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RECENT DEVELOPMENT IN PRODUCING BIODIESEL FROM NON-EDIBLE OIL FEEDSTOCKS

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Abstract- *Researchers around the globe are scrambling to develop biofuel feedstocks that would not divert food crops to energy as the world confronts a severe rising fuel price as well as food shortage. It has become discernible that biodiesel is destined to make a substantial contribution to the future energy demands of the domestic and industrial economies as well as the capability of reducing greenhouse gas emissions significantly. There are different feedstocks for biodiesel production, among them, non-edible vegetable oils which are known as the second generation feedstocks can be taken into account as promising substitutions for traditional edible food crops for biodiesel production. Thus it becomes imperative to search for dedicated non-edible feedstocks and their suitability for biodiesel production. This paper presents recent development in producing biodiesel from non-edible feedstocks. Edible and non-edible oil feedstocks are identified and assessed first, then the potential of non-edible biodiesel feedstocks for producing biodiesel are discussed. Furthermore, methods and steps used for oil extraction from non-edible feedstocks and transesterification process of converting oil into biodiesel are presented and discussed.*

Keywords: Non-edible oil, Biodiesel feedstock, Oil extraction technique, Transesterification method

1. INTRODUCTION

Petroleum based fuels are limited reserves concentrated in certain regions of the world. These fossil fuel sources are depleting day by day. Furthermore, the scarcity of conventional fossil fuels and growing emissions of combustion-generated pollutants have raised concern for environmental problems. Increasing costs of fossil fuels have made renewable energy sources more attractive [1, 2]. Biodiesel, an alternative fuel to petrodiesel, is attracting increasing attention worldwide as a blending component and/or as a direct replacement for diesel fuel in vehicle engines. Biodiesel is also considered as one of the promising alternatives for diesel engine especially from non-edible feedstocks [3, 4]. Biodiesel refers to a diesel-equivalent, processed fuel derived from biological sources. Biodiesel is defined as the monoalkyl esters of long chain fatty acids derived from a renewable lipid feedstock, such as vegetable oils, animal fats or waste cooking oil [5, 6].

Although the first generation biodiesel produced from edible oil has gained the attention, the edible oil based biodiesel faced the problem of fuel verses food debate and these factors have negatively affected on biodiesel production from edible oils. Besides, these oils could be more expensive to use as fuel [7]. Therefore, non-edible vegetable oils or the second generation feedstocks have become more attractive for biodiesel production. It is believed that non-edible oils can be one of the solutions to meet the world energy demand and reduce dependency on edible oils [6]. Zhang et al. [8] reported that the price of biodiesel is about US$ 0.5 per litre compared to US$ 0.35 per litre for petroleum diesel. It has been seen that the price of biodiesel mainly depends on the cost of feedstocks which makes up 70-95% of the total biodiesel cost [9-11]. The selection of non-edible vegetable oil is due to the high costs of edible biofuel feedstocks for biodiesel production [10, 11]. Therefore, the use of cost-effective non-edible oils can be a way to improve the economy of biodiesel production and its commercial production at an industrial scale. Due to variations in climate conditions different plant species dominate in different countries and these have been explored for biodiesel production [12]. However, due to the problems associated with food verses fuel, environmental and economic issues related to edible oils, the non-edible oil feedstocks [10] is gaining popularity for biodiesel production.

2. CHOICE OF NON-EDIBLE OIL OVER EDIBLE OIL

The non-edible oils represent potential sources for future energy supply. Non-edible oil resources are gaining worldwide attention because they are easily available in many parts of the world and they can be

grown in degraded lands that are not suitable for raising food crops, eliminate competition for food, reduce deforestation rate, are more efficient and more environmentally favourable, produce useful by-products and are likely to be more economical compared to edible oils [13]. The advantages of non-edible oils as a diesel fuel are liquid nature portability, ready availability, renewability, higher heat content, higher flash point, higher cetane number, lower sulphur and aromatic content as well as biodegradability [14, 15]. Therefore, the non-edible oils can be regarded as ideal feedstocks for biodiesel production over edible oil.

It has been seen that the use of edible oil for biodiesel production influences the global imbalance to the market demand and the food supply by their high prices, the reduction of food sources and the growth of commercial plant capacities. Thus, focus has to be shifted to non-edible resources that are not used as food items and could be grown in the barren lands. In addition, the oils produced from these resources are unsuitable for human consumption due to the presence of toxic components in the oils [12]. Besides low cost and impossibility of their use for human consumption, the other reasons for biodiesel production from non-edible oils include: (i) oil plants which produce large amounts of non-edible oils in nature all over the world; (ii) non-edible oil plants can be easily cultivated in lands unsuitable for human crops at a much lower costs than those of the edible oil crops [10]; and (iii) establishment of these plants reduce CO_2 concentrations in the atmosphere [12, 16]. However, there are some abridgement of non-edible vegetable oils as a diesel fuel such as higher viscosity, lower volatility, the reactivity of unsaturated hydrocarbon chains as well as higher percentage of carbon residue [14]. Furthermore, it has been shown that some of the non-edible oils contain a high amount of free fatty acids (FFAs), which ultimately increases the cost of biodiesel production [12].

3. POTENTIAL NON-EDIBLE BIODIESEL FEEDSTOCKS

In general, biodiesel feedstocks can be categorized into three main groups [16] ; vegetable oils (edible and non-edible), animal fats and waste materials (used oily materials). Additionally, algae oils have been considered as emerging fourth category of growing interest because of their high oil content and rapid biomass production [9, 15, 17, 18]. The various non-edible oil feedstocks are shown in Table 1 [15].

Table 1: Various non-edible oil feedstocks [15]

Type	Feedstocks
1.Non-edible oil seeds/kernels	Jatropha, Karanja, Mahua, Linseed, Cottonseed, Neem, Camelina, Putranjiva, Tobacco, Polanga, Cardoon, Deccan hemp, Castor, Jojoba, Moringa, Poon, Koroch seed, Desert date, Eruca sativa gars, Sea mango, Pilu, Crambe, Syringa, Milkweed, Field pennycress, Stillingia, Radish Ethiopian mustard, Tomato seed, Kusum, Cuphea, Camellia, Paradise, Terminalia, *Michelia champaca*, *Garcinia indica*, *Zanthoxylum bungeanum*.

There are a large number of oil plants that produce non-edible oils. Azam et al. [19] reported that 75 plant species contain more than 30% oil in their seeds and 26 species of them were found most suitable for the use as biodiesel that can be employed for the synthesis of fatty acid methyl ester (FAME). Table 2 [15] presents a summary of the oily plants with oil and the free fatty acid contents. The main sources for biodiesel production from non-edible oils are: Jatropha (*Jatropha curcas*), Karanja or honge (*Pongamia pinnata*), *Aleurites moluccana*, *Pachira glabra*, Nagchampa (*Calophyllum inophyllum*), Rubber tree (*Hevea brasiliensis*), Desert date (*Balanites aegyptiaca*), *Croton megalocarpus*, Rice bran, Sea mango (*Cerbara odollam*), *Terminalia belerica*, Neem (*Azadirachta indica*), Koroch seed (*Pongamia glabra*), Mahua (*Madhuca indica and Madhuca longifolia*), Tobacco seed (*Nicotiana tabacum*), Soapnut (*Sapindus mukorossi*), *Sterculia feotida*, Chinese tallow (*Sapium sebiferum Roxb*.), Silk cotton tree (*Ceiba pentandra*), Jojoba (*Simmondsia chinensis*), Babassu tree, *Euphorbia tirucalli*, and microalgae [5, 20]. The fatty acid compositions of some selected non-edible feedstocks are shown in Table 3 [21, 22].

Table 2: Various non-edible vegetable oil producing plants with oil and free fatty acid (FFA) contents [15]

Non-edible vegetable oil sources	Oil content		Free fatty acid (wt%)
	Seed (wt%)	Kernel (wt%)	
Polanga (*Calophyllum inophyllum*)	65	22	
Jatropha (*Jatropha curcas*)	20-60	40-60	13.5-14.5
Karanja (*Pongamia pinnata*)	25-50	30-50	8.3-20
Mahua (*Madhuca indica*)	35-50	50	20
Linseed (*Linum usitatissimum*)	35-45		
Rubber (*Hevea brasiliensis*)	40-60	40-50	17
Cottonseed (*Gossypium* sp.)	17-25		
Neem (*Azadirachta indica*)	20-30	25-45	
Putranjiva (*Putranjiva roxburghii*)	42		
Tobacco (*Nicotiana tabacum*)	36-41	17	
Cardoon (*Cynara cardunculus*)	25-26		
Deccan hemp (*Hibiscus cannabinus*)	13		
Castor (*Ricinus communis*)	45-50		
Jojoba (*Simmondsia chinensis*)	45-55		
Moringa (*Moringa oleifera*)	33-41	2.9	

Poon (*Sterculia foetida*)	50-55		
Koroch seed (*Pongamia glabra*)	33.6		
Desert date (*Balanites aegyptiaca*)	45-50		
Eruca sativa Gars	35		
Sea mango (*Cerbera odollam*)	54	6.4	
Pilu (*Salvadora oleoides*)	45		
Crambe (*Crambe abyssinica*)	38		
Syringa (*Melia azedarach*)	10-45	2.8	
Milkweed (*Asclepias syriaca*)	20-25	0.019	
Field pennycress (*Thlaspi arvense*)	20-36		
Stillingia (*Sapium sebiferum*)	13-32		
Radish (*Raphanus sativus*)	40-54		
Ethiopian mustard (*Brassica carinata*)	42	2.2-10. 8	
Tomato seed	32-37		
Kusum (*Schleichera triguga*)	10.65		
Michelia champaca	45		
Garcinia indica	45.5		
Zanthoxylum bungeanum	24-28	25	

Table 3: Fatty acid compositions of some selected non-edible feedstocks [21, 22]

Sources	Fatty acid composition (%)
Calophyllum inophyllum	16:0 (17.9), 16:1 (2.5), 18:0 (18.5), 18:1 (42.7), 18:2 (13.7), 18:3 (2.1), 24:0 (2.6)
Argemone mexicana	14:0 (0.8), 16:0 (14.5), 18:0 (3.8), 18:1 (18.5), 18:2 (61.4), 20:0 (1.0)
Jatropha curcas	14:0 (1.4), 16:0 (15.6), 18:0 (9.7), 18:1 (40.8), 18:2 (32.1), 20:0 (0.4)
Madhuca indica	14:0 (1.0), 16:0 (17.8), 18:0 (14.0), 18:1 (46.3), 18:2 (17.9), 20:0 (3.0)
Melia azedarach	14:0(0.1), 16:0 (8.1), 16:1 (1.5), 18:0 (1.2), 18:1 (20.8), 18:2 (67.7)
Pongamia pinnata	16:0 (10.6), 18:0 (6.8), 18:1 (49.4), 18:2 (19.0), 20:0 (4.1), 20:1 (2.4), 22:0(5.3), 24:0 (2.4)
Putranjiva roxburghii	14:0 (0.03), 16:0 (10.23), 16:1 (0.07), 17:0 (0.07), 17:1 (0.02), 18:0 (10.63), 18:1 (48.65), 18:2 (27.50), 18:3 (0.87), 20:0 (1.05), 20:1 (0.30), 22:0 (0.24), 22:1 (0.03), 24:0 (0.31)
Thevetia peruviana	16:0 (15.6), 18:0 (10.5), 18:1 (60.9), 18:2 (5.2), 18:3 (7.4), 20:0 (0.3), 22:0 (0.1)
Schleichera triguga	12:0 (0.31), 14:0 (15.54), 16:0 (10.35), 18:0 (11.11), 18:1 (27.08), 18:2 (6.14), 20:0 (15.79), 20:1 (6.17), 20:2 (0.08), 22:0 (0.01)

4. OIL EXTRACTION

The important step in the production of biodiesel is oil extraction. In this process, the oil contained in the kernels or seeds are extracted. The seeds or kernels are either ground using an industrial grade blender or grated using a small grate. The ground material is passed through a small sieve [22]. The prerequisite for oil extraction is seed preparation.

4.1 Seed Preparation

Preparation of seeds for oil extraction involves removal of outer layers fruits to expose kernels, and then drying the kernel to a desired moisture content [23]. The seeds are separated from fruits, and the fruits that do not dehisce are cracked open manually. The separated seeds or kernels are sieved, cleaned and stored at room temperature. Seeds of some species (e.g., *Syagrus*, *Calophyllum*) are nondehiscent. Hence, they are either fermented to remove the husk, or the entire fruit is dried. Once dried, the fruits are cracked open to collect kernel [22, 24]. The seeds that are procured from various sources are dried at 30-40^{0}C for 3-5 days either in the oven or sun, kernels are separated where necessary, and seeds and kernels are used in oil extraction [20, 22].

4.1.1. Kernel extraction

The first stage of preparation of seeds for oil extraction is removing the outer layers to expose the kernel. The seeds are cracked open in order to obtain kernels for further processing. There are two methods to crack the seeds namely, stompers and mallets. Kernel extraction is a time consuming and labour intensive process. It is possible to crack numerous seeds at a time by using stompers, and seeds are cracked either individually or several at a time by using mallets [23].

4.1.2. Kernel drying

The kernel or seed has to be prepared in such a way that it contains optimum moisture content for high oil yield. It has been observed that kernels with 15% moisture content provided the highest oil yields in both mechanical and solvent extraction methods [23]. The seeds have to be dried before the oil extraction takes place. The drying process is checked very carefully by weighing the trays several times in a day whenever possible and after reaching the desired dryness; the trays are stored in a refrigerated room. Both mulched and unmulched kernels are dried at 50^{0}C to 70^{0}C to determine the effect of mulching as well as temperature on drying rate [23]. Samples are dried for until the weights remain constant to determine absolute moisture content of kernels.

4.2. Extraction Techniques

There are three main methods that have been identified for extraction of oil: (1) mechanical extraction, (2) chemical or solvent extraction, and (3) enzymatic extraction. It has been observed that mechanical pressing and solvent extraction are the most commonly used methods for commercial oil extraction [20].

4.2.1. Mechanical extraction

The technique for oil extraction by mechanical presses is the most conventional one among the other

methods. In this method, either a manual ram press or an engine driven screw press is used. Jahirul et al. [23] used a Mini 40 screw press to extract the oil from beauty leaf seeds (*Calophyllum inophyllum*) as shown in Fig. 1 [23]. It has been observed that the ram press can extract only 60-65% while an engine driven screw press can extract 68-80% of the available oil [20]. Therefore, the design of mechanical extractor is very important in conventional mechanical presses techniques since the oil yield is affected by the type of mechanical extractor is used. Pre-treatment of the seeds by cooking process can increase the oil yield of screw pressing up to 89% after single pass and 91% after dual pass [13, 25, 26]. Different mechanical presses are used for different types of seeds. However, it has to be noted that oil extracted by a mechanical press needs further treatment of filtering process and degumming [20]. Mahanta and Shrivastava [26] reported that the mechanical presses are not efficient for extraction of oils because of the problems associated with non-edible seeds.

Fig. 1: Mechanical oil extraction through a screw press [23]

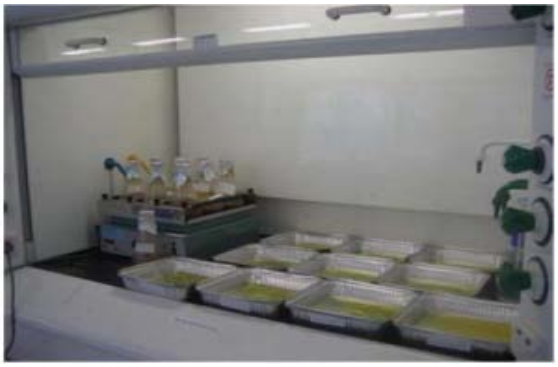

Fig. 2: Chemical oil extraction using n-hexane [23]

4.2.2 Chemical or solvent extraction

Solvent extraction is the process in which constituent is removed from a solid by means of a liquid solvent. It is also called leaching. The chemical extraction using n-hexane method results in the highest oil yield which makes it the most common method. Ashwath [22] and Jahirul et al. [23] also conducted the chemical oil extraction technique using n-hexane to extract the oil from beauty leaf seeds (*Calophyllum inophyllum*) which is shown in Fig. 2 [23]. According to Jahirul et al. [23], although the oil extraction technique using the mechanical screw press was obtained at a low cost but this is ineffective due to relatively lower oil yields. On the contrary, the chemical oil extraction technique was found to be very effective because of high oil yield and for its consistent performance which is shown in Fig. 3 [23]. It has been observed that there are many factors affecting the rate of extraction such as particle size, the type of liquid selected, temperature and agitation of the solvent [20]. Due to the greater interfacial area between the solid and liquid, the small particle size is preferable.

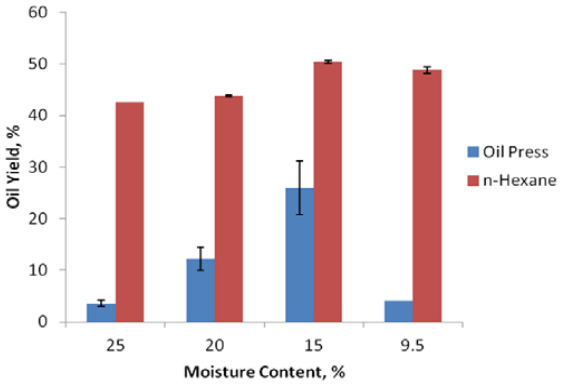

Fig. 3: Oil yield against moisture content for chemically and mechanically processed samples [23]

The liquid has to be chosen in such a way that it would be a good selective solvent and its viscosity would be sufficiently low to circulate freely. It has been found that the solubility of the material increases with increasing temperature. Agitation of the solvent also affects and increases the eddy diffusion and therefore, increases the transfer of material from the surface particles. For large scale production i.e., 50 tonne of biodiesel per day the solvent extraction is economical. Mahanta and Shrivastava [26], and Achten et al. [25] reported that solvent extraction with n-hexane can be used to extract the oil from the seeds of *Jatropha curcas* and *Pongamia pinnata*, and this method could produce about 41% and 95-99% of oil yield, respectively.

4.2.3. Enzymatic extraction

Enzymatic oil extraction method is a promising method for extraction of oil. Appropriate enzymes are used to extract the oil from crushed seeds. The main advantages in this process are: it is environment friendly and does not produce volatile organic compounds. Shah et al. [27] used a combination of ultrasonication and aqueous enzymatic method to extract oil from *Jatropha curcas* seeds. However, the main disadvantage associated with this method is, it takes very long time to complete the process [25].

5. TRANSESTERIFICATION METHOD OF BIODIESEL PRODUCTION

A lot of efforts have been made to develop and improve vegetable oil derivatives in order to approximate the properties and performance of hydrocarbons-based diesel fuel [15]. It has been remarked that high viscosity, low volatility and

polyunsaturated characters are the main barriers that prevent the use of direct vegetable oils in conventional diesel engines [5] . In addition, refinement is necessary in order to turn those vegetable oils into quality fuel. Transesterification is presently one of the most attractive and widely accepted technologies for biodiesel production. It is regarded as the best technique and the most promising solution to the high viscosity problem among other approaches due to its low cost and simplicity [28]. Moreover, this technique has been identified as a widely available technique for industrialized biodiesel production due to its high conversion efficiency and low cost [29]. The process consists of a number of consecutive, reversible reactions [13]. It has many advantages than the other processes e.g., it is achieved under normal conditions and it returns good yield of better quality biodiesel [30].

In transesterification process, alcohol is reacted with vegetable oil in the presence of appropriate catalyst. Generally, ethyl or methyl alcohol is used to produce ethyl/methyl esters. After completing the reaction, two distinct layers of liquids i.e., ethyl/methyl ester and glycerine appeared and are separated as shown in Fig. 4 [31]. The vegetable oil is reacted with methanol already mixed with catalyst and the results produced are crude biodiesel and crude glycerine. The glycerine is refined and disposed of for further use. The crude biodiesel is also refined and alcohol is separated from it which is reused in the cycle [31]. The technology of biodiesel production includes transesterification of oils (triglycerides) with alcohol which gives biodiesel FAME as the main product and glycerol as the by-product [3]. Fig. 5 shows the transesterification reaction of triglycerides [32]. The triglyceride is converted step-wise into diglyceride, monoglyceride, and finally, glycerol, during which one mole of alkyl ester is removed in each step [5]. There are many factors which affect the transesterification processes and they include: mixing intensity, the nature of the feedstock, reaction temperature, reaction time, reaction pressure, alcohol/oil molar ratio, type and concentration of catalyst, free fatty acids, moisture and water content, rate and mode of stirring, effect of using organic co-solvents, specific gravity as well as purity of reactants [28, 31].

5.1. Alkali-Catalyzed Transesterification Methods

The transesterification process is catalyzed by alkaline metal alkoxides, hydroxides, as well as sodium or potassium carbonates [28]. However, it has been observed that the sodium methoxide is the most widely used biodiesel catalyst and over 60% of industrial plants use this catalyst [33]. Alkaline catalysts require short reaction time and relatively low temperature and little or no darkening of colour of the oil [34].

Although Singh and Padhi [34] showed high performance for obtaining vegetable oils with high quality but the oils contain significant amount of FFAs which cannot be converted into biodiesel. The alkali-catalyzed transesterification of vegetable oils goes on faster than the acid-catalyzed reaction.

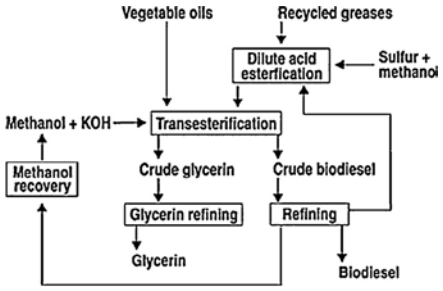

Fig. 4: Basic transesterification protocol [31]

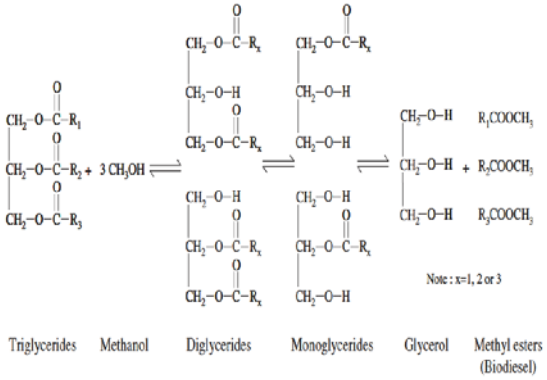

Fig. 5: Transesterification reaction of triglycerides with alcohol [32]

In the alkali-catalytic methanol transesterification method, the catalyst (KOH or NaOH) is dissolved into methanol by vigorous stirring in a small reactor [5, 28, 35]. The oil is then transferred into the biodiesel reactor and the catalyst/alcohol mixture is pumped into the oil. The final mixture is stirred vigorously for 2 h at 340 K in ambient pressure. A successful transesterification reaction produces two liquid phases: ester and crude glycerol. In general, alkali-catalyzed transesterification processes are completed at low temperatures and pressures (333-338 K and 1.4- 4.2 bar) with low catalyst concentrations (0.5-2 wt.%) [36].

5.2. Acid-Catalyzed Transesterification Methods

The main acid-catalyzed transesterification methods for biodiesel production are methanolic sulphuric acid, ferric sulphate, sulfonic acid, methanolic hydrogen chloride, and methanolic boron trifluoride [5, 28]. The catalysts are dissolved into methanol by vigorous stirring in a small reactor whereas the oil is transferred into the biodiesel reactor and then the catalyst/alcohol mixture is pumped into the oil [5]. Currently, the mostly used catalysts in biodiesel production are the organic acids, such as the derivatives of toluene sulfonic acid and, more often, mineral acids such as sulphuric acids, and hydrochloric acid [5, 28, 37]. There are some advantages of acid-catalyzed transesterification process: it directly produces biodiesel from low-cost lipid feedstocks, such as used cooking oil and greases, which commonly have FFAs levels of > 6% and insensitive to the FFAs content in the feedstock [38]. Liquid acid-catalysts are considered to overcome the limitations of liquid base-catalysts because of the liquid base-catalyzed transesterification process poses a lot of problems especially for oils or fats with high FFAs concentrations

[39]. However, the performance of the acid-catalysts is not strongly affected by the presence of FFAs in the feedstock.

Although the transesterification process using acid-catalysts is much slower than that obtained from the alkali-catalysis, which is typically 4000 times, due to the presence of high contents of water and FFAs in the vegetable oil, acid-catalyzed transesterification method can be used [28]. The acid-catalyzed reaction commonly occurs at temperatures above 373 K and requires reaction times of 3-48 h, except when reactions were conducted under high temperature and pressure. These catalysts give very high yields in alkyl esters in the transesterification process [28]. It has been identified that the alcohol/vegetable oil molar ratio is one of the main factors that influence the transesterification process [5]. However, acid-catalyzed reactions require the use of high alcohol to oil molar ratios in order to obtain good product yields in practical situations.

6. CONCLUSION

The non-edible oils have become the leading raw materials for obtaining biodiesel due to increase in world's energy demand, competition of edible oil sources for human use and biofuel production as well as environmental pollution. However, competition of edible oil sources as food vs. fuel makes non-edible oil an ideal feedstock for biodiesel production. The non-food lipid feedstock has the potential for the production of biodiesel that can partially supplement the growing demand for diesel. It has been found that the solvent extraction technique using n-hexane results in the highest oil yield which makes it to be the most common type amongst the three methods. The transesterification method is the most suitable method among the several possible methods for biodiesel production from non-edible oils. Base catalysts are preferable in the case of non-edible oils that contain low FFA content, while acid catalysts are suitable for oils having higher amounts of FFAs. However, priority should be given for the establishment of biofuel production system which must be economic, efficient and can take non-food feedstocks.

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