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Review

Environmentally benign solid catalysts for sustainable biodiesel production: A critical review

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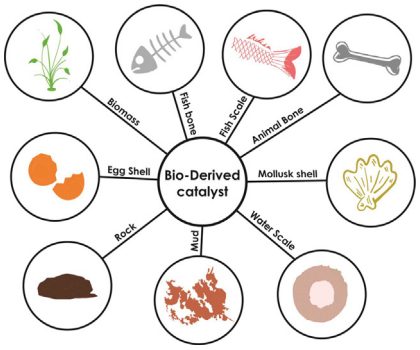
HIGHLIGHTS

- The capability of utilizing bio-derived catalysts for biodiesel production explored.
- Bio-derived catalysts have high potential and less harmful environmental impacts.
- Bio-derived catalysts not only reduces the cost but also promotes waste recycling.
- E-factor assessment confirmed that these catalysts have less environmental impacts.
- Bio-derived catalysts have prospects for large-scale biodiesel production.

GRAPHICAL ABSTRACT



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ABSTRACT

Versatile bio-derived catalysts have been under dynamic investigation as potential substitutes to conventional chemical catalysts for sustainable biodiesel production. This is because of their unique, low-cost benefits and production processes that are environmentally and economically acceptable. This critical review aspires to present a viable approach to the synthesis of environmentally benign and cost-effective heterogeneous solid-base catalysts from a wide range of biological and industrial waste materials for sustainable biodiesel production. Most of these waste materials include an abundance of metallic minerals like potassium and calcium. The different approaches proposed by researchers to derive highly active catalysts from large-scale waste materials of a re-usable nature are described briefly. Finally, this report extends to present an overview of techno-economic feasibility of biodiesel production, its environmental impacts, commercial aspects of community-based biodiesel production and potential for large-scale expansion.

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Contents

1. Introduction

> "*The use of vegetable oils for engine fuels may seem insignificant today, but such oils may become in the course of time as important as the petroleum and coal tar products of the present time*"
>
> [(Babu and Devaradjane, 2003)]

The transport sector is facing significant challenges with respect to the fuel utilized. Easily accessible fossil fuels will eventually deplete calling for long-term alternatives. Also substantial reduction of anthropogenic CO_2 emissions is more urgent than ever! In this perspective, the transport sector is considering several alternative fuels as substitutes for conventional fuels. They are made of gaseous and liquid fuels. Biofuels such as alcohols or vegetable oils are given more emphasis among liquid fuels because of their renewable nature. These biofuels are better than solid fuels because they can be stored (Agarwal, 2007; Hasan Altaie et al., 2017) and transported easily (Brito Cruz et al., 2013; Demirbas, 2010a; Ramsurn and Gupta, 2018).

Biomasses have proven an invaluable source for biofuels especially for developing economies (Agarwal, 2007; Demirel, 2018). In recent years, many studies have been carried out on the conversion of biomass into high value-added products (Haldar and Purkait, 2020; Rashid et al., 2019) including biodiesel (de Blasio, 2019; Akarte, 2004).

Biodiesel generally is defined as fatty acid methyl esters (FAMEs) or ethyl esters (FAEEs) obtained from natural triglycerides via transesterification (*Biodiesel*, 2007; Alenezi et al., 2013; Litinas et al., 2020). Despite years of development, the design of catalytic processes to manufacture FAMEs and FAEEs is still challenging (Banga and Varshney, 2010; Meira et al., 2015). Catalysts play a very important role in transesterification of vegetable oils or fats (Aderibigbe et al., 2020; Sharma et al., 2019). Developing efficient and low cost catalysts with an environmentally friendly approach is essential in order to resolve the current problems in homogeneous and heterogeneous chemical catalysts such as high cost, reusability, separation difficulty,

poisoning and leaching. A suitable catalyst will be a breakthrough in the biofuel industry if found for effective transesterification. Such a catalyst can contribute to biodiesel sustainability along with cost reduction (Ramos et al., 2019). In this regard, bio-derived catalysts are currently under investigation and have their intrinsic advantages and disadvantages. These catalysts are expected to be inexpensive and sustainable in large-scale applications. No previous literature explored the synthesis of environmentally benign solid catalysts from wide range of biological and industrial waste materials. In this outlook, we present a comprehensive overview focusing on the synthesis of environmentally benign solid catalysts from wide range of biological and industrial waste materials. The practical application of these catalysts in biodiesel synthesis is first considered. Then, the different approaches and techniques to improve the activity of the derived catalysts are discussed. We also provide current challenges and future directions to reach industrial application of bio-derived catalysts On the whole, this review might help in selecting suitable environmentally benign solid catalysts for sustainable biodiesel production.

2. Historical background of biodiesel

The fatty acid component for biodiesel can be obtained from vegetable oil (Lukić et al., 2016; Chavan et al., 2014; Purushothaman and Nagarajan, 2009) (edible or inedible) or animal fats (Sai Akhil and Alagumalai, 2019). The idea of using vegetable oil (originally peanut oil) in the engine was first formulated by Rudolph Diesel in 1900 at the Paris exhibition. Later, Diesel confirmed his appraisal of vegetable oils as alternative fuel (Knothe et al., 2005). During World War II, cotton oil was used as an emergency fuel in Brazil (Balat, 2009). After World War II, cottonseed oil and corn oil blends with diesel were used in America to replace imported diesel (Knothe, 2001). Almost two decades after the oil crisis in the 1970s, the real interest in vegetable oil fuels began. Still under the impression of the oil crisis, a conference entitled "Vegetable Oil fuels" was held in 1982 at Fargo, North Dakota. Several remarkable contributions were presented at the conference by researchers around the world. All the presentations at the conference were focused on the conversion of vegetable oils to biodiesel. The Persian Gulf war in 1991 also provoked the urgent need to use alternative fuels (Demirbas, 2010b). Today, biodiesel is produced worldwide from different feedstocks, and several countries have established their own biofuel policies (Sorda et al., 2010; *Advanced Biofuel Policies in Select EU Member States: 2018 Update*, n.d.; Miranda et al., 2011).

3. Biodiesel – a global scenario

The European Union (EU) advanced biofuel policy is to achieve 32% of energy utilized in select member state's transport fuel should return from the renewable energy sources by 2030 (*Advanced Biofuel Policies in Select EU Member States: 2018 Update*, n.d.).

In the U.S., soybean oil remains the most important feedstock for biodiesel production (Körbitz, 1999). Although the U.S. biodiesel production is growing, the policy impacts on biofuel development are lagging in the country.

In China, rapeseed has been identified as feedstock for biodiesel production. The recognition of biodiesel in China is growing year by year and therefore the projected usage of biodiesel in 2020 is estimated at 2 million tons. However, estimating the overall energy value of rapeseed-based biodiesel as a promising feedstock in China showed that the overall energy value had a negative energy return, emphasizing the changes to be made in the country's biodiesel policy (Chen and Chen, 2011).

In India, edible oils are hardly available and the country has to import up to 40% of its edible oil requirements (Singh et al., 2013). Therefore, the costs of edible oils are higher than that of petroleum diesel. The use of edible oils is also not a feasible idea because of the risk of converting food-based agricultural lands or crops for biodiesel production which leads to the detriment of the food supplies and therefore the use of non-edible oils is recommended for biodiesel production. For years it has been suggested to use Jatropha as a feedstock for biodiesel production in India. However, large-scale biodiesel production from Jatropha is facing many problem (Singh et al., 2013; Kumar et al., 2012; Axelsson et al., 2012). Also, the lack of promotion of sustainable agriculture is one of the major reasons that hinder large-scale biodiesel production in India. The several barriers in the promotion of sustainable agriculture are shown in Fig. 1.

4. Classification of biodiesel feedstock

In general, biofuels can be classified as first-generation, second-generation and third-generation biofuels (Aro, 2016; Nanda et al., 2018). First-generation biofuels are the typically derived from food crops and animal fats. Some of the examples are biodiesel, vegetable oil, and biogas. Second-generation biofuels are produced from waste biomass, and therefore lack many of the ethical and environmental issues associated to first-generation biofuels. Typical second generation biofuels comprise alcohols, such as ethanol, biodiesel from waste and diesel from wood. Finally, third-generation biofuels are produced from carbon dioxide mainly using algae technology. Carbohydrates produced in these photo bioprocesses are used to produce various fuels.

Thus, lipid or fat is the primary feedstock for biodiesel production. The selection of feedstock for biodiesel production is region specific because agricultural practices and climatic conditions in different nations offer different potential feedstock for biodiesel production (Atabani et al., 2017; Trombettoni et al., 2018; Avinash and Murugesan, 2018b). The potential feedstock available in major countries are shown in Fig. 2.

Globally, oilseed crops such as sunflower, soybean, Jatropha, palm and rapeseed are the main feedstock used in the production of biodiesel. Africa and Asia concentrate on the oilseed palm and Jatropha. In North America and Europe sunflower, soybean, and rapeseed prevail. In addition to oilseed crops, in some regions such as Ireland and Norway, fishery wastes form an important biodiesel feedstock.

5. Green catalysts for biodiesel production

As mentioned above, biodiesel production entails (trans) esterification of bio-derived fatty acid (esters) to obtain fatty acid methyl esters (FAMEs) as the classical biodiesel (Fig. 3).

To attain economically feasible transesterification rates, catalysis is inevitable. Catalysts not only accelerate the reaction rate but also allow performing the reaction at significantly lower reaction temperatures thereby saving energy-related costs and emissions.

A selection of established catalysts used for the synthesis of biodiesel is shown in Fig. 4.

In addition to these established, commercial (and rather expensive) catalysts, also some alternative, pot-ash-like catalysts are finding increased interest. In this study, various heterogeneous catalysts derived from wide range of biological and industrial waste materials and their performance in transesterification reaction is presented comprehensively. The schematic representation of potential materials considered in this study to derive bio-derived catalysts for biodiesel production is depicted in Fig. 5.

5.1. Biomass derived ash as catalyst

5.1.1. Banana peel ash

Banana peel has been regarded as trash for years. However, Pathak et al. (2018) research revealed that a possible low cost catalyst from the banana peel is possible. The authors derived catalyst from banana peel and used the catalyst for producing biodiesel from soybean oil. Initially, the catalyst was prepared by thoroughly washing the raw peel and drying it for a week. The dried peel was then grounded and burned completely to produce ash catalyst. It was observed that the effect of

Fig. 1. Barriers in promotion of sustainable agriculture (Avinash and Murugesan, 2018a).

increasing catalyst concentration from 0.3 wt%–0.7 wt% increased the biodiesel conversion and the maximum conversion of around 99% was observed at 0.7 wt%. However, beyond this the conversion rate decreased because of mass transfer effects and saponification reaction as reported by the authors. The authors also reported that the highest biodiesel yield could be achieved with banana peel ash because of the presence of potassium oxide (approximately 65% by a mass fraction) which exhibit high basic character. The authors also reported that the catalyst can be reused up to 4 reaction cycles after chemical washing and vacuum drying at 100 °C for 5 h. However, after fourth cycle, the yield dropped drastically to 52% due to a rise in carbon content (~77%) and a sharp decrease in potassium and oxygen content (16% and 5%, respectively).

5.1.2. Bamboo leaves ash

The bamboo plant is a perennial evergreen plant that is well cultivated in south-eastern Asia (especially in China and Indonesia) (Yang et al., 2008; Fatimah et al., 2018). There are about 1,000 bamboo kinds and about 200 of them are found in southeastern Asian province (Fatimah et al., 2018). As an alternative to low-cost catalyst for biodiesel production, Fatimah et al. (2018) suggested the use of zirconia supported bamboo leaves ash as a catalyst. In this work, the zirconia supported bamboo leaves ash catalyst was used for producing biodiesel from rice bran oil. The catalyst was produced in two steps: First by calcination and then by immobilization. In the first step, the bamboo leaves were calcinated at 500 °C for 4 h. In the second step, the resultant ash was mixed with precursor and then the solution was added drop wise

Fig. 2. Potential biodiesel feedstocks in major countries (Avinash and Murugesan, 2018b).

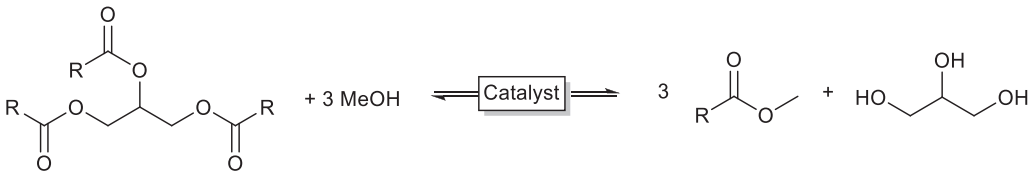

Fig. 3. Transesterification of natural triglycerides (fats and oils) to yield FAMEs.

into the ash suspension. Finally, the mixture was oven dried and the obtained a solid mass was subjected to calcination to obtain solid heterogeneous catalyst. This work concludes that subject to different reaction conditions, the produced catalyst has the same activity as the ZrO_2/SiO_2 catalyst in biodiesel conversion (Fig. 6).

5.1.3. Cocoa pod husk ash

As the principal source of potash, cocoa pod husk is a potential agricultural waste in Ghana, Nigeria and in the majority of the world producing cocoa (Oluyole et al., 2011; Rahim et al., 2019). Several investigations have shown that inorganic or mineral potash can be used as a biodiesel catalyst and the activity of such catalyst is high (Baroi et al., 2009). One such investigation was conducted by Ofori-Boateng and Lee (2013) using potash derived from cocoa pod husk as a catalyst for biodiesel production from soybean oil. In this research, a filtering system was employed to recover potash from the calcinated sample. The leached solution was then evaporated and further washed to produce potash crystals. The potash recovery was more than 52 g/l for five subsequent runs and the average recovery was noted to be 57.6 g/l. The transesterification reaction was conducted using unsupported and magnesium oxide (MgO) supported potash crystals. In this experimental work, it was finally revealed that 1 wt% loading of MgO supported potash to 6:1 methanol to soybean molar ratio maintained at 65 °C can produce ~99% biodiesel yield in 1 h reaction time. On the other hand, the unsupported potash under the same reaction conditions can yield ~91% for 2 h reaction time. The authors also observed that chemical characteristics of biodiesel produced using cocoa pod hush ash supported MgO catalyst were within the standard limits.

5.1.4. Coconut husk ash

Coconut husk is a low-cost, potassium-rich natural waste. The presence of a high level of inherent potassium makes the coconut husk derived catalyst as a highly active base to produce biodiesel even at room temperature (Vadery et al., 2014). To investigate the feasibility of biodiesel production at room temperature using coconut husk ash, an endeavor has been made by Vadery et al. (2014). It was noted that 350 °C was the optimum calcination temperature to produce a maximum of 5% ash from coconut husk. The authors reported that variation of catalyst quantity (3 wt%–8 wt%) significantly affected the fatty acid methyl ester (FAME) content and the optimum catalyst dosage to produce maximum yield was identified as 5 wt%. The study showed that more than 97% biodiesel yield can be produced by transesterifying Jatropha oil within 30 min of reaction time at 45 °C reaction temperature.

5.1.5. Palm bunch ash

Palm bunch is the main solid waste from palm oil processing mills (Berger, 1983; Fujii, 2002). In palm fruit bunches, the presence of potassium is a potential source of low-cost heterogeneous catalyst (Konsomboon et al., 2011).

Yaakob et al. (2012) investigated the use of palm-empty fruit bunch ash as a potential catalyst for biodiesel production from Jatropha oil. The study has shown that palm-empty fruit bunch ash can yield more than 98% biodiesel under optimum conditions of reaction (20 wt% catalyst loading, 15:1 molar ratio, 90 min of reaction, and 65 °C reaction temperature). Furthermore, the authors reported that impregnation of KOH on palm-empty fruit bunch ash can produce more than 99% yield under

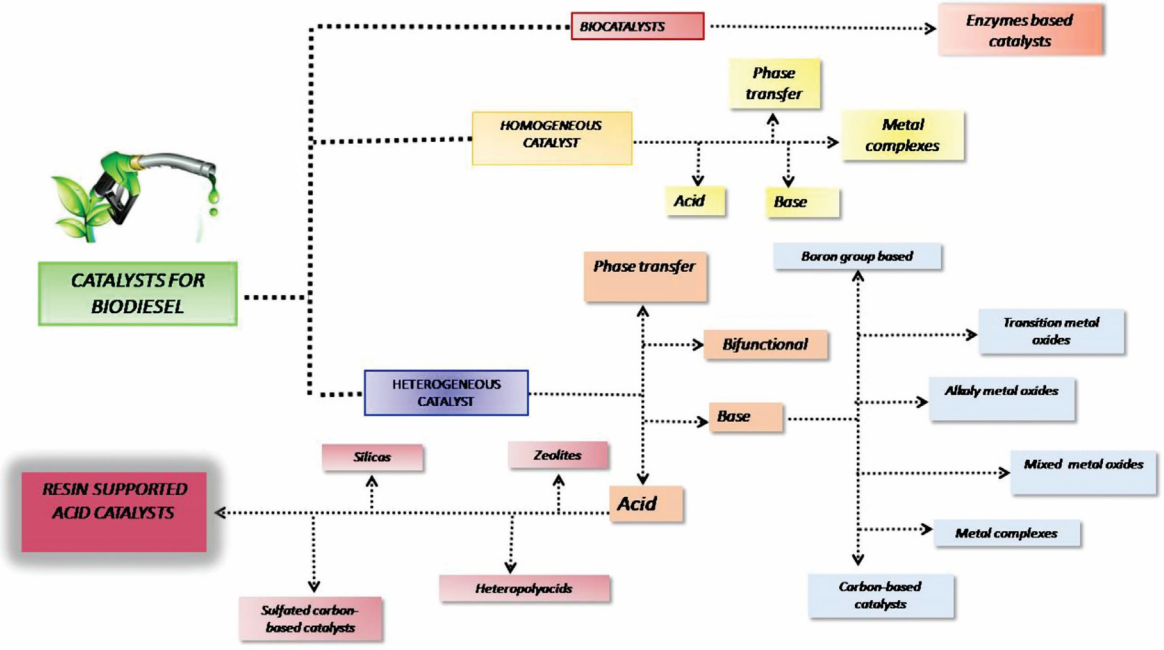

Fig. 4. Summary of the most common catalysts proposed for biodiesel production (Trombettoni et al., 2018). With permission from Elsevier, LN: 4971740260036.

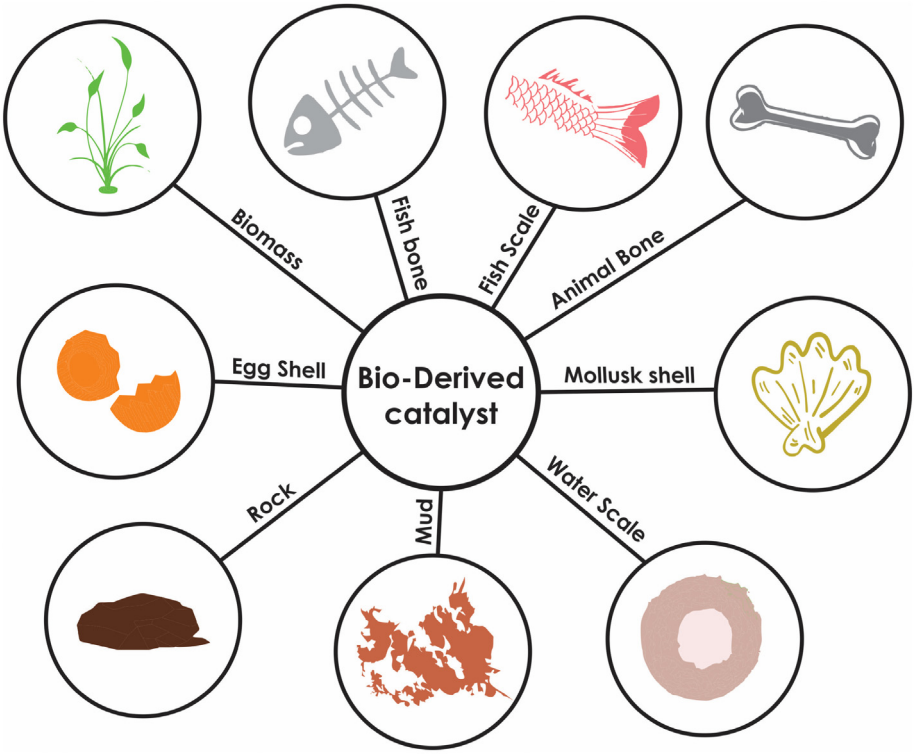

Fig. 5. Potential materials for bio-derived catalyst synthesis

milder reaction conditions (15 wt% catalyst loading, 15:1 molar ratio, 45 min of reaction, and 65 °C reaction temperature). It was also found that, even after reusability up to 5 reaction cycles, this catalyst can generate more than 85% biodiesel yield with palm-empty fruit bunch ash catalyst and more than 92% yield with KOH impregnated palm-empty fruit bunch ash catalyst.

Another research was carried out by Riadi et al. (2014) to explore the potential of palm bunch ash as a supporting catalyst for potassium hydroxide. The research has suggested that simultaneous ozonolysis and transesterification reaction (Fig. 7) can produce higher biodiesel yield by supplying ozone gas continuously for 180 min at 30 °C reaction temperature.

5.1.6. Peanut husk ash

Peanut husks are potential waste materials that are often burned or thrown away in agricultural lands. This farm waste is thought to have high silica content (Safarik and Safarikova, 2010). The use of heterogeneous catalysts with high silica content in biodiesel production can generally eliminate the process of purification and thus reduce the reaction time. To assess the potential of peanut husk in biodiesel production, Dai et al. (2014) produced highly active solid heterogeneous catalyst by calcinating the mix of grounded peanut husk and lithium carbonate (Li_2CO_3) at 900 °C for 2 h. The authors observed that variation of Li_2CO_3:peanut husk ash molar ratio significantly affected the conversion percentage (Table 1). The calcined catalyst (5 wt%) was then added to 12:1 methanol to soybean molar ratio and the reaction was carried out for 3 h at a constant reaction temperature of 65 °C. After 3 h, 97% biodiesel yield was achieved. Even after five reaction cycles, the catalyst has shown relatively good operational stability by yielding over 85%.

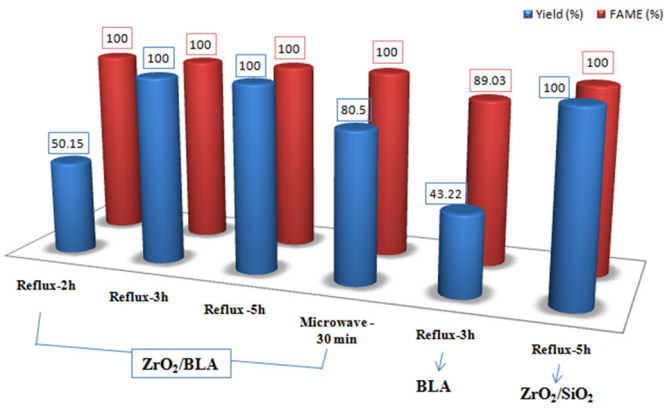

Fig. 6. Biodiesel conversion under different reaction conditions using bamboo leaves derived catalyst (Fatimah et al., 2018).

5.1.7. Rice husk ash

The improper disposal of burned rice husk is considered a major environmental threat in several developed and developing countries. This high silica content raw material has been explored by several researchers as a potential catalyst for biodiesel production (Witoon et al., 2008; Saputra et al., 2018). Research by Chen et al. (2015a) disclosed that more than 91% biodiesel yield can be produced by transesterifying palm oil to biodiesel using 800 °C calcinated rice husk ash. Furthermore, the catalyst reusability study showed that even after eight reaction cycles, the catalyst can yield more than 85%. The researchers also attempted to improve the catalytic activity of rice husk ash by adding lithium carbonate as an activator (Chen et al., 2013). This highly active catalyst was produced by adding 1.23 g of lithium carbonate in 1 g of rice husk ash and the mixture was calcinated at 900 °C for 4 h. The experimental results showed that the reprocessed catalyst could achieve 94% yield for the first time and more than 85% following five reaction cycles. Researchers have also made an effort to make nanocatalyst from rice hull/husk ash. Zeng et al. (2014) investigations revealed that nano-sized solid acid catalyst having a particle size of 50 to 100 nm can be synthesized from rice hull ash by acid activation (Fig. 8). This nanocatalyst, when added in appropriate quantity, can

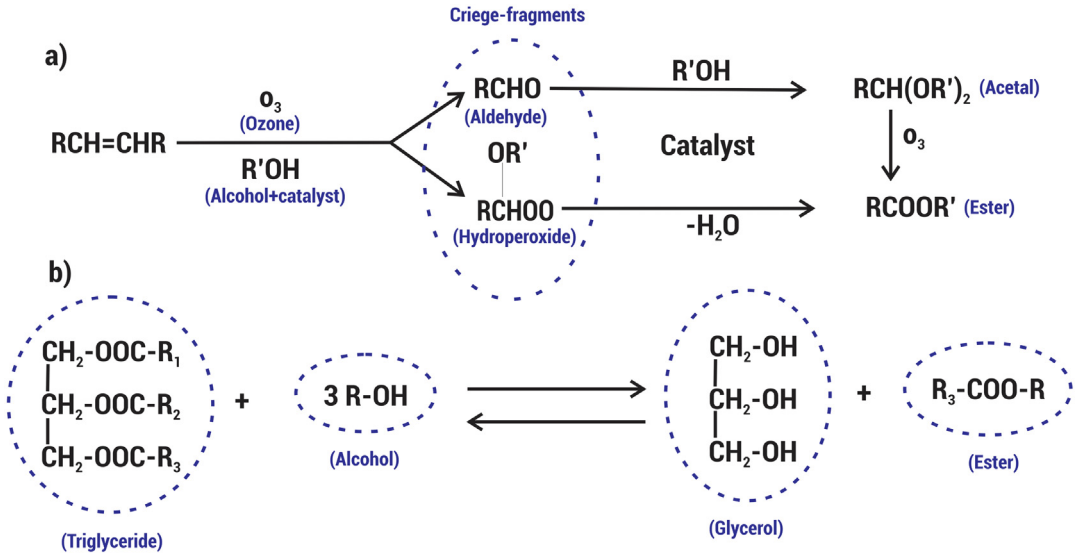

Fig. 7. a) Ozonolysis reaction b) transesterification reaction (modified) (Yaakob et al., 2012).

Table 1
Effect of Li_2CO_3:peanut husk ash molar ratio on conversion (Dai et al., 2014).

Li_2CO_3:peanut husk ash molar ratio	Basic strength	Conversion (%)
0 (Only peanut husk ash)	$H_- < 7.2$	0.36
1:0.5	$7.2 < H_- < 9.8$	65.8
1:1	$9.8 < H_- < 15.0$	88.5
1:2	$9.8 < H_- < 15.0$	89.5
1:4	$9.8 < H_- < 15.0$	77.9

produce biodiesel yield more than 90%. Also, this catalyst can be reused up to 10 reaction cycles without pretreatment but with a drop in yield (50%).

5.1.8. Wood ash

Forest fire has generated a huge amount of wood ash in recent days, creating environmental problems (Consigli et al., 2016). Although wood ash can be used for various applications, including farming and ceramics, the environmental agencies have formed stringent regulations to dispose this high alkaline waste (Campbell, 1990). In this regard, recent research studies have explored the use of wood ash as an alkaline catalyst for biodiesel production. Sharma et al. (2012) research has found that 97% biodiesel yield from calcined wood ash as a catalyst can be achieved using Jatropha as a feedstock. In addition, by chemically activating wood ash using potassium carbonate and calcium carbonate the authors note that the yield can be further improved by 2%. Through this study, the authors reported that thermally and chemically treated wood ash exhibited better catalytic activity during transesterification reaction. The observations recorded by researchers during transesterification reaction using thermally and chemically treated wood ash catalyst are provided in Table 2.

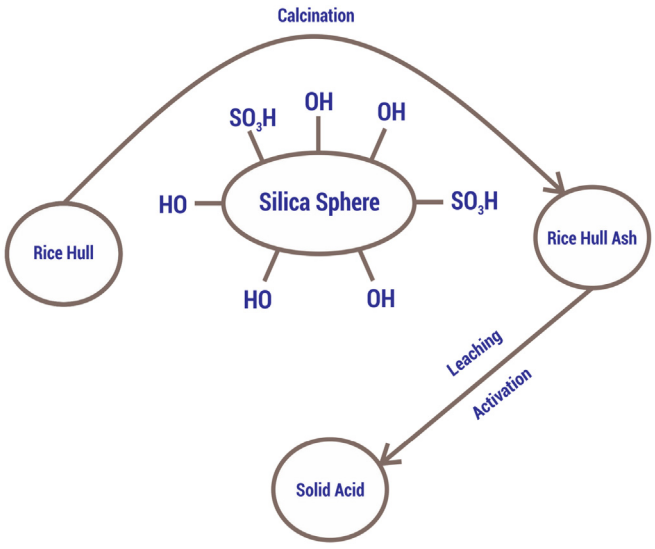

Fig. 8. Nano catalyst synthesized from rice husk hull by acid activation (modified) (Zeng et al., 2014).

Table 2
Observations recorded by Sharma et al. (2012) during the transesterification of Jatropha oil with thermally treated and chemically activated wood ash catalysts. With permission from Elsevier, LN: 4971830151270.

Catalyst	Ester conversion (%)	Observations
W_0	40.2	Slow reaction, require more settling time for catalyst and glycerine
CWC_{500}	42.2	Slow reaction, require more settling time for catalyst and glycerine
CWC_{800}	98.7	Fast reaction, required no refining after catalyst and glycerin separation
CWC_{1000}	98.2	Slow reaction, more reaction time, required no refining after catalyst and glycerin separation
CWC_{1200}	60.5	Slow reaction, required no refining
K_2CO_3	98.5	Very fast reaction, catalyst leached, act as homogeneous catalyst, product change to viscous material after glycerin separation
A_KWC_1	99.0	Fast reaction, fast glycerin and catalyst separation, no refining
$A_KWC_{0.5}$	99.0	Fast reaction, fast glycerin and catalyst separation, no refining
$A_KWC_{0.25}$	97.5	Slow reaction, fast glycerin and catalyst separation, no refining
$CaCO_3$	2.9	Slow reaction, emulsion formation, Vegetable oil remains unreacted
$A_{Ca}WC_{0.5}$	91.7	Slow reaction, emulsion formation, methyl ester refined by washing with brine

W_0 - wood ash catalyst, CWC - calcined wood ash catalyst, AWC - activated wood ash catalyst. Reaction conditions: oil to methanol ratio, 1:12; catalyst dosage, 5 wt%; reaction time, 3 h under reflux conditions.

Researchers' experimental studies also showed that supported wood ash catalyst can yield more than the unsupported wood ash catalyst. Uprety et al. (2016) research has revealed that the wood ash catalyst supported by alumina can produce 98% biodiesel yield, while for the unsupported catalyst the yield was 1% lower. Furthermore, the purity of glycerol recovered was 4% higher for supported ash catalyst compared to unsupported catalyst.

Overall, the influence of different biomass derived ash catalyst on biodiesel yield is portrayed in Table 3.

5.2. Industrial waste ash as catalyst

5.2.1. Coal fly ash

The fly ash obtained from coal-fired power plants is highly alkaline and pollutes the environment (He et al., 2012; Youcai, 2016). On the other hand, the fly ash of coal is considered to contain high oxides of silicon, calcium and magnesium. Thus, the use of this industrial residue as a biodiesel catalyst not only reduces costs but also controls pollution (Ram and Masto, 2014). Studies by Xiang et al. (2016, 2017) disclosed that hydrothermal treatment of coal fly ash can yield around 95% biodiesel by microwave radiation, and more than 95% by ultrasonic irradiation using waste cooking oil as a biodiesel feedstock. Furthermore, studies by Manique et al. (2017) disclosed that hydrothermal synthesis of coal fly ash zeolite can yield over 95% biodiesel. The steps involved in preparation of zeolite catalyst by hydrothermal process using coal fly ash are illustrated in Fig. 9.

Research studies have also shown that highly crystalline zeolites can be produced by optimizing the synthesis conditions (Bhandari et al., 2015). In addition to biodiesel production using zeolite derived from coal fly ash, a value-added chemical glycerol carbonate can also be synthesized by transesterifying crude glycerol with dimethyl carbonate in the presence of coal fly ash derived zeolite as a catalyst (Algoufi and Hameed, 2014).

In the direction of preparing heterogeneous catalyst using coal fly ash, limestone can be added as a supporting agent as reported by Widayat et al. (2017). This research has indicated that the prepared heterogeneous catalyst is reusable up to three reaction cycles without loss of biodiesel yield. In addition to adding support agents, ion exchange method to derive heterogeneous catalyst from coal fly ash can produce high-quality biodiesel as reported by Babajide et al. (2012).

Table 3
Comparison of yield for different biomass derived ash catalyst.

Feedstock	Catalyst	Yield	Reference
Soybean oil	Banana peel ash	98.95%	Pathak et al. (2018)
Rice bran oil	Bamboo leaves ash	100%	Fatimah et al. (2018)
Soybean oil	Cocoa pod husk ash	91.40%	Ofori-Boateng and Lee (2013)
Soybean oil	Cocoa pod husk ash-MgO	98.70%	Ofori-Boateng and Lee (2013)
Jatropha oil	Coconut husk ash	99.62%	Vadery et al. (2014)
Jatropha oil	Palm bunch ash	98.54%	Yaakob et al. (2012)
Jatropha oil	Palm bunch ash-KOH	99.45%	Yaakob et al. (2012)
Soybean oil	Peanut husk ash-Li_2CO_3	97%	Dai et al. (2014)
Palm oil	Rice husk ash	91.50%	Chen et al. (2015a)
Soybean oil	Rice husk ash-Li_2CO_3	94%	Chen et al. (2013)
Soybean oil	Rice husk ash (50–100 nm)	92%	Zeng et al. (2014)
Calophyllum inophyllum seed oil	Rice husk ash	87.68%	Saputra et al. (2018)
Jatropha oil	Wood ash	97%	Sharma et al. (2012)
Jatropha oil	Wood ash-K_2CO_3-$CaCO_3$	99%	Sharma et al. (2012)
Palm oil	Wood ash-Al_2O_3	98%	Uprety et al. (2016)

5.2.2. Palm oil mill boiler ash

The fly ash from the palm oil mill is a prominent inorganic waste, typically high in silica and alumina, which can be used as a low-cost catalyst for biodiesel production (Helwani et al., 2018). Maniam et al. (2013) used the decanter cake (feedstock) and boiler ash (catalyst) waste of a palm oil mill to produce biodiesel efficiently. They reported that more than 85% biodiesel yield is possible under optimal reaction conditions. For maximum biodiesel conversion, heterogeneous catalyst can be prepared by loading calcium oxide/calcined calcium carbonate onto palm oil mill boiler ash as reported by Ho et al. (2012, 2014). The authors reported that 15 wt% of calcined calcium carbonate at 800 °C loaded onto fly ash is the optimum catalyst quantity with which 94.5% conversion yield can be obtained.

5.3. Industrial slag as catalyst

5.3.1. Carbide slag

Carbide slag is a calcium-rich residue produced as a byproduct in ethyne gas production industry. Besides calcium, carbide slag contains other residual minerals like magnesium, iron and silicon (Cao et al., 2008; Liu et al., 2010). This slag is used as a primary raw material for polyvinyl chloride and cement processing industries (Sun et al., 2013; Namarak et al., 2018; Tan et al., 2018). Several studies have reported that calcium oxide-based catalyst can be derived from carbide slag. Several studies were carried out to test the catalytic performance of catalyst derived from carbide slag in biodiesel production.

Li et al. (2015) calcinated carbide slag at 650 °C and added 1 wt% of the prepared catalyst to 9:1 menthol to soybean molar ratio and carried out the transesterification reaction. It was noted from their investigation that the calcined carbide slag catalyst can yield more than 90% biodiesel within 30 min of reaction time. Similar observations were observed in the research conducted by Liu et al. (2014). The authors also observed that transesterification of peanut oil by calcined carbide slag (650 °C) followed the reaction orders 1.03, 0.84 and 1.02 at 57 °C, 60 °C and 63 °C, respectively. Furthermore, the kinetic model of transesterification of peanut oil by calcined carbide slag catalyst can be represented as shown in Eq. (1).

$$k = 1.75 \times 10^9 \; exp\left(-\frac{68.45}{RT}\right) \tag{1}$$

where, R is the gas constant and T is the absolute temperature.

5.3.2. Blast furnace slag

Blast furnace slag is a large volume byproduct obtained from iron processing industries. Although this material is recycled to cement, concrete and pavement materials, its high volume production and storage problems necessitate advanced recycling processes to convert this slag into other value added products including cement admixture (Gumieri et al., 2003) and adsorbents (Seggiani and Vitolo, 2003; Kuwahara et al., 2010). Recent studies revealed that hydrocalumite synthesized from waste slag (Fig. 10) can be used as catalyst in transesterification process. Kuwahara et al. (2012) attempted to derive mixed oxides of calcium and magnesium from blast furnace slag by calcinating it at 800 °C. The addition of 1 wt% of the prepared mixed oxide catalyst to methanol oil molar ratio of 12 at a constant reaction temperature of 60 °C for 6 h produced up to 97% yield. The recyclability test using the prepared mixed oxide catalyst confirmed that the yield significantly drops by 11% during the second cycle and 30% during the third cycle due to leaching of active elements.

5.3.3. Biomass gasifier slag

Gasified straw slag is a byproduct obtained from biomass gasifier. This gasified slag consists of mixed oxides of calcium, magnesium, sodium and potassium (Heinzel et al., 1998). These oxides contain potential active sites to be used as a solid heterogeneous catalyst for biodiesel

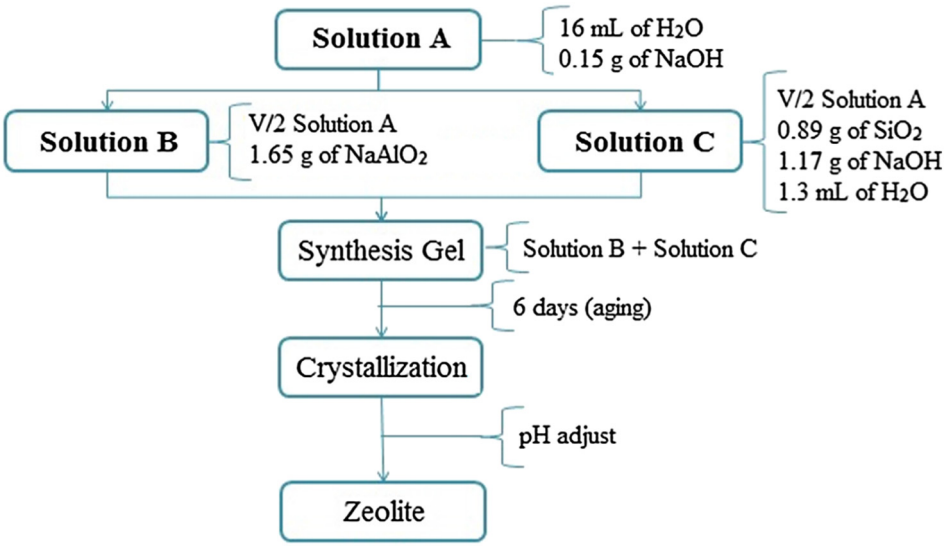

Fig. 9. Synthesis of zeolite from coal fly ash by hydrothermal method (Manique et al., 2017). With permission from Elsevier, LN: 4971740650371.

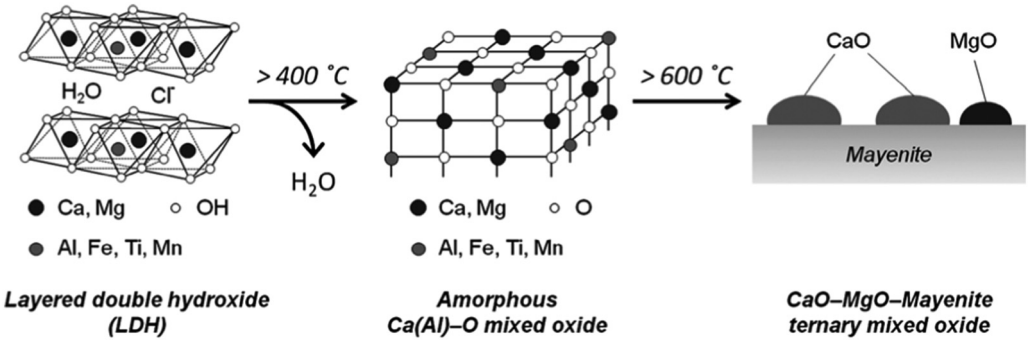

Fig. 10. Hydrocalumite synthesis from waste slag (Kuwahara et al., 2012). With permission from Royal Society of Chemistry, LN: 1085088-1.

production. Wang et al. (2017) assessed the catalytic activity of gasified straw slag in biodiesel production from rapeseed oil. The catalyst exhibited high catalytic activity and stability with better recyclability. It was noted from the experiment that more than 95% yield could be achieved during the first reaction cycle and up to 85% after 33 repetitive cycles (Fig. 11).

5.3.4. Magnesium plant slag

The waste slag from magnesium processing industry consists of abundant magnesium and calcium. The influence of these elements as a catalyst in the conversion of rapeseed oil was reported by Zhang et al. (Zhang and Huang, 2011). It was reported in the study that the rapeseed oil conversion rate increased on an average by 10% with every 1% increase in magnesium-calcium content and the highest conversion of 98% was reported for 55.5 wt% of magnesium-calcium content. After each reaction cycle, the reusability of the catalyst demonstrated a decrease of 25–30% in conversion rate. However, the conversion rate was significantly improved by calcinating the catalyst at 600 °C under nitrogen flow before the next use and up to 89% conversion was achieved after the third cycle.

Overall, the impact of industrial waste derived catalysts on biodiesel yield using various feedstock reported by researchers is described in Table 4.

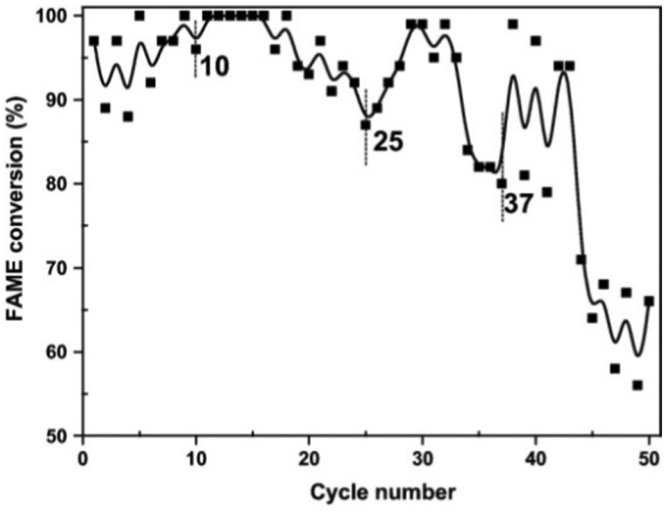

Fig. 11. Reusability of gasified straw slag in biodiesel production (the authors observed that FAME conversion (%) exceeded 95% and was maintained at 85% after 33 cycles without compensation of catalyst) (Wang et al., 2017). With permission from Elsevier, LN: 4971750151957.

Table 4
Comparison of yield for different Industrial waste derived catalyst.

Feedstock	Catalyst	Yield	Reference
Industrial ash			
Soybean oil	Coal fly ash	95.50%	Manique et al. (2017)
Waste cooking oil	Coal fly ash	96%	Xiang et al. (2016, 2017)
Soybean oil	Coal fly ash	91.20%	Bhandari et al. (2015)
Palm oil	Coal fly ash-limestone	83.60%	Widayat et al. (2017)
Sunflower oil	Coal fly ash	83.53%	Babajide et al. (2012)
Palm oil	Mill boiler ash-CaO	97.09%	Ho et al. (2012)
Decanter cake	Mill boiler ash	85.90%	Maniam et al. (2013)
Palm oil	Mill boiler ash-CaO	71.77%	Helwani et al. (2018)
Industrial slag			
Rapeseed oil	Carbide slag	93%	Liu et al. (2014)
Soybean oil	Carbide slag	91%	Li et al. (2015)
Soybean oil	Blast furnace slag	97%	Kuwahara et al. (2010)
Rapeseed oil	Gasifier slag	95%	Wang et al. (2017)
Rapeseed oil	Magnesium plant slag	98%	Zhang et al. (Zhang and Huang, 2011)

5.4. Mud based catalyst

5.4.1. Red mud

Red mud is a highly alkaline solid waste produced in the aluminum production industries during Bayer's process (Paramguru et al., 2005; Deelwal et al., 2014). This solid residue contains several minerals, including aluminum, silicon, iron and titanium in large quantities, and other residual minerals such as calcium and zirconium. Due to its high alkaline nature (pH > 10), it deteriorates the quality of the soil and pollutes the groundwater when disposed of as landfill. Several research studies have focused on judicious recycling of red mud for applications including brick manufacturing, filler material, coagulation, and adsorption (Brunori et al., 2005; Liu et al., 2009). Some researchers have also explored the use of red mud as a catalyst in biodiesel synthesis.

Liu et al. (2013) attempted to produce highly active and low-cost solid catalyst by calcinating red mud at 200 °C. In fact, calcination/cracking helps remove high iron oxides from red mud and turns red mud into an active catalyst (Senthil et al., 2016a; Senthil et al., 2016b). The study reported that addition of 4 wt% of the calcined catalyst to 24:1 methanol soybean oil molar ratio at a fixed reaction temperature of 65 °C for 3 h can produce more than 94% yield. Besides simple calcination, soda-lime calcination and loading various chemical and biological compounds to red mud would improve its catalytic activity as reported in the literature. Wahyudi et al. (2017) reported that by synthesizing the catalyst through the soda-lime calcination process, 99% biodiesel yield could be achieved. Research by Zhang et al. (2016) proved that adding potassium fluoride (KF) to red mud will also improve its catalytic activity. The authors observed that the new crystalline phase of $KFeF_4$ formed during the addition of KF to red mud as represented in Eq. (2).

$$Fe_2O_3 + 8KF + 3H_2O = 6KOH + 2KFeF_4 \quad (2)$$

Researchers have also tried to produce composite catalysts in recent studies by adding lipid waste to red mud by co-pyrolysis method (Yoon et al., 2019) as illustrated in Fig. 12. Their research concluded that composite catalysts can perform better than commercial silica catalyst in biodiesel conversion. It was reported in their study that the red mud-lipid composite waste catalyst can yield more than 90% biodiesel at 130 °C reaction temperature, whereas 350 °C is required to achieve the same yield using commercial silica catalyst.

5.4.2. Lime mud

During the alkali recycling process, lime mud is produced as a by-product in the papermaking industry. This paper mill residue can be calcinated at different temperatures (500 °C to 900 °C) to obtain basic heterogeneous catalyst as reported by Li et al. (2014a). The authors reported that the use of the lime mud derived catalyst (calcinated at 800 °C) could achieve more than 90% biodiesel yield. In addition, the catalytic performance of calcined lime mud catalyst was found to be on par with the laboratory grade calcium oxide up to five reaction cycles. Further research studies reveal that doping potassium fluoride with lime mud enhances its active sites and it is possible to achieve 99% biodiesel yield (Li et al., 2014b).

5.4.3. Lapindo mud

Lapindo is a name given to the volcanic eruption mud first discovered on 29 May 2006 in Indonesia Sidoarjo district. The mud's chemical analysis showed several metal oxides which exhibit active sites to be used as a catalyst. Researchers reported that potassium, calcium and silicon are the major elements present in lapindo mud (Talib et al., 2016). The synthesis of biodiesel from waste oils using lapindo mud revealed that lapindo:methanol:oil weight ratio of 0.03:0.44:1 can achieve more than 95% biodiesel yield. In addition, the catalyst's stable performance was seen up to the seventh reaction cycle with yield dropping by only 10%. The authors also noted that transesterification of waste cooking oil and palm oil using lapindo mud Langmuir–Hinshelwood first order kinetics and the minimum energy required to precede the reaction were calculated to be 55.7 kJ/mol and 59.8 kJ/mol for waste cooking oil and palm oil, respectively.

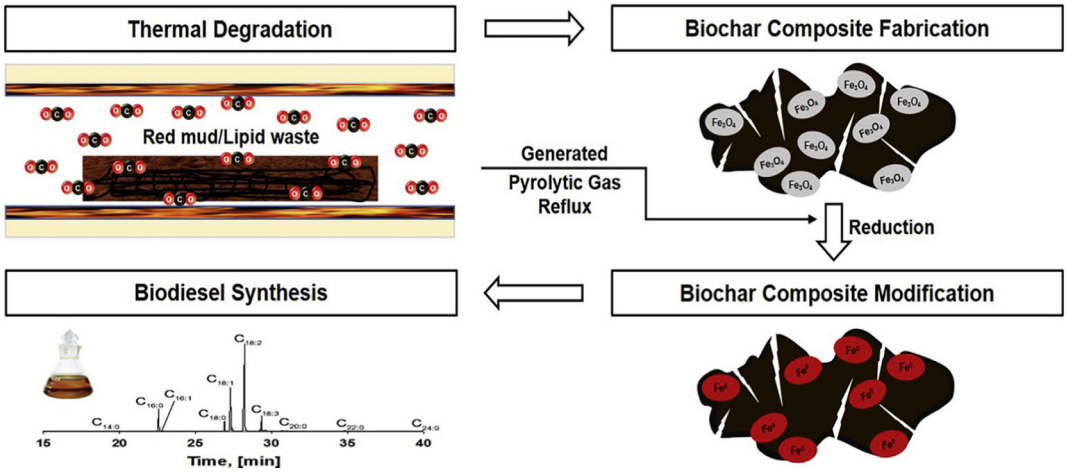

Fig. 12. Composite biochar fabrication from red mud and lipid waste (Yoon et al., 2019). With permission from Elsevier, LN: 4971750414734.

5.5. Rock based catalyst

5.5.1. Dolomite rock

Dolomite is a naturally abundant material mainly sourced from quarries. Dolomite rock is used in several industrial applications, including the manufacture of rubber, glass, iron, steel and plastics (Wilson et al., 2008). The structure of dolomite consists mainly of calcium-rich phase of magnesium calcite (Fig. 13). Several researchers have explored the utilization of this naturally occurring waste material as a heterogeneous base catalyst for biodiesel production.

Wilson et al. (2008) derived solid catalyst from natural dolomite rock obtained from the Northern Ireland deposit. By calcination at 900 °C for 3 h, the researchers extracted a solid base catalyst from fresh dolomite rock. Transesterification of olive oil with methanol was investigated to test the activity of the prepared catalyst. It was observed that the process of higher temperature calcination improved the basicity and surface area of the prepared catalyst which aided in better conversion of short and long chain fatty acids. Apart from high-temperature calcination, sulfonation of dolomite rock would also increase its surface area and pore volume. This procedure would, however, reduce the diameter of the pore which could limit the molecular diffusion of long chain fatty acids as reported by Vargas et al. (2019).

5.5.2. Igneous/volcanic rocks

Igneous rocks are porous volcanic rocks formed on the earth's surface by solidifying molten lava (Jaeger, 1968). These porous magmatic rocks find wider application in medical, textile, construction and cosmetic industries. The feasibility of biodiesel synthesis using volcanic rocks is reported in several research works. Samsudeen et al. (2017) has investigated how the commonly available pumice volcanic rock can be used as a catalyst to produce biodiesel from neem oil and ethanol. In their work, the pumicite (powdered pumice rock) was treated with potassium hydroxide to increase its catalytic activity. Under mild reaction conditions, 90% yield was recorded using this bifunctional catalyst. Other than pumice rock, scoria igneous rock can be used as catalyst support for biodiesel production as reported by Iranian researchers (Mohadesi et al., 2019). Based on their findings, it was observed that the high surface area of scoria could accelerate the transesterification reaction.

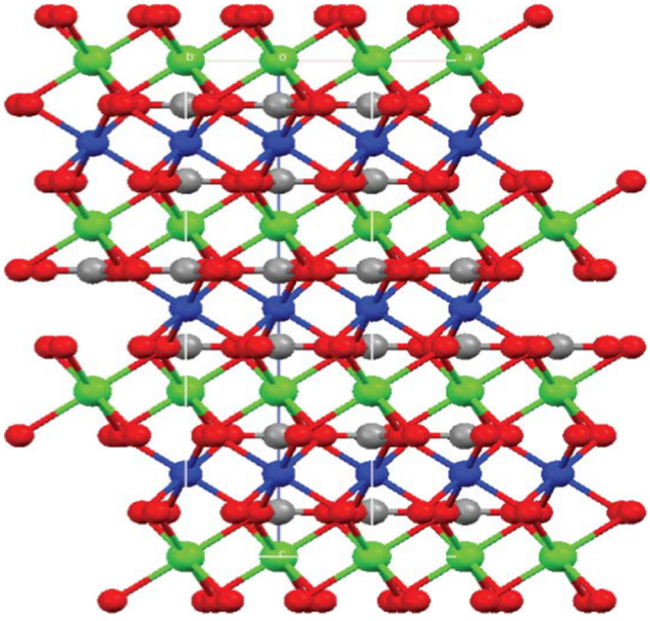

Fig. 13. Structure of dolomite $MgCa(CO_3)_2$ (C–grey, Mg^{2+}–blue, Ca^{2+}–green, O^{2-}–red) (Wilson et al., 2008). With permission from Royal Society of Chemistry, LN: 1085085-1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

5.5.3. Shale rock

Shale is a fissile sedimentary rock that consists of a mineral blend of clay, quartz and calcite. For years, shale was widely used for sequestration of heavy metals from waste water (Shawabkeh, 2009; Wei et al., 2013). To explore the catalytic potential of shale-derived catalyst in biodiesel synthesis, Doyle et al. (2017) extracted high-purity zeolite from surface shale rock by acid leaching followed by alkaline fusion and hydrothermal treatment. This high purity substance was used as a catalyst for oleic acid conversion. The conversion rate recorded with the prepared catalyst was found to be equal to commercial zeolite.

5.5.4. Kaolinite rock

Kaolinite is a mineral clay that is usually produced by chemical weathering. It consists mainly of basic aluminum silicate and is used in ceramics industries (Becerro et al., 2009). Novembre et al. (2018) collected kaolinite rock from North Sardinia Island in Italy and synthesized kalsilite by hydrothermal route. The extracted kalsilite was used as a heterogeneous solid catalyst for biodiesel synthesis from used frying oil. The researchers disclosed that kalsilite is an active catalyst for biodiesel production and hydrothermal route to extract kalsilite could significantly reduce the operating temperature by over 100 °C.

5.6. Waste scale as catalyst

5.6.1. Fish scale

Aquaculture is a booming sector in several developing and developed nations, but this also means effluent from the aquaculture industry is booming as well (Knuckey et al., 2004). Among different effluents from fish farms, the fish scale has got a high potential for recycling and reuse. The main constituent of fish scale is hydroxyapatite (calcium-rich substance). Literature data also indicate that calcium oxide derived from natural hydroxyapatite has significant basic strength to be used as a catalyst (Hamada and M, 2011). A group of researchers from India explored the viability of Labeorohita fish scale as a heterogeneous catalyst for biodiesel production. In India, Labeorohita is grown in large volumes among a variety of fish species (Chakraborty et al., 2011). As reported by researchers, calcinating Labeorohita fish scale at relatively high temperature (>900 °C) could effectively catalyze the reaction and yield ester content over 95%. The research group also reported that impregnation of nickel on the treated fish scale could further improve the yield by over 2% (Chakraborty and Das, 2012).

5.6.2. Waste water scale

Water scales are deposits of insoluble solid minerals produced primarily when carbonates/bicarbonates of calcium or magnesium containing water is heated. The representation of extracted water scale is represented in Fig. 14. Water scales contain several metal oxides with a high level of calcium oxide (Zhang et al., 2014). This calcium-rich residue could be used as a base catalyst for biodiesel production as reported by Zhang et al. (2014). Their research work attempted to derive calcium oxide based catalyst with water scales collected from home kitchens in eastern China. It was reported that the water scale derived catalyst is highly active and stable at 1000 °C. Their research revealed that about 93% biodiesel yield is achievable by using 1 wt% dose of derived catalyst in the methanolysis of soybean oil. In addition to water scales, water sludge obtained during defluoridation process can also be synthesized into solid base catalyst for biodiesel production as suggested by Gupta et al. (2017).

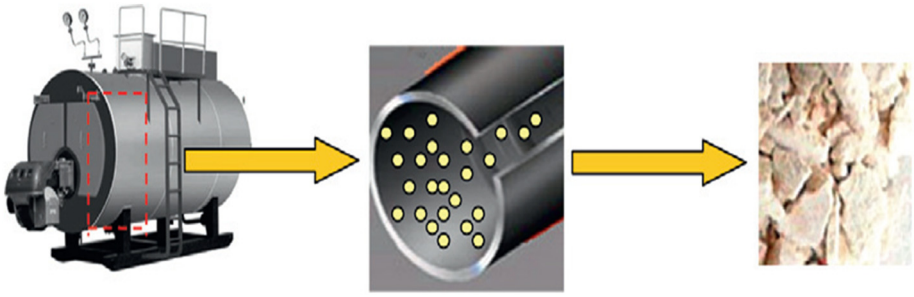

Fig. 14. Representation of water scale extraction (Zhang et al., 2014). With permission from Elsevier, LN: 4971751504575

5.7. Waste shell as catalyst

5.7.1. Eggshell

Eggshell is a semi-permeable membrane entirely made of calcium carbonate (in calcite form). This biowaste is an excellent material for recycling and reuse. The egg shells are used in several interesting applications including waste water treatment (Mittal et al., 2016) and biodiesel production (Mansir et al., 2018). The capability of eggshells as a bifunctional catalyst for biodiesel synthesis is reported in several research studies (Mansir et al., 2018; Ngadi et al., 2016). The synthesis of catalyst from egg shell is represented in Fig. 15.

Piker et al. (2016) investigated the potential of calcined eggshell (900 °C for 3 h) in the biodiesel synthesis from soybean oil and waste cooking oil. Their research indicated that high fatty acid methyl ester content of 98% using soybean oil and about 97% using waste cooking oil is achievable by using the eggshell synthesized catalyst. The synthesis of highly active and stable nano catalyst from waste egg shell is also reported in the literature.

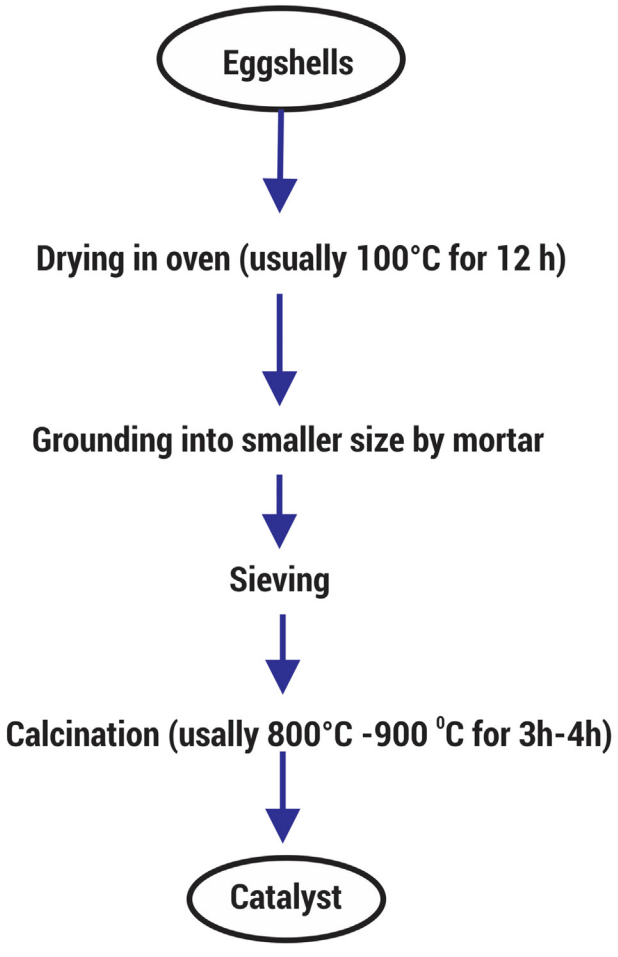

Fig. 15. Representation of catalyst synthesis from egg shell (Mansir et al., 2018).

The reusability tests with the derived egg shell catalyst showed that conversion yield gradually decreases after each run (Piker et al., 2016). Similar observations were reported in the work conducted by Pandit et al. (Pandit and Fulekar, 2017) and Farooq et al. (2018). Through the detailed analysis, the researchers concluded that the glycerol and other organic compounds cover the active sites and gradually decrease the catalytic activity after using the catalyst for several runs.

To improve the catalytic activity of eggshell derived catalyst, the researchers suggested incorporating metal precursors such as zinc, ferrous, manganese, aluminum and lithium (Joshi et al., 2016; Boro et al., 2014; Borah et al., 2019).

5.7.2. Mollusk shells

Mollusks are soft-bodied creatures usually enclosed in a calcium carbonate shell. Worldwide, more than one hundred thousand mollusk species have been identified and this green waste draws the attention of many researchers because of its potential to afford natural calcium (Suzuki and Nagasawa, 2013). Several researchers successfully derived calcium oxide based catalyst from different species of mollusk shell and tested its catalytic performance. In this regard, Viriya-empikul et al. (2010) extracted calcium oxide from two different mollusk shells such as apple snail shell and meretrix venus shell. These two shells were converted to calcium-rich catalyst by calcinating them at 800 °C. Between these shells, the catalytic performance of apple snail shell was found to be better in biodiesel conversion because of its higher surface area compared to meretrix venus shell. Among different mollusk species, Pila globosa (Agrawal et al., 2012), Anadara granosa (Hadiyanto et al., 2016a; Hadiyanto et al., 2016b), Paphia undulate (Hadiyanto et al., 2016a), and Margaritifera falcate (Madhuvilakku et al., 2013) were commonly used species by researchers for the extraction of calcium oxide. It has also been reported that the desired calcination temperature ranges from 800 °C to 900 °C to draw better catalytic activity from mollusk shells.

5.8. Waste animal bone as catalyst

Besides scales and shells, animal bones are the rich natural source of calcium. As indicated in the literature, bones contain 60–70% hydroxyapatite and 30–40% of other organic compounds (Mori et al., 2002). Waste bones attracted researchers because of their potential to be used as a catalyst, an absorbent and an ion exchange agent (Jazie et al., 2013; Obadiah et al., 2012). Researchers have attempted to derive solid base heterogeneous catalyst from waste bones of goat (Jazie et al., 2013), sheep (Obadiah et al., 2012), chicken (Farooq and Ramli, 2015), pig (Chen et al., 2015b), bovine (Smith et al., 2013), and fish (Sulaiman and Amin, 2016).

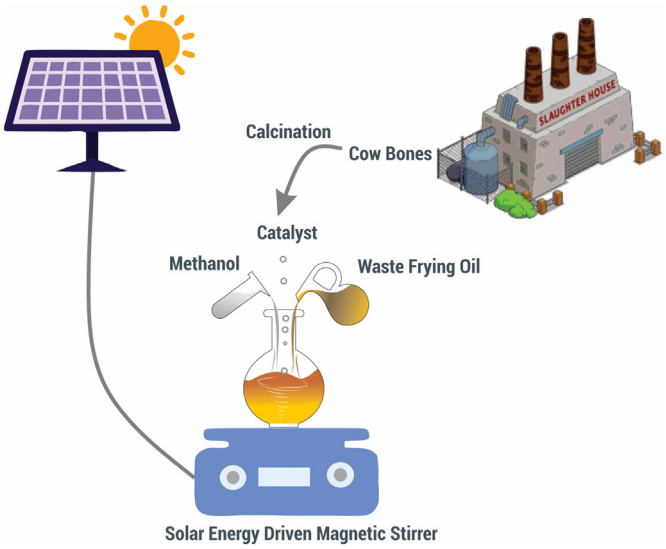

Fig. 16. Solar reactor used for biodiesel production (modified) (Corro et al., 2016).

The catalytic potential of calcined waste goat bone (900 °C) in biodiesel synthesis was assessed by Jazie et al. (2013). The authors produced peanut methyl ester and rapeseed methyl ester using the derived catalyst. It has been reported that the biodiesel prepared using the bone-derived catalyst has physical and chemical properties that are very similar to mineral diesel and comply with the available standards.

Further studies by researchers showed that a higher biodiesel yield (>90%) within low reaction time (≤5 min.) is possible by microwave irradiation method of biodiesel production using waste bone-derived catalyst (Buasri et al., 2015). Researchers explored, in addition to microwave heating, that solar radiation could be a potential renewable source of energy to produce biodiesel using bone-derived catalyst (Corro et al., 2016). Their research has shown that the use of solar heat for biodiesel production can save almost 7 kWh of total energy. The authors also suggested carrying esterification of fatty acids by animal bone derived catalyst followed by alkaline transesterification to obtain maximum conversion (>95%). They noticed that animal bone derived catalyst can be reused up to 10 reaction cycles without drop in efficiency. The representation of photovoltaic cell assisted reactor used for biodiesel production from used frying oil used cow bone derived catalyst is depicted in Fig. 16.

Overall, it is disclosed from the literature survey that most of the bio-derived catalysts discussed here consist of CaO as the main compound. The mechanism of transesterification using bio-derived CaO based catalyst is depicted in Fig. 17. One could observe that this reaction mechanism involves three major steps. First, the methoxide anion is attracted by the triglyceride carbonyl group (i.e., the carbonyl carbon nucleophilic attack takes place). Next, there is the formation of tetrahedral intermediate. At last, there is the formation of diglyceride anion and methyl ester.

6. Practical implications of this study

A large number of studies prove that bio-derived catalysts have potential as heterogeneous catalysts in the transesterification process. Among different bio-derived catalysts, biomass/industrial ash based catalysts are inexpensive and easy to use. Different methods have been developed for optimizing ash based catalyst's properties according to the specific use. Other underlying characteristics of these catalysts are the presence of surface functional groups and the existence of inorganic compounds, which may be beneficial for its use as a catalyst or as a support for catalyst.

On the other hand, CaO based catalyst derived from waste bones, scales and shells reveal that they exhibit high basic strength and low environmental impact when used as a catalyst for biodiesel production. However, the major challenges with CaO based catalysts are reusability in successive cycles, atmospheric poisoning, leaching into reaction medium, sensitivity of these catalysts under ambient conditions and the

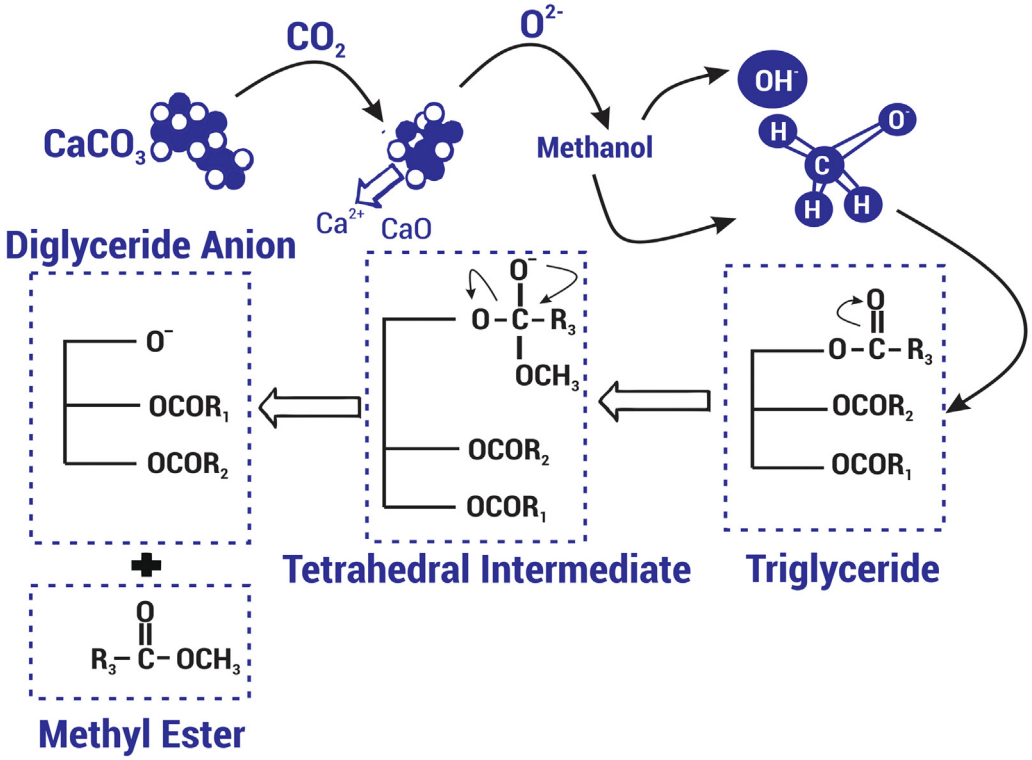

Fig. 17. Representation of reaction mechanism of triglyceride to methyl ester using bio-derived CaO based catalyst (Li et al., 2015; Chakraborty et al., 2011; Zhang et al., 2014).

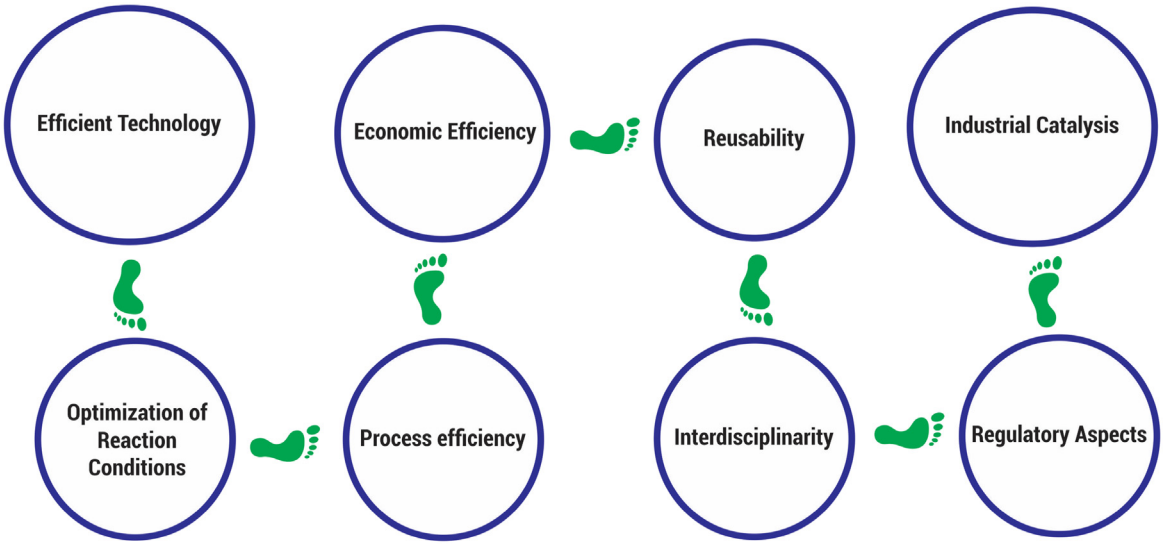

Fig. 18. Challenges towards industrial bio-derived catalysis.

need for pretreatment step when feedstock with high free fatty acid content is used. In addition to ash and CaO based catalysts, it is revealed from the literature that mud and rocks can be recycled to low cost and active catalysts.

The current literature proves that bio-derived catalysts have a potential for industrial biodiesel production. However, it is also necessary to further explore the challenges of catalyst synthesis from waste materials by highlighting the technical, economic and environmental facets as illustrated in Fig. 18. Finally, further research and development are required to exploit these waste-derived catalysts at the industrial level.

6.1. Environmental factor (E-factor) assessment of biodiesel production using bio-derived catalysts

In this review, an attempt has been made to calculate the E-factor of biodiesel production using bio-derived catalysts. The E-factor calculation is based on the expression proposed by Roger Sheldon (Sheldon, 2017) (Eq. (3)). The E-factor calculation was performed assuming biodiesel as the product and glycerol as waste.

$$\text{E}-\text{factor} = \text{Total waste (kg)}/\text{Product (kg)} \quad (3)$$

The calculated E-factor for different literature samples is presented in Fig. 19 and the details of samples are provided in Table 5. It can be seen that the calculated values are high for all considered cases. This is because of the assumption made earlier. On the other side, one can also see that the E-factor for biodiesel production using catalysts extracted from mud is more among the measured values. This shows that more waste is generated during the mud derived catalytic transesterification process. It was further noted that biodiesel production using catalysts extracted from waste such as banana peel, cocoa pod husk, fish scale and egg shells has less harmful impacts on the environment. This study implies that most of the waste derived catalysts have high potential and less harmful environmental impacts in biodiesel production process.

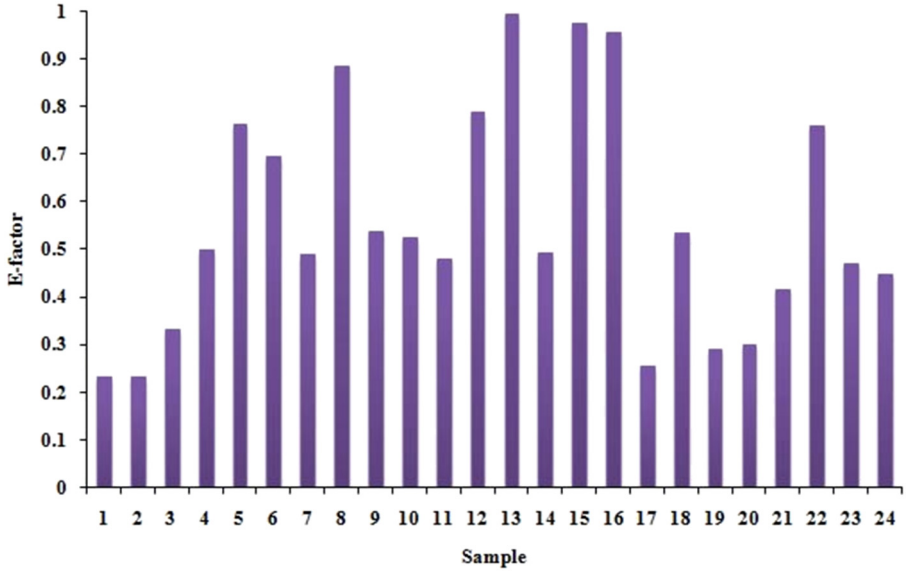

Fig. 19. E-factor plot for different literature samples.

Table 5
Details of samples used for E-factor analysis.

Sample	Feedstock	Catalyst
1	Soybean oil	Banana peel ash (Pathak et al., 2018)
2	Soybean oil	MgO doped cocoa pod husk ash (Ofori-Boateng and Lee, 2013)
3	Soybean oil	Cocoa pod hush ash (Ofori-Boateng and Lee, 2013)
4	Jatropha oil	Coconut husk ash (Vadery et al., 2014)
5	Jatropha oil	Palm bunch ash (Yaakob et al., 2012)
6	Jatropha oil	Palm bunch ash/KOH (Yaakob et al., 2012)
7	Soybean oil	Peanut husk ash (Dai et al., 2014)
8	Soybean oil	Li modified rice husk ash (Chen et al., 2013)
9	Palm oil	Rice hush ash (Chen et al., 2015a)
10	Soybean oil	Coal fly ash (Manique et al., 2017)
11	Waste cooking oil	Modified coal fly ash (Alkaline fusion) (Xiang et al., 2016)
12	Palm oil	Palm fly ash/CaO (Helwani et al., 2018)
13	Soybean oil	Red mud (Liu et al., 2013)
14	Canola oil	Soda-lime treated red mud (Wahyudi et al., 2017)
15	Jatropha oil	Red mud/potassium fluoride (Zhang et al., 2016)
16	Peanut oil	Lime mud (Li et al., 2014a)
17	Soybean oil	Fish scale (Chakraborty et al., 2011)
18	Soybean oil	Water scale (Zhang et al., 2014)
19	Soybean oil	Eggshell (Piker et al., 2016)
20	Waste cooking oil	Eggshell (Piker et al., 2016)
21	Waste cooking oil	Mollusk shell (Viriya-empikul et al., 2010)
22	Jatropha oil	Pork bone (Buasri et al., 2015)
23	Soybean oil	Blast furnace slag (Kuwahara et al., 2012)
24	Soybean oil	Carbide slag (Li et al., 2015)

Table 6
Production cost of biodiesel using different feedstocks.

Feedstock type	Biodiesel production cost (per liter)	Ref
Soybean oil	$0.53	(Haas et al., 2006)
Waste cooking oil	$0.80 to $0.90	(Karmee et al., 2015)
Jatropha oil	$0.99	(Ofori-Boateng and Lee, 2011)
Animal tallow	$0.22 to $0.63	(Nelson and Schrock, 2006)
Hemp seed oil	$1.35	(Samuel et al., 2020)
Tobacco seed oil	$1.36	(Samuel et al., 2020)
Microalgae	$2–4	(Delrue et al., 2012)

7. Trends and perspectives

7.1. Techno-economic feasibility of biodiesel production

Several studies were carried out to present the economics of biodiesel production using various feedstocks and processing routes. In general, the economic feasibility of biodiesel production mainly depends on the raw material cost (Cho et al., 2013). As reported by researchers more than 60% of manufacturing cost consists of raw material cost (Cho et al., 2013). In addition to the raw material cost, processing technology also plays an important role in the economy of biodiesel production (Zhang et al., 2003; Marchetti et al., 2008). The production cost of biodiesel using different feedstocks is summarized in Table 6.

Haas et al. (2006) developed a super-pro model to produce biodiesel from degummed soybean oil. In this work, two sequential catalytic transesterification methods have been employed to increase the reaction efficiency. It is reported that more than 85% of the total operating cost was allocated to raw materials and reaming 15% was shared by utility, facility and labor. The similar observations were revealed in the work conducted by Avinash and Murugesan (2017) to produce biodiesel from waste cooking oil by sequential catalytic transesterification method. Apart from classical catalytic transesterification, authors have adopted single-step non-catalytic transesterification, supercritical process, acid catalyzed and heterogeneous solid catalyzed processes to produce biodiesel (Cho et al., 2013; Marchetti et al., 2008; Marchetti and Errazu, 2008; Gebremariam and Marchetti, 2018).

Techno-economic viability study by Gebremariam and Marchetti (2018) in biodiesel production from acidic oil using sulfuric acid and calcium oxide as catalysts revealed that the total annual operating cost can be reduced by 13% when using calcium oxide as catalyst than sulfuric acid. Marchetti et al. (2008) and Marchetti and Errazu (2008) compared the economic feasibility of four different processes such as alkaline, acid, heterogeneous solid catalyzed and supercritical process for biodiesel production. They reported that alkaline catalyzed transesterification due to its low capital investment is the most economical process followed by heterogeneous solid catalyzed, acid catalyzed and supercritical processes.

7.2. Life cycle analysis of biodiesel

Life cycle analysis is popular among biofuel manufacturers to evaluate the environmental impact of the produced fuels with the available standards as illustrated in Fig. 20. This analysis generally takes place in four phases. The first phase is to analyze the purpose and scope, the second phase is to analyze the inventory, the third phase is to analyze the impact and the fourth phase is to interpret the results (Varanda et al., 2011). The first-ever report on life cycle analysis of biodiesel was published in May 1998 by Sheehan et al. (1998). Since 1998 several studies have been carried out using different feedstocks to analyze the life cycle of biodiesel in different countries.

Life cycle analysis of biodiesel production in Brazil revealed that biodiesel production from soybean oil can significantly mitigate the generation of carbon dioxide by 174 times compared to commercial diesel refining (Sieira et al., 2015). Researchers from Canada and Portugal indicated that among different feedstocks, waste cooking oil can reduce the total environmental impact by more than 70% (Varanda et al., 2011; Sajid et al., 2016; Ong et al., 2012). Liang et al. (2013) presented a comprehensive life cycle analysis of biodiesel production in China. The authors evaluated the three “e's” (energy, economics and environmental

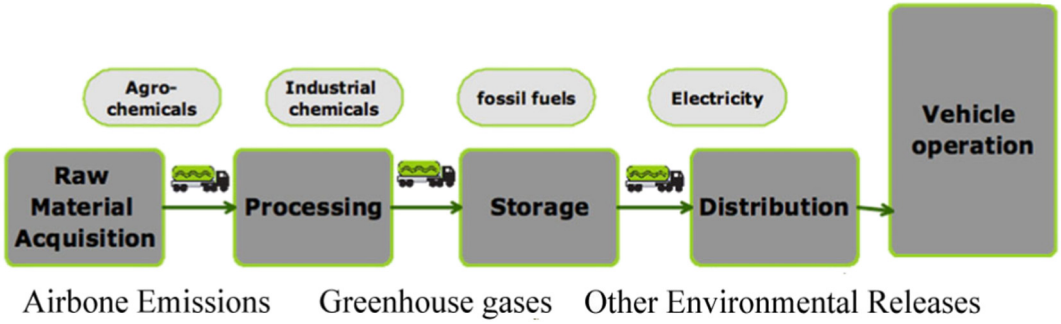

Fig. 20. Life cycle of biofuel.

Table 7
Comparison of lifecycle assessment results of biodiesel.

Impact category	Reference unit	Waste cooking oil (Chung et al., 2019)	Soybean (Hou et al., 2011)	Jatropha (Hou et al., 2011)	Microalgae (Hou et al., 2011)
Abiotic depletion potential	kg Sb eq	1.20E−01	1.6627E−04	9.8160E−05	1.0581E−04
Acidification potential	kg SO_2 eq	2.15E−02	1.4147E−03	9.0947E−04	6.5177E−04
Eutrophication potential	kg PO_4 eq	2.79E−03	3.1410E−04	1.7751E−04	1.4613E−04
Fresh water aquatic ecotoxicity potential	kg 1,4-DB eq	2.86E−01	1.9096E−02	1.3304E−02	6.2657E−04
Global warming potential	kg CO_2 eq	2.72E+01	3.4891E−02	1.7891E−02	1.6216E−02
Human toxicity potential	kg 1,4-DB eq	2.07E+00	1.2819E−02	9.2143E−03	8.0430E−03
Marine aquatic ecotoxicity potential	kg 1,4-DB eq	1.06E+03	4.0884E+01	2.6287E+01	2.2443E+01
Ozone layer depletion potential	kg CFC-11 eq	1.41E−06	1.2483E−11	3.0068E−12	1.9860E−12
Photochemical oxidation potential	kg C_2H_4 eq	1.39E−03	7.9646E−05	6.5995E−05	6.2964E−05
Terrestrial ecotoxicity potential	kg 1,4-DB eq	1.75E−02	3.4095E−04	2.2852E−04	2.8548E−05

performance) of seven different feedstocks such as soybean oil, Jatropha oil, vegetable seeds oil, castor oil, waste cooking oil, waste extraction oil and algae. They categorized feedstocks for short-term and long-term biodiesel production based on their assessment. Jatropha oil, castor seed oil, waste cooking and extraction oil were grouped under short-term biodiesel production and microalgae was described as a potential feedstock for the long-term biodiesel production. Table 7 presents the lifecycle assessment results of biodiesel produced from commonly used feedstocks.

7.3. Commercial aspects of biodiesel

The potential of sustainable biodiesel business in British Columbia is explored by Boyd et al. (2004). They identified the ability to balance supplies of feedstock, the ability to form sustainable trade relations, the ability to withstand market pressures and the ability to devise economically attractive business plans as factors affecting the British Columbian biodiesel market. To accelerate biodiesel production in the United States, Morrison et al. (2014) examined the commercial aspects of supply and demand for biofuels. Their analysis showed that if the constraints on supply and demand are removed, more than 3.5% of the total energy in the transport sector can come from biofuels in the United States by 2030.

A community-based biodiesel production facility has been successfully developed by Thai researchers in Chumporn region of Southern Thailand (Phalakornkule et al., 2009). Their research results showed that the Chumporn community will benefit from the developed system by directly utilizing the produced fuel in their agricultural engines. The estimated total capital investment for the construction of such biodiesel production unit is estimated at USD 3134.80. In addition to the biodiesel production unit, the researchers suggested the construction of biodiesel wastewater treatment plant at an additional cost of 940 USD to recycle and reuse the biodiesel treated wastewater.

Similar to prior work, Skarlis et al. (2012) investigated the feasibility of small-scale biodiesel production on the island of Crete. Their investigation revealed that at a low capital investment of € 4,000,000/plant, 10,000 tons of biodiesel can be produced annually with the available resources in the Island.

A case study by Iranian researchers has shown that a centralized model would assist Iran in using biodiesel resources effectively. The study highlights the need to address land management issues in certain regions of the country to protect farmlands and increase crop yield. Also, it is reported that microalgae is a promising raw material for biodiesel production in some parts of Iran (Avami, 2012).

Although Jatropha has been identified as a promising feedstock for biodiesel production in India, its large-scale extension is facing several issues, such as inconsistent annual yield (primarily due to vagaries of weather) and unsuitable pricing policy. Pohit et al. (2010) proposed an appropriate pricing policy for large-scale production of biodiesel from Jatropha in India. The authors conducted this study in the Indian state of Chandigarh and suggested the need to set different supply price and demand price in order to obtain an adequate return on investment.

8. Conclusion and future perspective

In this contribution, a comprehensive and critical overview focusing on the synthesis of environmentally benign solid catalysts from a wide range of biological and industrial waste materials is presented. The practical application of these catalysts in biodiesel synthesis is first considered. Then, different approaches and techniques to improve the activity of the derived catalysts are discussed. One must highly appreciate the abundant availability of attractive waste resources around us. While an enormous waste is available to synthesis catalyst for biodiesel production, its reusability and disposal after reuse are highly questionable. Even if metallic elements are added to increase the catalytic activity and recyclability, the concentration of certain organometallic compounds in biodiesel is measured to be higher than the specified limits which necessitate additional purification. After pretreatment, the activity of the derived catalyst is greatly affected which would eventually reduce the yield of the product. Further research is also needed to test the sensitivity of these waste-derived catalysts.

Furthermore, it is suggested through this review that commercial attractiveness and physio-chemical properties of biodiesel could be improved by overcoming the drawbacks in the conventional production route (i.e. transesterification). One of the major problems in biodiesel synthesis via transesterification is the presence of high oxygen content in the final product which leads to lower energy content and limits its uses in applications requiring high energy density such as compression ignition engines. In this perspective, we are actively looking for alternatives, particularly by decarboxylation, to produce diesel like hydrocarbon fuels with enzymes (Köninger et al., 2016; Sorigué et al., 2017; Huijbers et al., 2018; Ma et al., 2020).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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