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*Review*

# Biodiesel Is Dead: Long Life to Advanced Biofuels—A Comprehensive Critical Review

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**Abstract:** Many countries are immersed in several strategies to reduce the carbon dioxide ($CO_2$) emissions of internal combustion engines. One option is the substitution of these engines by electric and/or hydrogen engines. However, apart from the strategic and logistical difficulties associated with this change, the application of electric or hydrogen engines in heavy transport, e.g., trucks, shipping, and aircrafts, also presents technological difficulties in the short-medium term. In addition, the replacement of the current car fleet will take decades. This is why the use of biofuels is presented as the only viable alternative to diminishing $CO_2$ emissions in the very near future. Nowadays, it is assumed that vegetable oils will be the main raw material for replacing fossil fuels in diesel engines. In this context, it has also been assumed that the reduction in the viscosity of straight vegetable oils (SVO) must be performed through a transesterification reaction with methanol in order to obtain the mixture of fatty acid methyl esters (FAMEs) that constitute biodiesel. Nevertheless, the complexity in the industrial production of this biofuel, mainly due to the costs of eliminating the glycerol produced, has caused a significant delay in the energy transition. For this reason, several advanced biofuels that avoid the glycerol production and exhibit similar properties to fossil diesel have been developed. In this way, "green diesels" have emerged as products of different processes, such as the cracking or pyrolysis of vegetable oil, as well as catalytic (hydro)cracking. In addition, some biodiesel-like biofuels, such as Gliperol (DMC-Biod) or Ecodiesel, as well as straight vegetable oils, in blends with plant-based sources with low viscosity have been described as renewable biofuels capable of performing in combustion ignition engines. After evaluating the research carried out in the last decades, it can be concluded that green diesel and biodiesel-like biofuels could constitute the main alternative to addressing the energy transition, although green diesel will be the principal option in aviation fuel.

**Keywords:** biodiesel; advanced biofuel; straight vegetable oils (SVO); Gliperol; DMC-Biod; Ecodiesel; green diesel; pyrolysis; cracking; hydrocracking; less viscous and lower cetane (LVLC) vegetable oil blends



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## 1. Introduction

Nowadays, most of the countries worldwide are making an unprecedented effort to reduce anthropogenic greenhouse gas (GHG) emissions to carry out a decarbonization process, which significantly affects the energy sources applied. In this sense, the Treaty of Paris [1] and the European New Green Deal, as well as the REDII, aim to achieve a climate-neutral Europe by 2050 [2]. Therefore, several countries have also implemented their own energy and climate policy framework for 2030 and beyond, advancing in decarbonization and promoting innovation in order to achieve a viable new climate economy low in $CO_2$ emissions [3].

Considering that the choice of green hydrogen as the main energy vector for the decarbonization of the planet seems definitive, biofuels should receive a secondary role in the current research and development priorities of transportation, including cars, trucks, ships, planes, etc. However, the transition from current energy sources to this new technology requires a period of several decades, in accordance with the planning carried out by the same countries involved in these international agreements [4].

Notwithstanding the possibility of building a transport fleet operating with new technologies and being neutral in $CO_2$ emissions, it is mandatory to consider the temporary rate of the substitution of current vehicles working with internal combustion engines (ICEs) in order to avoid an economic chaos of unpredictable consequences. In this sense, the replacement of the enormous number of vehicles that operate with ICEs needs to be carried out in such a way that they can continue operating throughout their useful life with diesel fuels or, alternatively, with biofuels with similar properties. This fact does not constitute a trivial problem due to the very high number of vehicles currently in use and the fact that those vehicles that are being built now and in the next two or three decades must be added to the list [5]. Consequently, the reduction in emissions in this long transition period involves a reduction in fossil fuels and increase in other fuels that allow for their operation in ICEs, together with the incorporation of hydrogen-powered engines and other emerging technologies. In this way, a smooth transition to a scenario without fossil fuels could be foreseen [6].

In this sense, biofuels can be easily integrated into the logistics of the global transportation system. In fact, the goal pursued by EU is that biofuels constitute 30% of all fuels by 2030 [7]. Despite this goal being easy to achieve considering the technical issues, the substitution of fossil fuels with biofuels is considered unattainable in this deadline due to the impossibility of having enough agricultural land to carry out the necessary crops, since bioethanol and biodiesel (the most widely biofuels employed) require enormous agricultural resources to fulfill these purposes [8–12].

Therefore, it is also mandatory to introduce electric engines in the transport sector, since biofuels will not be able to completely replace all fossil fuels currently operating ICEs, considering that 20% of the global emissions of GHG are contributed by this sector [13–15].

In summary, to carry out the planned energy transition efficiently and sustainably, it is essential to have biofuels that are technically and economically feasible to not only be able to gradually replace the fossil fuels used by the current diesel engines but also to be used in a long indeterminate time horizon in trucks, ships, and especially airplanes, where the introduction of electric engines cannot yet be considered in a predictable time due to the technology immaturity [5,16–18]. Hence, regardless of the progress that will be obtained in the coming decades with respect to the introduction of electric engines in the transport sector, research on biofuels presents the maximum interest, not only to facilitate the necessary energy transition with the profitability of current conventional ICEs, but also for its application in more specific sectors, such as trucks, boats, and aircrafts, along a temporarily indefinite period.

On the other hand, the introduction of biofuels in the energy market dedicated to transport is in its early stages, since only 1.6% of biofuels are used currently, with respect to the total transport fuel worldwide spent [19], including the biodiesel production worldwide, which is around four billion liters. In this context, straight vegetable oils (SVO) and animal fats can be considered as the main option to achieve the substitution of fossil diesel fuel. In order to carry this out, SVO or animal fats are transformed into biodiesel by a transesterification reaction with methanol, producing a mixture of fatty acid methyl esters (FAMEs) [20]. This relatively simple process can reduce oil viscosity to the level of the values of conventional fossil diesel (4–5 $mm^2/s$) and constitutes the only currently applied industrial method to convert vegetable oils into biodiesel [21].

However, the transesterification process reveals a serious drawback at the industrial scale, which is associated with the production of a relatively high amount of glycerol generated as a by-product (10% by weight of the total biodiesel produced). Apart from reducing the performance of the process, the glycerol must be eliminated since the high temperatures reached in engines favor the formation of glycerol polymers, as well as acrolein, which has a high toxicity. In fact, glycerol production is considered the most important barrier, and has, so far, prevented the consolidation of biodiesel as the biofuel that can replace fossil fuels [22].

To overcome the glycerol production problem, different alternatives have been developed in the last decades to transform vegetable oils into high-quality biofuels without the production of glycerol. Thus, several oxygenated biofuels that integrate glycerol as soluble derivatives have been described, e.g., Gliperol, DMC-Biod, or Ecodiesel [23,24]. These biofuels are known as "biodiesel-like biofuels". In addition, biofuels obtained from triglycerides by different processes, such as cracking, pyrolysis, hydrodeoxygenation, and hydrotreating of vegetable oils, have also been described. These are high-quality renewable diesel fuels generally known as "green diesel" or "renewable diesel", exhibiting a similar composition to fossil fuel [25]. Finally, the possibility of using various additives in mixtures with SVO is also being evaluated to reduce the kinematic viscosity of the mixtures to the levels required by ICEs. For this reason, biofuels made up of mixtures of SVO and renewable solvents have been described. Since these compounds generally have low octane numbers, these biofuels are used in blends with oils, obtaining the so-called LVLC (less viscous and lower in cetane) fuels [26,27].

Figure 1 summarizes the different methods of transforming animal fats and vegetable oils into biofuels, avoiding the generation of glycerol, exhibiting all of the many advantages with respect to conventional biodiesel. This review is an overall view of the current research in fuel development alternatives to biodiesel, intending also to evaluate the strengths and weaknesses of these alternative processes described in Figure 1 in order to consider them as fuels in different transport sectors, such as heavy road vehicles, aviation, and/or maritime transportation sectors. In addition, this review aims to claim to the scientific community that the research in these alternatives to biodiesel must be made mandatory due to their strong dependence on fossil energy sources, since oil is the main energy source supplying approximately 95% of the sector's energy consumption [28].

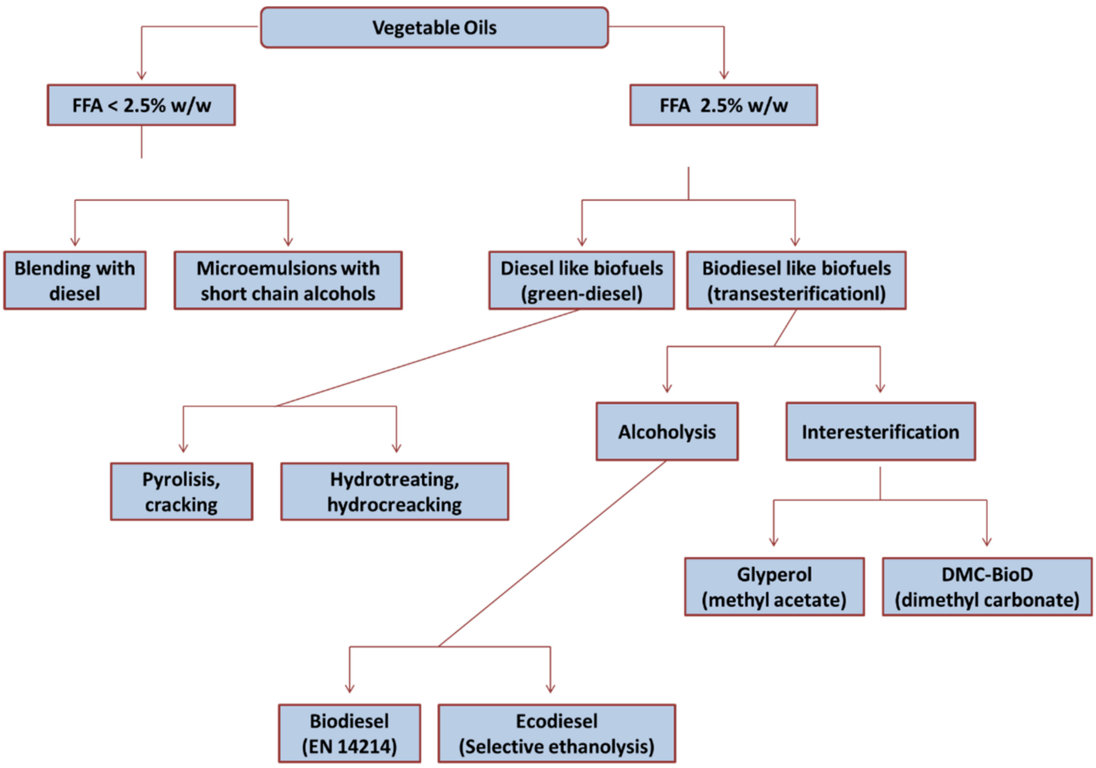


**Figure 1.** Different alternative methods of transforming animal fats and vegetable oils into biofuels, avoiding the generation of glycerol, adapted with permission of Ref. [24]. Copyright, 2014 Elsevier.

## 2. Strengths and Weaknesses of Biodiesel as Renewable Biofuel in Current Diesel Engines

Biodiesel is defined as a mixture of long chain fatty acid methyl esters derived from renewable lipid sources, such as vegetable oil or animal fat, that can be used in compression ignition engines with little or no modifications. Until now, the use of a homogeneous alkaline transesterification chemical process with methanol has been initially chosen to address the biodiesel production [29–32]. In fact, biodiesel is, to date, the liquid biofuel produced at a greater quantity, due to the simplicity of its chemical process and its rheological properties, like fossil diesel [33–35]. In addition, it can be produced from different feedstocks, depending on the availability of the crop in the region. Among other advantages, biodiesel exhibits biodegradability, non-toxicity, renewability, a high cetane number, a high flash point, and its high oxygen content allows for its complete combustion in engines, reducing the amount of particulate matter, hydrocarbons, and gases, such as carbon monoxide (CO), $CO_2$, and sulfur oxides (SOx). Furthermore, biodiesel has a very low sulfur content and very low aromatic components, as well as other pollutant emissions. Nevertheless, a slight increase in nitrogen oxide (NOx) emissions is usually described in comparison to diesel fuel [36–38]. Due to the high flashpoint that biodiesel exhibits, at around 150 °C, it is very safe for transportation and storage [39–41]. In addition, biodiesel perfectly fits into existing engines without any modification and it can be used in its pure form or blended with petroleum-based fuels without modification of existing engines or with only minor modifications [42–47]. Moreover, biodiesel exhibits better lubricant properties than fossil diesel, which allow for the extension of the engine life, and also allow for a reduction in carbon dioxide emissions by 78% in comparison with fossil diesel. In addition, the biodegradability of biodiesel is certainly high, ranging from 80.4% to 91.2% after 30 days, whereas the biodegradability of fossil diesel is only 24.5% [48]. Taking into consideration all of the advantages abovementioned, it is understandable that biodiesel has become a research hot spot during the last years, resulting in an increase in scientific publications and patents [49], as can be seen in Figure 2. Thus, for only microalgae biodiesel production, more than ten thousand patents have been published in the last 20 years [50–52]. Furthermore, in the last twenty years, almost forty-four thousand articles have been published, producing a growing increase year after year, demonstrating the growing interest in the problem.

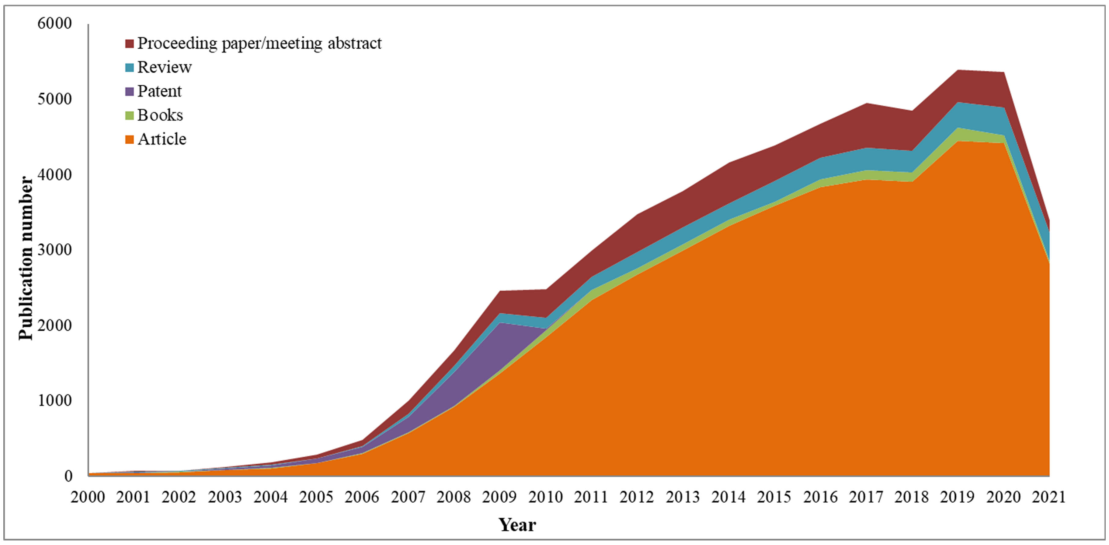


**Figure 2.** Publications found in the Web of Science database by the keywords "biodiesel" separated by document types from year 2000 to 2021.

Despite these efforts made to achieve better processes, better catalysts, and better sources of raw materials, it is currently concluded that the generation of glycerol represents a barrier that is difficult to overcome for the industrial production of biodiesel [53]. An alternative could be the reduction in the production cost of biodiesel. Nevertheless, the biodiesel industry strongly depends on the cost of the feedstock employed as a raw material. Despite the fact that some feedstock, such as non-edible oil and waste cooking oil, can be obtained at a good cost, they usually need a higher cost in their manufacture processing to produce standard-quality biodiesel [54–56]. The true magnitude of this problem has been proven in all of its consequences when the industrial-scale production of biodiesel has begun in the last three decades. The management of the huge amounts of wastes, where glycerol is the main component, is a problem with a very difficult solution [57,58], and there are still no industrial processes capable of integrating the enormous amount of glycerol. Furthermore, this glycerol obtained as a by-product also exhibits a very low quality, since it is in a mixture with other products, such as methanol, water, salts, and some amounts of monoglycerides [59].

Therefore, for being employed as a biofuel, the biodiesel obtained must be cleaned and separated from these by-products. The additional cleaning process is usually carried out by successive washing steps with water, so that a large consumption of water, energy, and time to obtain the glycerol elimination is required in order to obtain the limits established by the quality standard EN 14214 and the ASTM D6751, which are the European and the American ones, respectively [60–64]. These limits establish that the amount of glycerol should not exceed the 0.02% in the refined biodiesel in order to prevent its reaction with oxygen at high temperatures inside the engine, which would either produce acrolein or would polymerize generating deposits of carbonaceous compounds on the injector nozzles, pistons, and valves in the engines, consequently reducing the efficiency of the engine and its service life [65–67]. Therefore, it is clear that the industrial production of biodiesel requires a very complex design in order to avoid the presence of glycerol in the final biofuel [68], as is shown in Figure 3. In summary, the transesterification reaction is usually carried out in a batch reactor under constant stirring at 60 °C. Then, glycerol is separated together with the excess of methanol by decantation. Then, methanol is recovered by distillation. This crude biodiesel contains catalyst residues that must be neutralized and eliminated.

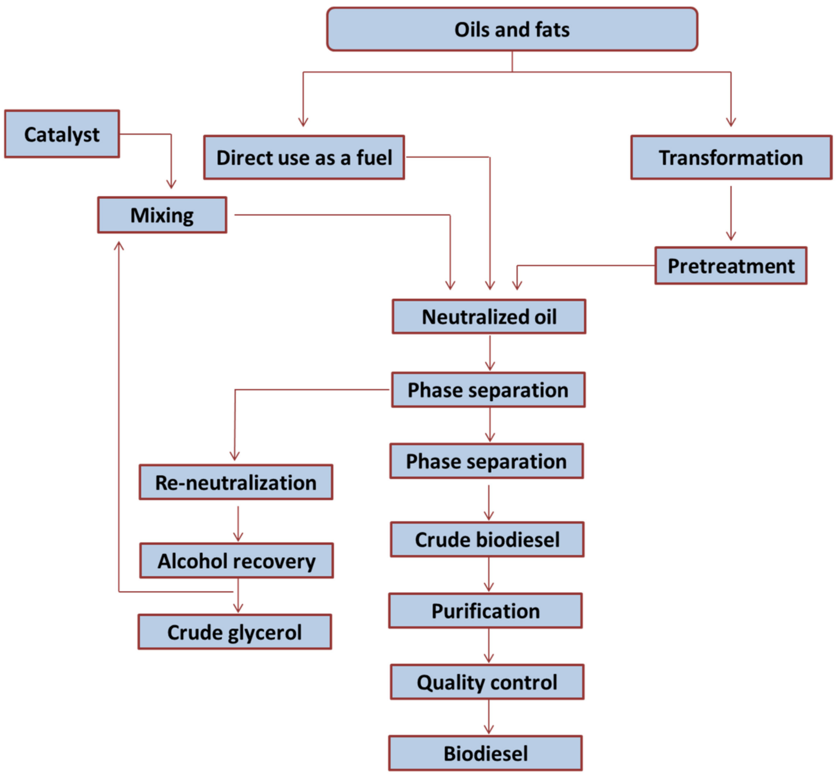


**Figure 3.** Standard flowchart of an alkali transesterification process in a conventional biodiesel plant, reproduced with permission of Ref. [68]. Copyright 2019 Elsevier.

As aforementioned, biodiesel must be subjected to several washing steps with water, although the purification process also requires a drying process in an evaporator to remove held residual water [60]. Alternatively, the purification of biodiesel may also be obtained by ultrafiltration and dry washing, employing fumed silica sorbent, molecular distillation, organic resins, and biomass-based adsorbent or starch and cellulose as adsorbents of impurities [69–74]. This vast number of studies devoted to obtaining methodologies that are economically viable show that this step is one of the main factors that lead to an unprofitable biodiesel production [75].

Consequently, there is not a practical solution for the problem associated with the destabilizing glycerol price in the global market, since there are no industrial processes capable of adsorbing the increasing glycerol production [76,77]. To minimize this problem, multiple investigations are being carried out in order to valorize this crude glycerol [78–80]; see Figure 4.

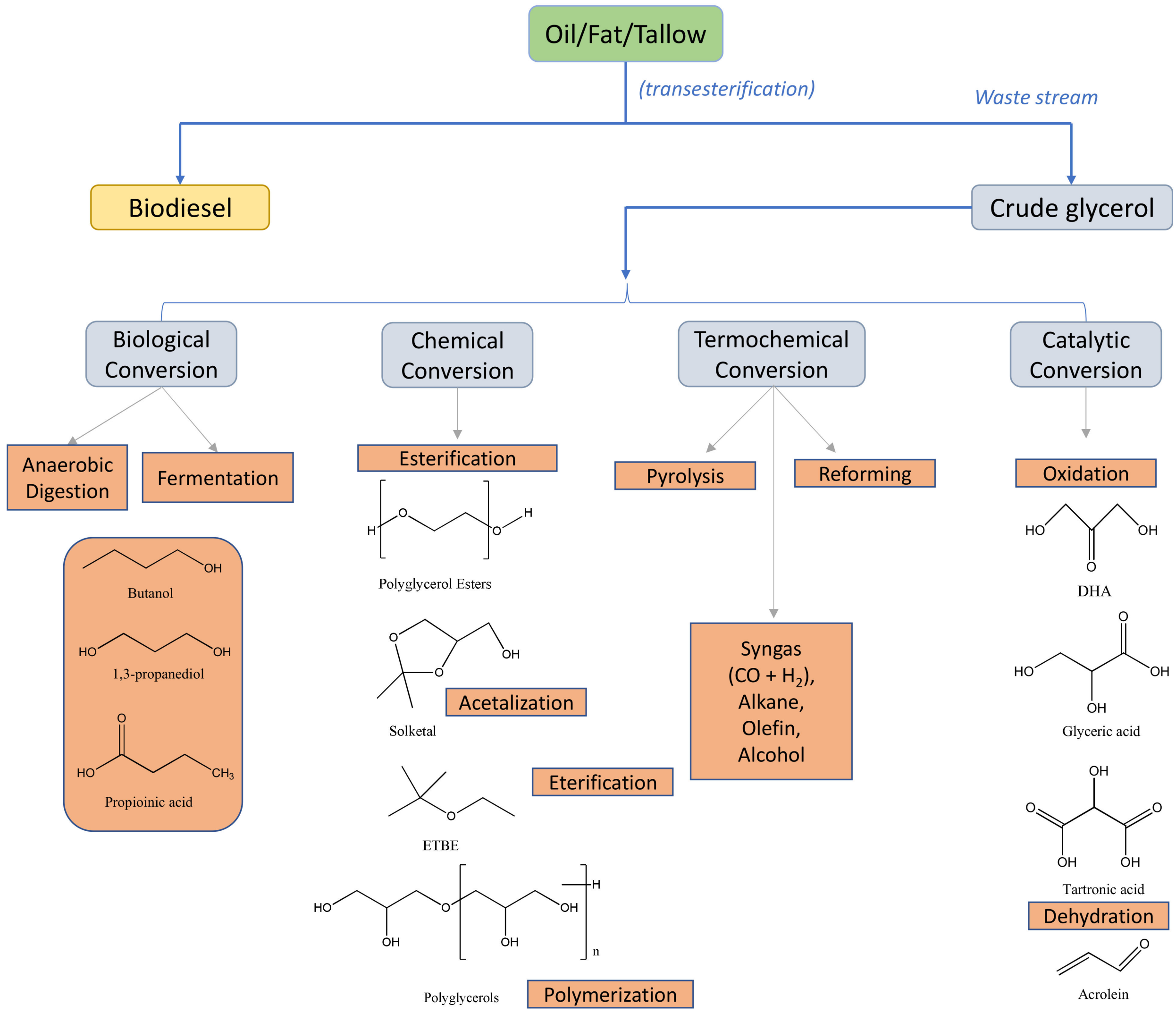


**Figure 4.** Different chemicals obtained to valorize crude glycerol generated in the industrial production of biodiesel, reproduced with permission of Ref. [78]. Copyright 2020 Elsevier.

Another element of vulnerability associated with the production of conventional biodiesel is related to the low atomic yield (or atomic efficiency) of the process. The atom yield is an important concept in green chemistry, and is far from the concept of chemical yield. In fact, a high-yielding process can still result in a substantial quantity of by-products, as is the case for biodiesel production. These green metrics are crucial for determining the sustainability and environmental impact of biodiesel production [81–83].

In summary, it is commonly accepted that the greatest contribution to determining the cost of biodiesel is determined by the price of feedstock, which occupies 70% of the biodiesel production cost [84–86]. Therefore, independently of looking at increasing the sources of triglycerides, available at appropriate prices for their transformation into biofuels, by optimizing the parameters influencing the production process of biodiesel, costs could be reduced by up to 30%. In addition, savings could be obtained by avoiding the management of residual glycerol, obtained together with conventional biodiesel, as well as the increase of at least 10% of the final product, if it is no glycerol is generated as a by-product. Thus, the search for different renewable biofuels integrating glycerol is still encouraged, while also avoiding several collective drawbacks, such as being energy-intensive, tedious in recovering glycerol, difficult in removing the acid or base catalyst from the product, the further treatment of alkaline wastewater, and the interference of free fatty acids and water in the reaction [87].

## 3. Green Diesel Fuels

Taking into account the complexity of the processes needed to perform the biodiesel purification, as well as the final management of the glycerol generated, the thermal conversion of fats and oils can constitute an alternative route for obtaining diesel-like biofuels, usually called green diesel, renewable diesel, or green fuels [25]. Green diesel is obtained directly from natural oils and fats via the UOP/Eni Ecofining$^{TM}$ process, providing a significant reduction in greenhouse gas emissions (GHGs) [88]. The production process implies a deoxygenation step and can be applied to different feedstocks, e.g., vegetable oils, animal fats, fatty acids, and waste cooking oils. Usually, the deoxygenation reaction occurs in the liquid phase following three pathways: decarboxylation, decarbonylation, and hydrodeoxygenation [89]. In this way, very similar hydrocarbons to those from crude oil are obtained when subjecting these compounds to different thermal treatments, such as cracking or pyrolysis, involving changes in the chemical structure of the triglycerides [90].

The thermal treatment can be also conducted in the presence of a catalyst, i.e., catalytic hydrocracking. This method requires an acid catalyst and a free-oxygen atmosphere. This method consumes less thermal energy and also produces a lower amount of coke that diminishes the catalyst deactivation by pore blockage and/or the catalytic poisoning. An important variation of the hydrocracking process consists of the hydroprocessing of triglycerides and petroleum gas oil simultaneously, by their co-processing in the same hydrotreating unit. In this section, we will focus on these processes.

Nowadays, the commercial plants for the production of renewable diesel have been installed all over the world. Currently, over 5.5 billion liters of renewable diesel is produced globally and is forecasted to grow up to 13 billion liters in 2024. Neste is the most important petroleum refining company, although some other oil and gas companies, such as ENI and Total, are also producing a significant amount of renewable diesel [91]. The current scenario of green diesel production worldwide is shown in Figure 5.

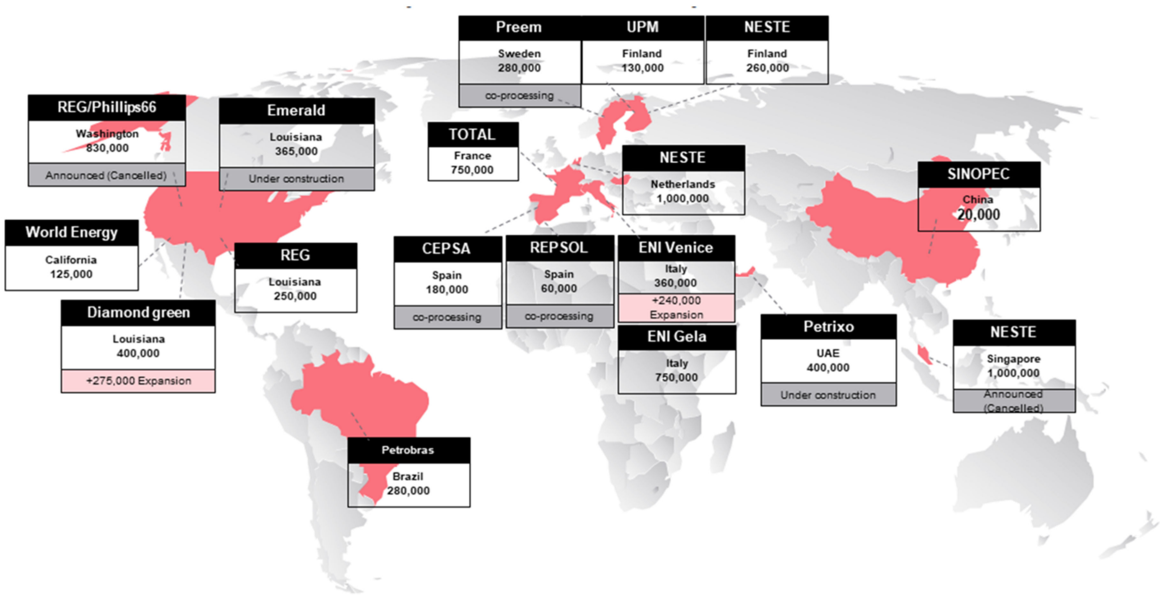


**Figure 5.** Current scenario of green diesel production worldwide. Source: Futurebridge analysis; EC Reports.

### 3.1. Pyrolysis or Cracking

In general, this technique involves a destructive distillation process that produces a change in the chemical structure of the compound through the irreversible breaking of chemical bonds, leading to smaller molecules. The thermal pyrolysis can occur either by the application of heat or in the presence of a catalyst (catalytic pyrolysis). The irreversible reaction is highly endothermic, requiring high heat transfer rates. These conditions can be reached by burning a fraction of the products to produce the thermal energy required for the reaction.

These processes are usually performed in an inert atmosphere, in a temperature range of 573–1573 K. Three different pyrolysis methods have been described, according to the reaction temperature, that influence the yield and reaction times. Conventional pyrolysis takes place in the temperature range of 550–900 K; the Fast pyrolysis in the range of 850–1250 K; and the Flash pyrolysis occurring within the temperature range of 1050–1300 K. Slow pyrolysis is also employed, although it is conducted under lower temperatures, taking a longer time to yield appropriate bio-oils [92].

The pyrolysis liquids obtained from different raw material containing triglycerides have different properties and their characteristics are strongly dependent on the reactor type used, the temperature employed, and the operational conditions. The green oil obtained through the pyrolysis of biomass can be used not only as biofuel but also as a raw material to synthesize other value-added chemicals. Furthermore, with this technique, low-quality oils and fats can be employed, contrary to what occurs in the transesterification process. For instance, an environmentally friendly renewable feedstock such as microalgae can be processed into an array of products via pyrolysis, yielding useful chemicals such as light olefins, alkanes, syngas, and biochar, as well as the bio-oils with less oxygen, more hydrocarbons, and higher gross heating values than the bio-oils derived from cellulosic biomass [93,94]. Even sewage sludge has been used to produce green diesel through a pyrolytic process [95].

Despite the cost of raw materials being around 80% of the total cost of bio-oil production, the pyrolysis method is much simpler and less expensive than other methods in producing green diesel, since it is carried out at atmospheric pressure and does not need hydrogen as a co-reagent, as abovementioned [25,96]. In summary, to produce green diesel through triglycerides, any of the three types of pyrolysis have lower costs than conventional transesterification. In addition, a wide variety of raw materials can be employed, including microalgae or waste materials, such as waste cooking oils [97–100].

### 3.2. Catalytic Cracking or Deoxygenation

Diesel-like hydrocarbons can also be obtained by the triglyceride catalytic cracking, comprising the deoxygenation (DO) or elimination of the oxygen atoms, obtaining hydrocarbon molecules with a lower molecular weight than the original molecules. Figure 6 shows a basic scheme of the triglyceride thermal cracking process in the presence of a catalyst. Three different fractions are generated during the process: the solid fraction is usually called coke, the liquid fraction is called bio-oil, and the gaseous stream is known as biogas. In this respect, the temperature and residence time are the key factors for this process.

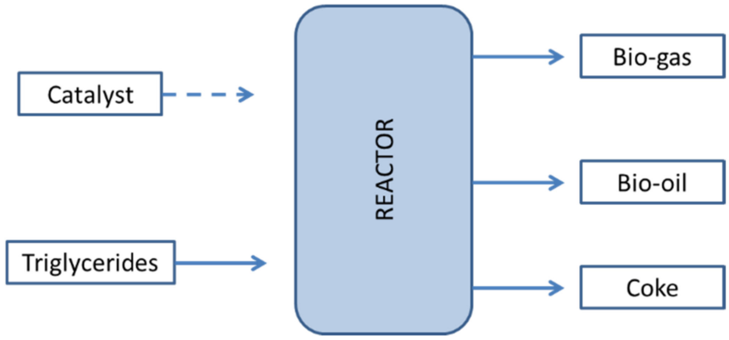


**Figure 6.** General scheme for the pyrolysis process of triglycerides.

Catalytic cracking occurs via a multiple-reaction process including steps such as dehydration, decarboxylation, and decarbonylation. This process produces a liquid fuel with great stability, fantastic combustion properties, and a suitable viscosity for being employed in current combustion engines. In addition, the catalytic process can be performed either in batch or continuous within a favorable temperature range of 300–450 °C [101]. The catalytic deoxygenation is carried out via hydrodeoxygenation, decarboxylation, and decarbonylation reaction pathways. Thus, oxygen is separated from the fatty acid structure of vegetable oils as $H_2O$, $CO_2$, and CO, respectively. The reaction pathways for fatty acid deoxygenation include both liquid and gas phase reactions. The liquid phase reactions consist of direct decarboxylation and decarbonylation, both occurring simultaneously. On the other hand, the deoxygenation of saturated fatty acids involving $H_2$ occurs via indirect decarboxylation and direct hydrogenation for the production of n-alkanes. The deoxygenation of fatty acids via decarboxylation/decarbonylation in the liquid phase produces $CO_2$, CO, $H_2$, and $H_2O$, which further undergo gas phase reactions, such as the methanation of $CO_2$ and CO and water-gas-shift reaction, as is shown in Figure 7 [102]. Decarboxylation/decarbonylation results in an advantageous process for the production of diesel-like hydrocarbons in comparison to hydrodeoxygenation because neither $H_2$ atmosphere nor any sulfide metal catalysts are employed.

Thus, it has been described that catalytic cracking allows for the efficient use of vegetable oils as biofuels through the deoxygenation of triglycerides. In addition, as can be seen in Figure 8, hydrodeoxygenation processes are obtained even in the absence of a hydrogen atmosphere [103]. These renewable hydrocarbon blends are chemically analogous to fossil petroleum-based fuels, having a good distribution (>32% bio-gasoline, >50% green diesel, and <11% heavy fraction). Moreover, they can be fractionated and used in different formulations depending on the types of desired fuels [104]. As the composition of the products may vary due to the solid catalyst used, many investigations evaluate the use of several catalysts. In addition, the coke formation limits the use of heterogeneous catalysts due to deactivation or poisoning, so this requires an additional regeneration process for its reuse, making the entire process very complex. On the other hand, liquid biofuels, depending on their energy density, have fundamental importance in the final energy consumption. In this way, most of the research is being conducted to maximize the amount of liquid bio-products.

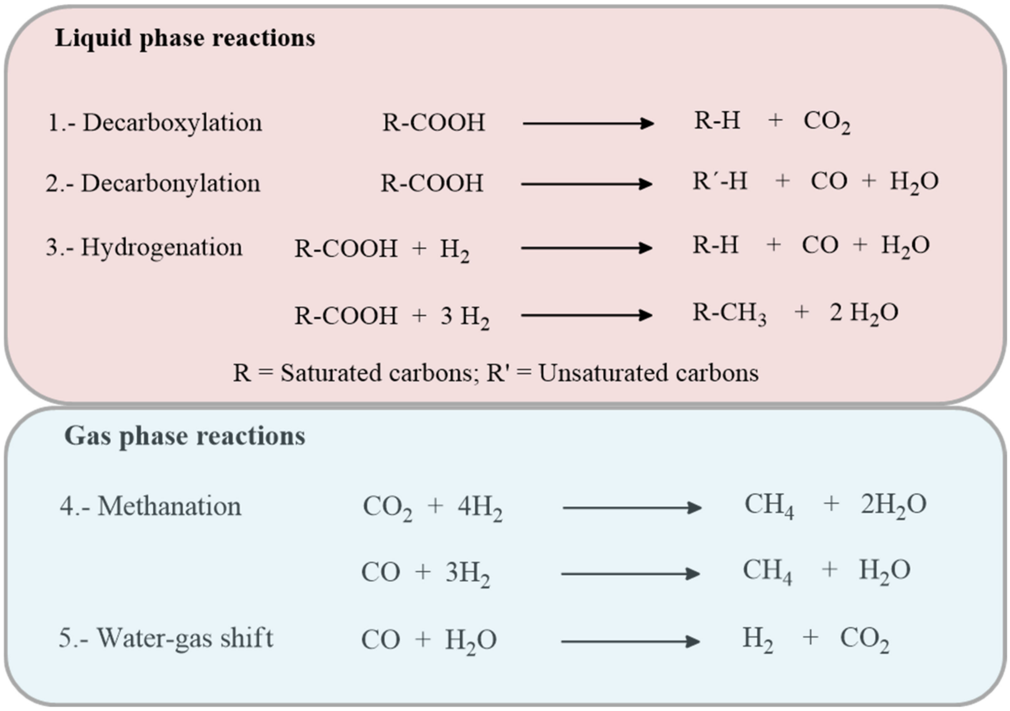


**Figure 7.** General saturated fatty acid deoxygenation reaction steps under inert atmosphere over supported metal catalyst, adapted with permission of Ref. [102]. Copyright 2015, Elsevier.

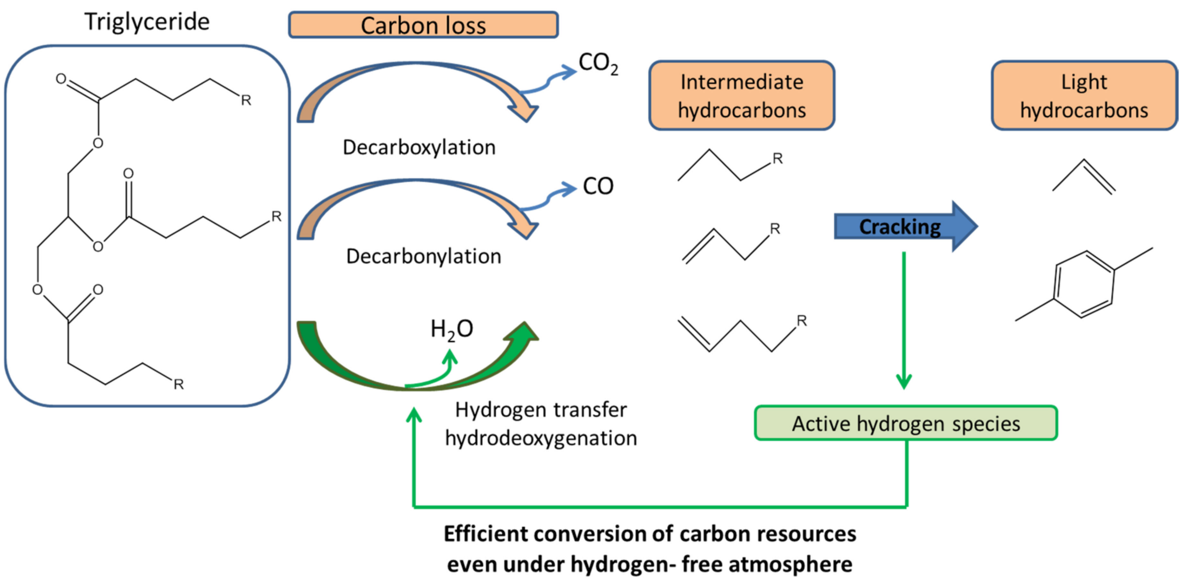


**Figure 8.** General scheme conversion of triglycerides to hydrocarbons in the absence of hydrogen by using a fluid catalytic cracking (FCC) process involving hydrogen transfer reactions, "Reprinted (adapted) with permission from Shimada et al. *Ind. Eng. Chem. Res.* **2017**, *56*, 75–86. Copyright 2017 American Chemical Society [103].

In this respect, many different solid catalysts have been described, including oxides, zeolites, mesoporous materials, and their composites, as well as commercial FCC catalysts [104–109]. Interestingly, the deoxygenation paths can be regulated by altering the synthesized methods of the same catalysts [110].

Among the catalysts tested, metal-supported mesoporous materials with a small particle size have been considered as optimum catalysts for the production of diesel-like hydrocarbons via the deoxygenation of fatty acids [89,111–115]. Furthermore, this process exhibits a higher selectivity to diesel-like hydrocarbons than that to fatty acid ester. The deoxygenation process, catalyzed by noble-metal-supported catalysts and employing a small amount of $H_2$, is recommended to obtain a higher yield of diesel-like hydrocarbons than that employing atmospheres rich in hydrogen, due to the formation of coke being favored at those conditions [111]. In the absence of hydrogen, the deoxygenation of triglycerides usually gives $CO_2$, CO, and $H_2O$, where, through the deoxygenation reaction paths, i.e., decarboxylation yielding $CO_2$ and decarbonylation yielding CO, it results in a partial loss of the carbon amount contained in the triglyceride feedstock [116,117]. However, hydrodeoxygenation, yielding $H_2O$, can convert most of the carbon content in the feedstock to hydrocarbons [118–120].

Regarding the production cost of green diesel, it is assumed that it is highly dependent on the synthesis procedure. The removal of oxygenate-bonded compounds via deoxygenation under a hydrogen-free atmosphere is more economic than hydrodeoxygenation and pyrolysis. The reason why hydrodeoxygenation is costly lies in the high consumption of hydrogen during the process. Regarding this part, pyrolysis has a lower cost, although the hydrocarbon product is mainly composed by light fractions. In addition, olefins could also be problematic, because they are associated with a lower stability due to the potential formation of gums or insoluble materials. Thus, to saturate the double bonds, the hydrorefining process or direct hydrocracking could be also an option.

In summary, despite the fact that the industrial application of pyrolysis and/or catalytic cracking still has some obstacles to overcome, the current refineries can be suitable for upgrading these green fuels production.

### *3.3. Catalytic Hydrocracking or Hydrodeoxygenation*

In addition to the pyrolysis or cracking processes, hydrocracking or hydrodeoxygenation constitutes a very suitable methodology to convert fats and oils into fuels similar to fossil diesel [118]. One of the main strengths of this procedure is that it can be carried out in the currently existing oil refineries [121–123]. Some results indicate that green diesel production by catalytic hydroprocessing, located in a petroleum refinery, appears to be the most cost-effective option compared to conventional biodiesel [124].

In this procedure, hydrocarbons, mainly n-paraffins, are obtained from triglycerides. The reaction temperatures range from 300 to 450 °C and the hydrogen pressures are above 3 MPa. Furthermore, CO, $CO_2$, and water are obtained as by-products. The hydroprocessing of triglycerides involves the hydrogenation of the double bonds of the side chains, the hydrogenation of the double bonds of the fatty acids, and the removal of oxygen in esters bonds [125]. The reactions involved in hydroprocessing can be classified into two groups: (a) hydrotreating and (b) hydrocracking. The hydrotreating of vegetable oils leads to C15–C18 hydrocarbons, which is so-called "green diesel", "renewable diesel", or "bio-hydrogenated diesel".

#### 3.3.1. Hydrotreating

As can be seen in Figure 9, the first step consists of the hydrogenation of double bonds of triglycerides. The removal of oxygen in the form of $CO_2$ (decarboxylation) and $H_2O$ (dehydration) occurs in the next step. This suggests that the hydrodeoxygenation requires a large amount of hydrogen due to the additional hydrogenation of double bonds existing in the triglycerides, attaining the total hydrogenation, which yields hydrocarbons and water as the only reaction products, since all oxygen atoms are eliminated as water.

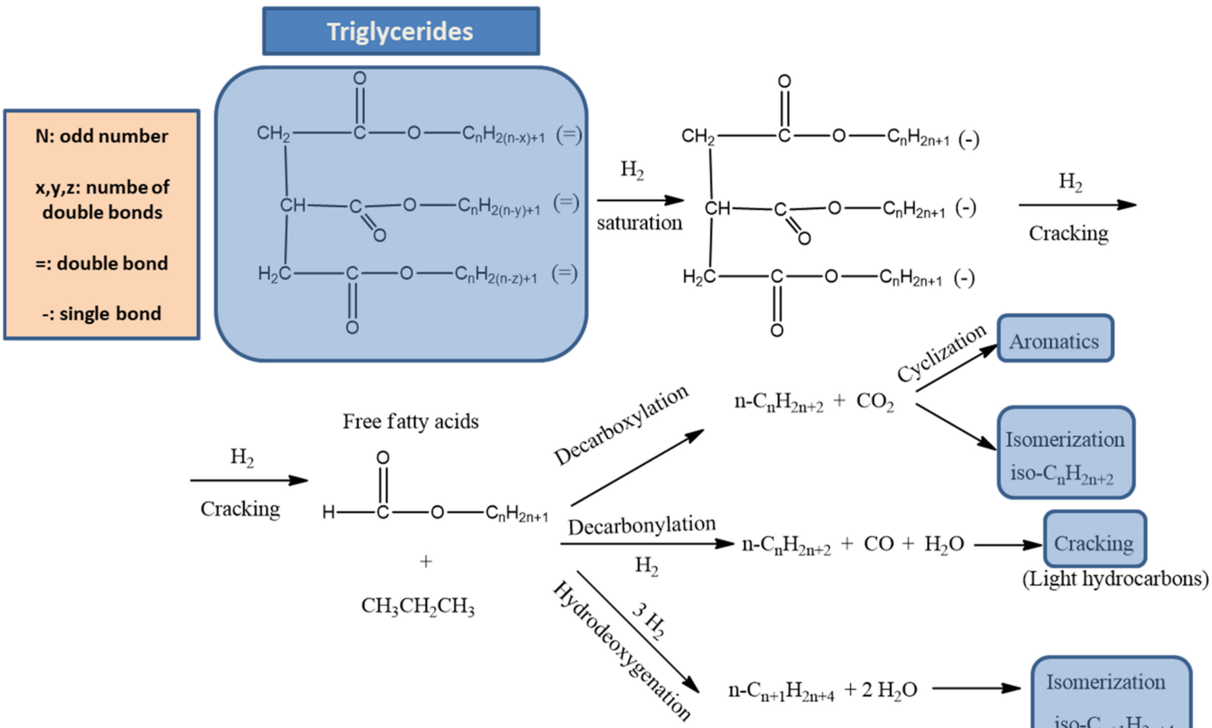


**Figure 9.** Reaction pathways of triglycerides reactions over hydrotreating catalysts, reproduced with permission of Ref. [126]. Copyright, 2014 Elsevier.

In the last decades, several heterogeneous catalysts capable of transforming vegetable oils into alkanes through a hydrotreating process have been described [127]. These hydrocarbons exhibit boiling points in the range of gasoline or diesel and, therefore, they can be used as fuels without any modification. These green fuels can be classified as naphtha, jet fuel, and diesel; see Figure 10 [90]. Green fuels are obtained from triglycerides by using the same process currently used in the hydrotreating of vacuum gas oil [128–132]. Thus, there is an increasing interest in developing the best catalytic systems, as well as the most favorable operating conditions with the most favorable techno-economic conditions, taking advantage of the facilities currently existing in refineries [133–139].

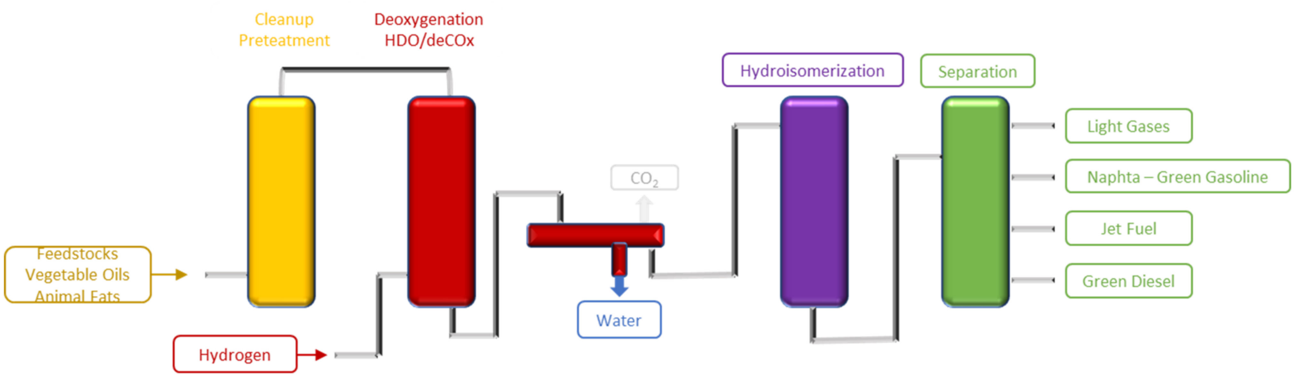


**Figure 10.** Green diesel production process by hydrotreatment of vegetable oils in a biomass hydroprocessing plant. Adapted from [90].

### 3.3.2. Hydrocracking

Regarding the hydrocracking or (hydro)decarboxylation process, all of the oxygen atoms in the triglyceride molecules are eliminated as carbon dioxide, so only hydrocarbons with odd carbon atom numbers can be obtained from the fatty acids. A hydro-prefix is employed to point out that hydrogen is involved in the reaction. Thus, it has been proposed that hydrogen is needed to break the fatty acid moiety loose from the triglyceride. Once the fatty acids are released, they undergo a subsequent decarboxylation step to yield hydrocarbon and $CO_2$ [140]. Both reaction pathways are schematically depicted in Figure 11.

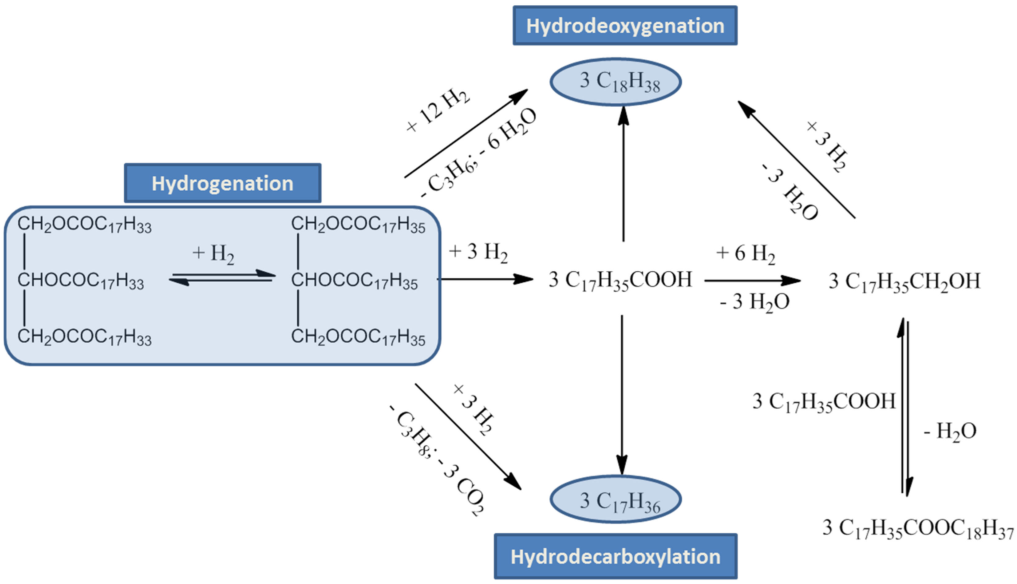


**Figure 11.** A schematic view on the transformation of triglycerides into hydrocarbons under hydrotreating conditions.

In contrast to that occurring in the hydrotreating process, where the reaction occurred on the metal acid sites, in the hydrocracking process, the reaction is carried out on acid sites of amorphous supports. Therefore, by modulating the balance between the acidic and hydrotreating centers in the catalyst, it is possible to achieve hydrocarbons with a boiling point in the range of jet fuel or gasoline. Moreover, the acidity of catalysts also increases the isomerization degree of the molecules, thus boosting the properties of the green liquid fuels, such as a lower pour point and a higher octane number.

Accordingly, various studies have shown that renewable liquid alkanes can be obtained by treating mixtures of vegetable oils and heavy vacuum gas oils (HVO) in hydrogen streams over conventional catalysts employed in the same hydrocracking units. These liquid hydrocarbons exhibit appropriate characteristics, such as a low acidity index, adequate density, viscosity, and a high cetane index. In addition, different by-products are obtained during the process, mainly consisting of hydrogen ($H_2$), CO, $CO_2$, oxygen ($O_2$), nitrogen ($N_2$), hydrogen sulfide ($H_2S$), methane ($CH_4$), and C2–C6 hydrocarbons. Despite quite a lot of by-products being obtained, the yield of liquid hydrocarbons is significantly high, at around 80% by weight [141–147]. In summary, the fossil fuel hydrotreatment process, initially developed to reduce the diesel sulfur amount to fulfill the specification of each country, has also been applied for obtaining a high-quality diesel biofuel from vegetable oils or animal fats [148–153].

Considering all of the advantages provided by the application of cracking treatments of vegetable oils, the research in this field has grown in recent decades, as can be seen in Figure 12. It is worth noting the growing relevance in the application of cracking techniques to produce biofuels for use in aviation [137,154–162]. Thus, the environmental impact and the dependence on fossil fuels in the aeronautical sector have promoted the demand for alternative and greener fuels. In this respect, while, in road transport, several biofuels or electricity can be used, in aviation, only high-quality paraffinic biofuels can be currently considered. Thus, biomass must be transformed into hydrocarbons that are fully compatible with the existing fossil fuel systems, so the implementation of renewable alternative green fuels is currently the main challenge for this sector. This current interest is shown in the high number of publications devoted to research on aviation green fuels production processes, as is also collected in Figure 12.

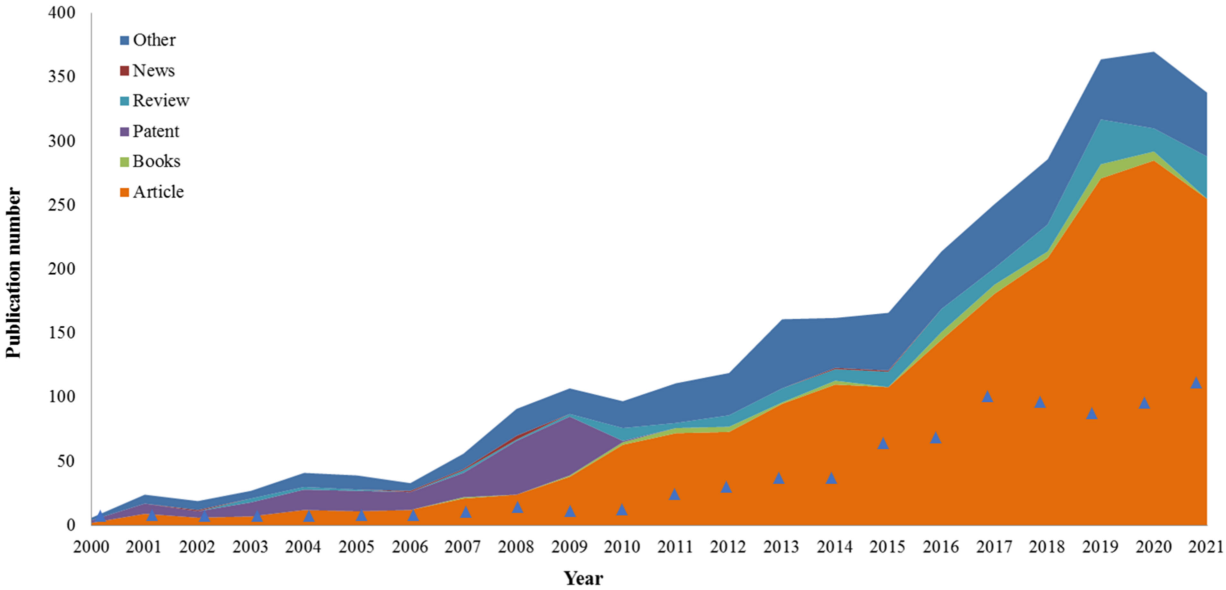


**Figure 12.** Publications found in the Web of Science database by the keywords "green diesel" from year 2000 to 2022. (Blue triangle indicates the number of publication found with the keywords "biofuel in aviation").

At present, short-term solutions, such as blending biofuels with jet fossil fuel, are gaining strength. With this in mind, the obtention of greener fuels that allow for a reduction in greenhouse gases and pollutant emissions without any significant changes in the existing fleets of the aviation companies is gaining strength. In fact, green diesel has the same chemical properties as fossil diesel, so it can be used in the current tanks, pipelines, trucks, and pumps without important infrastructure changes. Accordingly, during the last years, several companies supplying aviation fossil fuels have shown interest in the hydroprocessing of lipid feedstocks to produce renewable green liquid fuels. However, some technologies have already started to be commercialized for this purpose.

In this respect, UOP Honeywell Co. (Chickasaw, AL, USA), a refining technology company, is offering an alternative process to obtain green fuels from various vegetable oils and fats, consisting of converting the non-edible vegetable oils to green diesel [163]. Haldor Topsøe has also developed a proper hydrotreating technology, designated HydroFlex, to produce renewable fuels such as green diesel and jet fuel from non-edible raw grease material [164].

Similarly, many other hydroprocessing plants around the world are currently boosting jet biofuels production with several companies, such as SkyNRG Fly Green Fund in the Nordics, Project Solaris in South Africa, Initiative Towards sustainable Kerosene for Aviation (ITAKA) project in Europe, and Commercial Aviation Alternative Fuels Initiative (CAAFI) Petrobras, ConocoPhillips, Haldor Topsøe, or BP [165]. In general, they all have adopted a co-processing method, where the biofuel is directly mixed with petroleum feedstocks. In this way, after the hydrotreating of the mixture, a fuel product with a lower sulfur content is obtained, reducing the carbon footprint.

Likewise, various studies based on the technological preparation of biofuels have shown that the hydroprocessing of fatty acids and esters (HEFA) route has many advantages with regard to the production facilities and final properties of the biofuels, being one of the four alternative fuels approved in the ASTM D7566, with a maximum blending proportion allowed of 50% [166].

Despite the fact that the production costs of aviation biofuels are higher than conventional fossil fuels, limiting their use on a commercial scale, there is a great number of companies producing renewable fuels with a growing market, so it seems that biofuel for aircraft has a promising economic future [167].

For a better comparison between green diesel, biodiesel, and fossil diesel, some of the most important data have been collected in Table 1.

**Table 1.** Comparison between green diesel, biodiesel, and fossil diesel.

| Properties | Green Diesel | Biodiesel EN 14214 | Fossil Diesel |
|---|---|---|---|
| Cetane number | 75–90 | 50–65 | 40–55 |
| Energy density, MJ/kg | 44 | 38 | 43 |
| Density, g/mL | 0.78 | 0.88 | 0.83–0.85 |
| Cloud Point, °C | −10 | 20 | −5 |
| Lubricity [a] | >700 | - | 226–354 |
| Energy Content, BTU/gal | 123 K | 118 K | 129 |
| Sulfur | <10 ppm | <5 ppm | <10 ppm |
| NOx emissions [b] | −10 to 0 | +10 | Baseline |
| Viscosity, $mm^2/s$ | 2–4 | 2.9–11 | 1.9–4.1 |
| Global warming, $gCO_2eq/MJ$ | −7.32 | 61.35 | 79.93 |
| Acidification, $gSO_2eq/MJ$ | 0.396 | 0.7 | 0.547 |
| Ozone layer depletion, mgCFC-11eq/MJ | 0.003 | 0.006 | 0.012 |

[a] Measure by wear scare diameter (μm); [b] Percentage in comparison to fossil diesel. Data taken from [90,168–170].

### 3.4. Environmental and Economic Impact of Green Diesel

Considering the green diesel emissions in internal combustion engines, the company Neste has reported the results of exhaust emission tests that have been performed in trucks, buses, and also in passenger cars, some with neat green diesel and some with a blend of 85% green diesel and 15% petroleum diesel. The results showed that those engines operating with green diesel reduced the emissions of CO, $CO_2$, unburned hydrocarbons (HC), nitrogen oxides (NOx), and solid particulates [90,170]. CO emissions were, on average, 27, 38, and 45% lower than the EN 590 levels in the case of trucks and buses. The reduction in CO usually means higher $CO_2$ due to a more complete combustion, but Neste claims that $CO_2$ emissions were also reduced due to the higher H/C atomic ratio of the green diesel fuel. Therefore, most of the communications published to date manifest a reduction in greenhouse emissions using green diesel in comparison to those obtained with biodiesel or fossil diesel.

Regarding the economic issues, despite the fact that most of the papers are focused on the chemistry and engineering behind these (bio)fuels, companies are already seeing the economic benefits of switching to renewable fuels. According to a recent study [168], a green diesel fuel tested in a Class 8 truck reduced the lifecycle emission by 66%, saving 1217 t of carbon after one million miles. According to the authors, approximately USD 0.021/mi—USD 0.015/mi are saved from reduced exhaust replacement parts and downtime spent clearing diesel particulate filters (DPFs), USD 0.005/mi from a 75% oil cost reduction, and the remainder resulting from reducing the amount of diesel particulate filters (DPFs) required.

Despite these data seeming insignificant, saving USD 0.021/mi using renewable diesel, considering that there are around three million of these trucks just in US roads, each truck would have saved an average of USD 1317.77/year, or USD 5.15 B/year in savings. In terms of emissions reductions, if all Class 8 trucks had used this biofuel, it would have saved more than 297 million metric tons of carbon dioxide ($CO_2$) per year according to the U.S. Energy Information Administration.

## 4. Biodiesel-like Biofuels

Another emerging alternative to obtain conventional biodiesel, with a higher atom efficiency than transesterification, consists of obtaining some derivatives of glycerol in the same transesterification process, i.e., the glycerol is integrated into the blend as a derivative. Thus, the atomic yield increases to 100%, and, above all, the separation and cleaning of residual glycerol dissolved in the FAME mixture are avoided. To achieve this, some acyl acceptors (ester molecules) instead of short chain alcohols must be employed. Hence, the corresponding glycerol ester is obtained together with the FAME (or FAEE). The reaction products are constituted by lipophilic compounds completely miscible with fossil fuels, attaining a new biofuel that is very similar to biodiesel, but avoiding the presence of free glycerol, as aforementioned [171,172].

It is also interesting to note that these processes that avoid the production of glycerol, using other acyl donor compounds instead of methanol, can be performed with the same catalysts as those employed in the conventional transesterification processes, such as several acid or alkali catalysts, under homogeneous or heterogeneous conditions, as well as with several free or immobilized lipases, or without a catalyst, under supercritical conditions.

### *4.1. Biodiesel-like Biofuels Integrating the Glycerol as Glycerol Triacetate*

To improve the biodiesel manufacturing process, the Industrial Chemistry Research Institute patented a new type of biofuel by the interesterification of triglycerides with methyl acetate in the presence of a strong acid catalyst [173]. The reaction products consist of a mixture of FAMEs and glycerol triacetate (triacetin) and was called Gliperol (Figure 13) [173–175].

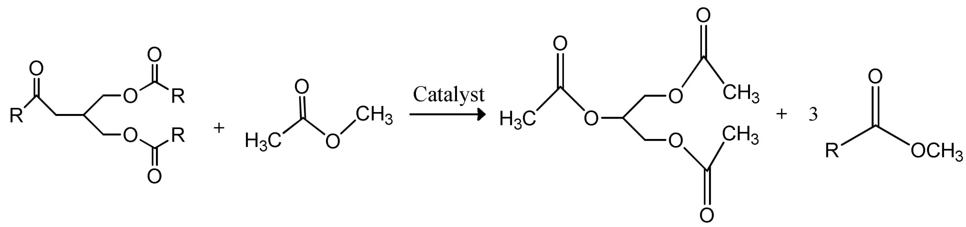


**Figure 13.** General reaction scheme of Gliperol® production, obtained by interesterification reaction of a triglyceride molecule with methyl acetate, obtained by a conventional catalyst, producing a mixture of FAMEs and one molecule of glycerol triacetate (triacetin).

After that, several studies optimized the reaction conditions, i.e., oil/methyl acetate molar ratio (from 1:3 to 1:9), reaction temperatures (from 40 to 200 °C), etc. The results obtained have shown that Gliperol exhibits similar fuel properties to biodiesel, although the process itself improves not only the yield but also factors such as the efficiency and the economic feasibility. Thus, the production cost is around 30–35% lower than the biodiesel production cost, but the incorporation of glycerol in the mixture also reduces the ecological costs associated with the biodiesel purification and processing. In addition, the combustion of Gliperol improves the greenhouse gases emissions [176].

Likewise, different types of catalysts have been investigated, from homogeneous basic catalysts, such as potassium hydroxide, potassium methoxide, and polyethylene glycolate, etc. [177–184], to different heterogeneous catalysts [185–194]. In addition, the use of lipases as biocatalysts, in solvent-free systems [195–202], in ionic liquids [203–207], supercritical conditions [208–219], or ultrasound-assisted interesterification has also been studied [183,208,220–227].

Another possibility is the use of ethyl acetate as an acyl acceptor instead of methyl acetate. In this case, triacetin and a mixture of ethyl esters of fatty acids or FAEE are obtained. This biofuel has been considered as a biofuel-like Gliperol, although with a greener character, since ethyl acetate has a renewable character. However, despite this acyl acceptor exhibiting a similar behavior to methyl acetate in the interesterification process, it is still less studied [228–232].

According to various studies, the presence of triacetin improves the biodiesel behavior, since triacetin act as an anti-knocking additive when it is used along with biodiesel in diesel engines, improving the performance and reducing pollutant emissions [233–246]. Other studies have shown that a 10% by weight of triacetin in triacetin/biodiesel or triacetin/diesel blends exhibited the best results in the engine [247–252]. In addition, the triacetin also exhibits a positive effect on the cloud point and cold filter plugging point [253–259].

Regarding the cost analysis, the fact that Gliperol obtains the triacetin in the same process of FAMEs synthesis makes this product as viable from a technical and economic point of view, according to many studies that support the economic viability of the interesterification of triglycerides [260–266].

Therefore, it can be concluded that Gliperol exhibits clear techno-economic advantages compared to conventional biodiesel, being a suitable methodology to obtain a biofuel with a certain amount of a well-recognized additive, the triacetin, that improves the quality of biodiesel and practically meets the quality standards ASTM D6451 and EN 14214.

### *4.2. Biodiesel-like Biofuels Integrating the Glycerol as Glycerol Carbonate*

Regarding the use of dimethyl carbonate (DMC) as an acyl acceptor, FAME blends with glycerol esters from lipids can be obtained, yielding alternative co-products in biodiesel solutions. This reagent is especially attractive since it is cheap, neither toxic for human health nor for the environment because it is neutral and odorless, non-corrosive, non-toxic, inexpensive, an effective non-flammable solvent, and less toxic than methanol [267–269]. In addition, the reagents to produce DMC, methanol, and carbon monoxides can be obtained from the synthesis gas [270,271]. These facts make DMC one of the most widely used reagents in fine chemistry. Among all of the options available for DMC, the production of glycerol carbonate has gained great attention, since glycerol carbonate is an added-value product that can be employed as an additive to fossil diesel but also allows for the valorization of glycerol [272–279]. However, the use of the residual glycerol obtained in the synthesis of biodiesel does not suppose an adequate solution to the main problem that this glycerol generates, which focuses on the need to intensely clean the biodiesel produced, which must contain a maximum of 0.04% of this impurity, according to standard N14014.

On the contrary, a biofuel produced with DMC and vegetable oil or fats as raw materials must be considered as an alternative to biodiesel because it is totally derived from renewable resources, improves the atomic yield, and avoids the formation of glycerol. Thus, through an interesterification process in which dimethyl carbonate operates as an acyl acceptor, a new biodiesel-like biofuel integrating glycerol carbonate is directly produced, whose abbreviation is DMC-BioD; see Figure 14 [280]. However, FAME and DMC are not the only products obtained in the interesterification of triglycerides with DMC, since cyclic esters of glycerol carbonate fatty acids molecules (FAGC), a small amount of glycerol dicarbonate (GDC), and glycerol carbonate (GC) are also obtained [281]. These mixtures, including glycerol derivative molecules, exhibit physical and rheological properties that allow for their use as a biofuel.

As can be seen in Figure 13, the difference between DMC-BioD and conventional biodiesel is the presence of some amounts of fatty acid glycerol carbonate monoesters (FAGCs), glycerol dicarbonate (GDC), and glycerol carbonate (GC), together with the corresponding FAMEs that constitute biodiesel [282–286]. Regarding the techno-economic analysis of processes for biodiesel coproduction with glycerol carbonate, they all indicate the profitability of the process because not only is the atomic yield improved (the formation of glycerol is avoided) but also the same basic catalysts described in the production of biodiesel, such as KOH, sodium methylate, sodium hydride, some amines, or different alkaline solids, can also be employed for this procedure [287–294]. Moreover, the DMC-

BioD production has also been studied under supercritical conditions [295–298], employing lipases as a biocatalyst [299–306] and even ionic liquids [307–309].

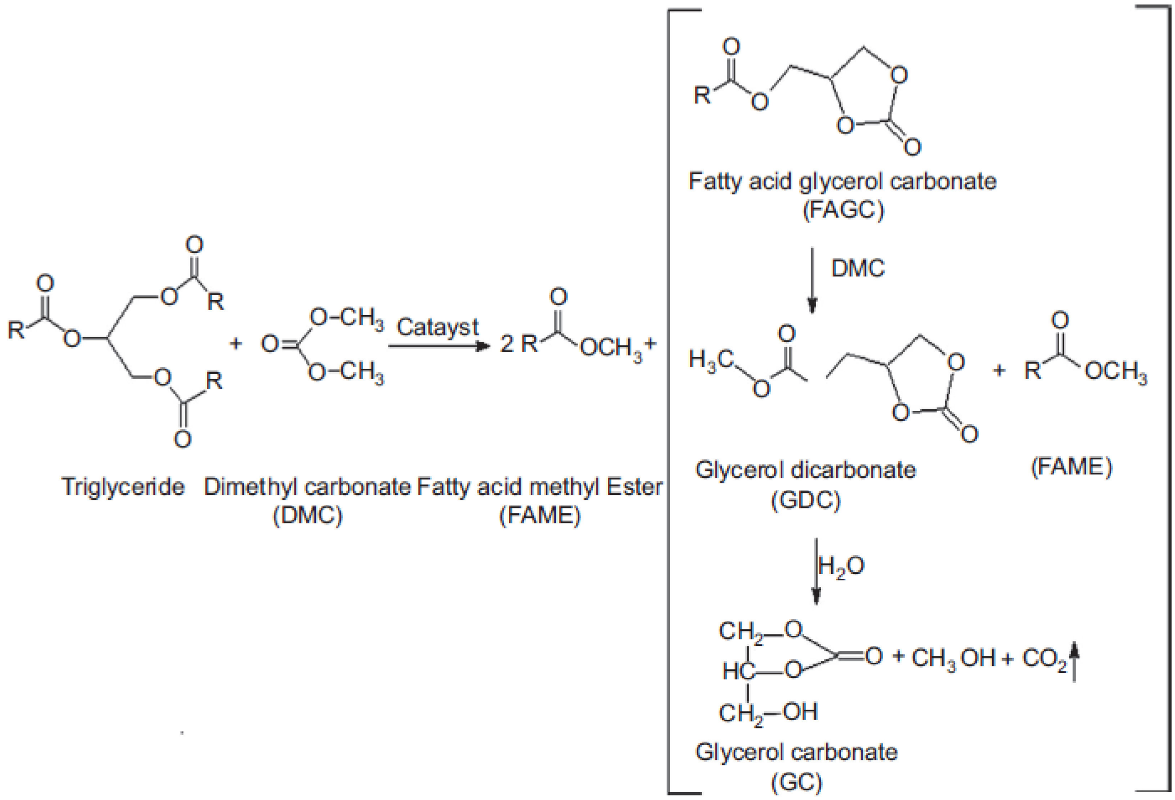


**Figure 14.** Reaction scheme of biodiesel-like biofuel "DMC-BioDs", patented by Polimeri Europa (Italy). Obtained by reacting oils with DMC under alkaline conditions, obtaining a mixture of 2 mol of FAMEs and 1 mol of FAGC.

In summary, the substitution of methanol with DMC in order to produce DMC-Biod instead of biodiesel could simplify the synthesis process and increase the atomic yield by up to 100. In addition, all of the reaction products can be used as biofuel, not being necessary for the separation of nonreacted DMC, because it is an effective additive for diesel engines due to its high oxygen content [310–317].

With respect to diethyl carbonate (DEC) as an acyl acceptor in the substitution of DMC, it must be said that it has been much less investigated, although more analogous results than those with DMC have been obtained, since ethyl esters (FAEEs) can be successfully employed as biofuels [298,318–324].

### *4.3. Biodiesel-like Biofuels Obtained by Incorporating Glycerol as Monoglycerides in the Selective Transesterification Process of Oils and Fats*

Another strategy for the preparation of a new type of biodiesel-like biofuel that also integrates glycerol in its composition has been developed in the last decade. In this case, the reaction takes place through the 1,3-regiospecific transesterification of triglycerides, generating only two equivalent molecules of FAMEs (or FAEEs), maintaining the third fatty acid as monoglyceride (MG). This method was initially applied using lipases as a biocatalyst, given its 1,3-selective character, called Ecodiesel-100® [325]; see Figure 15. At the beginning, several research studies were described using pig pancreatic lipase (PPL), either in free form [326,327] or immobilized [328,329].

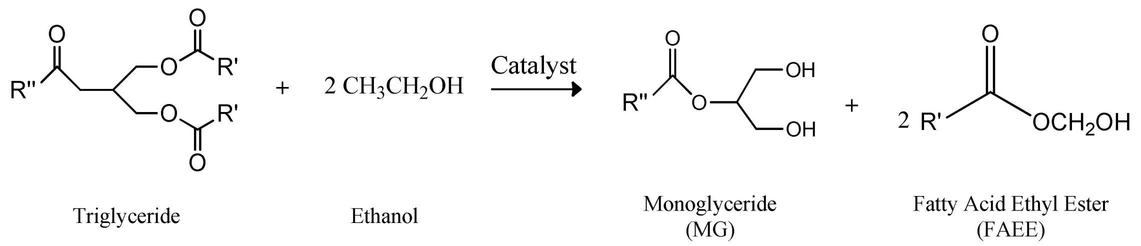


**Figure 15.** Reaction scheme of biodiesel-like biofuel "Ecodiesel-100®", obtained by enzymatic technology patented by the University of Cordoba (UCO).

This reaction allowed for a reduction in the triglycerides' viscosity and also displayed important advantages in comparison with the conventional biodiesel production. Analogously to what happened in the obtention of the abovementioned biodiesel-like biofuels, 100% of atomic efficiency is achieved, avoiding the formation of glycerol. In addition, it is worth mentioning that the operating conditions of the enzymatic process are much softer than those employed for biodiesel production, and acidic or alkaline impurities are not generated, reducing the environmental impact of the process [330]. Moreover, a specific feature of the enzymatic process is the use of ethanol, instead of methanol, so that a blend of fatty acid ethyl esters (FAEEs), together with MGs are obtained.

Ecodiesel, like biodiesel, has excellent advantages, such as renewability, a high octane number, high flash point, good lubricity, low viscosity, high biodegradability, being able to reduce important amounts of the environmental pollutant emissions, and being able to be used in internal combustion engines without any modification [331–334]. In fact, monoacylglycerides (MG) enhance the lubricity of biodiesel, as was demonstrated by recent studies [335–338]. In addition, all of the reagents used in the synthesis of Ecodiesel remain in the final blend that constitutes the biofuel. This represents great simplicity in the synthesis process, since there are no residues or final refining process of biofuel, as well as a total atomic yield of 100%, which provides a cost reduction that results in the technical and economic feasibility of the process.

The main handicap of this procedure is related to the economic difficulty of carrying out this process on an industrial scale, because of the high price of pig pancreatic lipases, even in immobilized form on an inorganic support. That is why, in recent years, a high number of low-cost lipases, both in free and immobilized form, have been tested, demonstrating the technical efficiency of the process [339–349], but without solving the economic difficulties associated with the high cost of the different lipases investigated.

To overcome this problem, some heterogeneous catalysts have also been investigated, e.g., KF [350] or CaO [351,352], as well as homogeneous alkaline catalysts such as sodium methoxide, operating at low enough temperatures, which contribute to a low cost of the procedure [353]. Nevertheless, in order to reach the same selectivity as that obtained with enzymatic catalysis, it is necessary to operate under the kinetic control of the chemical process, i.e., a less basic catalyst than the alkali metals must be employed and/or operate under softer reaction conditions, a lower temperature, and/or a lower concentration of ethanol. In addition, in these cases, methanol can also be used, obtaining the FAMEs blend, together with monoglycerides. According to the results, the weaker surface basic sites of heterogeneous catalysts are strong enough to perform the transesterification of primary alcohols (such as those in positions 1 and 3 of glycerol) but are not enough strong to obtain the methanolysis of secondary alcohols (such as those in positions 2 of glycerol), which is much more difficult to achieve [350–352].

Regarding the behavior of this biofuel in the engine, results obtained with 30% of Ecodiesel in diesel/Ecodiesel blends have shown that there are no differences with pure fossil diesel in terms of power generation, attaining a significant reduction in the emission of pollutants, at around 40% less, although a small increase in fuel consumption was obtained [353].

In summary, Ecodiesel can be obtained with similar catalysts to those conventionally employed in transesterification but operating at much softer conditions. In addition, there is no need to perform any additional purification operation, since glycerol is not produced.

Table 2 collects the technical and economic advantages that the three processes reviewed to obtain a comparison between biodiesel-like biofuels and biodiesel.

**Table 2.** Summary sheet of the pros and cons of different existing methodologies for obtaining biodiesel-like biofuels, integrating the glycerol as a derivative in the same transesterification process. Data taken from references of Sections 4.1–4.3.

| | **Biodiesel EN 14214** | **Biodiesel-like Biofuels** | | |
|---|---|---|---|---|
| | | **Gliperol** | **DMC-Bio** | **Ecodiesel®** |
| Reactive | Methanol or ethanol | Methyl acetate | Methyl carbonate | Methanol or ethanol |
| Catalyst | NaOH or KOH | Acid, basic, or lipases | Basic or lipases | Lipases |
| Products | 3 FAME or 3 FAEE | Glycerol triacetate + 3 FAME | Fatty acid glycerol carbonate | Monoglycerides + 2 FAEE |
| By-products | Glycerol | No waste | No waste | No Waste |
| Separation process and cleaning | Complex | Not needed | Not needed | Not needed |
| Investments facilities | Medium | Low | Low | Low |
| Free fatty acids and/or water in the starting oil | Free fatty acids are transformed to soaps | Free fatty acids are transformed to biofuel | Free fatty acids are transformed to biofuel | Free fatty acids are transformed to biofuel |
| Catalyst cost | Low | Low | Low | Low |
| Environmental impact | High. Alkaline and saline effluents are generated. Wastewater treatment is needed | Low | Low | Low |

## 5. Straight Vegetable Oils (SVO) Blending with Less Viscous and Lower Cetane (LVLC) Biofuels

Pure vegetable oils (SVO) could constitute an ideal alternative to fossil diesel due to the similar properties that they exhibit. In addition, SVOs are renewable, non-toxic, biodegradable, and do not contain sulfur, which could make them very suited to replace fossil diesel. In addition, vegetable oils can be obtained from agricultural or industrial sources, avoiding the costs associated with the transesterification to obtain Biodiesel. The main drawback in the use of SVO is their high viscosity, which dramatically alters fuel spray characteristics, atomization quality, and volatility, leading to severe carbon deposits, fuel injector clogging, and rapid wearing of fuel pump components [26].

To solve this problem, the incorporation of SVO into blends with diesel in optimal proportions could be an option. This method can operate along the current strategy of reducing the level of $CO_2$ emissions as much as possible. Thus, a recent study supports the possibility of using 10% of vegetable oils of different sources in blends with diesel [354].

However, to achieve higher percentages of fossil diesel substitution using SVO, another method consisting of blends of SVO with other lower viscosity biofuel, with the goal of reaching viscosity values similar to fossil diesel, has been proposed. In this way, an effective and inexpensive method to obtain dual biofuels able to be blended efficiently with diesel fossil is being investigated. Thus, blends of a light biofuel a with low viscosity, based on Melaleuca Cajuputi oil (MCO) and SVO, to obtain higher substitution values of diesel fossil, have recently been proposed [355–357].

Since low-viscosity biofuels also have a low cetane number, they are often referred to as low viscosity low cetane (LVLC) compounds. Thus, biofuels composed of alcohol such as methanol, ethanol, or butanol, and plant-based light biofuels such as eucalyptus and pine oil, have been chosen and classified as LVLC fuels [358]. The high-viscosity values of some of these oils, e.g., castor oil (226.2 cSt) and pine oil (1.3 cSt), are balanced with the low viscosity of alcohols, achieving a suitable viscosity value for blending in different proportions and attaining a final viscosity in the range of 2.0–4.5 cSt, which are the values imposed by the EN590 normative. However, these blends exhibit a lower cetane number, so a limit usually exists in which they can be blended in order to be correctly employed in an engine [27]. Analogously to pine and castor oil, many other compounds, such as eucalyptus oil, orange oil, or camphor oil, have been studied as LVLC fuels for use in double or triple blends with biodiesel, or even with fossil diesel [27,359–366]. These low viscous vegetable oils improved the performance of the biodiesel-fueled CI engine, described as operating as biofuels either by themselves or in blends with fossil diesel [367–369]. Moreover, some of these ternary blends (vegetable oil, alcohols, and fossil diesel) have demonstrate that they are able to reduce the emission pollutants [370–378].

In addition to vegetable oils and some other low-viscosity natural products, triple mixtures with molecules obtained by synthesis from renewable commodities are also being evaluated, so they can also be considered as renewable compounds. Thus, diethyl ether (DEE), which can be synthesized from bioethanol, has also been considered as a biofuel. DEE is a butanol isomer that has shown good properties for being blended with diesel, which are as follows: a high cetane number, reasonable energy density, high oxygen content, low autoignition temperature, broad flammability limits, and high miscibility with vegetable oils and diesel fossil. In addition, DEE has been reported as a low-emission renewable fuel and high-quality combustion improver when it is used in blends with diesel fossil and several vegetable oils [366,379–388]. The incorporation of DEE in the triple blend, diesel/vegetable oil/DEE, allows for the substitution of fossil diesel by up to 40% by volume, improving the engine power with fewer emissions, as well as improving the cold flow behavior of fuel.

As with DEE, the possibility of using acetone (ACE) in triple blends with fossil diesel and straight vegetable oils as alternatives has also been investigated. ACE is an oxygenated additive that also complies with the requirements for being blended with vegetable oils and fossil diesel, i.e., acetone exhibits a very low kinematic viscosity that balances the high viscosity of SVOs. Likewise, the oxygen content, the low autoignition temperature, and the very low cloud point and pour point values, make acetone a good candidate for being employed in triple blends. On the other hand, although acetone is currently produced from fossil resources, it can also be obtained from renewable resources, either from ethanol [25] or from cellulose through a typical acetone–butanol–ethanol (ABE) fermentation process. Regarding the results obtained in the engine with a triple blend of diesel/vegetable oil/acetone, in which, acetone is at around 16–18% by volume, a considerable reduction in emissions of air pollutants, as well as a good power engine, were attained. Nevertheless, the fuel consumption was slightly higher than with fossil diesel [389]. Acetone has also been described as a fuel additive in biodiesel–diesel blends [390].

Another organic compound object of study for these purposes is the ethyl acetate (EA). It has been tested in blends of diesel/sunflower oil/EA or diesel/castor oil/EA. The results obtained indicated that triple blends composed of up to 24% of EA in the case of sunflower oil and 36% of EA in the case of castor oil allow for the substitution of 60–80% of fossil diesel, providing engine power values that are very similar to conventional diesel [391]. In addition, the EA properties make it a fuel that is very safe for transportation [392–396].

Diethyl carbonate (DEC) has also been reported as an effective oxygenated additive, lowering soot and NOx emissions with an improvement in the engine performance [397–402]. The main advantage of DEC is that it can be synthesized from bioethanol and $CO_2$, which could contribute to the reduction in the atmospheric $CO_2$ to a large extent [403]. Very recently, DEC has been addressed as a solvent for vegetable oils for use in diesel engines in triple blends [404], as well as dimethyl carbonate (DMC) [405–411]. In addition, it has been tested in blends with biodiesel [412] and in biodiesel/diesel blends [413]. Results have shown that, in all cases, the use of DMC notably improves the engine performance and exhaust emissions from C.I. engines [413]. To move forward in the substitution of fossil fuels with others of renewable character, the strategy has even been applied in triple mixtures with gasoline. In this way, these triple blends allow for the substitution of up to 40% of fossil diesel with sunflower oil, and up to 25% with castor oil, with a significant reduction in the emission of pollutants also being obtained with these triple blends [414,415].

Therefore, we can conclude that, in just a decade, it has been possible to verify a suitable strategy to reduce the viscosity of vegetable oils to the values required by current CI engines by their blending with low-viscosity solvents (LVS). In this way, SVO/LVS/fossil diesel triple blends can be obtained with a suitable composition to comply with regulations of the EN 14,214 standard. In addition, to achieve a higher fossil diesel replacement, compounds derived from renewable sources represent the better option. On the other hand, the use of oxygen-rich compounds as viscosity reducer solvents allows for a better combustion process and reduced emissions. In this respect, up until this moment, some light vegetable oils (orange, camphor, eucalyptus, and pine oil) and higher alcohols (1-propanol, 2-propanol, 1-butanol, 2-butanol, and 1-pentanol), as well as several renewable oxygenated compounds, such as diethyl ether, acetone, ethyl acetate, diethyl carbonate, and dimethyl carbonate, have been described as viscosity reducers of SVOs. Overall, the exhaust emissions were significantly reduced with the use of these blends, resulting in a similar or slightly lower engine performance than that exhibited by conventional diesel. Moreover, the behavior of blends at low temperatures is usually improved by using these less viscous oxygenated compounds.

## 6. Summary and Concluding Remarks

Very recently, ambitious targets were established by the European New Green Deal and the REDII, aiming to achieve a climate-neutral Europe by 2050, with the transport sector being the most critical area to decarbonize. Given the urgency of the deadlines imposed by policy makers, it is time to determine which biofuels have the necessary maturity to be incorporated into this substitution process. In this respect, a wide range of biofuels are under development. Although some of them have been extensively commercialized, the so-called conventional biofuels (starch, sugar-based ethanol, biodiesel, etc.), some others, generally called non-conventional, are far from commercialization; in particular, biofuels produced from lignocellulosic residues (i.e., agricultural and forestry residues, e.g., straws, stoves, bagasse, woody biomass). However, all renewable biofuels are considered important for the long-run decarbonization of the transportation sector due to incompatibilities that other low-carbon fuel, such as electricity or hydrogen, exhibit when being applied to heavy-duty fleets and air transports.

In this respect, most of the research in the literature focuses on the production of biodiesel to supplement petroleum-based diesel. However, as has been discussed in the present review, biodiesel presents techno-economic difficulties that prevent its massive application on an industrial scale, mainly due to the production of glycerol as a by-product. Thus, it is understandable that transesterification is not adequate on its own to take advantage of vegetable oils as substitutes for fossil fuels in ID engines.

Therefore, in this review, only those methodologies that can be incorporated immediately into the process of substituting fossil fuels with biofuels obtained from vegetable oils are addressed. We do not consider the “drop-in” fuel production from lignocellulosic sources as viable, which will probably be an important procedure in the medium-long term, but cannot be applied immediately.

Independently of the current energy scenario, the complete replacement of petroleum-derived fuels with biofuels is practically impossible in the short-medium term, since the production of all of the raw materials needed is impossible, not only due to economic difficulties and the high prices of vegetable oils but also because of the shortage of agricultural land suitable for it. Thus, in this review, three alternatives to obtaining biofuels that do not produce glycerol have been evaluated and compared with the conventional biodiesel, as can be seen in Table 3.

**Table 3.** Summary sheet of the main technologies currently available to produce renewable liquid fuels from vegetable oils, able to operate correctly in current internal combustion engines, as well as in spark-ignition engines or/and aviation.

| Parameters for Comparison | Type of Biofuel | | | |
|---|---|---|---|---|
| | **Biodiesel** | **Biodiesel-like Biofuel** | **Green Diesel** | **LVLC Blended with Vegetable Oils** |
| Atomic efficiency | 85% | 100% | 85% | 100% |
| By-products/waste generation | Dirty glycerol (15%) | No wastes | CO, $CO_2$, and $H_2O$ (15%) | No wastes |
| Cleaning process | Complex, high-water consumption | Not needed | Not needed | Not needed |
| Cetane index | Slightly lower than diesel | Slightly lower than diesel | Like diesel | Slightly lower than diesel |
| Lubricity | High | High | Low | High |
| Industrial production | Complex | Simple | Simple | Very simple |
| Environmental impact | High | Low | Low | None |

In addition, these advanced methods (biodiesel-like biofuel, green diesel, and/or LVLC solvents blended with vegetable oils) could be applied without any problem from a technical point of view. It is a matter of discriminating which of them are more viable from a techno-economic point of view. Another important aspect is the need to produce adequate fuels for air transport. In this respect, hydrocracking would constitute the ideal solution, since it can simultaneously produce high-quality biojet and biogasoline.

Finally, it can be concluded that, according to the checked bibliography in this comprehensive review, any of the alternative methods proposed in Table 2 are able to compete advantageously with conventional biodiesel in order to achieve the gradual replacement of fossil fuels by some other fuels of renewable nature, operating in the car fleet currently in use. Thus, any of the selected advanced biofuels must always be within the acceptable limits prescribed by ASTM D 6751, currently in force for conventional fossil diesel fuel. However, publications from the academic world have not yet become aware of this fact. Thus, it can be verified in Figure 2 that the increase in the publications in the Web of Science database with the keywords "biodiesel" from year 2000 to 2021 grows continuously year after year, without noticing any change in this trend. This is despite the fact that, from 2015, there was a sharp increase in the number of publications dedicated to the study of so-called green diesel, as shown in Figure 12. In addition, we can verify the sudden eruption from this date, which was at the beginning of several commercial plants for the production of renewable diesel all over the world; see Figure 5. Therefore, it seems that the process of replacing fossil fuels continues, but that conventional biodiesel is no longer the chosen candidate for this process.

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