

Review

Biodiesel Production from Waste Cooking Oil: A Perspective on Catalytic Processes

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Abstract: Presently, the use of fossil fuels is not ecologically sustainable, which results in the need for new alternative energies such as biodiesel. This work presents a review of the classification of the lipidic feedstocks and the catalysts for biodiesel production. It also presents the pros and cons of the different processes and feedstocks through which biodiesel is obtained. In this context, cooking oil (WCO) has emerged as an alternative with a high potential for making the process sustainable. A detected limitation to achieving this is the high content of free fatty acids (FFA) and existing problems related to homogeneous and heterogeneous catalysts. To overcome this, the use of bifunctional catalysts is being evaluated by the scientific community. Thus, this work also explores the advances in the study of bifunctional catalysts, which are capable of simultaneously carrying out the esterification of free fatty acids (FFA) and the triglycerides present in the WCO. For the sake of an improved understanding of biodiesel production, flow diagrams and the mechanisms implied by each type of process (enzymatic, homogenous, and heterogeneous) are provided. This article also highlights some of the challenges in catalyst development for sustainable biodiesel production from low-grade raw materials.

Keywords: biodiesel; waste cooking oil; bifunctional catalysts; transesterification



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1. Introduction

1.1. Biodiesel Global Scenario

The energy model that currently prevails is highly dependent on the usage of fossil fuels and supports various sectors such as transportation, industry, and agriculture, among others [1]. However, this model has become less and less viable due to the reduction in the non-renewable energy source, its increasing price, and the fact that this type of fuel favors an elevation in greenhouse gas emissions, some of which have been shown to have an impact on people's health as they are related to various types of cancer [2–4]. As stated by the United Nations' Sustainable Development Goals (SDG), regarding sustainable energy access (SDG7) and climate change (SDG13), several countries have started to take actions to reduce carbon emissions by at least 43% by 2030 and to not reach an increase in global temperature of 1.5 °C between 2030 and 2050 (IPCC, 2018) [5].

The aforementioned problems have motivated the development of other cleaner energy options. An example is biodiesel, because of its capability to be produced from vegetable and/or animal fats while also being able to be used in diesel engines without major changes because it poses physicochemical characteristics similar to those of high-performance diesel [6–8]. This biofuel improves engine performance in addition to having cleaner combustion because it has an oxygenated molecule that allows it to reduce CO₂ emissions by up to 80% [6,9].

In the last decade, there has been an increase in biodiesel production of approximately 4–14%, which is an economic advantage due to its growing demand [10]. Figure 1 shows the distribution of biodiesel production per geographical area in 2021 (42.7 billion liters) [11]. It can be observed in this figure that Europe produces the highest percentage of biodiesel (34%) around the globe. Biodiesel production from 2023 to 2027 is expected to grow from 50 to 52.5 billion liters, respectively [12].

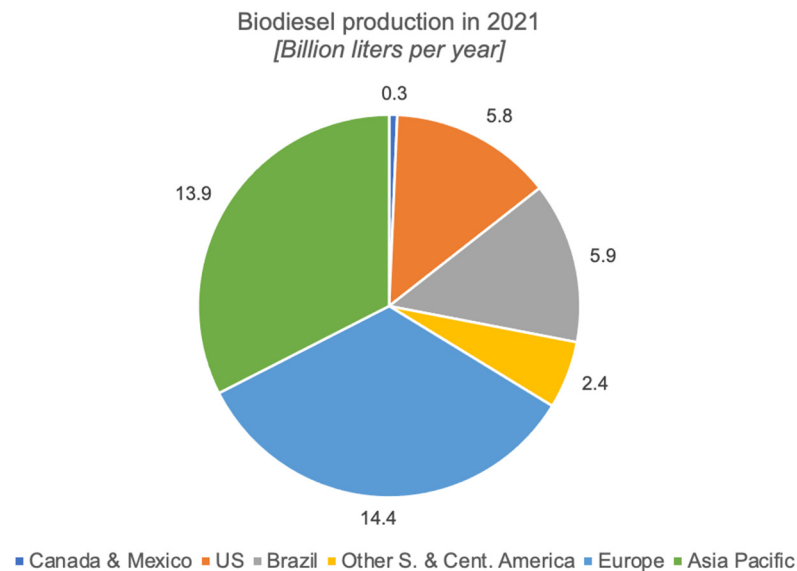


Figure 1. Global distribution of biodiesel production in 2021 with data from [11].

However, the production of this biofuel still cannot match its demand in multiple countries, mainly due to its production cost, which is derived from 60–80% of the cost of the lipid raw material [4,9]. Figure 2 shows biodiesel consumption in 2021 (45.6 billion liters) [11]. It is not surprising that the highest consumption percentage also lies in Europe (35%), and Canada and Mexico are some of the lowest consumers (1.4%).

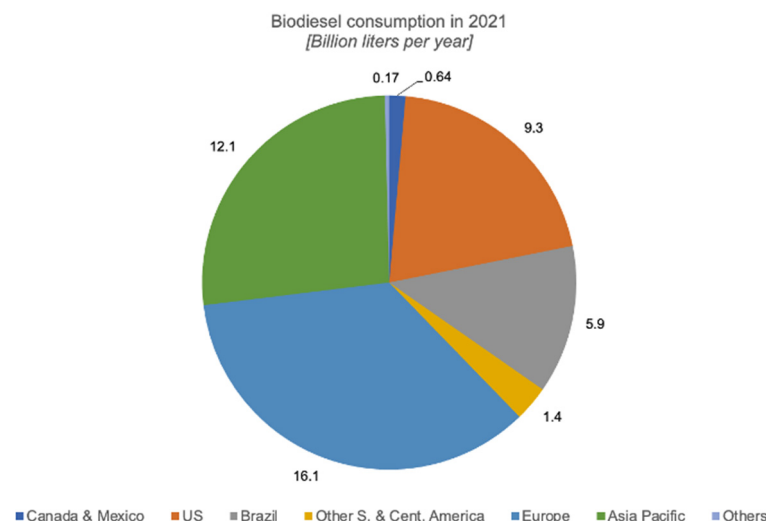


Figure 2. Global distribution of biodiesel consumption in 2021 with data from [11].

In addition, there are several countries where the legislation to promote the production of biodiesel is nonexistent and there are no established regulations about the composition of biodiesel with petroleum diesel mixtures, as has been the case in other countries in Europe and America.

In recent years, many biodiesel-producing companies have used feedstock that is more competitive and accessible than edible seed oils, such as waste oils. Furthermore, since the beginning of the Ukraine–Russia conflict, the price of biofuels has increased due to the shortage or increase in price of raw materials [13], and several European countries have been forced to use cheaper oils or waste cooking oil (WCO) to obtain biodiesel.

1.2. Biodiesel Feedstock

Figure 3 shows the feedstocks from which this biofuel can be produced. Furthermore, the context of their use will be discussed.

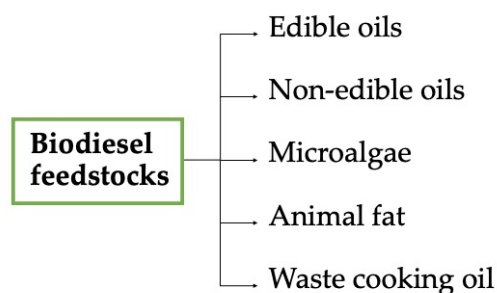


Figure 3. Feedstocks for biodiesel production.

- **Edible Oils (EO):** This raw material has been used and studied for several decades mainly because, of its purity [14]. Nevertheless, its use is currently in great controversy, mainly due to the ethical dilemma derived from its nutritional value in the market [6]. In addition, the use of this type of raw material increases the cost of biodiesel production [14]. Some of the most commonly used refined oils are soybean, peanut, corn, and sunflower [6].
- **Non-edible Oils (NEO):** The need to discover low-cost raw materials that do not compete with the food market has led to various investigations for biodiesel production from non-edible oils and reusable oils [6,14]. NEO has shown a reduction in biodiesel production costs and is highly available in several parts of the world [6]. Around the world, in different areas (mainly Africa and Asia), various species of plants are known to have an oil content of $\geq 20\%$ within their seeds, which makes them potential sources of edible oils [14]. However, the main disadvantage of using NEO as a raw material comes from their high free fatty acid (FFA) content since they would saponify when in contact with a basic catalyst [6]. To obtain a high-quality biodiesel from this feedstock, it is necessary to conduct esterification prior to the transesterification reaction [15]. However, carrying out two processes considerably increases the final cost of biodiesel production. Among the non-edible oils used to obtain biodiesel are: *Jatropha curcas*, *Croton megalocarpus*, *Ricinus communis*, *Cerbera odollam*, *Celastrus paniculatus*, *Lepidium perfoliatum* Linn, *Ailanthus altissima* (Mill.), *Capparis spinosa* L., *Calophyllum inophyllum*, *Carthamus lanatus* L., neem, jojoba, date seed, palm kernel oil, medlar seeds, karanja, and mahua [4,6,16–21]. Different from these oils, there are others such as that from the *Raphanus raphanistrum* L. seed, which is easy to grow and whose oil FFA content is very low, which allows the transesterification reaction to be carried out in a single step, which would solve the aforementioned drawbacks [17].
- **Microalgae:** This group of unicellular organisms can be cultivated in multiple climatic conditions and can be a great source of biomass with a high fat content (between 40–60%) [22,23]. This raw material has some considerable advantages, such as its rapid growth, its ability to grow in wastewater bodies, and its high lipid content. However, some of their main disadvantages are the need for large amounts of nutrients such as nitrogen and phosphorus, and some species of microalgae produce harmful toxins [22,24] and the need for large areas of land for their cultivation [25]. Some examples of microalgae that produce biodiesel are *Chlamydomonas*, *Chlorella vulgaris*,

Chlorophyceae, Chrysophyceae, Cryptocodinium cohnii, Cyllindrothec, Dinophyceae, Isochrysis, Monallanthus salina, Nannochloropsis, Rhodophyceae, and Xanthophyceae [14,26–30].

- **Animal Fat (AF):** This feedstock is available in large quantities and generally is taken from waste generated by slaughterhouses or food processing industries [31]. This residue is considered an economically viable raw material, being used mainly in Europe, the United States, and Brazil, where it is considered the 2nd most used raw material for biodiesel production [31–33]. As with reusable oils and non-edible oils, AF has high fatty acid contents (5–40%) [34,35]. Therefore, it is necessary to use a catalyst capable of handling the high fatty acid contents or a process of two steps (esterification and transesterification) to obtain commercial-quality biodiesel [22,32]. Among the most commonly used animal fats are chicken fat, lard, tallow (sheep or beef), and mixtures of all of the above [22].
- **Waste Cooking Oil (WCO) or Used Cooking Oil (UCO):** The usage of this feedstock allows for a reduction in the production cost of biodiesel by 70–80%, in addition to being a raw material with high availability [2,36,37]. This lipid raw material can be considered a viable option, mainly because it is a waste without nutritional value generated by restaurants, households, and food processing industries [38]. WCO can be classified as a combination of triglycerides and free fatty acids that have undergone physicochemical changes, which occur when the oils are subjected to high temperatures and humidity for food preparation [37].
- The use of WCO to obtain biodiesel has a double benefit: economically, it allows a decrease in the cost of its production, and environmentally, it allows a reduction in environmental contamination in water bodies and soils derived from its incorrect disposal [39]. To obtain biodiesel from WCO, it is necessary to carry out three stages: pretreatment, transesterification reaction, and biodiesel purification.

When EO or microalgae oils are used as lipid feedstocks, it is possible to obtain high-quality biodiesel through the use of a conventional basic catalyst in the transesterification reaction. The above is attributed to the low content of free fatty acids (FFA) in the raw material. Nonetheless, when unrefined or waste raw materials are used, it is not possible to omit a pre-treatment of the raw material. The objective of the pre-treatment is to eliminate solid particles and contaminants from the cooking process. Depending on the catalyst to be used, it may be imperative to perform acid esterification to reduce the amount of FFA present in the lipid raw material, followed by basic transesterification. However, this two-step process implies major energy consumption and higher production costs [14].

Recently, other unconventional feedstocks to produce biodiesel have begun to gain relevance, such as those obtained from instant coffee production (spent coffee grounds, SCG) or fat derived from insect biomass (black soldier fly larvae, *H. illucens*, BSF) [40–44]. Worldwide, it is estimated that 60 million tons of spent coffee grounds (SCG) are generated [45]. The coffee residue contains a high amount of fat (10–20%), and from this percentage, 80–90% are glycerides, becoming a potential raw material to produce biodiesel [46]. With SCG oil, biodiesel yields of 97.11–97.18% have been achieved [40,44]. Insects have a high fat content (34–58%) compared to vegetable oils, which range from 15–46% [43]. For this reason, fats derived from insects are attracting more interest in the production of biodiesel. Some of the insects that have been studied are mealworm beetles, blowflies, meat flies, houseflies, black soldier flies, and superworms [43,47–50]. Recently, the larva of the black soldier fly *H. illucens* (BSFL) is the one that has generated the most expectations because it has a high amount of fat (50%) [51,52], which can vary depending on the diet of the BSFL; reporting important percentages of fatty acids: lauric acid (47.47%) [53], oleic acid (41.90%), and palmitic acid (39.83%) [54]. Various methods have been used to obtain biodiesel from BSF: acid-catalyzed esterification followed by alkali-catalyzed transesterification [55–60]; enzymatic catalysis (Novozym 435) [61], direct transesterification (the lipid extraction and transesterification are carried out in one step) [62,63], and non-catalytic transesterification (without lipid extraction, direct conversion of biodiesel from dry BSFL) [59]. Depending on

the process for obtaining biodiesel and the diet that the BSFL have had, the biodiesel yield ranges from 90–98%.

Waste Cooking Oil as a Feedstock for Biodiesel Production

Since 2014, the main exporting countries of WCO to the EU have been increasing, being mainly China, Indonesia, Malaysia, Russia, the United States, and Saudi Arabia [64]. In 2019, WCO represented the second-most important feedstock, which translates to 21% of the total feedstock for biodiesel production. In the European Union, the greatest biodiesel producers were Germany, the Netherlands, Portugal, the United Kingdom, Spain, and Austria, representing 90% of the use of WCO. However, in 2020, WCO collection decreased during the COVID-19 pandemic as many countries in the EU closed or restricted restaurant services [65].

According to Claeys et al. [66], almost one fifth of all European biofuels are made from WCO, which has also seen the highest growth compared to any other biomass-based diesel raw materials in Europe and North America in recent years. Globally, 6.6 million tons of WCO biofuel were consumed in 2021, representing 5% of the total biofuels market [66]. The size of the global WCO market was \$6.1 billion in 2022 and is projected to attain \$8.9 billion by 2028, registering a 6.3% compound annual growth rate (CAGR) from 2023 to 2028 [67].

Due to the aforementioned, used cooking oil represents a viable alternative to satisfy the demand for biodiesel, in addition to contributing to the proper handling and disposal of this residue. This process contributes to the global politics of establishing a circular economy; furthermore, because of the use of residue as a raw material, the environmental impact categories are positively affected.

However, it is imperative to take into consideration the quality of the WCO since the quality of the biodiesel obtained depends on it. During the frying process, oils are heated to temperatures above 100 °C and can be used repeatedly, which leads to a degradation of the quality of the oil because, during this process, thermal, hydrolytic, oxidative reactions, polymerization and cracking [68,69], chemically modify the original oil. These changes alter the properties of the WCO, presenting a higher content of free fatty acids (FFA), which may affect the transesterification reaction, favoring the formation of soaps (saponification) when basic catalysts are used. Other properties that are affected by the frying process are viscosity, change in surface tension, flash point, color, and moisture content [69,70]. For this reason, it is compulsory to conduct an adequate characterization of the residual cooking oil to guarantee that the biodiesel produced meets quality standards and contributes to the achievement of the sustainable development goals. Tables 1 and 2 show the fatty acid composition and the properties of several WCO samples, respectively. As can be observed, the composition and properties hinge on the type of oil and the handling it has had.

Table 1. Fatty acid composition of WCO.

Type of Fatty Acid	% [71] ^a	% [72] ^b	% [72] ^c	% [73] ^d	% [74] ^e	% [75] ^f	% [76] ^g	% [77] ^h
lauric (C12:0)	0.03	-	-	-	-	-	-	-
myristic (C14:0)	0.16	-	-	0.77	-	1.00	-	-
palmitic (C16:0)	12.03	0.36	5.98	31.88	11.00	39.00	8.48	18.14
palmitoleic (C16:1)	0.17	-	-	-	-	-	-	-
margaric (C17:0)	0.12	-	-	-	-	-	-	-
stearic (C18:0)	4.40	-	-	6.45	4.00	4.50	2.73	4.73
oleic (C18:1)	23.58	0.8	2.74	41.04	24.00	44.60	66.79	38.86
linoleic (C18:2)	52.48	0.10	33.89	17.98	54.00	10.90	20.14	36.45
arachidic (C20:0)	0.33	-	-	-	-	-	1.86	-
linolenic (C18:3)	6.65	-	-	0.43	7.00	-	-	1.82

Table 1. Cont.

Type of Fatty Acid	% [71] ^a	% [72] ^b	% [72] ^c	% [73] ^d	% [74] ^e	% [75] ^f	% [76] ^g	% [77] ^h
erucid (C22:1)	-	0.26	-	-	-	-	-	-
caprylic(C8:0)	-	-	-	-	-	-	-	-
undecylic (C11:0)	-	-	0.52	-	-	-	-	-
Others	-	0.20	-	-	-	-	-	-

^a Waste oil from local food industry in Toluca, Mexico. ^b Waste sunflower oil from restaurants (fish and chips) in Durban, South Africa. ^c Waste sunfoil from restaurants (chips) in Durban, South Africa. ^d Waste oil from local restaurants in Bushehr, Iran. ^e Waste oil from canteen of Malaviya National Institute of Technology, Jaipur India. ^f Waste oil from local source-UTP Cafeteria in Seri Iskandar, Perak. ^g Waste oil from local restaurant in Mérida, Yucatán. México. ^h Waste oil from a restaurant in Malaysia.

Table 2. Properties of waste cooking oil samples.

WCO Properties	[68] ^a	[72] ^b	[72] ^c	[74] ^d	[75] ^e	[76] ^f	[78] ^g	[79] ^h
Acid value (mg KOH/g)	0.31	2.29	1.44	1.2	2.04	7.06	2.8	2.7
Viscosity at 40 °C (mm ² /s)	49.40	31.38	35.23	54.00	51.04	42.98	13.45	-
Water content (wt%)	0.14	0.36	5.98	-	0.12	0.04	0.09	0.3

^a Waste oil from a restaurant in Toluca, Mexico. ^b Waste sunflower oil from restaurants (fish and chips) from Durban, South Africa. ^c Waste sunfoil from restaurants (chips) from Durban, South Africa. ^d Waste oil from canteen of Malaviya National Institute of Technology, Jaipur India. ^e Waste oil from local source-UTP Cafeteria, Seri Iskandar, Perak. ^f Waste oil from local restaurant in Mérida, Yucatán. México. ^g Waste oil from home activities, Suez, Egypt. ^h Waste oil from fast food.

As previously mentioned, during the frying process, the properties of the oils are modified. Saturated fatty acids like stearic acid, palmitic acid, and monounsaturated fatty acids like oleic acid increase in relation to polyunsaturated fatty acids like linoleic acid [72,80]. This is important because biodiesel obtained from feedstocks with a high content of saturated or monounsaturated fatty acids has superior resistance to oxidation. The oxidation rates for fatty acids (C18) are: linolenic > linoleic > oleic. A high oxidation rate can cause damage to fuel pumps and injectors [81].

This work will discuss biodiesel synthesis methods, emphasizing esterification and transesterification; the different types of catalysts that can be used in biofuel synthesis will also be discussed, as well as some of their advantages and disadvantages. The present work seeks to highlight the advances in the study of the development of bifunctional catalysts and the challenges for sustainable biodiesel production from waste cooking oil (WCO), although some results with other raw materials like refined oil and oil from microalgae are presented to highlight the relevance of those results obtained with WCO.

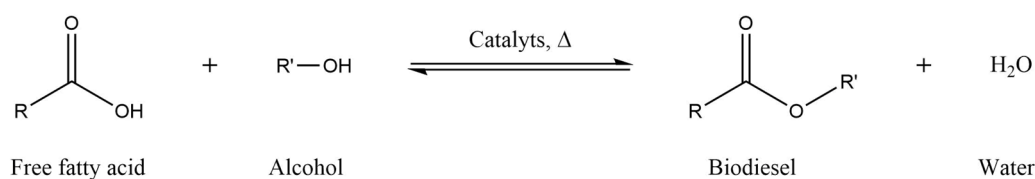
2. Biodiesel Preparation Methods and Strategies

2.1. Pyrolysis

Pyrolysis, otherwise called thermal cracking, is a process that consists of decomposing organic matter by heating it at high temperatures in an atmosphere devoid of air or oxygen [82]. The resulting properties of the fuel obtained by this method are very similar to those of petroleum diesel; however, the equipment used for thermal cracking is expensive and releases gases into the environment, eliminating the environmental advantage of biodiesel [83].

2.2. Esterification

This reaction is generally used as the pretreatment of lipid feedstocks with high FFA contents, such as WCO. As you can see in Scheme 1, the esterification reaction involves the production of fatty acid monoalkyl esters (FAMES) from the reaction of FFAs with alcohol and the use of a catalyst [4].

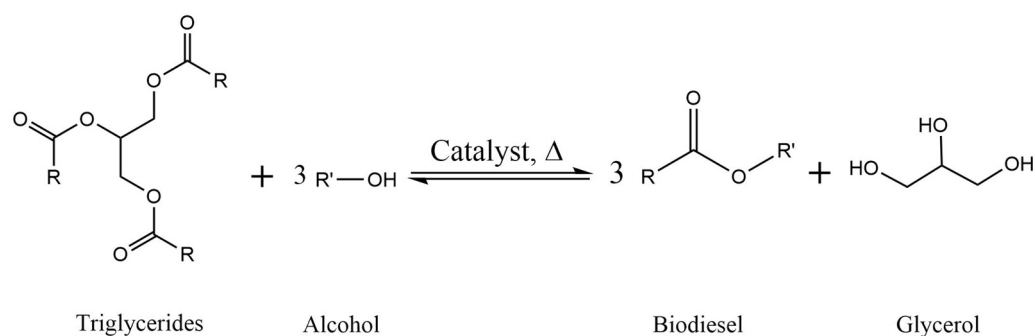


Scheme 1. Esterification reaction of free fatty acid from WCO.

Commonly, this reaction is favored by acid catalysis, with sulfuric acid (H_2SO_4) being the most widely used catalyst [4].

2.3. Transesterification

This is the process that has been used the most for biodiesel production on an industrial scale because only three raw materials are required to produce it: oil, alcohol, and a catalyst [32,84]. Scheme 2 shows this reaction, which consists of the transformation of the triglycerides contained in oils, an alcohol, and a catalyst to obtain fatty acid monoalkyl esters (FAMES) [4]. This procedure has proven to be capable of producing good-quality biodiesel, depending on the lipid feedstock used. The transesterification reaction requires the use of triglycerides, which results in the need for a lipid raw material with a high degree of purity, mainly due to the high sensitivity of most catalysts to the presence of FFA in percentages greater than 2% [32,84]. The transesterification reaction consists of three steps: the conversion of triglycerides (TG) into diglycerides (DG), these into monoglycerides (MG), and finally obtaining glycerol (G).



Scheme 2. Transesterification reaction of triglycerides.

To obtain quality biodiesel from a lipid raw material with a high free fatty acid content (>2% by weight), it is suggested to carry out a pretreatment process, such as esterification, thus ensuring the quality of the final product [85]. However, this double process implies a meaningful increase in the production cost of biodiesel.

Direct Transesterification

The direct (or in situ) transesterification reaction occurs when the extraction and transesterification of lipids from the biomass are carried out in the same step. This reaction is further enhanced when a co-solvent is used [62,86–88]. The co-solvent increases the solubility of alcohol in lipids. Originally, in direct transesterification, methanol was used as a reagent and solvent at the same time, which implied an excess of methanol that caused the activity of the catalyst in the reaction to decrease as well as its effect as a disruptor agent of the cell wall, so the yield of biodiesel also decreased [44,89]. Generally, organic solvents such as chloroform, n-hexane, n-pentane, acetone, diethyl ether, isopropanol, and petroleum ether have been used in direct transesterification [46,62,89], with the aim of reducing the amounts of methanol and avoiding the aforementioned problems. However these types of chemical reagents are toxic and can cause serious environmental problems [44,90]. For this reason, several studies have reported the use of more environmentally friendly

co-solvents such as 2-methyltetrahydrofuran (2-MeTHF), cyclopentyl methyl ether (CPME), and 1,8-Diazabicyclo [5.4.0]undec-7-ene (DBU) solvents [91–93].

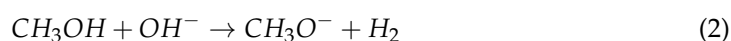
2.4. Electrolysis

The electrolysis method allows the use of feedstocks with a high content of FFA and water [94–96]. Some of the advantages of electrolysis are that no pretreatment is required to decrease free fatty acids and lipid moisture [97]. In addition, methoxide ions keep forming rapidly in the electrolysis cell [94]. At the cathode, hydroxide ions are obtained from the electrolysis of water molecules (Equation (1)), which then react with the methanol molecules to produce methoxide ions (Equation (2)). OH^- ions are formed at the cathode, while H^+ ions form at the anode (Equation (3)) [96,98], which ensures that esterification and transesterification can be carried out in the same electrolytic cell [99]. By adding NaCl to the mixture (Equation (4)) [96], the reaction rate rises due to the increase in conductivity [100]. The transesterification reaction requires the presence of methoxide ions that attack the carbon of the carbonyl group to obtain methyl esters [101,102].

Cathodic Reaction:



Proton Transfer Reaction:



Anodic Reaction:



The biodiesel produced by any of the aforesaid methods, depending on purity, can be used directly in the engines or in blends. Blending or dilution consists of mixing biodiesel or vegetable oil with diesel. This strategy aims to reduce the utilization of fossil fuels in addition to the decrease in the viscosity of the mixture, which improves the efficiency of compression engines [82,103]. Fossil fuel dilutions are made with some additives (biodiesel, animal fat, vegetable oil, bioethanol, etc.). Some of the most common blends are B10 (90% additive and 10% diesel) and B20 (80% additive and 20% diesel) [82]. In this context, biodiesel is known as a bioadditive. This process, however, presents important disadvantages, such as the generation of gums when oils with high FFA content are used and the generation of carbon deposits inside the tanks and engines. In addition, for a diesel engine to be able to use vegetable oils, significant changes in the materials of the pipes and injectors need to be made [82]. To avoid these problems, some authors suggest mixing the oils before carrying out the transesterification reaction, which would not only solve the problem of insufficient raw materials but also improve the quality of the biofuel [104].

3. Catalysis in the Production of Biodiesel

It is well known that the transesterification reaction occurs at slow reaction rates, and for biodiesel production to become sustainable, it is necessary to reduce reaction times and reaction temperatures, for which the use of catalysts is required [105,106]. The reduction of these two variables implies lower energy consumption and therefore lower environmental impacts [107] and costs.

The catalysis studied in biodiesel production has three main categories: (1) homogeneous catalysis, (2) enzymatic catalysis, and (3) heterogeneous catalysis. Next, each of the categories and their subdivisions will be discussed.

3.1. Homogeneous Catalysis

In homogeneous catalysis, reactants and the catalyst are in the same phase, which is generally liquid. These catalysts are frequently used in industrial-scale processes because they present higher reaction rates than heterogeneous catalysis.

However, they have some disadvantages, for example, the impossibility of recovering the catalyst when the reaction is finished, in addition to the need for a purification process where large amounts of residual water are generated [108]. Figure 4 presents the biodiesel production process using homogeneous catalysts.

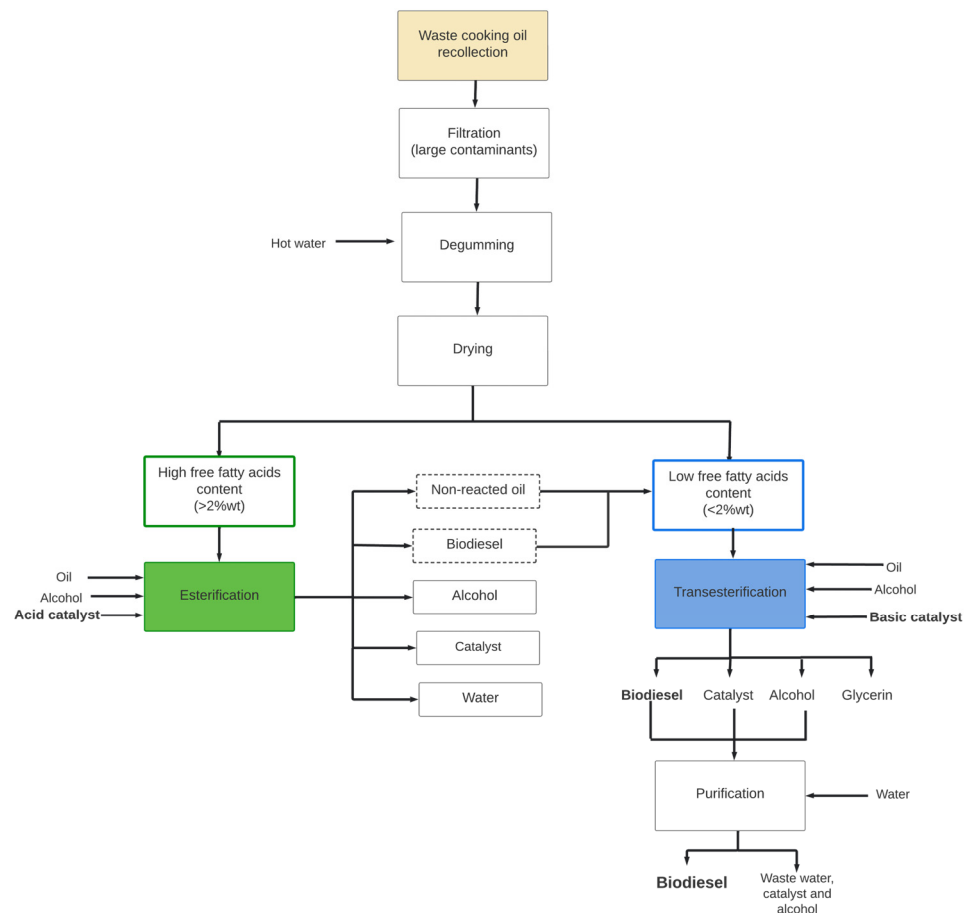


Figure 4. Schematic of biodiesel production process using homogeneous catalysts.

Next, the subdivisions of this type of catalysis are discussed: (1) Homogeneous acid catalysis and (2) Homogeneous basic catalysis.

3.1.1. Homogeneous Acid Catalysis

This type of catalyst admits the use of unrefined lipid raw materials because they present a high tolerance to FFA. Though this process is slower than basic catalysis.

Next, Table 3 shows some examples of homogeneous acid catalysts, in addition to the type of feedstock used for the production of biodiesel, the reaction conditions used, and the %FAMEs obtained. It can be seen in Table 3 that the preferred acids to be used as catalysts are hydrochloric, sulfuric, and phosphoric acids.

Despite the fact that this kind of catalyst has a high tolerance to the FFA content within the lipid raw material, its use has not been extended at an industrial level, mainly because of its corrosion hazards, so its use poses environmental and safety risks [22]. In addition, to remove this type of catalyst from the final biodiesel, it is necessary to carry out multiple washes with water, which implies a considerable environmental impact.

The reaction temperatures required in this type of catalysis can range from 60 °C to 200 °C, depending on the process used for biodiesel production. Conventional batch processes generally require higher temperatures and longer reaction times. However, some research papers suggest the use of microwave-assisted reaction systems or ultrasound to reduce these disadvantages [122].

Table 3. Examples of homogeneous acid catalysts used in biodiesel production.

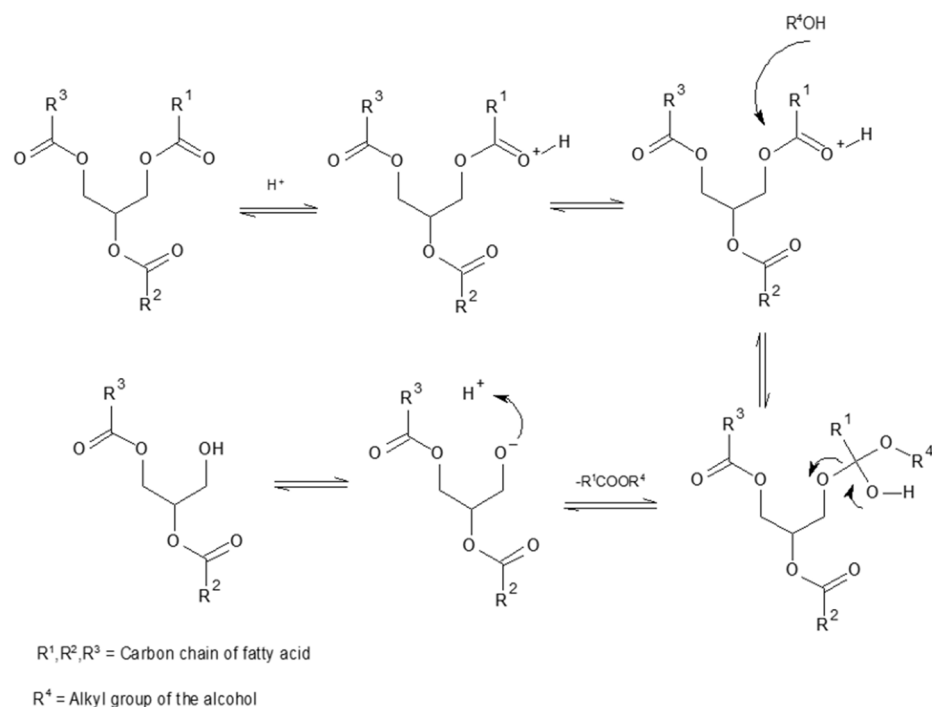
Catalyst	Oil	Reaction Conditions	Methyl Esters Content (%FAMES)	References
HCl	Microalgae	T = 76.67 °C; 0.54 M; M:o = 7.92:1; t = 1.73 h	98.19	[109]
H ₂ SO ₄	Chrysophyllum albidum	T = 65 °C; CC% 2%wt; M:o = 12:1; t = 0.33 h	98.15	[2]
Orthophosphoric acid H ₃ PO ₄	<i>Calophyllum inophyllum</i> L.	T = 60 °C; CC% 0.8%wt; M:o = 9:1; t = 1.25 h	97.14	[110]
H ₂ SO ₄	S. obliquus lipids	T = 60 °C; CC% 10%wt; M:o = 30:1; t = 4 h	96.68	[111]
H ₂ SO ₄	Microalgae	T = 80 °C; CC% 3%wt; M:o = 9:1; t = 8 h	96.5	[112]
H ₂ SO ₄	WCOEsterification	T = 60 °C; CC% 5%wt; M:o = 12:1; t = 3 h	95.4	[113]
Superphosphoric acid H _(n+2) P _(n) O _(3n+1)	Palm oil	T = 70 °C; CC% 9%wt; M:o = 12:1	95	[114]
H ₂ SO ₄	Jatropha oil	T = 60 °C; CC% 4%wt; M:o = 7:1; t = 1 h	92.4	[115]
H ₂ SO ₄	Corn oil	T = 200 °C; CC% 0.2%wt; E:o = 18:1; t = 0.5 h	92	[116]
H ₂ SO ₄	Palm oil	T = 60 °C; CC% 5%wt; M:o = 9:1; t = 4.5 h	91.1	[117]
H ₂ SO ₄	Soybean oil	T = 60 °C; CC% 3.5%wt; M:o = 9:1; t = 1 h	90.6	[118]
H ₂ SO ₄	Oleic acid	T = 60 °C; CC% 5%wt; M:o = 3:1; t = 2 h	89.3	[119]
H ₂ SO ₄	WCO	T = 80 °C; CC% 1.5–3.5 %mol; M:o = 50:1; t = 4 h; 170–180 kPa pressure	97.0	[120]
HCl	Waste Coconut Oil Esterification	T = 80 °C; CC% 3%wt; M:o = 10:1; t = 1 h	90.45	[121]

T—Reaction temperature, CC—Catalyst weight, M:o—Methanol:oil molar ratio, E:o—ethanol:oil molar ratio, t—Reaction time.

In addition, as can be observed in Table 3, the percentage contents of FAMES obtained are very close to the one required by the European Union Quality Standard (UNE-EN 1403), which requires a minimum of 96.5% FAMES. It is also important to emphasize that the lipid raw materials used in this research are mainly refined oils. However, we must not forget the environmental and safety disadvantages that arise during the use and storage of these catalysts.

Generally, this type of catalysis is used in conjunction with basic catalysis, being used as a pretreatment for unrefined lipid raw materials or with a free fatty acid content greater than 1%. First, an acid esterification is performed to decrease the FFA content (<1%), and later, a basic transesterification is carried out.

The mechanism of this process is shown in Scheme 3. It begins with the protonation of the carboxylic group of the ester (this step is the most important in the catalyst-reactive relationship), followed by the action of the acid catalyst (H⁺), followed by the nucleophilic attack of the alcohol, producing a tetrahedral intermediate. This intermediate breaks down to form a diacylglyceride ion and the ester alkyl of the fatty acid; these steps continue until three fatty acid esters are formed and, subsequently, the glycerol is released.



Scheme 3. Homogeneous acid transesterification mechanism [71].

3.1.2. Homogeneous Basic Catalysis

This type of catalysis is the most extensively utilized at the industrial level, as homogeneous basic catalysts are low-cost and easy to access [106]. It is also due to their high reaction rates, which implies shorter reaction times, lower methanol:oil molar ratios than acid catalysis, and mild reaction temperatures. Furthermore, the use of this catalysis allows the elimination of corrosion problems related to the use of acid catalysts. The transesterification reaction employing basic catalysts is faster than with homogeneous acid catalysts [123,124]. The most commonly used basic catalysts are KOH, NaOH, and CH₃ONa. Some examples of these catalysts can be seen in Table 4.

Table 4. Examples of basic homogeneous catalysts used in biodiesel production.

Catalyst	Oil	Reaction Conditions	Methyl Esters Content (%FAMES)	References
NaOH	WCO	T = 62.4 °C; CC% 1.16%wt; M:o = 9.4:1; t = 0.017 h Esterification (1.56%wt FFA) Transesterification (0.35%wt FFA)	99.7	[125]
CH ₃ ONa	Refined palm oil	T = 55 °C; CC% 0.32%wt; M:o = 5.48:1; t = 0.67 h	98	[126]
KOH	Black mustard oil	T = 57.1 °C; CC% 0.4%wt; M:o = 20.39%wt; t = 0.9 h; 0.8%wt FFA	97.3	[127]
KOH	Jatropha curcas-WCO	T = 50 °C; CC% 1%wt; M:o = 6:1; t = 2 h; 1%wt FFA	97.1	[128]
CH ₃ ONa	WCO	T = 65 °C; CC% 0.75%wt; M:o = 9:1; t = 0.13 h; 2.4%wt FFA	97.1	[129]
NaOH	Black mustard oil	T = 59.5 °C; CC% 0.5%wt; M:o = 21.5%wt; t = 1 h; 0.8%wt FFA	96.9	[127]
KOH	Waste Cotton oil	T = 50 °C; CC% 0.65%wt; M:o = 7:1; t = 0.16 h	96.44 (Microwave)	[130]

Table 4. Cont.

Catalyst	Oil	Reaction Conditions	Methyl Esters Content (%FAMES)	References
KOH	WCO	T = 65 °C; CC% 1.2%wt; M:o = 6:1; t = 1 h; 1.25%wt FFA	93.2	[131]
NaOH	Mango oil	T = 60 °C; CC% 1%wt; M:o = 6:1; t = 3 h; 0.06%wt FFA	92.7	[132]
KOH	WCO	T = 60 °C; CC% 1.2%wt; M:o = 5:1; t = 2 h; 0.41%wt FFA	92	[133]
CH ₃ ONa	WCO	T = 25 °C; CC% 0.75%wt; M:o = 6:1; t = 0.05 h; <2%wt FFA	87.0	[134]
NaOH	WCO	T = 56.5 °C; CC% 0.75%wt; M:o = 12:1; t = 3.25 h; 0.92%wt FFA	82	[135]
KOH	WCO	T = 60 °C; CC% 1%wt; M:o = 6:1; t = 0.5 h; 0.93%wt FFA	94.01	[136]
CH ₃ OK	WCO	T = 60 °C; CC% 1%wt; M:o = 6:1; t = 0.5 h; 0.93%wt FFA	99	[136]
KOH	WCO	T = 60 °C; CC% 1%wt; M:o = 8:1; t = 2 h	92.5	[137]
NaOH	WCO	T = 65 °C; CC% 0.8%wt; M:o = 12:1; t = 0.033 h	98.2 (Microwave)	[138]
KOH	Soybean WCO	T = 60 °C; CC% 0.5%wt; t = 2 h	93.2	[139]

T—Reaction temperature, CC—Catalyst weight, M:o—Methanol:oil molar ratio, t—Reaction time.

By contrasting Tables 3 and 4, it can be concluded that both acid and basic catalysis allow the production of biodiesel at mild temperatures (~ 60 °C). The obtained yields with basic catalysis tend to be higher with lower reaction times. The methanol:oil ratio (M:o) is also observed to be lower under basic catalysis. This does not represent an improvement only in the process cost but also in energy consumption and therefore in environmental impacts like global warming potential (carbon footprint), which is expected to decrease [107].

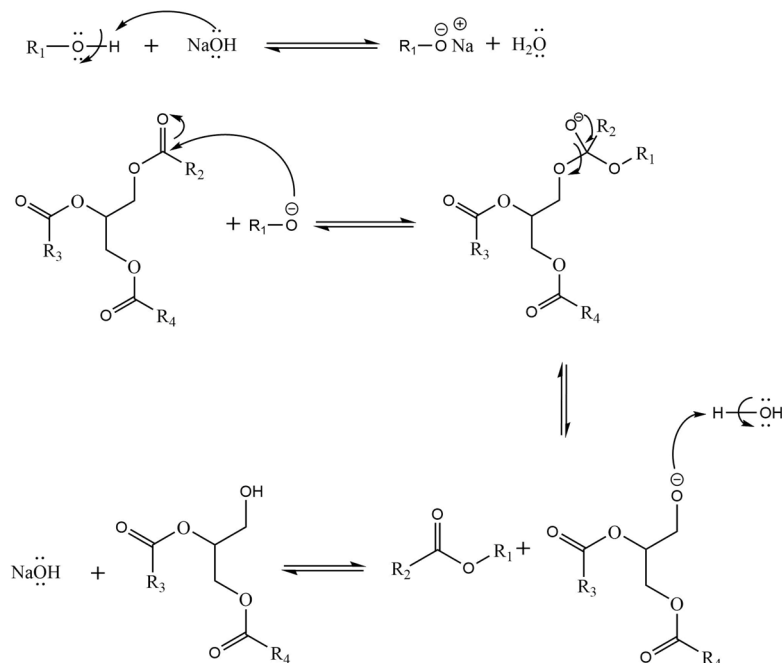
Some of the disadvantages of homogeneous basic catalysts include the need to perform washes to remove the catalyst at the end of the reaction, as well as not being able to recover the catalyst [9,140]. The most important disadvantage of this type of catalyst, however, lies in its high sensitivity to the presence of water or FFA within the lipid raw material, since this type of catalyst needs to use refined feedstocks (<1%wt) or with a content of FFA <2% weight [141–143].

Generally, to reach high-quality biodiesel from WCO, it is necessary to perform a previous acid esterification (Scheme 1). In esterification, FFA present in oils is transformed into biodiesel, which leads to a reduction of the percentage content of FFA in the lipid raw material, making it possible to carry out a basic transesterification without saponification.

Nevertheless, recent research has shown that the use of systems assisted by ultrasound allows for high-quality biodiesel production using WCO with elevated FFA content and basic catalysts. This is due to the fact that ultrasound waves influence heating at the molecular level, allowing internal heating to be distributed evenly [130].

Table 4 shows that percentage contents of FAMES higher than the minimum required by the UNE-EN 1403 standard (96.5% FAMES minimum) can be obtained. However, in most cases, the acidity values reported for the used cooking oils were low (<2%wt), which is considered the limit for obtaining high-quality biodiesel. It can also be observed that in the case of feedstocks with a FFA with a higher content than refined oils (>1%wt), it is necessary to conduct a prior esterification to obtain biodiesel with a standard quality. The above-mentioned agrees with that reported by Hsiao and Mohadesi, who required carrying out prior esterification to obtain high-quality biodiesel.

The mechanism using basic catalysts (Scheme 4) can be accomplished by the attack of the alkoxide ion on the electrophilic carbon of the triglyceride, forming an alkyl ester (tetrahedral intermediate). Subsequently, the catalyst is deprotonated, and the proton is joined to the diglyceride anion until esters and glycerol are formed.



R_1 : Alkyl group of the alcohol

R_2, R_3 y R_4 : Carbon chain of fatty acid

Scheme 4. Homogeneous basic transesterification. Adapted with permission from [101]. Copyright 2010 Elsevier.

As mentioned above, the transesterification reaction using homogeneous catalysts involves 3 stages: the triglyceride (TG) reacts with alcohol to obtain diglycerides (DG) $TG + ROH \leftrightarrow DG + R'CO_2R$, this reacts with alcohol to form monoglycerides (MG) $DG + ROH \leftrightarrow MG + RCO_2R$ and finally, it reacts with alcohol to produce methyl esters and glycerol (GL) $MG + ROH \leftrightarrow GL + RCO_2R$.

For the determination of the kinetics of transesterification, DG and MG can be omitted since methyl esters are the final product of this reaction, allowing us to use a simple mathematical model that expresses total conversion as a single step. With respect to the reversible reactions, they can also be depreciated due to the excess of methanol in the product. Furthermore, in this reaction, there are no mass transfer problems, the kinetic process is chemically controlled [144], and the kinetics of transesterification can be considered irreversible pseudo-first order [145].

3.2. Enzymatic Catalysis

Recently, this type of biocatalyst has gathered the attention of researchers due to its potential to produce quality biodiesel from high FFA content feedstocks with yields close to 100% and a reduction of impurities in the final product. In addition, the formation of soap is eliminated, the enzymes have a high tolerance to water content, have minor energy consumption since the reaction is conducted at low temperatures, can be recycled when immobilized, and are easily separated and purified from the products at the end of the reaction [146]. However, it still presents limitations on an industrial scale as a result of the high cost, deactivation of the enzyme, and low reaction rate [147]. Furthermore, as can be seen in Table 5, the yields achieved are not as high as those achieved by basic catalysis (see Table 4).

Enzymatic catalysts are mainly divided into three types: extracellular lipases, intracellular lipases, and free enzymes (see Table 5) [148,149].

Table 5. Examples of enzymatic catalysts used in the biodiesel production.

Catalyst	Oil	Reaction Conditions	Methyl Esters Content (%FAMES)	References
Callera TM Trans Lipase	Soybean	T = 35 °C; CC% 1.45%wt; M:o = 4.5:1; t = 24 h	96.9	[150]
Lipozyme (<i>Thermomyces lanuginosus</i>)	Wasted fenix oil	T = 31 °C; CC% 9.7%wt; M:o = 4.3:1; t = 6.9 h	93.8	[151]
<i>Pseudomonas cepacia</i>	Jatropha	T = 8 °C; CC% 5%wt; M:o = 4:1; t = 50 h	98.0	[152]
Rice bran lipase	Rice bran oil	T = 40 °C; CC%; M:o = 6:1; t = 288 h	83.4	[153]
Novozym 435	Waste Fish oil	T = 35 °C; CC% 50%wt; E:o = 35.45:1; t = 8 h	82.91	[154]
Novozym® 435	WCO	T = 50 °C; CC% 40%wt; M:o = 6:1; t = 14 h	72.0	[155]
<i>Chromobacterium viscosum</i>	Jatropha curcas	T = 30 °C; CC% 10%wt; M:o = 4:1; t = 4 h	51–65	[156]
<i>B. stearothermophilus</i> and <i>S. aureus</i> lipases (Immobilized)	WCO	T = 55 °C; CC% 1%wt (50% of each lipase); M:o = 6:1; t = 24 h	97.66	[157]
<i>Oreochromis niloticus</i> lipase	WCO	T = 45 °C; CC% 30 kUnit; M:o = 4:1; t = 28 h	96.5	[158]
<i>Candida rugosa</i> and <i>Rhizomucor miehei</i> lipases (Immobilized)	WCO	T = 45 °C; CC% 1%wt (50% of each lipase); M:o = 6:1; t = 24 h	96.5	[159]
<i>Burkholderia cepacia</i> lipase (Immobilized)	WCO	T = 35 °C; CC% 25%wt; M:o = 6:1; t = 25 h	85.2	[160]
<i>Candida</i> sp. lipase	WCO	T = 40 °C; CC% 1%wt; t = 12 h	80	[161]
Lipase from porcine pancreas	WCO	T = 40 °C; CC% 7.5%wt; M:o = 9:1; t = 10 h	92.33	[162]

T—Reaction temperature, CC—Catalyst weight, M:o—Methanol:oil molar ratio, E:o—ethanol:oil molar ratio, t—Reaction time.

In the case of extracellular lipases, they offer great selectivity, preventing the generation of by-products such as soaps and baits. However, they are expensive to produce due to their complicated synthesis process, they can be easily deactivated by the wrong alcohol selection, their reaction times are higher than basic catalysis (3–288 h), and enzymatic catalysts have a high affinity for glycerin, which implies a drawback in their separation [22]. Intracellular lipases can be used directly as catalysts, which makes them cheaper since expensive processes such as extraction and purification are omitted [22]. In 2021, Acherki et al. [163] reported the use of the Eversa® Transform 2.0 liquid enzyme with jatropha oil and butanol, obtaining 83% biodiesel conversion (T = 42 °C, 9.79%wt catalyst). One of the advantages of this enzyme is its low cost, the handling of oils with high FFA contents, and the fact that no purification steps are required. A Finally, it is known that free enzymes or liquid enzymes offer better miscibility and mass transfer, although they cannot be reutilized.

Figure 5 represents a process scheme using enzymes. When using WCO, it is necessary to carry out a filtration to separate the solid particles. Later, it is recommended to dry the oil in order to eliminate excess water since the presence of a lot of water may be undesirable, without forgetting that water is necessary to maintain the catalytic activity of enzymes [101].

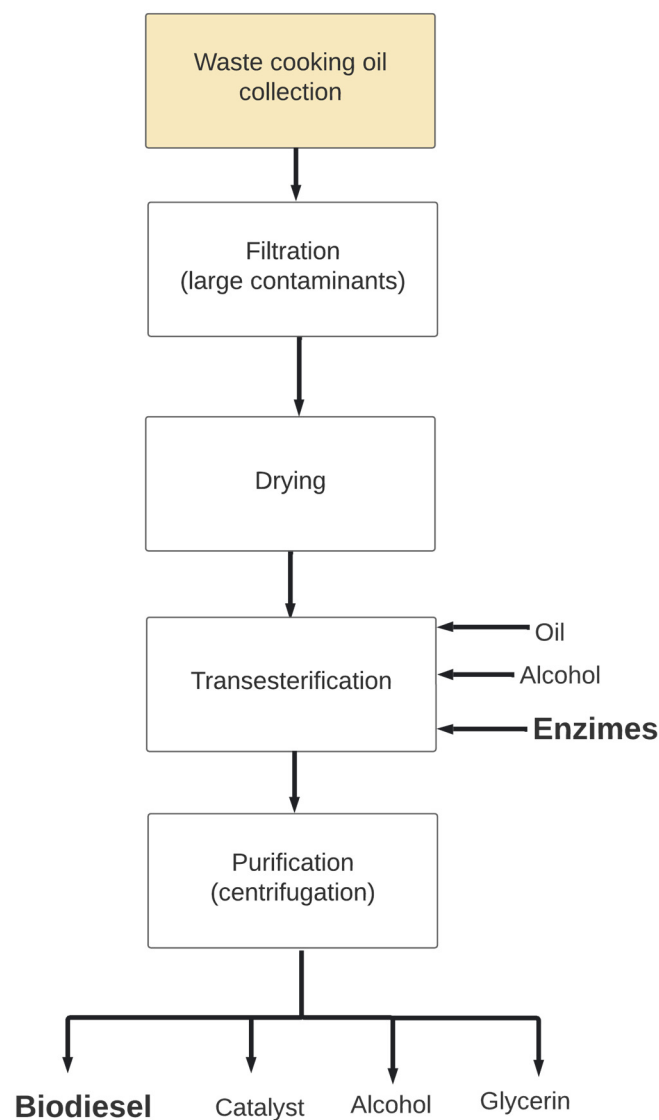


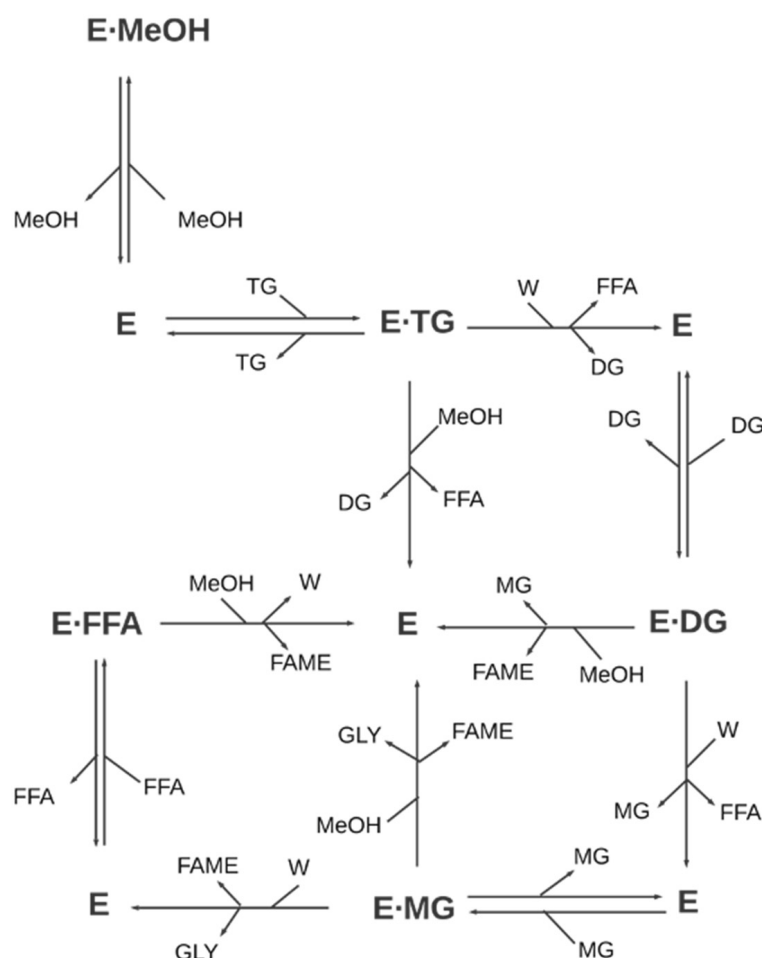
Figure 5. Schematic of biodiesel production process using enzymatic catalysts.

The transesterification mechanism using an enzymatic catalyst involves two steps: 1. Hydrolysis of the esters to produce FFA; 2. Esterification to obtain esters (FAME). Furthermore, it is assumed that the transesterification reactions with alcohol (methanol) are simultaneous with the hydrolysis reactions [164–166]. Scheme 5 shows the mechanism proposed by Andrade et al. [165] for enzymatic transesterification of castor oil, where: TG = triglycerides, DG = diglycerides, MG = monoglycerides, FAME = fatty acid methyl esters, FFA = free fatty acids, MeOH = methanol, W = water, GLY = glycerol, and E = enzyme.

3.3. Heterogeneous Catalysis

This kind of catalysis occurs when the reagents and the catalyst are in different phases, the most common being that the catalyst is in solid form. Heterogeneous catalysis arose from the need to reduce production costs implied by homogeneous catalysts.

These catalysts caught the attention of researchers because they can be separated and reused many times before losing their catalytic activity, which allows cost reduction in biodiesel production by reducing post-treatment complexity [167]. This is the reason heterogeneous catalysis is considered the most effective way to produce biodiesel from lipid raw materials with high FFA content [167].



Scheme 5. Mechanism for the enzymatic transesterification of castor oil. Adapted with permission [165]. Copyright 2017 American Chemical Society.

However, the main objection to the use of these catalysts lies in the contamination of the final product derived from the leaching of the active sites and in the mass transfer problems derived from the solid-liquid biphasic reaction [22]. Figure 6 shows the processes of heterogeneous catalysis.

Next, the subdivisions of this type of catalysis are discussed: (1) Heterogeneous acid catalysis and (2) Heterogeneous basic catalysis.

3.3.1. Heterogeneous Acid Catalysis

This type of catalyst is effective for biodiesel production from lipids with high FFA content [167]. This type of catalyst can simultaneously perform the esterification of the FFA and the transesterification of the triglycerides present in the lipid raw materials since these catalysts have Brönsted acid sites and Lewis acid sites capable of promoting esterification reactions and ester exchange reactions [168,169]. Although transesterification can also be conducted on acid sites, this proceeds at a very low rate [170].

The most commonly used acid heterogeneous catalysts are mixed metal oxides, heteropoly acid derivatives, ion exchange resins, sulfonated carbon-based catalysts, and sulfated catalysts [170,171]. Table 6 below shows some examples of heterogeneous acid catalysts, as well as the raw material from which biodiesel was produced, the reaction conditions used, and the %FAMEs obtained.

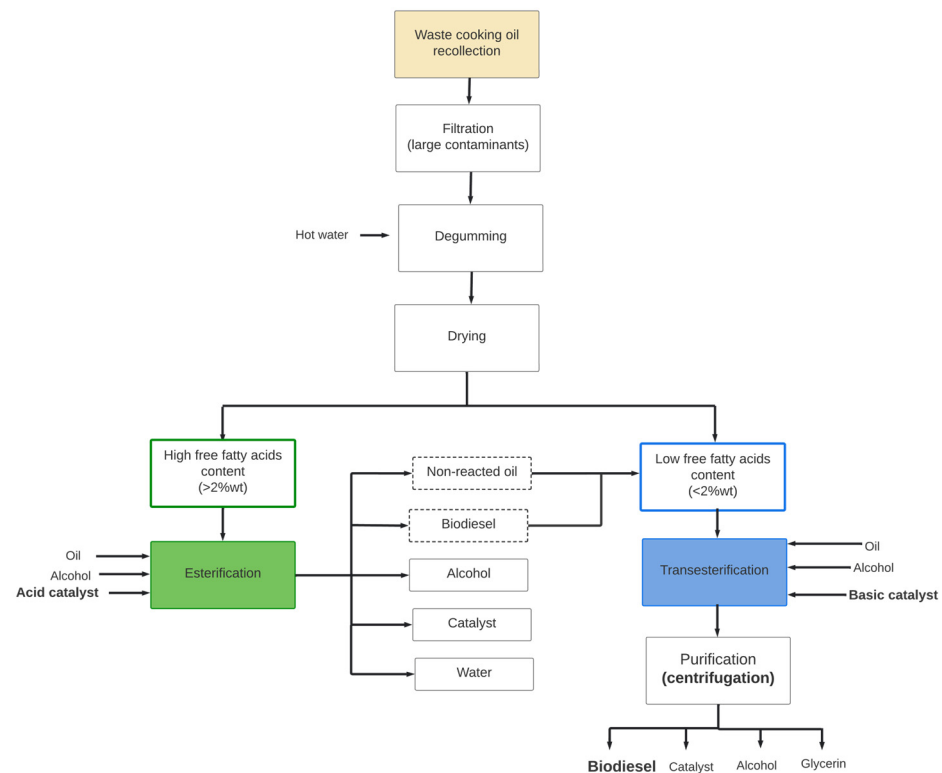


Figure 6. Schematic of biodiesel production process employing heterogeneous catalysts.

Table 6. Examples of heterogeneous acid catalysts used for biodiesel production.

Catalyst	Oil	Reaction Conditions	Methyl Esters Content (%FAMES)	References
4-BDS	Palm oil	T = 110 °C; CC% 20%wt; M:o = 30:1; t = 7 h	98.1	[172]
(ZS/Si) Zinc stearate	WCO	T = 200 °C; CC% = 3%wt; M:o = 18:1; t = 10 h; 15%wt FFA	98	[173]
Sulfonated hypercrosslinked exchange resin	WCO	T = 60 °C; CC% 5%wt; M:o = 12:1; t = 2 h	97	[174]
C-SO ₃ H	Oleic acid	T = 80 °C; CC% 8%wt; M:o = 21:1; t = 1 h	96.77	[175]
TPA/Bentonite	WCO	T = 100 °C; CC% 0.7 g; M:o = 10:1; t = 4.5 h, 11.2%wt FFA	96	[37]
SO ₄ /Fe-Al-TiO ₂	WCO	T = 90 °C; CC% = 3%wt; M:o = 10:1; t = 2.5 h; 2%wt FFA	96	[176]
S-TiO ₂ /SBA-15	WCO	T = 200 °C; CC% = 1%wt; M:o = 15:1; t = 0.5 h; 2.92%wt FFA	94.96	[177]
WO ₃ /ZrO ₂	S. obliquus lipids	T = 100 °C; CC% = 15%wt; M:o = 12:1; t = 3 h	94.58	[111]
Carbon acid catalyst	WCO Prior esterification	T = 100 °C; CC% 5%wt; M:o = 22:1; t = 3 h	92.3	[178]
H ₂ SO ₄ /Bamboo ashes	Oleic acid	T = 65 °C; CC% 0.3 g; Metanol: 2.7 g; t = 8 h	92.1	[84]
Carbon acid catalyst	Chicken fat	T = 200 °C; CC% 3%wt; M:o = 9:1; t = 6 h; 50%wt FFA	90.8	[178]
RS-SO ₃ H	WCO	T = 70 °C; CC% 5%wt; M:o = 18:1; t = 1 h	90.38	[78]

Table 6. Cont.

Catalyst	Oil	Reaction Conditions	Methyl Esters Content (%FAMES)	References
Bi ₂ SiO ₅	Oleic acid	T = 70 °C; CC% 10%wt; M:o = 20:1; t = 6 h; 2.8%wt FFA	90.0	[179]
Xylose derived sulfonated carbon catalyst	WCO	T = 80 °C; CC% 0.3 g; M:o = 120:1; t = 2 h	89.6	[180]
SO ₄ ²⁻ /ZrO ₂	WCO	T = 150 °C; CC% 10%wt; M:o = 10:1; t = 4 h; 2.7%wt FFA	86	[181]
MgF _x (OH) _{2-x}	WCO	T = 150 °C; CC% 5%wt; M:o = 30:1; t = 5 h; 13%wt FFA	75.29	[182]

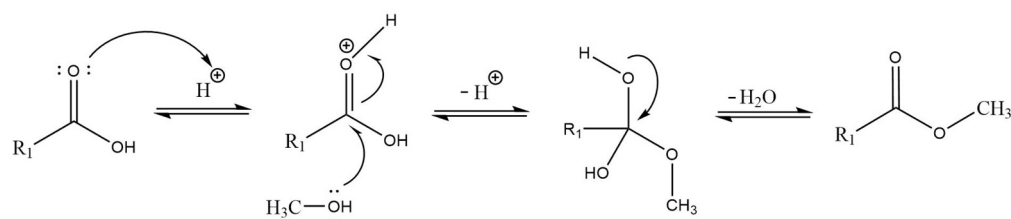
T—Reaction temperature, CC—Catalyst weight, M:o—Methanol:oil molar ratio, t—Reaction time.

As can be seen in Table 6, these catalysts are capable of producing biodiesel with a high amount of FAMES from lipid raw materials whose FFA content is higher than refined oils (such as animal fats and WCO). For conventional batch systems, this type of catalyst requires longer reaction times as well as higher reaction temperatures (60–220 °C) [101,183]. Some of their main advantages are found in their ease of recovery, their ability to be reused, the reduction of their corrosive nature (compared with homogeneous), and the possibility of esterification and transesterification taking place simultaneously.

The works cited in Table 6 show that most of the WCOs used have an FFA content greater than 2%wt, which means those lipidic raw materials are considered low-quality oils. Despite the above, most of the catalysts manage to obtain >90% FAMES at the end of the reaction; nonetheless, relatively long reaction times (2–7 h) were required to reach these percentages. In addition, many of these catalysts require MeOH:oil molar ratios greater than 10:1. This leads to mass transfer problems (catalyst separation problems), increased production costs [68,105] and a negative effect on environmental impact categories such as global warming potential and photochemical oxidation [107].

The foregoing has motivated the search for catalysts that are capable of reducing such reaction conditions, in addition to seeking to reduce the environmental impact derived from the corrosive nature of this type of catalyst.

The mechanism of the esterification reaction (Scheme 6) by acid catalysts, begins with the donation of an H⁺ to the carboxylic acid, later it undergoes the nucleophilic attack by the hydroxyl group of the methanol to continue with the reaction until the water is eliminated [184].



Scheme 6. Mechanism of acid catalyzed esterification of FFA [71].

According to several experimental studies using heterogeneous acid catalysts with high FFA content oils, the experimental data approximates a pseudohomogeneous irreversible reaction [168,185]; first order for both FFA content and methanol concentration. The above agrees with Zeng et al. [184], who found that when heterogeneous acid catalysts are used, the esterification reaction follows a second-order mechanism.

3.3.2. Heterogeneous Basic Catalysis

This type of catalyst has been extensively investigated as it shows attractive catalytic activities under mild reaction conditions as well as easy recovery and the ability to be reused [106,186].

Nonetheless, using these catalysts during the transesterification reaction of oils with a high FFA content might result in a reduction in the quality of the biodiesel obtained because the saponification reaction is favored [167]. This not only lowers the biodiesel yield but also implies two main consequences: it makes it harder to separate glycerol from biodiesel in addition to consuming the added catalyst [187]. Furthermore, this type of catalyst is highly sensitive to poisoning when exposed to air (absorbing CO₂ and humidity) [186].

The most frequently used basic heterogeneous catalysts are zeolites, transition metal oxides, hydrotalcite-based catalysts, mixed metal oxides, and alkaline earth metal oxides [170]. These catalysts have good reaction kinetics and adsorption capacities and are widely available in nature, which makes them low-cost [187].

Table 7 shows some examples of basic heterogeneous catalysts, as well as the sort of raw material from which biodiesel was produced, the reaction conditions used, and the %FAMES obtained at the end of the reaction.

Table 7. Examples of basic heterogeneous catalysts used in biodiesel production.

Catalyst	Type of Oil Used	Reaction Conditions	Yield (%FAMES)	References
K ₂ CO ₃ /Sepiolite	Turnip oil	T = 70 °C; CC% 2%wt; E:o = 12:1; t = 4 h	99.9	[188]
Na ₂ ZrO ₃	<i>Jatropha curcas</i> L. oil	T = 65 °C; CC% 5%wt; M:o = 65:1; t = 8 h	99.9	[189]
CaO	WCO	T = 80 °C; CC% 2%wt; M:o = 9:1; t = 0.17 h	98.7	[190]
CaO	WCO	T = 55 °C; CC% 6%wt; M:o = 8.3:1; t = 0.67 h	98.62	[191]
CaO	(Mixture of blended oil)	T = 61.61 °C; CC% 4.5%wt; M:o = 8:1; t = 1.08 h	98.0	[192]
KOH/limestone	WCO	T = 65 °C; CC% 5.36%wt; M:o = 12.26:1; t = 0.97 h	97.15	[193]
MgO-NaOH	WCO	T = 50 °C; CC% 3%wt; M:o = 6:1; t = 6 h	97	[194]
CH ₃ ONa/Bentonita	Sunflower oil	T = 55 °C; CC% 2%wt; M:o = 12:1; t = 1.17 h	94.33	[195]
MgO	WCO	T = 65 °C; CC% 2%wt; M:o = 24:1; t = 1 h	93.3	[196]
SrO	WCO	T = 65 °C; CC% 3%wt; M:o = 9:1; t = 0.07 h	93	[197]
CaO	Waste Cotton Oil	T = 50 °C; CC% 1.3%wt; M:o = 9.6:1; t = 0.16 h	89.94	[130]
α-Fe ₂ O ₃ -Al ₂ O ₃	WCO	T = 65 °C; CC% 1%wt; M:o = 15:1; t = 3 h	87.78	[198]
KOH/Diatomita	Palm oil	T = 75 °C; CC% 5%wt; M:o = 9:1; t = 2 h	84.56	[199]

T—Reaction temperature, CC—Catalyst weight, M:o—Methanol:oil molar ratio, t—Reaction time.

Nevertheless, a major disadvantage of these catalysts involves the decrease of their catalytic activity derived from the adhesion of reaction by-products, which results in the deactivation of the catalytic materials [200].

The results observed in Table 7 indicate that when there is a raw material with a greater FFA content than a refined oil (such as non-edible oils and WCO), they require higher temperatures of reaction (to increase the collisions among molecules and to improve the reaction rate), higher methanol:oil molar ratios, and a higher catalyst loading to obtain biodiesel with a higher FAME content.

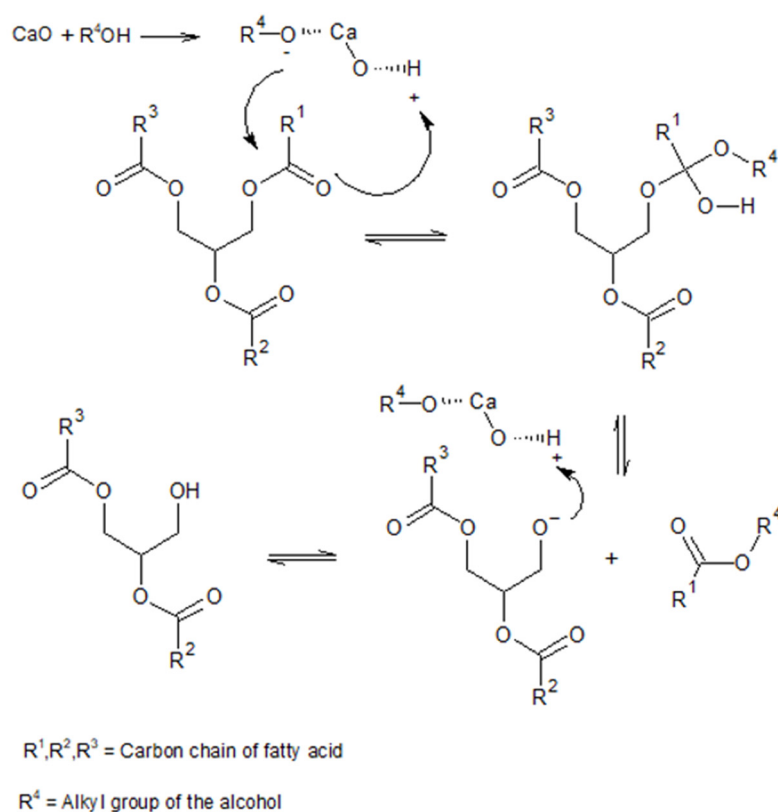
The aforementioned reaction conditions are linked to the basicity of the material used and possible mass transfer problems. As an example of the above, it is well known that using alcohol in excess will result in mass transfer problems [201].

Most of the works cited in Table 6 use WCO with FFA contents lower than 2%wt, which allows this type of catalyst to obtain 85–99.9% FAMES after the transesterification reaction. Though we must remember that international standards require a 96.5% FAMES content to consider that the biodiesel has quality enough for its usage in engines.

On the other hand, it is essential to remember that basic catalysts have a high sensitivity to the FFA content, which is why, in the case of a low-quality raw material (>2%wt FFA), it is necessary to reduce said content of FFA, for which an acid esterification reaction is usually used as a pretreatment.

In addition, it is necessary to carry out process optimization. Some investigations have suggested the use of microwave-assisted or ultrasonic systems to overcome the mass transfer problems that occur in conventionally stirred processes, thus reducing reaction times [170].

The mechanism of transesterification using a basic heterogeneous catalyst (Scheme 7) begins with the formation of the methoxide anion, followed by the attack of the carbon of the carbonyl group by the methoxide anion, forming an intermediate, to obtain the ester and a diglyceride anion, which are compounds more stable. Subsequently, the attack of the cation by the diglyceride anion occurs, forming the diglyceride regenerating the catalyst (CaO). The mechanism is repeated until the methyl esters (biodiesel) and glycerol are formed.



Scheme 7. Heterogeneous base-catalyzed mechanism for the transesterification of triglycerides [71].

Nain et al. [202] analyzed two kinetic models based on Langmuir-Hinshelwood-Hougen-Watson (LHHW) and Eley-Rideal (ER) mechanisms of the transesterification reaction of canola oil and quicklime (CaO) as a heterogeneous catalyst. They concluded that the best model, according to their experimental data, is the Eley-Rideal mechanism and that the controlling step is the surface reaction [202]. This is consistent with the ER model, especially when a highly basic catalyst is used, like CaO, BaO, or SrO [203]. Likewise, they observed that there was no adsorption of triglycerides on the surface of the catalyst, this being caused by the rapid adsorption of methanol [202].

3.3.3. Bifunctional Catalysis

As previously mentioned, biodiesel's final quality is clearly related to the content of FFA in the feedstock. In feedstocks with a low content of FFA (<2%wt), a transesterification

reaction is good enough to obtain a high-quality product. Furthermore, the high sensitivity of the transesterification reaction to the FFA content results in its saponification or in obtaining low-quality biodiesel. To avoid the above drawbacks and ensure the quality of the final product, it is necessary to carry out a process consisting of two steps, which consist of a prior esterification reaction (to transform the FFA into FAMES) and later a transesterification of the triglycerides (see Figure 7).

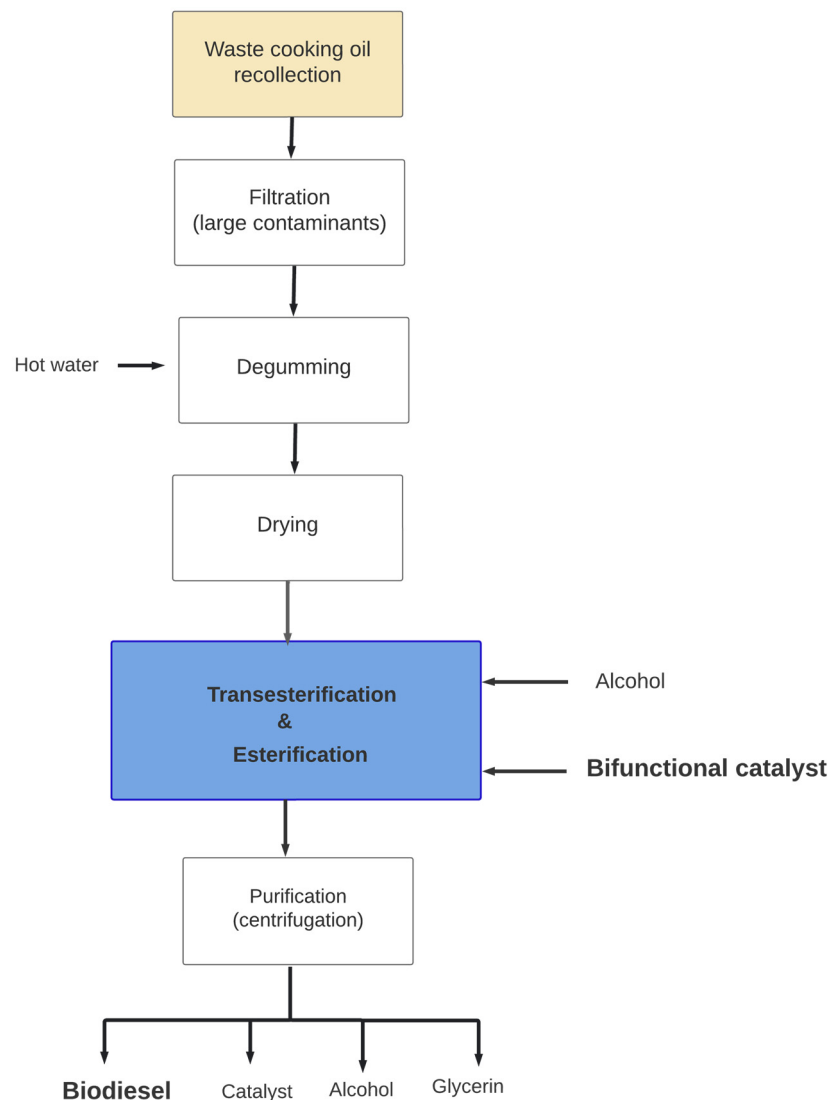


Figure 7. Schematic of biodiesel production process using bifunctional catalysts.

In the related literature, there is a broad variety of bifunctional catalysts used for biodiesel synthesis. These catalysts generally consist of mixtures of some support and metal oxides. Nevertheless, it is also possible to find bio-based catalysts [204,205]. Additionally, it is precisely these catalysts based on biomass that have gained more attention in recent years due to their economic origin and because they are friendly to the environment [206].

The obstacles mentioned above for catalysts in transesterification reactions have allowed the development of research focused on obtaining sustainable technology for biodiesel production. Within these investigations, the attention of researchers has been centered on the use of bifunctional catalysts, also known as acid-base catalysts. These catalysts involve the conjugation between a metal and metal oxides, which gives them both acid and basic sites [207,208].

Below, Table 8 shows some examples of bifunctional catalysts, as well as the sort of lipidic raw material employed, the reaction conditions used, and the %FAMES obtained at the end of the reaction.

Table 8. Examples of bifunctional catalysts used in biodiesel production.

Catalyst	Oil Used	Reaction Conditions	Methyl Esters Content (%FAMES)	References
Fly ash	WCO	T = 59 °C; CC = 11.2%wt; M:o = 3.1:1; t = 6 h	100	[209]
CaO(10%)-Fe ₂ O ₃ (10%)	WCO	T = 65 °C; CC = 3%wt; M:o = 18:1; t = 3 h	98.3	[210]
TiO ₂ /PrSO ₃ H	WCO	T = 60 °C; CC = 4.5%wt; M:o = 15:1; t = 9 h	98.3	[211]
RHC/K ₂ O-20%/Ni-5%	WCO	T = 65 °C; CC = 4%wt; M:o = 12:1; t = 2 h	98.2	[212]
CaO/Al ₂ O ₃	WCO	T = 60 °C; CC = 2.5%wt; M:o = 12:1; t = 3 h	98.23	[213]
MgO/MgSO ₄	WCO	T = 50 °C; CC = 6.4%wt; M:o = 10.8:1; t = 0.8 h	98.8	[214]
LiNbO ₃	WCO	T = 65 °C; CC = 2%wt; M:o = 24:1; t = 6 h	98.08	[140]
CaO/Al ₂ O ₃	WCO	T = 65 °C; CC = 1%wt; M:o = 11:1; t = 4 h	98	[215]
KOH/AC	WCO	T = 45 °C; CC = 1%wt; M:o = 18:1; t = 1 h	97.8	[216]
SrTi _{0.85} Fe _{0.15} O ₃	Palm oil	T = 170 °C; CC = 5%wt; M:o = 18:1; t = 3 h	97.52	[217]
Sn-CaO	WCO	T = 85.15 °C; CC = 2.2%wt; M:o = 16.1:1; t = 3.42 h	97.39	[218]
SrO-ZnO/Al ₂ O ₃	WCO	T = 75 °C; CC = 15%wt; M:o = 10:1; t = 5 h	95.7	[219]
10W/BV	WCO	T = 65 °C; CC = 8%wt; M:o = 6:1; t = 7 h	96	[220]
Sulfonated RHC	WCO	T = 50 °C; CC = 3.5%wt; M:o = 13:1; t = 0.84 h	96	[221]
2.6SrO-ZnO/Al ₂ O ₃ (2.6SZA)	WCO	T = 75 °C; CC = 15%wt; E:o = 10:1; t = 5 h	95.7	[219]
PKSAC-K ₂ CO ₃ (30%)CuO(5%)	WCO	T = 75 °C; CC = 15%wt; M:o = 10:1; t = 5 h	95	[222]
WO ₃ -Zr ₂ O ₃ (7WZ)	WCO	T = 80 °C; CC = 2%wt; M:o = 15:1; t = 1 h	94.4	[167]
SrTiO ₃	Palm oil	T = 170 °C; CC = 6%wt; M:o = 15:1; t = 3 h	93.14	[223]
BBFC	Neen seed oil	T = 61.9 °C; CC = 2.58%wt; M:o = 14.76:1; t = 1.21 h	92.89	[224]
CaO-CeO ₂	WCO	T = 70 °C; CC = 4%wt; M:o = 9:1; t = 1.2 h	90.14	[225]
CaO-Ca ₂ Fe ₂ O ₅ -CaFeO ₃	WCO	T = 60 °C; CC = 5%wt; M:o = 12:1; t = 2 h	90	[71]
Cu/Zn/γ-Al ₂ O ₃	Low grade	T = 65 °C; CC = 10%wt; M:o = 20:1; t = 2 h	88.82	[226]
MgO-SnO ₂	WCO	T = 60 °C; CC = 2%wt; M:o = 18:1; t = 2 h	88	[227]
7% SR/ZrO ₂	WCO	T = 70 °C; CC = 1%wt; M:o = 15:1; t = 0.34 h	85	[228]

T—Reaction temperature, CC—Catalyst weight, M:o—Methanol:oil molar ratio, E:o—Ethanol:oil molar ratio, t—Reaction time.

In Table 8, it can be seen there is a wide variety of materials that can be used to synthesize bifunctional catalysts. To dope the support, metals such as tungsten [167], lanthanum [229,230], strontium [219], iron [71,229], and zirconium [231,232]. It is also important to note that each metal will have its own catalyst characteristics, such as the crystalline phases present and acid strength. The catalytic features will depend on oxidation states, the synthesis method used, and the catalyst precursor [71].

The combination of Lewis and Brønsted acid sites within the bifunctional catalyst gives them a greater tolerance to FFA content in the lipid feedstock than basic catalysts, in addition to eliminating the low catalytic activity of acid catalysts [140]. They are also easily recoverable, which simplifies the end of the need for takeout washings [140]. The above leads to a reduction in the process's total cost.

Two of the main advantages of bifunctional catalysts are: (1) they are able to concurrently carry out the esterification as well as the transesterification [140]; and (2) these catalysts can be designed for specific needs, which allows them to have high selectivity [183]. In the case of biodiesel production, this permits the use of low-purity oils without complex production and purification processes [9].

The supports used in the synthesis of these catalysts are also very important since they can be obtained from various metal oxides such as Li_2CO_3 [140], CaO [213,225], ZrO_4H_4 [167], and Al_2O_3 [219], among others. These supports will provide different densities of basic sites, pore sizes, and surface areas, which will affect the final properties of the catalyst and directly influence the percentage content of FAMES obtained at the end of the reaction.

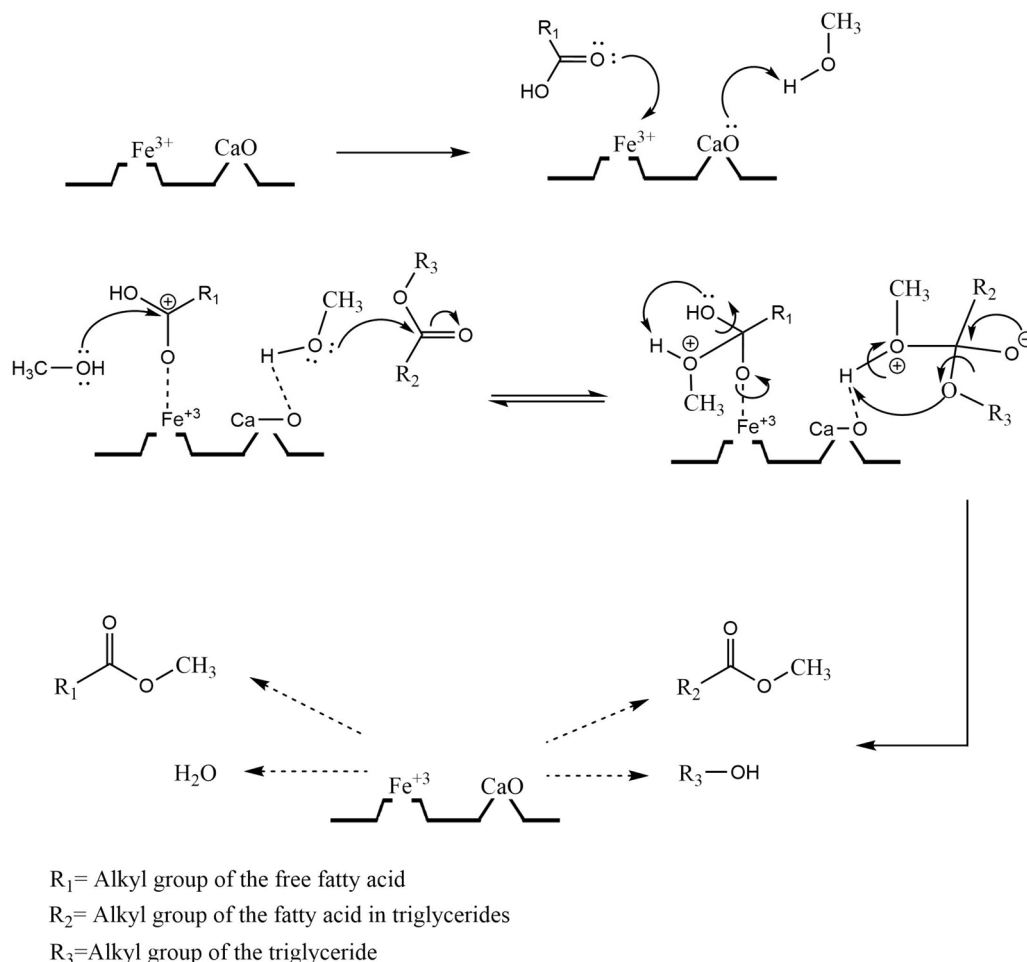
In the search for cheap raw materials, multiple materials were discovered that are sources of metal oxides; some of them are considered waste, which makes them low-cost raw materials as CaO sources such as eggshell [233] and recycled waste oyster shells [234]. As a result of the high tolerance to the FFA content of the bifunctional catalysts, we can see that most of those listed in Table 8 managed to obtain a high FAME content (>90%) at the end of the reaction.

Despite having reaction times ranging from 20 min to 9 h, it is possible to obtain high-quality biodiesel from a low-quality raw material in a single-step process considering that FFA esterification and triglycerides transesterification are carried out concurrently, which allows the reduction of production costs and the environmental impact of energy consumption.

In addition, the works presented in Table 8 show that with this type of catalyst, in most cases, it is possible to work with mild reaction temperatures. Depending on the catalyst used, using higher reaction temperatures allows the catalyst to use feedstocks with higher water contents (8%wt) without affecting the %FAMES obtained, as reported by Li [235].

As can also be observed in Table 9, the use of nanocatalysts/magnetic catalysts. Several investigations of nanocatalysts, most of them bifunctional, have been reported in recent years. Nanocatalysts' advantages are their high efficiency, minimal generation of chemical residues, safety, reduced global warming, economic efficiency, and energy efficiency [236]. However, one of the most important disadvantages of this type of catalyst is the difficulty of separating it from the reaction mixture. Therefore, they have been synthesized using magnetic metals to facilitate their separation and using biomass materials in several of them. A nano-magnetic catalyst ($\text{K}/\text{ZrO}_2/\text{g-Fe}_2\text{O}_3$) was synthesized by Liu et al. [237] for the transesterification of soybean oil, obtaining promising results with 5% by weight of catalyst, a methanol-oil molar ratio of 10:1, a temperature of reaction of 65 °C, and 3 h of reaction. A yield of 93.6% of biodiesel was obtained, presenting good magnetic characteristics that allowed the adequate separation of the reaction products. Liu et al. [237] synthesized a bifunctional magnetic catalyst based on bamboo charcoal, potassium, and iron ($\text{K}/\text{BC-Fe}_2\text{O}_3$) to obtain biodiesel using soybean oil. This catalyst presented excellent catalytic activity and good recoverability, reaching 98% yield with 2.5% catalyst, a methanol:oil 8:1 molar ratio, a reaction time of 1 h, and a temperature of 60 °C.

In Scheme 8, the mechanism of the transesterification reaction using bifunctional catalysts can be observed. It is carried out in three steps: (a) adsorption of the FFA from the WCO on the acid sites and adsorption of methanol on the basic sites of the catalyst; (b) reaction on the catalyst surface; and (c) desorption of methyl esters, glycerol, and water. In this scheme, Enguilo et al., using a Fe/CaO bifunctional catalyst, postulate that iron (the proton donor) provides the sites for both FFA esterification and triglyceride transesterification.



Scheme 8. Bifunctional transesterification mechanism of used cooking oil [71].

An area of opportunity to further our knowledge regarding the use of bifunctional catalysts to produce biodiesel is being able to establish the catalyst composition related to a specific FFA content. In addition, it is crucial for the sustainability of the process to conduct life cycle assessments, not only of the processes but also of the catalyst making, since several authors have reported this to be one of the most important stages contributing to the environmental impact [107,238].

In addition, it is crucial for the sustainability of the process to conduct life cycle assessments, not only of the processes but also of the catalyst making, since several authors have reported this to be one of the most important stages contributing to the environmental impact.

Table 9 shows the advantages, disadvantages, and economic feasibility of catalysts used for biodiesel production.

Table 9. Pros, cons and economic feasibility of catalyst type used for biodiesel production.

Catalyst	Pros	Cons	Economic Feasibility	References
Homogeneous acid	<ul style="list-style-type: none"> High tolerance to the presence of water and FFA Ability to use low-cost raw materials 	<ul style="list-style-type: none"> Environmentally harmful Safety risky Corrosive nature Needs higher methanol:oil molar ratios Higher reaction temperatures Slower reaction rates Longer reaction times Generation of large amounts of wastewater 	<p>The high quality of the raw material significantly affects the production cost of biodiesel.</p> <p>The corrosive nature of these catalysts results in an economically unfeasible process.</p>	[22,122,140,239]
Homogeneous basic	<ul style="list-style-type: none"> Lower cost. High availability Lower methanol:oil ratios Mild reaction temperatures No corrosive problems Relatively short reaction times 	<ul style="list-style-type: none"> High sensitivity to the presence of water and FFA Need of refined raw materials The need of a previous esterification to manage raw materials with elevated FFA content High reaction rate Generation of large amounts of wastewater 	<p>The high quality of the raw material significantly affects the production cost of biodiesel.</p> <p>It can be considered as an economically viable option to produce biodiesel from high-quality oils.</p>	[9,106,123,124,140–143,239]
Enzymatic	<ul style="list-style-type: none"> No catalyst leaching is generated Easy recovery. Mild reaction conditions High tolerance to the presence of FFA Lower reaction temperatures Ability to be recycled Easy recovery Great selectivity 	<ul style="list-style-type: none"> High cost of most enzymes Easily deactivated by wrong alcohol selection Slower reaction rates Enzyme deactivation 	<p>The high quality of the raw material significantly affects the production cost of biodiesel.</p> <p>These catalysts are good examples to produce biodiesel from low-quality feedstocks in one step.</p>	[23,239–241]
Heterogeneous acid	<ul style="list-style-type: none"> High tolerance to the presence of FFA Reduction of corrosive nature Ability to carry out esterification and transesterification simultaneously Ability to use cheaper raw materials Ability to be easily separated and recycled Reduce purification costs 	<ul style="list-style-type: none"> Active sites leaching Mass transfer problems Slow reaction rate for transesterification reaction Needs higher methanol:oil molar ratios Higher reaction temperatures Longer reaction times 	<p>High profit at minimal investment.</p> <p>The high quality of the raw material significantly affects the production cost of biodiesel.</p>	[101,170,183,206,239]
Heterogeneous basic	<ul style="list-style-type: none"> Ability to be easily separated and recycled Reduce purification costs Mild reaction conditions Lower cost High availability Lower methanol:oil rates Shorter reaction times 	<ul style="list-style-type: none"> Active sites leaching Mass transfer problems High sensitivity to the presence of FFA Sensitive to poisoning by absorbing humidity and CO₂ Deactivation derived from adherence of reaction by-products 	<p>High profit at minimal investment.</p> <p>The high quality of the raw material significantly affects the production cost of biodiesel.</p> <p>It can be considered as an economically viable option to produce biodiesel from high-quality oils.</p>	[106,167,186,206,239]
Bifunctional and Nanocatalysts/magnetic	<ul style="list-style-type: none"> High tolerance to the presence of FFA Ability to use cheaper raw materials Ability to be easily separated and recycled Ability to carry out esterification and transesterification simultaneously Reduce purification costs Mild reaction conditions Acid-base conjugation eliminates low activity of acid catalysts High selectivity They can be designed with specific characteristics 	<ul style="list-style-type: none"> Active sites leaching Mass transfer problems Higher synthesis cost depending on the used metal Complicated synthesis 	<p>The high quality of the raw material significantly affects the production cost of biodiesel.</p> <p>These catalysts are good examples to produce biodiesel from low-quality feedstocks in one step.</p>	[9,140,183,237,239]

4. Reactors for Biodiesel Production

In recent years, biodiesel production has focused on trying to reduce production costs by using various feedstocks that make them decrease, since the raw material represents 60–80% of the production cost. Therefore, the development of catalysts as well as the types of reactors have been essential to optimizing and developing new sustainable technologies. In the previous section, different types and characteristics of the catalysts used to obtain biodiesel were shown. As for the reactors, they have an important role in the yield of biodiesel since they must be optimized to achieve greater sustainability, technical advantages, and economic viability. In the process of obtaining biodiesel, vegetable edible and non-edible oils are mainly used, although oil-containing materials such as seeds, rice bran, spent coffee grounds, fat derived from insect biomass, animal fat, waste cooking oil, and products from edible oil have also been employed as feedstocks [6,19,26,38,43,47]. In some studies, pure acids such as oleic, caprylic, and capric acids, as well as pure triolein, have been utilized [242]. The reactors that are typically used for biodiesel production are shown in Figure 8.

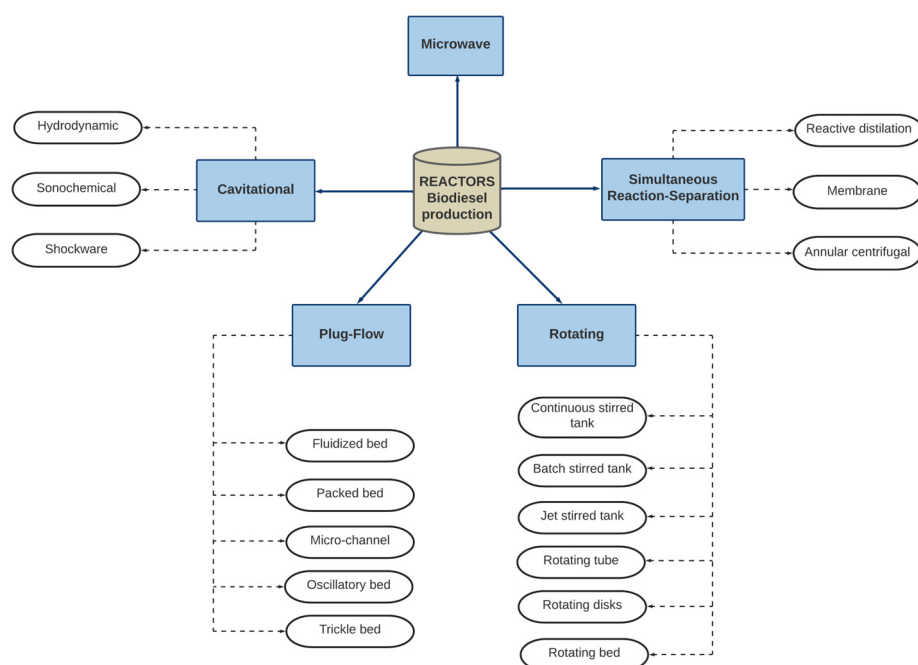


Figure 8. Types of reactors for biodiesel production. Adapted with permission [243] Copyright 2019 Elsevier.

In plug-flow reactors, products have consistent quality. They use liquids with diverse viscous ranges and have low maintenance and capital costs.

Rotating reactors have tubes, discs, and impellers that require different energy and rotational power, directly affecting the biodiesel quality. These reactors are expensive due to the large amount of energy needed [22].

The most widely used plug flow reactor is the tubular, or PFR, where the reagents are fed, at a constant speed, through one end of the reactor with an adequate residence time to allow the reagents to mix while they flow towards the outlet. When handling viscous liquids, PFRs present laminar flows, so it is necessary to use injection devices (a T mixer) or mechanical mixers. Some of the advantages of PFRs are that they do not require constant maintenance, are built in small spaces, and have low capital costs [243,244].

The cavitation reactors usually used to obtain biodiesel are hydrodynamic and sonochemical. Cavitation reactors are multiphase reactors that intensify the process using acoustic or flow energy, causing a cavitation phenomenon and, therefore, an increase in

turbulence, thereby improving mass transfer. This cavitation phenomenon achieves high values of temperature and pressure [243–245].

Simultaneous reaction-separation reactors perform the chemical reaction, extraction, and separation of the products in a single stage. This type of reactor achieves better quality and a higher biodiesel yield because there is an optimal mixture. Of these reactors, the membrane ones are the most used in the production of biofuels since they do not require additional units in the process. Reactive distillation has also been used for the simultaneous reaction-separation process, which presents advantages over other reactors, such as short reaction times and improved yields because the products are extracted immediately after production. Costs decrease since additional operating units are not required [243,244].

Table 10 shows some research regarding the use of chemical reactors to obtain biodiesel from waste cooking oil. To intensify the production of this biofuel, techniques such as supercritical conditions [31], microwaves [170,246,247], ultrasonic [248], hydrodynamic cavitation [245], and spinning disk [249] have been used to eliminate resistance to mass transfer between alcohol and oil and have short periods and low energy consumption compared to industrial processes that are currently used [245].

Table 10. Examples of bifunctional catalysts used in biodiesel production.

Type of Reactor	Oil	Catalyst	Catalyst Load (%)	Alcohol:Oil Ratio	Reaction Temperature (°C)	Yield (%FAMES)	References
Continuous Flow-Microwave	WCO	SrO/SiO ₂	41	12:1	65	99.2	[247]
Hydrodynamic cavitation	WCO	KOH	1	6:1	60	98.1	[245]
Ultrasonic	WCO	KOH	1	7.4:1	60	96.5	[250]
Microreactor	WCO	KOH	2	12:1	50	>95.00	[251]
Rotating flask oscillatory flow	Coconut CWO	KOH	1	6:1	60	93.72	[252]
Reactive distillation	WCO	CaO	Bed height of 150 mm	4:1	65	93.48	[253]
Microtubes (prior esterification)	WCO	H ₂ SO ₄	1	9:1	65	91.76	[254]
Microwave Irradiation	WCO	KOH	0.5	5:1	-	91.63	[246]
Batch stirred tank	WCO	Fe/CaO	7	12:1	60	90.0	[71]

As seen in Table 10, moderate reaction temperatures (60 °C) and several homogeneous and heterogeneous catalysts are used in different types of chemical reactors, obtaining yields of at least 90% in all of them with WCO.

Tangy et al. [247] reported the highest yield using WCO (99.2%) in a continuous flow reactor for microwave-assisted transesterification and heterogeneous catalysts (SrO/SiO₂). However, it is necessary to improve the process and see other catalyst synthesis options to prevent SrO leaching and make the FFA content fed to the reactor more efficient.

On the other hand, microreactors, whose advantages include small diffusion distances, increased specific surface area, high heat and mass transfer rates, and greater safety, have driven the study of capillary reactors for biodiesel production [251,254]. Tanawannapong et al. [254] reported biodiesel production from waste cooking oil using a microtube reactor with a 1.2 cm length and an internal diameter of 0.508 mm along with a T mixer at the inlet of the reactor; a methyl ester content of 91.7% was obtained in a 5 s reaction time; this study verifies that the transesterification reaction takes a shorter time as compared to the batch reactor. Although satisfactory initial results are observed, the technology still requires advancements in certain areas. Further research is needed to develop operating procedures that include the usage of feedstock, possessing increased levels of free fatty acids, and exploiting heterogeneous catalysts, in addition to high costs and complexities in the fabrication of glass and silicon microdevices [255].

In summary, the cost-effectiveness of the transesterification reaction to produce biodiesel rests heavily on many process parameters, namely the oil type and composition, type and amount of the catalyst, type and quantity of alcohol, residence time and temperature, and process intensification method. Actually, the last parameters depend upon catalyst type, and therefore its cost and activity greatly affect not only the overall cost of the process [107,206,256] but also the environmental impacts [107]. Actually, it has been demonstrated by Alanis et al. that the metal precursor used to prepare the catalyst has a significant impact on the production cost and energy consumption because the catalyst activity is modified. Therefore, in order to minimize environmental impacts and the overall cost of the catalyst preparation and the biodiesel production process, an optimization of the catalyst synthesis must be conducted.

The batch operation remains the most common process for the production of biodiesel. However, continuous production has advantages, such as small spaces for their installation, reduced operating costs, low investment, and ensuring a product of consistent quality. This approach is ideal when a fast response is required and offers higher heat transfer and selectivity than the batch method. As a result of all of this, the biodiesel industry now has its eyes on the continuous process model, hoping to solve batch production problems.

5. A Perspective on Biodiesel Production in Different Countries and Biodiesel Commercialization Improvement Strategies

In the last decade, the production and consumption of biodiesel in different countries in the world have been increasing due to the increase in energy demand derived from the growth of the human population [257]. This has allowed these countries to embrace policies related to biodiesel production.

- In the US, biodiesel is commonly used by the transportation sector through blends with fossil fuels, such as B10 or B20. Demand for biodiesel in the US is expected to increase as fossil fuels are replaced. As an example, biodiesel production in 2023 is expected to be 37% higher than in 2022 due to the implementation of economic incentives by the government [258].
- The Canadian government has improved biodiesel production through subsidies to decrease the cost of biofuel production and comply with the agreements established in the 2030 Agenda. As an example, biodiesel production in 2026 is expected to be 313 M liters, compared to the 147 M liters awaited this year [258].
- In Latin America, the biodiesel production expected in 2023 is 635 M liters. In Brazil, the variety of climates and the large expanse of soil allow it to cultivate various oilseeds. Aside that, this country has policies that promote this biofuel usage [258].
- In Europe and the UK the biodiesel production expected in 2013 is 37% higher than in 2022 [258]. Due to the implementation of different policies in the EU, biodiesel will be more accessible in order to reduce the use of fossil fuels.

Despite the abundance of available studies about biodiesel production from waste cooking oil, there are still various strategies that could improve its commercialization. These are presented below.

- (a) Implementing efficient collection systems according to local regulations for the correct disposal of lipid waste of animal or vegetable origin. Then the need for these regulations arises because large quantities of this residue are produced worldwide, which increases proportionally to the human population's increase. To promote the recollection and correct disposition of WCO, several countries have implemented different incentives. For instance, China's government offers several subsidies, such as training, a collecting system focused on the market, and professional disposal. In Japan, the subsidies offered to biofuel producers allow for greater economic competitiveness by reducing production costs. In Spain, there is a WCO recollection program where collection bins are set up in some neighborhoods of Barcelona and Valladolid [259].
- (b) Designing industrial-scale processes based on a circular economy perspective: In recent years, this concept has been introduced with the idea of reducing waste to

a minimum, which implies the employment of renewable energy sources to use energy more efficiently and reuse process water [260,261].

- (c) Life Cycle Sustainability Assessment (LCSA) of existing processes. This is mandatory to estimate the environmental impacts of biodiesel production through different processes, which ultimately will aid the government and industry in making decisions about the less harmful process [259,262,263].

This tool has been applied to evaluate different biodiesel production processes, such as the one conducted by Talens Peiró et al. [264] which found that the transesterification process contributed 68% to the total impact. Lombardi et al. [265] reported a comparison against several methods of biodiesel production, and it shows that NaOH-catalyzed transesterification of WCO can lead to decreased environmental impacts [265]. Alanís et al. [107] compared the environmental impacts of biodiesel production from WCO catalyzed by a heterogeneous bifunctional catalyst synthesized with two different iron salt precursors [107].

6. Conclusions

Biodiesel can be obtained by the esterification and transesterification of waste cooking oil (WCO) as a raw material by homogeneous or heterogeneous catalysis. The type of biodiesel production process impacts the quality of biodiesel. In this context, transesterification is highly preferred. The feedstock and catalysis type used to produce biodiesel determine the stages of the process. The homogeneously catalyzed process implies two more steps than the latter, and those are esterification under acid conditions (usually adding H_2SO_4) and elimination of the transesterification catalyst (usually a base like NaOH or KOH) by washing with water. Enzymatic catalysis to conduct the process is not a viable option due to the long reaction times and cost of the enzyme. Bifunctional catalysts are solids that contain both acid and basic sites and are able to reduce the number of process stages by simultaneously conducting esterification of free fatty acids and triglyceride transesterification. This eliminates the need to add an acid and the high consumption of water to clean the biodiesel.

The challenges to overcome in biodiesel production using WCO and bifunctional catalysts are reducing costs and energy consumption, using catalysts whose synthesis does not demand high loads of energy, and conducting life cycle assessments in order to establish the environmental impacts of each process and provide a relevant criterion to make the decision of electing or not electing a specific production process.

From the assessed processes to conduct the transesterification of WCO, the ones including microwaves, microreactors, and reactive distillation seem to be the most promising ones in terms of yield, reaction time, and number of involved stages.

Biodiesel from WCO usually does not meet all the standards, mainly that of methyl esters content and viscosity. In order to use biodiesel from WCO, it is suggested to use it in blends, to use co-solvents or to improve the pre-treatment of the raw material.

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