catalysts

Review

Bio-Derived Catalysts: A Current Trend of Catalysts Used in Biodiesel Production

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Abstract: Biodiesel is a promising alternative to fossil fuels and mainly produced from oils/fat through the (trans)esterification process. To enhance the reaction efficiency and simplify the production process, various catalysts have been introduced for biodiesel synthesis. Recently, the use of bio-derived catalysts has attracted more interest due to their high catalytic activity and ecofriendly properties. These catalysts include alkali catalysts, acid catalysts, and enzymes (biocatalysts), which are (bio)synthesized from various natural sources. This review summarizes the latest findings on these bio-derived catalysts, as well as their source and catalytic activity. The advantages and disadvantages of these catalysts are also discussed. These bio-based catalysts show a promising future and can be further used as a renewable catalyst for sustainable biodiesel production.

Keywords: alternative fuel; bio-derived catalyst; biodiesel; ecofriendly benefit; (trans)esterification

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1. Introduction

Global industrialization leads to the extensive use of fuel-based energy for transportation, which consequently causes the depletion of fossil fuels and global warming. Therefore, renewable fuels are considered an alternative energy to solve the problem of fuel depletion and environmental pollution. Biodiesel, a biomass-derived fuel, is a promising bioenergy, which is increasingly produced worldwide to replace fossil fuels because of its combustion efficiency, compatibility with diesel engines, and low carbon dioxide emissions [1,2]. As a result, studies have been focusing on developing an efficient approach for biodiesel production.

Biodiesel is mainly synthesized from oils/fat (derived from plants, animals, and microorganisms) through the (trans)esterification process [3,4]. The efficiency of this process is mainly affected by several factors including the quality of feedstock (level of free fatty acids), the type of acyl acceptor (e.g., alcohols or methyl acetate), and the type of reactions (e.g., noncatalytic reaction, chemical-catalyzed reaction, and enzymatic reaction) [5,6]. To enhance the reaction efficiency, most studies focused on developing efficient catalysts for the (trans)esterification reaction. Consequently, different types of catalysts, such as alkali catalysts, acid catalysts, and enzymes have been studied [7,8]. Commonly, chemical catalysts (such as KOH, NaOH, H_2SO_4, and HCl) are used for biodiesel production processes [9]. Although these chemical catalysts efficiently catalyze the reaction, they retain several limitations regarding their reusability, negative effect on the environment, and complicated purification steps in the downstream process [2]. Therefore, studies have shifted to using bio-derived catalysts for biodiesel production.

Bio-based catalysts have increasingly attracted attention for biodiesel production due to their availability and environmentally friendly nature [8]. Those catalysts are derived from natural sources and divided into 3 types: alkali catalysts, acid catalysts, and biocatalysts (enzymes). Each type of catalyst has its advantages and disadvantages for biodiesel production. To synthesize these catalysts, various biomass materials and synthesis methods have been reported [8]. Catalysts derived from different sources possess different catalytic activity. This review aims to summarize the bio-derived catalysts for biodiesel production. The natural sources used for catalyst synthesis are reported. The advantages and disadvantages of each type of catalyst are also discussed in this review.

2. Bio-Derived Alkali Catalysts for Biodiesel Production

Biodiesel is commonly produced through transesterification of vegetable oil or animal fat with short-chain alcohols (e.g., methanol and ethanol) in the presence of liquid/homogenous alkali catalysts (e.g., NaOH, KOH). The homogenous alkali catalyst-catalyzed transesterifications achieve high biodiesel yield within a short reaction time (30–45 min) and proceed at atmospheric pressure [10]. However, emulsification, difficulty in separation of the catalyst after reaction, and generation of excess wastewater are major problems associated with those catalysts [11]. To address these issues, heterogeneous alkali catalysts have been increasingly developed as alternative catalysts for biodiesel production. The use of heterogeneous alkali catalysts for biodiesel production simplifies the operation, easily removes and recovers catalysts from the reaction mixture, results in better biodiesel refining, and lowers environmental problems as compared to homogenous catalysts [10]. Furthermore, heterogeneous alkali catalysts can be synthesized from various cheap materials, thus reducing the production cost [12]. However, heterogeneous catalytic reactions are typically time-consuming and require a higher reaction temperature than the homogenous alkali catalyst-catalyzed transesterification due to diffusion problems owing to the formation of three phases of the reactants (methanol–oil–solid catalyst) [8]. Therefore, studies have focused on developing solid catalysts with high catalytic activity to produce biodiesel under mild reaction conditions and short reaction time [13].

Alkali catalysts derived from biomass have attracted considerable interest in biodiesel production due to their ecofriendly nature, low cost, and the availability of biomass as a material for the synthesis of catalysts. Moreover, the use of biomass for catalyst synthesis can solve the environmental problems caused by surplus biomass waste. Therefore, various types of bio-derived alkali catalysts have been studied for transesterification reactions. Biomass-derived calcium oxide (CaO) is one of the most promising solid alkali catalysts used for biodiesel production [14]. The availability of CaO has been recorded in different types of waste/low-cost materials, mainly from animal-derived biomass, including eggshell [15], *Turbo jourdani* shell [16], oyster shell [17], *Pomacea canaliculata* shell [18], *Turbonilla striatula* shell [19], crap shell [20], mussel (*Perna varidis*) shell [21], *Grooved razor* shell [22], conch shell [23], *Malleus malleus* shells [24], and animal bone [25]. The catalytic activities of the synthesized CaO catalysts vary, depending on the materials used and the synthesis method. Among the materials used, eggshell seems to be one of the most suitable materials and attracted extensive investigations for the synthesis of CaO catalyst since it contains a high level of $CaCO_3$ and is easy to obtain [26]. Yaşar [26] reported the synthesis of a CaO catalyst from waste eggshell. The transesterification reaction catalyzed by eggshell-derived CaO resulted in 96.81% biodiesel yield, compared to 95.12% biodiesel produced by commercial CaO, under the reaction conditions of 4% catalysts, 1 h reaction time, and 60 °C reaction temperature [26]. The synthesis method also seems to affect the catalytic activity of the catalyst; therefore, catalysts are derived from the same materials, but they exhibit various catalytic activities (yielding 90–97% biodiesel) [26,27]. For example, Gollakota et al. [28] used eggshell-supported pyrolysis residue as a solid alkali catalyst for transesterification of waste cooking oil (WCO). This study also compared the catalytic activity of unsupported eggshell catalysts and the supported catalyst. Results revealed that the biodiesel yield reached over 95% at 65 °C using 10% supported catalyst with a methanol to oil molar ratio of 12:1 in 3 h. In comparison with unsupported eggshell catalysts, the synthesized catalyst shows improved surface area and catalytic activity [28]. Goli et al. [29] reported biodiesel production from soybean oil using a CaO catalyst that was derived from chicken eggshell waste, yielding 93% biodiesel, whereas Kirubakaran et al. [27] also used a waste chicken eggshell-derived CaO catalyst for biodiesel production and reported 90.41% biodiesel yield under optimal reaction conditions.

In addition to CaO, other calcium (Ca)-based catalysts synthesized from biomass have been reported as potential alkali catalysts for biodiesel production [15,30]. Gupta et al. [15] synthesized eggshell-CaDG catalyst for biodiesel production. The transesterification of WCO was conducted to compare the catalytic activity of eggshell-CaOC-H-D and eggshell-CaDG. Under optimized reaction conditions (catalyst loading of 1.5%, reaction time of 50 min, methanol:oil molar ratio of 10:1, temperature of 60 °C, and agitation speed of 300 rpm), the eggshell-CaDG catalyzed reaction provided 96.07% biodiesel. The eggshell-CaDG demonstrated higher catalytic activity than eggshell-CaOC-H-D (93.10% biodiesel yield under the optimal reaction conditions of temperature of 65 °C, catalyst loading of 3%, methanol:oil molar ratio of 12:1, 400 rpm, and reaction time of 90 min) [15]. Both catalysts (eggshell-CaOC-H-D and eggshell-CaDG) could be reused for up to five cycles for biodiesel production [15]. In addition, alkali catalyst obtained from the plant materials through calcination method such as calcined *Musa acuminata* peduncle [30], calcined waste cupuaçu (*Theobroma grandiflorum*) seeds [31], calcined banana peel [32], calcined elephant-ear tree pod husk [33], calcined kola nut husk pod [34], calcined *Brassica nigra* plant [35], ZrO_2-supported bamboo leaf ash [36], calcined *Sesamum indicum* ash [37], calcined *Tectona grandis* leaves [38], calcined fig (*Ficus carica)* leaves [39], calcined ginger (*Zingiber officinale*) leaves [40], calcinated *Carica Papaya* stem [41], and calcined *Musa balbisiana* Colla peel [42], also efficiently converted oil into biodiesel with conversion rates of higher than 95.1% (Table 1).

Activated carbon-based and biochar catalysts derived from biomass are another type of alkali catalyst, which shows promise for biodiesel production. These catalysts are mainly synthesized from plant materials through the carbonization process. Recently, Naeem et al. [43] reported the use of KOH/corn cob activated carbon catalyst for biodiesel production with a biodiesel yield of 97.8%, whereas the nano-bifunctional catalyst from rice husk resulted in 98.6% biodiesel yield [44]. Due to their high catalytic activity, the synthesis of these bio-based activated carbon catalysts and other bio-based alkali catalysts is still an objective of investigation for biodiesel production.

Table 1. Several bio-derived alkali catalysts for biodiesel production.

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Eggshell-derived CaO	Rapeseed oil	4	9:1	60	60	95.12	3/93.24	[12]
Chicken eggshell-derived CaO	WCO	1.5	10:1	50	60	96.07	5/81.15	[15]
Chicken eggshell-derived CaO	Chicken fat	8.5	13:1	300	57.5	90.41	5/85	[27]
Chicken eggshell-derived CaO	WCO	10	12:1	180	65	95	-	[28]

Table 1. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Chicken eggshell-derived CaO	Soybean oil	7	10:1	180	57.5	93	4/75	[29]
Eggshell-derived CaO	*Phoenix dactylifera* L. seed oil	5	12:1	90	65	93.5	6/>80	[45]
Eggshell-derived CaO/SiO_2	WCO	8	14:1	60	60	91	10/>85	[46]
Chicken eggshell-derived CaO	*Jatropha curcas* oil	2	6:1	120	90	98	-	[47]
Chicken eggshell-derived CaO	*Chlorella vulgaris* biomass	1.39	10:1	180	70	92.03	3/>85.2	[48]
Fe_3O_4 nanoparticles impregnated eggshell	*Pongamia pinnata* oil	2	12:1	120	65	98	7/98	[49]
Chicken eggshell-derived CaO	*Terminalia belleric* seed oil	2.25	9:1	90	62.5	97.98	-	[50]
Chicken eggshell-derived CaO	Palm kernel oil	4	10:1	60	50	97.1	5/>90	[51]
Al_2O_3 impregnated on calcined eggshells	Rubber seed oil	3	12:1	240	65	98.9	-	[52]
Chicken eggshell-derived CaO	Rubber seed oil	5	9:1	240	65	97.84	-	[53]
Eggshell-derived CaO supported on a fly ash-based zeolitic material	Sunflower oil	6	6:1	30	60	99.2	5/97.9	[54]
Chicken eggshell-derived CaO	WCO	5	9:1	165	65	87.8	-	[55]
Palm mill fly ash-supported CaO derived from eggshells (CaO/PMFA)	Palm oil	6	10:1	180	70	86.2	5/70	[56]
KOH impregnated eggshell	*Reutealis trisperma* oil	5	12:1	60	60	94	-	[57]
Eggshell-derived CaO supported W-Mo mixed oxide	WCO	2	15:1	120	70	96.2	5/90	[58]
KF/eggshell-Fe_3O_4	Neem oil	6	15:1	120	65	97	5/>75	[59]
Chicken eggshell-derived CaO	Sunflower oil	5	11:1	180	60	83.2	4/>80	[60]

Table 1. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
La_2O_3/CaO derived from eggshell	Palm oil	10	12:1	250	60	92.3	-	[61]
Ostrich eggshell-derived CaO	WCO	1.57	11:1	114	65	97.54	-	[62]
Chicken eggshell-derived CaO	WCO	1.61	11.4:1	114	65	94.7	-	[62]
Fe_3O_4/CaO derived from eggshell	Palm oil	6	10:1	120	70	90	-	[63]
Chicken eggshell-derived CaO	WCO	1.47	7.85:1	144	43	90.13	3/73.3	[64]
Eggshells-derived CaO	Rubber seed oil	4	12:1	180	65	99.6	7/86.4	[65]
Chicken eggshell-derived CaO	Soybean oil	3	9:1	240	65	94.2	-	[66]
Quail eggshell-derived CaO	Soybean oil	3	9:1	240	65	94.8	-	[66]
CaO@MgO nanocatalyst derived from chicken eggshell	Waste edible oil	4.571	16.7:1	424.8	69.37	98.37	-	[67]
SrO/CaO derived from eggshell	Jatropha oil	4.77	27.6:1	89.8	65	99.71	5/>60%	[68]
Na-K doped CaO derived from calcined eggshell (Na_1K_1/CaO)	Canola oil	3	9:1	180	50	97.6	4/66.0	[69]
Egg shell-derived nano-CaO	*Chlorella pyrenoidosa* oil	2.06	30:1	180	60	93.44	6/85.2	[70]
Duck eggshell-derived CaO	*Momordica charantia* oil	10		80	65	96.8	-	[71]
Na impregnated calcined eggshell	*Madhuca indica* oil	5	9:1	60	60	81.56	5/>70	[72]
Zn doped eggshell-derived CaO	WCO	5	20:1	240	65	96.74	5/64.5	[73]
Zn doped eggshell-derived CaO	Eucalyptus oil	5	6:1	150	65	93.8	5/>88	[74]
Chicken eggshell-derived CaO	WCO	1.5	10:1	210	50	91.42	5/48	[75]
Chicken bone-derived CaO	Algal oil	5	9:1	180	65	95	4/>80	[75]

Table 1. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Chicken eggshell-derived CaO	Algal oil	5	9:1	180	65	94	4/70	[76]
Chicken manure-derived catalyst	Algal oil	5	9:1	180	65	85	4/>60	[76]
Chicken eggshell-derived Ca-based catalysts	Waste cooking palm oil	3	15:1	180	80	90.1	3/>70	[77]
Turbo jourdani shell-derived CaO	Palm oil	10	3:1	420	80	99.33	8/>75	[16]
Oyster shell-derived CaO	WCO	6	9:1	180	65	87.3	-	[17]
Pomacea canaliculata shell-derived CaO	Palm oil	0.8	12:1	360	65	95.2	4/90.7	[18]
Activated carbon supported CaO from *Turbonilla striatula* shell	WCO	11	40:1	420	120	96	5/96	[19]
Crap shell-derived CaO	Waste fish oil	2.5	12:1	90	65	96.6	5/80	[20]
NaOH impregnated activated carbon/CaO derived *Perna varidis* shell	Palm oil	7.5	0.5:1	180	65	95.12	-	[21]
Grooved razor shell-derived CaO	WCO	5	15:1	180	65	94	5/87	[22]
Conch shell-derived CaO	*Moringa oleifera* oil	8.022	8.662:1	130	65	97.06	-	[23]
Malleus malleus shells derived CaO	WCO	7.5	11.85:1	86.25	65	93.81	-	[24]
Calcined sheep bone impregnated fly ash catalyst	Mustard oil	10	5.5:1	360	65	90.4	7/80.3	[25]
Snail shell-derived CaO nanocatalyst	Scum oil	0.89	12.4:1	145.15	61.6	98.93	5/>90	[78]
	Hydnocarpus wightiana oil	0.87	12.7:1	119.68	58.6	96.93	-	
Snail shell-derived CaO	Soybean oil	6	9:1	210	65	90	5/80	[79]
KOH impregnated snail shell	Soybean oil	6	9:1	210	65	96	5/90	[79]
Snail shell-derived CaO	Soybean oil	3	6:1	420	28	98	8/90	[80]
Quail eggshell-derived CaO	Sunflower oil	2	10.5:1	120	60	99	3/78.26	[81]

Table 1. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
CaO-based catalyst derived from eggshell-snail shell-wood ash mixed	Mixture of *Irvingia gabonensis*, *Pentaclethra macrophylla*, and *Elais guineensis* oil	4.5	8:1	64.71	61.61	98	5/79	[82]
Ram bone supported Cr catalyst	Used frying mustard oil	4	8:1	30	60	96.85	5/95.56	[83]
Lithium based chicken bone composite	Canola oil	4	18:1	180	60	96.6	5/82	[84]
Lithium/zinc supported on chicken bone catalyst	Waste canola oil	4	18:1	210	60	98	7/>96	[85]
Goat bone-derived nano-CaO	*Scenedesmus* algal oil	2	11:1	180	60	92	-	[86]
KOH impregnated CaO derived from goat bone	WCO	6	9:1	300	65	84	-	[87]
Chicken and fish bone-derived CaO	WCO	1.98	10:1	92	65	89.5	5/<50	[88]
Struthio camelus bone-derived CaO	WCO	5	15:1	240	60	90.56	5/>80	[89]
Poly- glycidyl-methacrylate grafted flax fibers	Cottonseed oil	2.5	33:1	120	60	88.6	3/72.5	[90]
Calcined cupuaçu (*Theobroma grandiflorum*) seeds	Soybean oil	10%	10:1	480	80	98.36	3/>20	[31]
K_2O-KCl derived from calcined banana peel	Soybean oil	1.5	15:1	60	65	95.1	4/75.5	[32]
Calcined husk of *Enterolobium cyclocarpum* pods	Oil blend	2.96	11.44:1	5.88	65	98.77	4/74.68	[33]
Calcined kola nut husk pod	*Hevea brasiliensis* seed oil	3.5	6:1	75	65	96.97	-	[34]
Calcined *Brassica nigra* plant	Soybean oil	7	12:1	25	65	98.79	3/>96	[35]
ZrO_2 supported on bamboo leaf ash	Soybean oil	12	15:1	30	50	92.75	-	[36]
Calcined *Sesamum indicum* ash	Sunflower oil	7	12:1	40	65	98.9	3/94.2	[37]
Calcined *Tectona grandis* leaves	WCO	2.5	6:1	180	RT	100	4/>80	[38]
Calcined *Ficus carica* leaves	WCO	1	6:1	120	60	90.75	-	[39]
Calcined ginger (*Zingiber officinale*) leaves activated by KOH	Sunflower oil	1.6	6:1	90	60	93.83	-	[40]

Table 1. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Calcined *Carica papaya* stem	WCO	2	9:1	180	60	95.23	6/85.4	[41]
Calcined banana peel	WCO	2	6:1	180	60	100	3/66.66	[42]
KOH/corncob-derived activated carbon	WCO	1	18:1	60	45	97.8	2/35	[43]
Supermagnetic catalyst derived from rice husk doped with K_2O and Fe	WCO	4	12:1	240	75	98.6	5/>80	[44]
CaO/zeolite-based catalyst derived from chicken eggshell and coal fly ash	Sunflower oil	6	6:1	30	60	97.8	-	[54]
Orange peel ash	Soybean oil	7	6:1	420	RT	98	5/85	[91]
Rice husk biochar supported CaO	Palm oil	8	9:1	180	65	93.4	10/85	[92]
Silica impregnated CaO derived from eggshell	Virgin cooking palm oil	3	20:1	120	60	87.5	6/>80	[93]
Sugarcane leaf ash	*Calophyllum inophyllum* oil	5	19:1	180	64	97	10/74	[94]
SiO_2-rich sugarcane bagasse ash	Palm oil	6	20:1	180	65	93.8	5/70.3	[95]
Calcined barnacles shell	*Aglaia korthalsii* seed oil	4.7	12.2:1	180	65	97.12	4/95.83	[96]
Calcined banana peduncle	*Ceiba pentandra* oil	1.978	9.2:1	60	65	98.69	-	[97]
Silica-supported CaO derived from goat bone	WCO	6	15:1	120	60	94	7/40	[98]
Calcined quail beaks	Rapeseed oil	7	12:1	240	65	96.7	6/>90	[99]
Calcined walnut shell	Sunflower oil	5	12:1	10	60	98	4/>95	[100]

RT: room temperature.

3. Bio-Derived Acid Catalysts for Biodiesel Production

Alkali-catalyzed transesterification is efficient for producing biodiesel from refined oils (containing a low level of free fatty acids (FFA)). However, the biodiesel yield is significantly reduced when the oil contains a high level of FFA (>1%, *w/w*) because alkali catalysts cannot convert FFA into biodiesel and the liquid alkali catalysts can react with FFA to form soap [2]. Therefore, acid-catalyzed esterification/transesterification is commonly proposed to produce biodiesel from high FFA-containing oils. Acid catalysts simultaneously catalyze the esterification of FAA and transesterification of oil (triglyceride) into biodiesel; therefore, they are insensitive to the quality of the raw material. In addition, the use of the acid catalysts for biodiesel production prevents the saponification reaction, which is commonly found in the homogenous alkali-catalyzed transesterification reaction. Homogenous acid catalysts (such as HCl, H_2SO_4, H_3PO_4) are widespread in biodiesel production because they efficiently convert FFA and triglyceride into biodiesel [9,10]. However, there are lots of associated problems in the downstream process, which is costly and requires complicated steps for product purification and separation of the catalyst. In addition, the use of these homogenous acid catalysts causes corrosive damage to the equipment and negatively affects the environment. These liquid catalysts are also difficult recover and reuse. To address these obstacles, heterogenous/solid acid catalysts have been increasingly considered as promising alternative catalysts to facilitate a cleaner, safer, simpler, and cheaper process for biodiesel production [101,102].

In recent years, biomass-derived acid catalysts have gained much interest in biodiesel production due to their ecofriendly properties, potential reusability, and the availability and low cost of materials used for catalyst synthesis. Recently, several forms of heterogeneous acid carbon-based catalysts have been developed for biodiesel production from high-FFA oils. The carbonization followed by sulfonation method is commonly used to synthesize various solid acid catalysts such as sulfonated carbon from corn cobs [103], sulfonated starch [104], sulfonated carbon from vegetable oil asphalt [105], sulfonated carbon from cacao shell [106], sulfonated rice husk [107], sulfonated bamboo [108], sulfonated sugarcane bagasse [109], sulfonated biochar derived from cassava peel [110], and sulfonated biochar derived from sugarcane bagasse, corncob, coconut shell, and peanut shell [111]. Different materials result in different catalytic activities of the synthesized catalysts. The catalysts prepared from these materials demonstrated good catalytic efficiency towards esterification of high-FFA oils, with FFA conversions ranging from 71% to 98% [109,110]. Among the materials used, waste shells, such as cacao shell [106], wing shell [112], and coconut shell [113], show promise for the synthesis of solid acid catalysts. More acid catalysts used for biodiesel production are shown in Table 2. In comparison with alkali catalysts, the bio-based acid-catalyzed reaction commonly requires a longer reaction time and higher temperature for biodiesel production. Therefore, the acid-catalyzed reaction is only suggested for producing biodiesel from feedstock containing a high level of FFA.

Table 2. Several bio-derived acid catalysts for biodiesel production.

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Sulfonated-carbonized bamboo	Oleic acid	5	8:1	60	65	97.31	5/<40	[108]
Sulfated angel wing shells	Palm fatty acid distillate	2	6:1	15	290	98	7/>80	[112]
Sulfonated-carbonized coconut shell	Palm oil	6	30:1	360	60	88.15	-	[113]
Sulfated-carbonized *Jatropha curcas* seed	*Jaropha curcas* oil	7.5	12:1	60	60	99.13	4/81.03	[114]
Carbonaceous solid acid magnetic catalyst from empty fruit bunch	Palm fatty acid distillate	4	16:1	180	100	98.6	6/79	[115]
Sulfonated cow dung-derived carbon-based catalyst	Palm fatty acid distillate	4	18:1	60	90	96.5	7/75	[116]
CaO-based calcined angel wing shell sulfated catalyst	Palm fatty acid distillate	5	15:1	180	80	98	4/>40	[117]

Table 2. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Sulfonated-carbonated coconut meal residue	Waste palm cooking oil	5	12:1	180	150	95.5	4/82	[118]
Sulfonated carbon derived from coconut meal residue	Waste cooking oil	6	9:1	300	65	96	-	[119]
Sulfated Ce supported activated carbon derived from coconut shell	Chicken fat oil	3	12:1	60	90	93	5/90	[120]
Sulfonated and magnetic catalyst derived from palm kernel shell	Waste cooking oil	3.66	13:1	102	65	90.2	4/73.63	[121]
Sulfonated carbon-based catalysts from murumuru kernel shell	Oleic acid	5	10:1	90	90	97.2	4/66.3	[122]
Sulfonated carbon-based catalyst from Murumuru kernel shell	*Jupati* oil	6	30:1	240	135	91.8	4/>80	[123]
Sulfonated biochar derived from sawdust	*Pongamia pinnatta* oil	2	9:1	120	85	95.6	4/85.7	[124]
Sulfonated-carbonized *Zanthoxylum bungeanum* seed	*Zanthoxylum bungeanum* seed oil	8	30:1	240	140	95.6	5/57/9	[125]
Sulfonated-calcined kenaf seed cake	Palm fatty acid distillate	2	10:1	90	65	97.9	5/>90	[126]
Palm biochar-based sulfated zirconium	Palm fatty acid distillate	3	15:1	180	75	94.3	5/80.2	[127]
Sulfonated activated carbon derived from Monk fruit seed (*Siraitia grosvenorii*)		4		360	120	98.5	4/84.4	[128]
Sulfonated-derived tea waste	Palm fatty acid distillate	4	9:1	90	65	97	5> 80	[129]
Sulfonated-carbonized *Hura crepitans* seed pod	High-FFA vegetable oil	10	9:1	60	90	94.81	4/93.37	[130]
Sulfonated-carbonized cotton stalk	*Madhuca indica* oil	5	18:1	300	60	89.2	7/83.4	[131]

Table 2. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield (%)	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (min)	Temp. (°C)			
Sulfonated activated carbon derived from *Mesua ferrea* shell	*Mesua ferrea* oil	10	6:1	120	55	95.57	-	[132]
Sulfonated biochar derived from palm empty fruit bunch	Palm fatty acid distillate	20	30:1	420	110	98.1	-	[133]
Sulfonated carbon derived from corncob residue	Palm fatty acid distillate	3	15:1	120	70	85	5/60	[134]
Sulfonated carbon derived from coconut meal residue	Waste palm oil	5	12:1	720	65–70	92.7	4/>80	[135]
Sulfonated-carbonized spent coffee grounds	Oleic acid	10	10:1	420	80	91.2	4/26.41	[136]
Sulfonated pine needle-derived carbon	Levulinic acid	5	5:1	480	80	96.1	4/>60	[137]
Sulfonated rice husk	Oleic acid	5	5:1	20	28	99.8	3/70	[138]
Sulfonated rubber de-oiled cake	Waste cooking oil	8.18	12.8:1	60	63	91.2	3/80	[139]
Magnetic carbonaceous acid derived from *Jatropha hulls*	*Jatropha* crude oil	7.5	18:1	450	180	95.9	5/94.3	[140]
Sulfonated carbon derived from potato peel	Oleic acid	5	12:1	150	80	97.2	5/68	[141]
Sulfonated waste yeast residue	Waste cooking oil	1	10:1	360	60	96.2	6/<80	[142]
Sulfonated-carbonized cacao shell	Oleic acid	5	7:1	1440	45	94	4/<50	[143]
Sulfonated coconut coir husk	Waste palm oil	10	12:1	180	130	89.8	4/<80	[144]
Sulfonated lignin-derived from olive cake	Waste vegetable oil	10	35:1	360	65	57	10/75	[145]
Sulfonated soaked palm seed cake derived catalyst	Palm fatty acid distillate (PFAD)	2.5	9:1	120	60	97.8	-	[146]
Sulfonated-calcined corncobs and calcined poultry	Neem seed oil	2.58	14.76:1	72.65	61.90	92.89	4/76	[147]
Sulfonated brewer's spent yeast	Palm fatty acid distillate	8	21:1	180	65	87.8	-	[148]

4. Enzyme

With an increasing demand for environmental protection, green processes have been rapidly developed for chemical production. Consequently, various ecofriendly processes have been proposed for producing biodiesel to reduce the adverse environmental effects [5,149]. Particularly, the enzyme-catalyzed reaction is one of the most promising processes for biodiesel production due to the ecofriendly and reusable nature of the enzyme. Notably, the enzymatic process proceeds at mild reaction temperature and pressure, thus lowering the energy consumption [150]. For this approach, biodiesel can be produced via lipase-catalyzed transesterification or lipase-catalyzed hydroesterification processes (hydrolysis of oils into FFA followed by esterification of the produced FFA with short-chain alcohols). The lipase catalyzes the esterification and transesterification simultaneously; therefore, the enzymatic process is insensitive to high-FFA oil [150]. Because of such benefits, enzymatic processes have been widely developed for biodiesel production from various feedstocks [150].

The efficiency of the enzymatic process mainly depends on the activity of lipases. Therefore, a great effort has been made to use lipase from different sources (microorganisms, plants, animals) for biodiesel production [151,152] (Table 3). The most common source of the lipase is microorganisms such as *Candida antarctica* [153,154], *Thermomyces lanuginosu* [155,156], *Rhizomucor miehei* [157,158], *Pseudomonas cepacia* [159,160], *Candida rugosa* [161,162], *Aspergillus oryzae* [163], *Burkholderia cepacia* [164,165], *Adansonia grandidieri* [166], *Rhizopus oryzae* [167], *Pseudomonas fluorescens* [168], *Lactobacillus plantarum* [169], and *Aspergillus terreus* [170]. Lipases from microorganisms are mainly used for biodiesel production due to the availability of sources and rapid growth rate of microorganisms for enzyme production [171]. Lipase activity depends not only on the source of the enzyme, but also the type of enzyme used (immobilized form or liquid form) [171]. Immobilizing lipase on the support material can enhance the stability of the enzyme, making the enzyme less susceptible to the pH, temperature, and impurities of reactants [171]. Notably, the supports and/or immobilization protocols can greatly modulate the specific activity of lipase, affecting biodiesel yield. Tacias-Pascacio et al. [172] immobilized different lipases on different supports and used them for biodiesel production. They found that the specific activity of lipases and biodiesel yield greatly depended on the support, solvent used, and media [172]. In addition, the immobilized enzyme is easy to reuse. Consequently, lipase immobilized on various supporting materials has been studied for biodiesel production. Recently, Iuliano et al. [173] reported that lipase from *C. rugosa* was physically attached to Mg modified Fe_2O_4 nanoparticles and used to turn brewers' spent grains into biodiesel. After 48 h at 45 °C, a remarkable yield of 98% was achieved using a 1:4 oil/methanol molar ratio. In addition, lipases were immobilized on other materials such as graphene oxide [174], polyhydroxyalkanoate [175], alginate-polyvinyl alcohol (PVA) [167], polydopamine coated iron oxide (Fe_3O_4_PDA_lipase) [170], modified polyporous magnetic cellulose support [153], Co^{2+}-chelated magnetic nanoparticles [168], core-shell structured Fe_3O_4@MIL-100(Fe) composites [162], Fe_3O_4/Au nanoparticles [176], waste-derived activated carbon support [177], genipin cross-linked chitosan [178], and other materials [160]. Several immobilized lipases have been commercialized and used for biodiesel production such as Novozym® 435 (lipase B from *C. antarctica*) [179–181] and Lipozyme TL IM (lipase from *T. lanuginosus)* [182]. Nevertheless, the immobilized lipase-catalyzed reaction rate is relatively low due to the mass transfer limitation between the substrate and enzyme [183]. Notably, the immobilized lipases are expensive, thus limiting their industrial applications.

Table 3. Several lipases used for biodiesel production.

Catalyst	**Feedstock**	**Reaction Conditions**				**Conversion/ Biodiesel Yield (%)**	**Time of Reuse/Corresponding Biodiesel Yield**	**Ref.**
		Catalyst Loading (%)	**Alcohol: Fatty Acid Molar Ratio**	**Time (h)**	**Temp. (°C)**			
C. antarctica lipase B (CALB) immobilized on modified polyporous magnetic cellulose beads	Yellow horn seed oil	15	1.6:1	2	60	92.3	5/85	[153]
C. antarctica lipase A (CALA)	Palm oil	5.5	7:1	22	30	94.6	-	[154]
Novozym® 435 (CALB immobilized on macroporous acrylic resin)	Residual babassu oil	0.14 g	18:1	4	48	96.8	10/90.96	[179]
Novozyme® 435	Castor oil fatty acid	10	3:1	5	60	88.64	-	[180]
Novozyme® 435	*Spirogyra* oil	1	4.5:1	42.5	35	93.2	-	[184]
Novozym® 435	Black soldier fly larvae oil	17.58	14.64:1	12	39.5	96.97	20/>95	[185]
CALA	Soybean oil	5	7:1	26	38	92.4	-	[186]
CALB immobilized on methacrylic resin	Waste animal fat	14	10:1	6	40	87	-	[187]
CALB immobilized on magnetic nanoparticles	Palm fatty acid distillate	8	1.6:1	10	50	82.74	5/80.19	[188]
CALB immobilized magnetic nanoparticles	Microalgal oil	1	10:1	3	30	91.4	4/90	[189]
67% CALB + 33% lipase from *R. miehei*	Residual chicken oil	15	5:1	3	30	89.95	-	[190]
CALB	Soybean oil	3	3:1	15	40	64.7	-	[191]
CALB immobilized on silica nanoflowers	Waste oil	33.24 mg	2.63:1	8.11	45.97	98.5	15/76.68	[192]
CALB and *Rhizomucor miehei* lipase co-immobilized on epoxy functionalized silica gel	Palm oil	4.9 U/mg	5.9:1	33.5	35.6	78.3	-	[193]
Lipozyme TL100L (*T. lanuginosu* lipase)	Waste phoenix seed	9.7	4.3:1	6.9	31	93.8	-	[155]
Lipozyme TL IM (immobilized *T. lanuginosus* lipase)	Rapeseed oil	5	5:1	5	25	98.76	-	[156]
Lipozyme TL IM	*Ankistrodesmus* sp. oil	9.6	8:1	12	42	97.69		[194]

Table 3. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield	Ref.
		Catalyst Loading (%)	**Alcohol: Fatty Acid Molar Ratio**	**Time (h)**	**Temp. (°C)**			
T. lanugionous immobilized on Fe_3O_4 nanoparticles	Soybean oil	9	4:1	28	41	82.2	10/71.23	[182]
Lipase NS 40116 (liquid lipase formulation derived from *T. lanuginosus*)	Residual chicken oil	0.3	4:1	36	35	93.16	-	[195]
Lipozyme TL IM	Rapeseed oil	5	9:1	7	30	99.89	-	[196]
Lipozyme TL100L	Phoenix tree seed oil	10	5:1	6.98	30	98.8	-	[197]
Lipase NS 40116	Soybean oil	0.7	6.3:1	8	35	97.1	-	[198]
Lipase NS 40116	Soybean oil	0.5	4.5:1	12	35	94.3	5/90	[199]
T. lanuginosus lipase immobilized on Immobead 150	None-edible oils	3.55	7.64:1	2	36	90	-	[200]
T. lanuginosus lipase immobilized on Fe_3O_4/Au nanoparticles	Tomato seeds oil	20	6:1	24	45	98.5	5/68.95	[176]
Liquid formulation of *T. lanuginosus* lipase	Palm oil mill effluent	2100 U	4:1	24	40	97.43	-	[201]
Eversa Transform lipase (liquid lipase from *T. lanuginosus*)	Oleic acid	11.98	3.44:1	2.5	35.25	96.73	5/<30	[183]
R. miehei lipases	Oleic acid	20	2:1	4	40	85	4/74	[157]
R. miehei lipase immobilized on magnetic nanoparticles	Babassu oil	5	1:1	6	40	81.7	-	[158]
C. rugosa immobilized on polyhydroxybutyrate + *R. miehei* immobilized on polyhydroxybutyrate	WCO	1	6:1	24	45	96.5	10/28.95	[175]
Mixture of polyhydroxybutyrate-immobilized *C. rugosa* and *R. miehei* lipases	Mixed chicken waste oil	2.5	6:1	12	40	97.1	15/10	[202]
P. cepacia lipase immobilized on bio-support beads.	Hybrid non-edible oil	10	6:1	24	50	78	12/19.5	[159]
P. cepacia lipase immobilized on bio-support beads	Non-edible hybrid oil	9.46	5.93:1	24.32	49.7	84.58	10/>70	[160]
P. cepacia lipase immobilized on hybrid PVA/AlgNa	Crude castor oil	10	6:1	24	50	78	6/70	[203]

Table 3. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (h)	Temp. (°C)			
C. rugosa lipases immobilized on immobead 150	*Acutodesmus obliquus* oil	15	3:1	8	50	95.36	5/90.07	[161]
C. rugosa lipase immobilized lipase on core-shell structured Fe_3O_4@MIL-100(Fe) composites	Soybean oil	25	4:1	60	40	92.3	5/83.6	[162]
C. rugosa lipase immobilized on magnetic Fe_3O_4-poly (glycidyl methacrylate-co-methacrylic acid) composite	Soybean oil	25	4:1	54	40	92.8	5/79.4	[204]
C. rugosa immobilized on Mg modified Fe_2O_4 nanoparticles	Brewers' spent grains oil	30	4:1	48	45	98	4/87	[173]
Eversa® Transform 2.0 (liquid lipase from *T. lanuginosus*)	Palm oil	0.2	4:1	24	40	97	-	[205]
A. oryzae ST11 lipase immobilized on polyacrylonitrile coated magnetic nanoparticles	Palm oil	30	3:1	24	37	94.7	5/65	[163]
B. cepacia lipase immobilized on hydroxyapatite coated magnetic nanoparticle	WCO		7:1	48	40	98	4/82	[164]
B. cepacia lipase immobilized on mesoporous silica/iron oxide magnetic core-shell nanoparticle	WCO	36	6.2:1	25	34	92	3/81	[165]
B. cepacia lipase	Sunflower oil	10	3.4:1	1	50	>99	-	[206]
A. grandidieri lipase	Sunflower oil	25	2:1	96	40	95	-	[166]
R. oryzae lipase immobilized on alginate-polyvinyl alcohol	Sludge palm oil	2	3:1		40	91.3	15/>91	[167]
P. fluorescens lipase immobilized onto Co^{2+}-chelated magnetic nanoparticles	WCO	7.5	4:1	12	50	95	10/83	[168]
Immobilized *L. plantarum* lipase	Olive oil	5	6:1	2	37	81	4/>65%	[169]

Table 3. *Cont.*

Catalyst	Feedstock	Reaction Conditions				Conversion/ Biodiesel Yield (%)	Time of Reuse/Corresponding Biodiesel Yield	Ref.
		Catalyst Loading (%)	Alcohol: Fatty Acid Molar Ratio	Time (h)	Temp. (°C)			
Lipase immobilized on graphene oxide	*Karanja* oil	3	8:1	24	25	88	-	[174]
Oreochromis niloticus lipase	WCO	30 kUnit	4:1	28	45	96.5		[151]
A. terreus AH-F2 lipase immobilized on polydopamine coated iron oxide	WCO	10	6:1	30	37	92	5/>80	[170]
Steapsin lipase immobilized on waste-derived activated carbon support	Rubber seed oil	3	6:1	5	20	83.9	7/>77	[177]
Steapsin lipase immobilized on Immobead-350	WCO	14	4:28	14	40	88.33		[207]
Proteus sp. NH 2-2 lipase	soybean oil	0.5	4:1	36	40	91.5		[208]
Garbage lipase	*Naganishia liquefaciens* NITTS2 oil	20	6.4:1	16	35	97.13		[209]
Lipase (from porcine pancreas) immobilized on genipin cross-linked chitosan beads	WCO	7.5	9:1	10	40	92.33	4/>80	[178]

Liquid lipase formulations or free lipases have been considered as a substitute for immobilized lipase for biodiesel production due to their high catalytic activity and significantly low cost (30 to 50 times lower) as compared to immobilized lipase [183,210]. Recent studies have demonstrated a promising use of several liquid lipases for biodiesel production such as *C. antarctica* lipase A [154] and liquid lipase formulations from *T. lanuginosus* (Eversa® Transform, Eversa® Transform 2.0, and NS-40116) [195,205,211]. The use of liquid lipase facilitates the homogenous reaction, thus overcoming the mass transfer limitation presented in the immobilized lipase-catalyzed reaction. However, liquid lipase is sensitive to the reaction environment. Studies have reported that high water content (from the feedstock and/or generated from the esterification of alcohol and fatty acid) not only promotes the reverse reaction but also negatively affects the lipase activity (including the formation of lipase-lipase aggregates in aqueous media), thus reducing the biodiesel production efficiency [183]. To address this obstacle, several adsorbents such as superabsorbent polymer, silica gel, alumina, and molecular sieve have been used to remove the water from the reaction mixture, enhancing the reaction efficiency [183,212,213].

Similarly, the type of acyl acceptor used also affects the lipase-catalyzed reaction. Studies have reported that lipase is deactivated using a high amount of methanol or ethanol, lowering the biodiesel yield [181,185]. In addition, the use of methanol or ethanol as an acyl acceptor for biodiesel production resulted in the formation of by-product glycerol [181]. This by-product also inhibits the activity of lipases, especially immobilized lipases because it can easily accumulate on the surface of immobilized lipases [181]. To address this obstacle, methyl acetate is proposed as another alternative acyl acceptor for biodiesel production [185]. The use of methyl acetate prevents the inhibition of lipase caused by methanol/ethanol and by-product glycerol (no glycerol produced in the reaction), thus enhancing the reaction rate [185]. Besides this method, ultrasounds [214,215] or very hydrophobic supports [216,217] can be used as another approach to lower the negative effect of glycerol on the enzyme. Studies have reported that ultrasounds can stir the enzyme particles from the inside and avoid the formation of the glycerin/water phase [215,218].

Another concern for each specific lipase in biodiesel production is associated with the oil source [218]. Fats/oils are a very heterogenous substrate, which are mainly comprised of triglycerides, low levels of mono and diglycerides, and some FFA [218]. Therefore, enzyme specificity affects the enzyme activity over each substrate [218]. To address this issue, the combination of different lipases (combi-lipase) has been proposed for biodiesel production [190,219]. There are several types of combi-lipase, which include co-immobilized lipases (different lipases immobilized on the same support), a mixture of individually immobilized lipases, and a mixture of free lipases [218]. Guan et al. [219] firstly reported the use of *R. miehei* lipase and *P. cyclopium* lipase mixture (in a liquid form) for biodiesel production from soybean oil. The result showed that the *R. miehei* lipase (individual enzyme) resulted in 68.5% biodiesel yield, but the yield increased to 95% when using the mixture of *R. miehei* and *P. cyclopium* lipases [219]. This was due to the use of lipases with different specificities [218,219]. In another study, the individual use of *R. oryzae* lipase and *C. rugosa* lipase resulted in 94.36% biodiesel yield at a reaction time of 9 h and 92.63% biodiesel yield at a reaction time of 30 h, respectively [220]. However, the biodiesel yield reached 98.16% (at a reaction time of 6 h) by using the mixture of both enzymes [220]. Similarly, various combi-lipases such as lipase cocktail (67% *C. antarctica* lipase B and 33% *R. miehei* lipase) [190]; immobilized *C. rugosa* and *R. miehei* lipases [175,202], co-immobilized *R. miehei* lipase and *C. antarctica* lipase B [193]; a mixture of 10% *T. lanuginosus* lipase, 75% *C. antarctica* lipase B, and 15% *R. miehei* lipase [221]; a mixture of lipases from porcine and *T. lanuginosus* (in both liquid and immobilized forms) [222]; a mixture of immobilized *C. rugosa* and *R. oryzae* lipases [223], and co-immobilized *C. rugosa* and *R. oryzae* lipases [224,225] were also tested for biodiesel production. These combi-lipases showed a higher biodiesel yield than the individual enzymes [175,190,193]. Mixtures of the same enzyme immobilized using different protocols/support materials also affect the biodiesel yield. Toro et al. [226] immobilized the same lipase (*T. lanuginosus* lipase) on two different supports (Purolite®ECR1604 and Lewatit®VPOC1600) and used them for biodiesel production from palm olein. The biodiesel yield reached 70.3% (for lipase immobilized on Purolite®ECR1604) and 78.2% (for lipase immobilized on Lewatit®VPOC1600). Notably, the biodiesel yielded increased to 86.1% when the mixture of the two individually immobilized lipases was used [226]. This could be explained by the fact that the enzyme features (flexibility of their active site and their mechanism of action) can be modulated by changes in the immobilization protocol [172]. Consequently, the changes in the support feature influence the stability, activity, and specificity of the lipase [172,218].

Generally, although both immobilized and liquid lipases (individual lipases or combi-lipases) show effectiveness for converting oil into biodiesel, their industrial application is still limited due to the high cost of the enzyme as compared to chemical catalyst [227,228]. Therefore, further studies on lipase-catalyzed biodiesel production are still required to improve the efficiency and economic feasibility of the process.

5. Catalyst Reusability

For biodiesel conversion, the catalyst's effectiveness is not only determined by its catalytic activity but also its recoverability and reusability. Since homogenous catalysts cannot be reused for the next batch of production, heterogeneous catalysts play an important role in reducing production costs. Their recyclability not only lowers production costs but also maximizes environmental protection [229]. As compared to homogenous catalysts, one of the benefits of heterogeneous catalysts is that they can be reused several times. Furthermore, these catalysts may be regenerated or used for other purposes after losing their catalytic activity, such as construction materials, soil stabilizers, cement industries, and phosphate adsorbents [230].

Most of the bio-derived acid and alkali catalysts can be reused 4–7 times to yield biodiesel of 65–85% (Tables 1 and 2). da Luz Corrêa et al. [122] prepared sulfonated carbon-based catalysts from murumuru kernel shell and used them for FFA conversion. The first use of the catalyst resulted in 95.1%, but the FFA conversion was reduced to 84.5% and 66.3% after the second and third catalyst reuses, respectively. The reusability of a solid base oxide catalyst derived from chicken eggshell was investigated by performing transesterification using the same catalyst for 10 cycles, and the yield was found to be marginally reduced after the seventh cycle, which may be due to catalyst pores being blocked, reducing reactant adsorption and desorption [49]. Kirubakaran and Arul [27] also investigated the reusability of a heterogeneous catalyst derived from eggshell. The catalyst could be reused five times to yield 85% biodiesel. After that, the biodiesel yield reduced significantly, suggesting that the catalyst's stability had deteriorated. This is due to the presence of active Ca(OH)$_2$ phases which reacted partially with the homogenous mixture in the transesterification reaction. In comparison with solid bio-based acid and alkali catalysts, several immobilized lipases show better reusability. Several immobilized lipases can be reused for up to 20 cycles without loss of enzyme activity [181,185]. However, the use of immobilized lipase for biodiesel production is still under lab-scale investigation because of the high cost of the enzyme. Therefore, to be used for industrial biodiesel production, further studies are still required to improve the catalytic activity, stability, and reusability of bio-based catalysts. In addition, a pilot-scale investigation is also needed to evaluate the potential use of these catalysts for biodiesel production before being used for industrial applications.

6. Environmental and Economic Evaluation

Catalyst selection is one of the crucial issues in biodiesel production with the aim to minimize energy consumption, waste generation and treatment, and reduce production costs [185]. The use of bio-based catalysts (alkali catalysts, acid catalysts, and enzymes) lowers the environmental effect since these catalysts are derived from natural sources (plants, animals, or microorganisms). These catalysts are also easy to separate from the reaction mixture and reuse, reducing the generation of wastewater and chemical residues in the downstream process, especially the purification step. Consequently, the fee for the purification step and waste treatment can be reduced, lowering the production cost.

Several studies have also evaluated the economic feasibility of different biodiesel production processes [231,232]. The bio-derived alkali- and bio-derived acid-catalyzed processes are more economically feasible than the conventional process (H_2SO_4- or KOH-catalyzed process) for biodiesel production since the cost of those catalysts (and total biodiesel production cost) is considered lower than that of conventional chemical catalysts (KOH or H_2SO_4) [232–234]. Among these two processes, the alkali-catalyzed transesterification seems to be superior to the acid-catalyzed process because the former proceeds at a lower temperature, has a shorter reaction time, and requires a lower molar ratio of alcohol to oil as compared to the latter, as shown in Tables 1 and 2 [232,235]. In addition, the bio-based alkali and bio-based acid catalysts can be synthesized from the same natural source, but the synthesis of bio-based acid catalyst commonly requires one more step (sulfonation) [232]. Consequently, in some cases, the cost of bio-derived acid catalysts can be higher than that of bio-derived alkali catalysts. However, the cost of each specific catalyst depends on various factors including the source, synthesis method, and its reusability [232]. Therefore, it is difficult to compare the cost of all different types of catalysts. Different from bio-derived acid and alkali catalysts, the enzyme is expensive, especially the immobilized enzyme, making the lipase-catalyzed biodiesel production less competitive [236,237]. To reduce the enzyme cost, free lipase (or liquid lipase formulation) has been proposed for biodiesel production [183]. However, the reusability of liquid lipases is limited [183]. Therefore, the enzymatic process is still under investigation to improve its industrial application. Generally, among the three processes, the bio-derived alkali- and bio-derived acid-catalyzed processes are more economically feasible than the enzymatic process [236].

However, no individual studies have been conducted to compare the economic feasibility of the bio-derived acid-, bio-derived alkali-, and enzyme-catalyzed biodiesel production processes. Therefore, more studies are still required to evaluate and compare the economic feasibility of these processes.

7. Future Prospects and Conclusions

The use of biomass-derived catalysts has become a recent interest to make biodiesel production more sustainable. In addition, the use of these catalysts is promising to reduce the current high cost of biodiesel production, making biodiesel competitive with petro-diesel fuels. Research is therefore aimed to develop environmentally friendly, cost-effective, and efficient biomass-derived catalysts for biodiesel production. Consequently, different natural sources (animals, plants, microorganisms) have been used for synthesizing bio-based catalysts including acid catalysts, alkali catalysts, and enzymes. The catalytic activity of these catalysts varies among them. The use of acid or alkali catalysts depends on the quality of the feedstock. Besides, enzymes can be used as an alternative to both acid and alkali catalysts for biodiesel production. These catalysts show their advantages and disadvantages when they are used for biodiesel production. These catalysts show promise for biodiesel production, but these investigations have been stopped at lab-scale investigations. More investigations on these catalysts are therefore needed, especially large-scale investigations to prove the potential use of these catalysts for industrial biodiesel production.

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