30               | 0.005               | 101.02         | Esterification and Transesterification | 95.60        | [224]      |
| Pomelo peel          | KOH activation                     | 278.2               | 0.154               | -              | Transesterification | >82.00       | [225]      |

a = Determined by GC-FID analysis.
4. Biodiesel Production Techniques

Biodiesel production from vegetable-based feedstock can be performed using various techniques. This section will explain various biodiesel production techniques and their feasibility. Some of the techniques identified in this section include catalytic transesterification or esterification using microwave irradiation, methanol reflux, and ultrasound conditions. Methanol supercritical technique is the only non-catalytic transesterification process used for biodiesel production.

The most common method for producing biodiesel is catalytic transesterification of vegetable oil or animal fats in the presence of a catalyst using alcohol such as methanol [226]. Noted, other alcohol species including (ethanol, propanol, and butanol) [227]. From an economic standpoint, both methanol and ethanol are regarded as the best alcohols for biodiesel generation [227]. It is worth noting that during the transesterification reaction, the alcohol and triglycerides are rarely miscible enough to form a single phase mixture. Owing to the limited contact between the reactants, the reaction is therefore lower than the reaction rate. Interestingly, catalysts can promote more contact of reactants and hence optimize catalytic activity and yielded more biodiesel product [228]. Contact between reactants can be categorised into three types of mechanisms: mass transfer, kinetic, and equilibrium [229,230]. Indeed, mass transfer becomes slower in multiphase reactants due to the poor miscibility of methanol and triglycerides. The mass transfer continues between the phases until it becomes complete. Once the process is complete, it will then continue with kinetic process, in which the forward and backward reaction continues until the reactant phase becomes in equilibrium state, which stems from the alcohol and oil molar ratios. Increased temperatures and intense mixing can facilitate both mass transfer and kinetic process (Figure 3) [231].

The techniques used in intensifying the transesterification process such as microwave and ultrasound have been developed to improve mixing, heat and mass transfer, and separation of products [73]. These techniques improve the miscibility of oils and methanol and thus increase the yield of the product during transesterification reaction process. Immiscible liquids can be emulsified through the use of low frequency ultrasonic irradiation at an industrial scale. Meanwhile, microwave irradiation reactants can be efficiently and rapidly heated at a small scale [73]
4.1. Catalytic Transesterification and Esterification

4.1.1. Microwave Irradiation

Microwave irradiation, or microwave-assisted transesterification, is a heating technique using an electronic microwave to heat up a system. Low electromagnetic wave frequency ranging from 0.3 GHz–300 GHz is used to produce energy [231,232]. Microwave irradiation generates heat inside the particles and heats the entire particles at approximately the same rate. The energy is rapidly transferred to the sample and received by particles in the system. The bonds are neither broken nor formed. Microwave irradiation is integrated in a chamber contained of a mixture of methanol, feedstock, and catalyst (Figure 4) [232].
Microwave-assisted catalytic transesterification is widely studied since it is able to promote high biodiesel conversion while producing premium quality of product. Due to the homogeneous distribution of heat generated by the microwave, heating generation occurs rapidly, so this technology is extremely energy efficient [73]. Recently, Patil et al. studied microwave-assisted catalytic transesterification over alkaline catalysts (BaO and KOH) to evaluate the efficiency of microwave irradiation in biodiesel production [233]. This study established that microwave heating utilises approximately 10% less energy than conventional heating. As previously stated, microwave heating has many appealing characteristics that make it effective for use in biodiesel production including high yield of biodiesel, cleaner output, and a lower catalyst-to-methanol ratio [232]. This is consistent with current findings [234], which study the catalytic transesterification of chicken waste through microwave radiation for biodiesel production. Indeed, a high yield of biodiesel (95%) was obtained only with a lower power consumption (1.4 kW) and a minimal volume of solvent usage (pDES) of 8% at an optimal time of 25 min.

4.1.2. Reflux System

Reflux method is a low-temperature, open-system method used for biodiesel production process at laboratory scale [235]. The technique is usually adopted for catalyst testing on biodiesel reaction for the purpose of research. The reaction takes place in a heated round bottom flask container coupled to an open-atmosphere reflux condenser system. The vaporized solvents are condensed back into the reaction medium [236].

Methanol-to-oil molar ratio is an important factor in a catalytic transesterification reaction. Heating a mixture that contains methanol solvent during transesterification may cause them to vaporise in the atmosphere. While the molar ratio of methanol to oil should be controlled throughout the process, small changes in the concentration of the mixture via vaporisation could affect the methanol-to-oil molar ratio in the reaction. As a result, applying reflux method in catalytic transesterification would prevent the solvent loss when heating.
Al-jaberi et al. reported the use of heterogeneous catalyst of MnO-NiO-SO$_4^{2-}$/ZrO$_2$ for esterification of palm fatty acid distillate, PFAD, under conventional reflux system to synthesise biodiesel. PFAD is a low grade of palm oil that contains water, impurities, and free fatty acids [237]. Therefore, a single run using catalyst to obtain biodiesel is difficult. In the experiment, esterification of PFAD took place at 70 °C for 3 h with 15:1 methanol:oil ratio with catalyst loading 3 wt% using normal conventional reflux reactor. The yield recorded from this experiment was 97.7 ± 1.02%.

Lin et al., 2015 in their experiment examined potassium hydroxide, KOH, homogeneous catalyst for transesterification of chicken oil in the reflux system and allowed to reflux to produce fatty acid methyl/ethyl (acyl) esters mixture. It successfully produced a fatty methyl/ethyl esters mixture from the best molar ratio of methanol:ethanol:oil as 3:3:1 within an hour. It was concluded that this ratio offered an alternative of transesterification of chicken oil [238].

4.1.3. Ultrasound Conditions

Ultrasound techniques employ a sound spectrum from 20 kHz to 100 MHz in order to improve the mass transfer speed between immiscible liquid-liquid phases; the reactants undergo emulsification in response to the microturbulence produced by the radial movement of cavitation bubbles [239]. Cavities are induced via sustained molecular vibration; high soundwave frequency compresses and expands the molecular distribution within the medium with the result that microfine bubbles are the product of abrupt stretching and irregular inward motions that disturb the phase interface.

Soybean oil transesterification as a technique for biodiesel synthesis has been the subject of research [239]. In this study, an alkali-based catalyst was employed, together with power ultrasound with a wavelength of about 20 kHz. A brief reaction period was permitted, which led to a significant harvest; the underlying mechanisms encompassed emulsification and cavitation of the immiscible liquid-liquid substrate. The researchers determined that the use of ultrasound yielded more rapid processing and had a lower energy requirement than traditional physical mixing. Apposite admixing, generation and collapse of the tiny bubbles within the medium were produced by the use of ultrasonic techniques; the reaction generated high temperatures and pressures. The method markedly diminished the time required, the molar proportions of methanol and oil, and the quantity of necessary catalyst. As a result, the combination of calcined hydrotalcite catalyst and ultrasonic radiation is now acknowledged as a novel efficacious method.

A further study demonstrated that in an ultrasonic reactor, a solid alkali-based catalyst comprised of hydrotalcite-derived particles with a 3:1 molar ratio of magnesium and aluminium together with a range of variables could be used in conjunction with Jatropha oil feedstock [240]. The influence of the power from the ultrasound on the biodiesel was assessed; the largest harvest of biodiesel, 94.5%, was obtained with a supersonic power of 210 W (Figure 5).

![Figure 5. Effect of biodiesel yield for ultrasonic power. Reproduced with permission from Deng et al., Production of biodiesel from Jatropha oil catalyzed by nanosized solid basic catalyst; published by Elsevier, 2011 [240].](image-url)
A study involving the transesterification of waste frying oil was performed by Hingu et al. [240]. These researchers employed methanol in conjunction with uniform KOH as a catalyst; a low ultrasonic frequency of 20 kHz was utilised. The effect of a range of procedural variables on the degree of oil transformation was assessed, i.e., alcohol-oil molar proportions of 6:1, 1 wt.% catalyst concentration, ultrasound power of 200 W, 40-min period of irradiation, temperature of 45 °C, pulse and horn position. A 150 W power yielded a 66% conversion rate; this was elevated to 89% with a power of 200 W. This increase was determined to be the result of improved admixing and emulsification of the immiscible reaction components. Any further rise in power diminished the transesterification transformation yield. In this instance, a cushioning phenomenon occurred, which led to a reduction in the energy shift into the system which produced diminished cavitation behavior [241]. The findings of this study are illustrated in Figure 6.

![Figure 6. Effect of ultrasonic power dissipation on the extent of conversion of biodiesel Reproduced with permission from Hingu et al., Synthesis of biodiesel from waste cooking oil using sonochemical reactors, Elsevier, 2010 [241].](image)

The use of pulsed ultrasound, i.e., intermittent on-and-off delivery phases of a few seconds’ duration, has given rise to interesting outcomes. For 2-s on and 2-s off phases, the transformation rate was 62%; 65.5% was obtained with a 5-s on followed by a 1-s off sequence. When an on phase of a minute together with a 5-s off pattern was employed, the conversion rate reached 89.5%, implying that the immiscible liquid-liquid strata had been fully segregated.

It was noted that the overall shift of triglycerides between the oil phase and the methanol-oil boundary was tardy, a reaction hastened by the ultrasonic cavitation. In contrast to the traditional physical mixing method, the latter yielded a conversion rate of 89.5% as opposed to 57.5%; the reaction duration was 40 min. The process was mass transfer controlled; the cavitation bubbles gave rise to microturbulence which facilitated the conversion process owing to the greater surface area at the liquid-liquid interface. The authors surmised that ultrasound can stimulate efficacious emulsification and mass transfer, thus enhancing the speed of esterification.

### 4.2. Non-Catalytic Transesterification and Esterification

#### Methanol Supercritical

Interestingly, that non-catalytic methanol supercritical conditions can also yield biodiesel [242]. According to reports, supercritical biodiesel manufacturing necessitates a high molar ratio of alcohol to oil (42:1), as well as high pressure and temperature without the need of a catalyst. [229,232]. As a result of the high pressure and high temperature
used in this process, it is capable of transesterified the high free fatty acid feedstock to biodiesel [73]. Contrary to catalytic transesterification, supercritical method favours the presence of water as it promotes mechanism of the reaction although it is expensive method as it requires high volume of alcohols. This method is environmentally friendly because wastewater from this method does not contain acid or alkali, which is usually produced in catalytic transesterification. This method involved simple post-production process, because the separation of the used catalyst and saponified products from methyl esters is eliminated.

When judged against catalysed transesterification processes, this technique requires a briefer reaction period and the yield is excellent. In terms of increased acid worth and water content of the oil, this modality converts biodiesel with ease. The drawbacks encompass the significant pressures and temperature required, the high proportion of alcohol to oil and risk of biodiesel breakdown [243]. It has been underlined that the surplus alcohol should be collected assiduously once the reaction has concluded since any contamination of esters with methanol can be confirmed via flash point temperatures in ASTM D93 [227]. Moreover, any such impurities may cause the biodiesel not to meet the criteria for the minimum fuel flash point. Inadequate ester purification was the principal cause of impurities following the transesterification process. The viability of a range of manufacturing regimes for biodiesel with differing variables is presented in Table 4.

Table 4. Comparison of different technologies in producing biodiesel. Modified from [229].

| Variables                  | Acid Catalyst | Base Catalyst | Supercritical Alcohol | Heterogeneous Catalyst |
|----------------------------|---------------|---------------|-----------------------|------------------------|
| Reaction temperature       | 55–80         | 60–70         | 239–385               | 180–220                |
| Water in the feedstock     | Interfere with reaction | Interfere with reaction | Not sensitive              |                        |
| Free fatty acid in feedstock | Esters     | Saponified products | Esters               | Normal                 |
| Yields of methyl esters    | Normal        | Normal        | Good                  | Normal                 |
| Purification of methyl esters | Repeated washing | Repeated washing | Nd                    | Easy                   |
| Recovery of glycerol       | Difficult     | Difficult     | Nd                    | Easy                   |
| Production cost of catalyst| Cheap         | Cheap         | Medium                | Potentially cheaper    |

5. Properties of Biodiesel

5.1. Physical Properties

Physical properties of biodiesel that are comparable to those of petroleum-derived diesel include a high flash point, viscosity, cetane, and energy values, respectively (Table 5). The two forms of diesel can be combined and deployed in traditional diesel ignition engines with only a small number of adaptations; none may, in fact, be required [77,244]. Key benefits of biodiesel are that it is neither incendiary nor volatile; it has a lower toxicity, is biodegradable, and can be recycled [245]. The emission profile of biodiesel is positive for the environment, and it has superior combustion efficacy, cetane number, flash point, and lubrication than diesel synthesized from petroleum [7,246–248].

An updated summary of the major manufacturers of biodiesel included industries in Asian nations, e.g., Thailand, Malaysia, China, the Philippines, India, and Japan [249], together with Australia [250], Turkey [251], Bangladesh [252], and Africa [253]. These geographical territories comprise a wide surface area and they are rich in intrinsic feedstock sources. However, there are few data available with respect to biodiesel plants in developing nations with less land and who are dependent on trading for most of their energy supplies. It may be challenging for them to pursue contemporary practices of deploying biodiesel, together with additional biofuels founded on domestic reservoirs, so as to offset their reliance on fossil fuels and to diminish their carbon footprint.
Table 5. Comparison of biodiesel and diesel according to the American Standard for Testing and Materials (ASTM).

| Property of the Fuel        | Biodiesel                          | Diesel                                    |
|-----------------------------|------------------------------------|--------------------------------------------|
| Standard method             | ASTM D6751                         | ASTM D975                                 |
| Fuel composition            | FAME(C_{12}-C_{22})                 | Hydrocarbon(n-C_{10}-n-C_{21})             |
| Density (g/cm³)             | 0.878                              | 0.848                                     |
| Pour point (°C)             | −15 to 16                          | −30 to −15                                |
| Cloud point (°C)            | −3 to 12                           | −15 to 5                                  |
| Flash point (°C)            | 100–170                            | 60–80                                     |
| Cetane number               | 48–60                              | 40–55                                     |
| Water (vol %)               | 0.05                               | 0.05                                      |
| Carbon (wt. %)              | 77                                 | 87                                        |
| Hydrogen (wt. %)            | 12                                 | 13                                        |
| Oxygen (wt. %)              | 11                                 | 0                                         |
| Sulphur (wt. %)             | 0.05                               | 0.05                                      |

Biodiesel is considered the best candidate for petroleum-derived fuel substitute in diesel engines (compression-ignition engines) due well-known advantages [254]. Biodiesel has good combustion properties and is able to reduce noxious emissions such as CO, CO₂, HC, PM, Sox, and PAH due to its high oxygen content (11–15%) [255]. In addition, lifecycle analysis indicates that biodiesel has 78% lower net carbon dioxide emissions than conventional diesel and has a lower smoke emission factor as a consequence of minimal free soot [256,257]. And biodiesel is a better lubricant than conventional diesel and therefore a better promoter of engine efficiency. Biodiesel has a higher flashpoint than conventional diesel (Table 4), which is a safety feature, reducing likelihood of explosive combustion. Biodiesel offers the additional environmental benefit with the potential to recycle food oils, removing them from the waste stream. Biodiesel (B20) in low blends can be used directly (without modification) in engines, although some engine modification may be required to use higher ratios of biodiesel.

5.2. Analytical Technique to Determine Chemical Properties of Biodiesel

Various analytical approaches have been developed to analyse biodiesel formation. Chromatography procedures are commonly utilised because they provide extensive perception and detailed information needed for product quality control. However, baseline drift, overlapping signals, and standards are needed, and samples can destructively affect the GC accuracy. Another drawback of the GC analysis is some components of the biodiesel aren’t volatile enough to be evaporated and quantified by the GC analysis. Nuclear Magnetic Resonance (NMR) spectroscopy and Fourier Transform Infrared (FTIR) spectrometry have lately been employed to monitor biodiesel production. NMR is a powerful method that is currently underutilised in biodiesel analysis [258]. Although FTIR is one of the well-established methods for analysing biodiesel, it is less sensitive for detecting tiny components than Gas Chromatography (GC). It can, however, be used to check the quality of biodiesel fuel by comparing it to current GC or other analytical data [259]. The FTIR approach is simpler to implement and faster than the GC method. The spectral analysis of biodiesel, feedstock oil, and samples during intermediate conversion can be used to establish a biodiesel formation or biodiesel management strategy [260]. Other studies in the literature describe the application of FTIR analysis to chemical detection and quantification. Indeed, high performance liquid chromatography (HPLC) also can be used for biodiesel characterization, yet HPLC analysis is less employed in biodiesel analysis. It worthy to mention that HPLC technique can be applied to biodiesel from different feedstock and a variety of detectors can be used; the most commonly used ones are UV/ VIS absorption detector (UV/DAD) and Mass Spectroscopy (MS). The two procedures, GC and HPLC, rely substantially on the use of standards for each component of the biodiesel. As a result, chemical changes and the production of new products during biodiesel storage would be difficult to detect using those two approaches.
6. Current Status of Biodiesel Production

In 2019, the world has produced 40,783 Mmt of biodiesel, led by Indonesia (7000 Mmt) and followed by USA (5712 Mmt) and Brazil (5075 Mmt), as presented in Figure 7 [261]. The success of developing countries such as Indonesia and Brazil as biodiesel world-leading producers are mainly due to the availability of feedstock, government support, and financial incentives [262]. Government of Brazil through its supportive policy has increased 1% of biodiesel blend into diesel fuel at pump stations from 11% to 12% [263]. In another recent study, the mandate of B20 for biodiesel implied a positive impact on the economic, social, and environment of Indonesia [264]. On the other hand, Figure 7 German's biodiesel production have fallen 21% as compared to its five-year average due to poor growing conditions [265].

Currently Argentina is expanding its biodiesel production into second generation biodiesel whereby the biodiesel plant is able to turn sewage waste into biodiesel [266]. The conversion of sewage effluents into deacidified residual oil can reduce up to 90% of greenhouse gas emissions [267].

The scenario of top biodiesel producer of Asian countries including Indonesia, Thailand, Malaysia, China, Philippines, India, and Japan [249] and other countries such Australia [250], Turkey [251], Bangladesh [252], and Africa [253] have been summarised. Most of the countries appear to have abundant natural resources, including ample oily feedstock for biodiesel manufacturing. However, data on biodiesel generation in those countries is still scarce, as the majority of their energy is imported [268].
Loa PDR, formerly known as Laos, imports 100% fossil fuel from Thailand and Vietnam and uses it for transportation except for rural areas. Biodiesel development in Loa PDR is still behind compared to its neighboring countries especially Thailand. Private sectors initiated the biodiesel development in Loa PDR [269]. Non-edible has been used for biodiesel raw materials including Jatropha [270], Vernicia Montana [271]. Lao PDR has less developed plantation compared to other developing country like Thailand. Thus, Loa PDR Vernica Montana has seen as the most suitable raw material for biodiesel production in Laos since it can be cultivated under low-temperature/frosty conditions, non-edible, possess 40–60% oil in each seed and has low free fatty acid [272]. In addition to that, reforestation program enabled the plantation of Vernicia montana across Laos and Vietnam, which in turn to prevent land erosion and also to support the generation of oil for BDF production.

Biodiesel production in India is slow in comparison to Ethanol due to limited access to feedstock limited production capacity, a rudimentary supply chain, and import restrictions [273]. However, the consumption of biodiesel in India started rising back in 2016 with 119 million L. The steady progress of biodiesel consumption is 4% and a slight reduction has been seen in 2019. India currently produces 650 million road litres biodiesel per year for oil marketing companies, the Indian railways, State Road Transport Corporation, fleet owners of transport companies, and port authorities [274]. The source of raw materials for biodiesel in India are mainly from imported palm stearin, non-edible oils, used cooking oil (UCO), and animal fats [275]. The government of India encourages the use of UCO as the feedstock for biodiesel production and increases its procurement as stated in India National Policy on Biofuels [276]. Production of biodiesel from UCO seems promising. In a recent study on biodiesel production from UCO’s, only 5% of the total production cost was required. Total production cost was estimated at 28.55 INR/L, which is equivalent to 0.39 USD/L. These figures were based on the reflux method setup and foreseen to break even at 3.5 years [277]. Not only will economic sectors benefit but also the environment, waste management, and health of India’s people.

Raw materials are an important aspect to consider by the developing country. The selection of the appropriate feedstock will increase the value of the biodiesel. A few developing countries such as India, Mexico, Ethiopia, and Ghana found that Jatropha cultivation for biodiesel fuel faced challenges. For example, India found the investment of Jatropha was such a failure after a decade of cultivation whereby the average seed yield was actually 0.5–1.4 Mg ha$^{-1}$ yr$^{-1}$ after five years of plant growth in a multi-locations trial [278]. The reports showed that pest and disease of jatropha affects the seed yield in Burkina Faso [279], cultivation in India [280], and the death of trees in Brazil [281]. However, the pest and disease damage were not the main factor of Jatropha abandonment by farmers in Mexico. Their decision to abandon jatropha cultivation was based on the deterioration in perception of jatropha profitability, the wealth position of the household, and the non-payment of expected subsidies [282].

Initiatives instigated by governments have enhanced the biofuel market by facilitating the expenses related to their synthesis [255]. At present, politicians in the United States, European Union, Brazil, and some developing nations have implemented a range of support programmes for biofuels, e.g., tax benefits for manufacturers, countrywide blending mandates, and import fees [283]. In several instances, the costs of the ethanol and gasoline in fuel admixtures have been diminished, thus promoting the use of fossil fuels [284]. The policymakers need to assess their strategies and their underlying rationale in order to circumvent offering motivations that might hinder the ultimate objectives of mitigating climate change and decreasing reliance on fossil fuel importation. These political manoeuvres and the consequences of inflated oil costs have provided incentives for the swift growth of biofuel manufacture seen over recent years and reflect a likely future pattern.

Biodiesel fuel manufacture may be expensive and potentially of detriment to the environment. It has financial implications related to the type of feedstock utilised and potential rivalry generation with other industries, e.g., where edible feedstocks are considered. Deforestation may also be accelerated by the harvesting of vegetation.
Growth of the biodiesel industry is accelerating globally, owing to energy safeguarding and additional issues pertaining to the environment. The administrations of developed nations are being leaned on to instigate compulsory legislation relating to biodiesel and traditional fossil-derived diesel blending. The requirement for biofuels is likely to rise significantly in the future as a result of political recommendations to change to recyclable energy sources, the rise in costs of petroleum products, and environmental contamination.

Although biodiesel has a number of advantages, there are still issues to surmount in order to upscale biodiesel manufacture to an industrial level. It should be stressed that a large proportion of the financial burden relating to biodiesel synthesis relates to the feedstock used; thus, apposite feedstock choice is a key decision. Positive feedstock options encompass those that have no rivalry with the food industry, do not cause deforestation and engender a diminished carbon footprint.

7. Current and Future Challenges of Biodiesel Fuel Productions

Since the numbers of humans inhabiting the planet is rising swiftly, additional arable areas will be necessary for food production. A paucity of available ground will unquestionably cause biodiesel manufacture to become a costly process. This is a current issue in Asia, where vegetable oils are already expensive. It is likely that this pattern will be copied globally, thus representing the greatest difficulty to be overcome with respect to biodiesel synthesis. Cheap, non-edible oils, microalgae, vegetation that is genetically manipulated, and several waste oils may offer a more sustainable way forward for future biodiesel manufacture. Nevertheless, feedstock harvesting will need to be well-designed and organised. Further studies should concentrate on non-edible vegetation that offers profitable yields. Feasible and encouraging feedstocks include jatropha, microalgae, and genetically engineered species, e.g., poplar and switch grass.

The two catalysts with the highest potential encompass heterogeneous and enzymatic agents rather than the homogeneous type. Heterogeneous catalysts can be segregated, rescued, and recycled with ease following the reaction, increasing efficacy and reducing manufacturing expense. Enzymatic agents have comparable advantages but facilitate a successful reaction in less extreme processing conditions; this also applies to cheap oil, rich in FFA. Thus, these two forms of catalyst have the potential to contribute to the developing desired processing mechanisms for biodiesel synthesis.

Contemporary robust and innovative intensification methods for biodiesel manufacture have emerged, e.g., microwave heating, ultrasonic irradiation, membrane reactor, motionless mixer, and reactive distillation, amongst others. These all diminish the necessary energy as a result of reaction rate improvement and increased harvest of biodiesel. Nevertheless, the majority of these techniques are in their infancy. Further refinement is required through optimisation research and quantitative modelling studies in order to gather information on the kinetic profiles of the reactions, efficacious reactor blueprints, expense and expansion for the commercial market. The present situation pertaining to these methods together with their benefits and drawbacks are listed in Table 6.

With the exception of the difficulties alluded to, social compliance with biodiesel is a further problem to be surmounted which is linked with the population’s faith. The concept of biodiesel cannot be forced onto the community; instead, converting individuals to this energy source requires genuine endeavours from the sector and administrative bodies. When a precis is formed of the previous discourse, a number of difficulties yet to be surmounted impact the future direction of biodiesel: (i) the discovery of de novo and plentiful feedstock from cost-effective reservoirs that do not generate rivalry with the food industry; (ii) the design of financially acceptable, straightforward and rapidly acting heterogeneous catalysed and immobilisation techniques for lipids; (iii) an improvement in the current methods of biodiesel synthesis with careful utilisation of intensification processes apt for cheap feedstock and catalysts; and (iv) the promotion of the use of biodiesel as a recognised substitute for diesel derived from petroleum.
Table 6. Biodiesel production methods.

| Techniques                  | Concept/ Principle                                                                 | Advantages                                                                                                                                                                                                 | Disadvantages                                                                                                                                                                                                 | Current Status  | References |
|-----------------------------|-----------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------|------------|
| Ultrasonic method           | Ultrasound radiation promotes phase miscibility through microturbulence-induced emulsification | Heightened interface contact area for the immiscible components                                                                                                                                           | High likelihood of erosion of probe ultrasonic reactor’s horn tip                                                                                                                                             | Industrial scale | [285–292] |
| Microwavemethod             | Ongoing changes in microwave magnetic field orient polar alcohol molecules, leading to molecular friction-generated heat | Diminished period of reaction Lower quantity of catalyst necessary Reduced standards of feedstock can be utilised, e.g., animal fats and waste oils Decreased need for catalyst and methanol by up to half Lower waste therefore less toxic to environment Lower thermal gradient Mitigates excessive surface temperature rise Superheating can occur locally Improved kinetics of the reaction Reduced time required for segregation and removal of impurities Environmentally friendly Low energy requirement | Microwaves only infiltrate a depth of several centimetres Requires internal stirring Local superheating needs minimising Mass transfer is restricted Difficulties associated with scale-up | Laboratory scale | [68,286,293–295] |
| Co-solvent method           | Deployment of a secondary solvent, which can dissolve in alcohol and oil liquids, thus improving reactant miscibility and diminishing resistance of the first mass transfer | Faster reaction rate Reaction temperature, pressure and molar proportions of oil: alcohol diminished Efﬁcacious segregation and cleansing owing to a lower number of rinsing phases Lower glycerine content compared to biodiesel generated in a batch reactor Temperature and pressure requirements reduced Ester conversion augmented Reduced processing expense No parts in motion Likely diminished power requirements | Extraction of cosolvent from reaction medium Additional quantity of wastewater created Increased expense of manufacture owing to cosolvent cleansing required | Industrial scale | [286,287,296–299] |
| Membrane reactor            | Membrane deployed for selective transport of components at varying mass transfer speeds Reaction and segregation occur in a chamber Mechanism involves the immiscibility of oil and alcohol and the diverse surface dynamics | Efficacious segregation and cleansing owing to a lower number of rinsing phases Temperature and pressure requirements reduced Ester conversion augmented Reduced processing expense | Soap conﬁguration in the reactor may contaminate the costly membrane Accurate governance of process Strict variables maintained to inhibit membrane contamination | Pilot scale     | [288,300,301] |
| Motionless mixer            | Static helical mixing component within hollow tube Offers dynamic and efficacious mixing | Upkeep straightforward Expense and area necessary reduced Low energy requirement Increased reaction rate Higher FAME harvest Eradicates the need for extrinsic recycling flows from segregation units and therefore diminishes the requirement for contact with waste Reduced power requirements | Admixture has a brief period in the mixer Accurate and timely quantities of constituents necessary | Laboratory scale | [286,293] |
| Reactive distillation       | Chemical reaction and distillation take place within one entity                   | Brief reaction duration Large transformation rate and biodiesel harvest Easy segregation of products Operation time, solvent, amount and total expense diminished | High energy requirement Transformation impacted by catalyst efﬁcacy | Pilot scale     | [99,298,302,303] |
| In-situ biodiesel production| Concurrent extraction and transesteriﬁcation                                    | Biodiesel puriﬁcation is more complex and expensive | | Laboratory scale | [301,304–306] |

8. Conclusions

There are notable benefits to biodiesel processing in several contexts. However, its feasibility with respect to industrial use is still the subject of debate. The methods, analyses and findings of current research studies are not always of immediate relevance for the upscaling of biodiesel manufacturing techniques. The world’s contemporary requirement for energy means that it is exigent to discover other energy resource options. Fossil fuel, on
which the globe is heavily reliant, is not an ongoing method of choice as it is not renewable. Furthermore, its use is accelerating climate change.

Energy forms that are potentially recyclable, e.g., biofuels known as biodiesel, are currently deemed to represent the optimal substitute for traditional fossil fuels owing to their ability to biodegrade, lower toxic profile, and equivalent physical parameters. Upscaling biodiesel synthesis to a level that will satisfy the world’s energy needs is the sole answer to the contemporary energy quandary. Nevertheless, in its present form, biodiesel manufacture is not sustainable in view of its production expense when feedstocks are comprised of superior grade oil; additionally, the traditional catalysts used have segregation issues. These issues are only rectifiable if synthesis costs can be decreased and catalytic methods improved.

The typical feedstock used for biodiesel manufacture is a superior grade oil which contains <1% FFA; this can be transformed into biodiesel in a straightforward manner with only mild reaction parameters. In this scenario, approximately 80% of the overall manufacturing expense of biodiesel is related to the feedstock. If waste cooking oils were used instead, the price of biodiesel synthesis would be reduced by 50%.

A more optimal catalyst for industrial biodiesel manufacture is likely to be a homogeneous agent rather than the heterogeneous solid-based form. This is owing to the simple technique required for catalyst segregation and extraction following the reaction.

In terms of characteristics such as combustion heat and octane number, the two forms of diesel are equivalent. However, the particulate, carbon dioxide and sulphur released from biodiesel use are notably less.

Biodiesel production involves a straightforward process of triacyl glycerol and fatty oil transesterification enhanced by alkaline or acidic catalytic agents. The latter have a number of limitations; enzymes that have less deleterious consequences on the environment have been studied. These offer a lower intensity energy release and are restricted by by-product formation. If feedstocks, such as Jatropha or palm oil, are substituted with waste cooking oil, the expenses associated with biodiesel manufacture can be notably diminished; it additionally provides a disposal solution for the latter.

The chemical transesterification reaction requires methanol or alcohol for initiation. However, the former may have toxic effects on complete cell or cell-free enzymes; it is additionally difficult to recycle following the reaction’s conclusion. Ethanol is a more apposite choice.

Establishing an efficacious method for biodiesel synthesis is likely to be a dominant factor in economic growth. Thus, studies that investigate the difficulties surrounding feedstock resources and process designs, such as reactor assembly, a range of operating variables and the generation of financially viable enzymes, are merited.

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