# Comparative Study of Biofuel Potentials of *Adansonia digitata, Citrullus lanatus* and *Lagenaria siceraria* seedoils

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## ABSTRACT

*Alternative fuel is an important issue globally due to efforts sought to combat emissions which currently pose strong threat to a clean environment. This study was carried out to expose the Biofuel potentials of lagenaria, adansonia and citrullus seeds oil compared with Jatropha curcas oil. The investigation covered physicochemical characterization of their triglycerides and production of mono-alkyl esters. Oils extracted from these seeds were dissolved in ethyl acetate and analyzed for fatty acids composition using GC-MS. The results showed percentage oil yield of > 44% for all the three seeds. Some of the important fuel parameters such as flash point, Diesel Index, sulfur content, acid value, Iodine value and other properties compared favorably with those of Jatropha curcas and showed compliance with ASTM standard specification. Oleic acid is dominant in adansonia oil, while linoleic is most dominant in lagenaria and citrullus seed oils. Other pronounce fatty acids present in all the oils include palmitic, stearic, eicosanoic and some amounts of non-fatty compounds. The fatty acids observed in the seed oils are similar to those of soybean, rape, sunflower and jatropha seeds oil. Thus, the results indicate that, the studied seed oils are suitable for mono-alkyl esters production.*

**Keywords:** Lagenaria siceraria**,** mono-alkyl ester, Diesel Index, Biofuel, non-fatty acids compounds.

## INTRODUCTION

Use of energy is considered the most fundamental requirement for human existence. Among different types of fuels, petroleum constitutes the world's major energy supply. It plays a significant role in industry, agriculture and transportation as well as meeting many other basic human needs[1, 2]. As reported by [3]; the global demand for petroleum is expected to increase to 40% or more by 2025. However, petroleum is finite and nonrenewable energy source, which has been implicated to have serious environmental issues. Thus, a renewable, sustainable, affordable and environmentally friendly alternative to petroleum is urgently required. Liquid biofuels are made from biomass and have qualities that are similar to gasoline, diesel or other petroleum-derived fuels. The two dominant liquid biofuels are bioethanol and biodiesel that together meet about 4% of the global transport fuel demand and so far are produced using only 2—2.5% of the global arable land. Bioethanol can be produced from sugarcane, sugar beets, potatoes, cassava etc. The largest producers of bioethanol are the United States, Brazil and China. Biodiesel is made from vegetable oils, derived from rapeseed, palm seeds, Jatropha as well as from animal fat or waste oils. The largest producers of biodiesel are the European Union, United States, Brazil and Argentina. The most important advantage of biofuels is that they can substantially reduce greenhouse gas (GHG) emissions in the transport sector between 70% and 90% compared to petroleum with only slight changes to vehicle technology and the existing fuel distribution infrastructure [4, 5]. The future of biofuels depend on some factors; one of which is largely the price of biomass from which the feedstock is derived. The technology of production should be geared towards biofuels from cellulosic feedstock which could be sourced from a broader range of non-food biomass such as agriculture and forestry waste which could ease feedstock supply and prices and address certain sustainability issues [6].

Biodiesel is one of the most attractive alternative to conventional petro-diesel which is composed of mono-alkyl esters of long chain fatty acids derived from a renewable lipid feedstock that conforms to the Standard Specification for Biodiesel (ASTM D6751). “Bio” indicates an energy that is biologically sourced and can be renewed; while “diesel” means it can be used only in diesel engines. Biodiesel can be used directly in its pure form known as B100 (100% biodiesel) or in a blend with various proportions of conventional diesel fuels [7, 8]. Biodiesel has a number of advantages compared to conventional diesels which include; it is renewable, non-toxic, and biodegradable since feedstock is originally from plants or animals. Use of biodiesel can reduce air pollution because of low levels of emissions of carbon monoxide (CO), polycyclic aromatic hydrocarbons (PAH), particulate matter (PM), Sulfur oxides ($SO_X$) Nitrated PAH (nPAH), soot and other byproducts [9]. One of the most important disadvantage of biodiesel is its high cost which is one and half times that of petrodiesel. The cost of biodiesel depends on a number of factors; which include: cost of reactant and feedstocks, purification method, storage and so on. Of these, cost of feedstock accounts for 70—90% of the total production cost [10]. It became imperative to use more economical feedstocks such as oils from wild crops and waste cooking oils and fats that can significantly decrease the biodiesel cost [11].

*Lagenaria* and *Citrullus* are wild weeds and *Adansonia*, all are wild plants in some areas and domesticated crops in other places in Northern Nigeria. These plants contained seeds embedded within their fruits that have no established application or appear on the food menu. In most cases, seeds from this resource are thrown away or dumped as refuse. The dumped seeds decay and decompose as garbage by the road site. Their decomposition produces odoriferous gases that are unpleasant to the surroundings. Consequently, to find application to these seeds, they are being assessed for use as biodiesel production source. This is to convert waste to use and generate additional route to energy supply. The desire for reduction of over dependence on fossil fuel and support for agricultural industries to ensure sustainable economic development through the exploitation of abundant wild crop resources as well as sustenance of our environment are the factors responsible for initiating this work [12]. The objectives of this work is to undertake a comparative study of fuel potentials of the aforementioned seed oils by analysing the chemical composition of their oils and derived alkyl esters. These seeds are from a cheap source thereby keeping the production cost relatively low.

## MATERIALS AND METHODS

### *Sample seeds sources*

Plant seeds of *Adansonia*, *Citrullus* and *Lagenaria* were purchased dry from Damaturu, Yobe State.

### *Sample preparation and oil extraction*

The seeds collected were separated manually, washed with water for any adhering flesh and sun dried for a week at ambient temperature. The seeds were dried in an oven at 50$^{o}$C for 72 hours until constant weights were obtained and crushed to powder using mortar and pestle. Crude lipid was extracted separately from each seed using Soxhlet extractor on a water bath with n-Hexane as extracting solvent.

### *Transesterification of oils*

The oils of adansonia, lagenaria and citrullus contained 1.10% FFA, 2.6% FFA and 9.14%FFA respectively, were used as raw materials in this study. Oils of adansonia and lagenaria were transesterified to mono-alkyl esters using direct alkali transesterification reaction with methanol and sodium hydroxide as catalyst. While oil from citrullus (due to high FFA's) was subjected to acid esterification method using paratoluene sulphonic acid and methanol as first step to reduce the high free fatty acids level prior to alkali transesterification; hence conversion to mono-alkyl esters.

### *Analysis of Adansonia, Lagenaria and Citrullus Seed Oils*

The physico-chemical analysis of crude seed oils and their derived methyl esters were carried out according to [13] and [14] standard methods. ASTM [15], and EN 14214 (2003) Standard methods were employed for the evaluation of methyl ester fuel properties.

### *Analysis of the acquired mono-alkyl esters using GC-MS*

The method reported by [16] was adapted to quantify the fatty acids methyl esters produced using a 6890N model (AGILENT) GC-MS machine. One microliter (1µl) of the methyl ester sample was introduced into the gas chromatograph at injection temperature of 250$^{O}$C. The column temperature was programmed from 60$^{O}$C to 260$^{O}$C at a linear flow rate of 5$^{O}$C/min, hold 5 min. at 260$^{O}$C. The chromatographs obtained were scanned, then the methyl ester components were identified based on software matching with mass spectra.

## RESULTS AND DISCUSSIONS

The crude lipid yields of the studied samples and the free fatty acids (FFA) content of these oils were measured by titration using 0.1M KOH solution and phenolphthalein indicator. Jatropha curcas are as presented in Table 1. Oil content of plant seeds is an important factor for assessing the economic viability of a feedstock for biodiesel production. The results showed oil content values of 45.54±0.35%, 48.50±0.92%, 45.50±0.45% and 46.00±0.84% for adansonia, lagenaria, citrullus and jatropha respectively. The results revealed that adansonia and citrullus seeds contain oil quantity very close to Jatropha while lagenaria seeds have higher oil yield than jatropha seeds. All the results fall within the range of the percentage oil content (30—55%) reported elsewhere [27]. Oil content from seeds of these plants is highly appreciable and compare well considering the oil content of Jatropha and other domesticated plants seeds, hence the seeds contain appreciable amounts of oil enough to be extracted for commercial scale production of biodiesel.

**Table 1: Percentage Yields and Properties of Oils Extracted from Seeds of *Adansonia*, *Lagenaria* and *Citrullus* with corresponding values of *Jatropha curcas* seed oil**

| Parameter | *Adansonia* | *Lagenaria* | *Citrullus* | *Jatropha** |
|---|---|---|---|---|
| Crude Lipid Yield (%) | 45.54±0.35 | 48.50±0.92 | 45.50±0.45 | 46±0.84 |
| Density ($g/cm^3$) @ $20^oC$ | 0.90±0.00 | 0.91±0.00 | 0.92±0.01 | 0.914±0.02 |
| Viscosity @$40^oC$ (cSt) | 32.45±0.60 | 32.10±0.15 | 26.41±1.15 | 36.0±4.24 |
| SV(mgKOH) | 192.84±1.84 | 197.23±2.05 | 201.61±1.81 | 182.8±11.43 |
| FFA (%) | 1.10±0.03 | 2.6±0.0.12 | 9.14±0.02 | 3.40±0.46 |
| IV ($gI_2$/100g) | 79.28±0.84 | 61.28±0.84 | 101.52±6.86 | 101±6.0 |

**SV=Saponification Value, IV= Iodine Value, FFA= Free Fatty Acid content**

*****source: [17, 18, 19, 20] and [21].

The free fatty acid content of an oil is an important parametric factor that affects the optimal conversion of the vegetable oils to alkyl esters and determines the selectivity of a suitable catalyst for the transesterification reaction [28]. Free fatty acids values obtained in *Adansonia*, *Lagenaria* and *Citrullus* oils are as depicted in Table 1. The FFA's values for adansonia and lagenaria are within the category of oils that could yield alkyl esters on single step alkaline transesterification reaction [16], while citrullus oil contains high FFA's value, as such can't be directly transesterified, hence the need for pre-treatment to reduce FFA's to less than 2%. Thus, esterification of the oil must be carried out to reduce the acid level prior to alkaline transesterification and this could give an optimal yield. Except for citrullus oil, the two remaining oil's FFA's compare favorably with or even little less than Jatropha oil's value. The FFA's recorded in this study are lower than palm oil (5.3%) and Jatropha oil (6.85%) as reported elsewhere [29].

Iodine value is an indicator of the chemical stability of a fuel. It's a measure of the average amount of unsaturation of fats and oils. Oils exhibit high iodine value due to high of unsaturation level. Low iodine value implies high stability of the oils. *Adansonia d., Lagenaria s.,* and *Citrullus l.* each had iodine values of 79.28±0.84, 61.28±0.84 and 101.52±6.86 respectively (Table 1). The values obtained for adansonia and lagenaria are lower than that of Jatropha used as control in this study. However, citrullus recorded a value which is very close to that of *Jatropha* (101±6.0). All the results obtained here are lower than what was reported (156.00) for melon seed oil [29].

**Table 2: Fuel Properties of mono-alkyl esters derived from *Adansonia, Lagenaria and Citrullus* with corresponding values of *Jatropha curcas* methyl ester**

| Fuel Parameter | *Adansonia* | *Lagenaria* | *Citrullus* | *Jatropha** |
|---|---|---|---|---|
| Viscosity @40$^o$C (cSt) | 6.0±0.05 | 6.0±0.12 | 5.4±0.35 | 5.65±0.47 |
| Flash point ($^o$C) | 150±3.06 | 154±2.45 | 155±5.48 | 176±11.00 |
| Free glycerol (wt. %) | 0.18±0.010 | 0.23±0.03 | 0.22±0.03 | 0.03±0.00 |
| Sulfur content (wt. %) | 0.02±0.00 | 0.02±0.00 | 0.03±0.00 | 0.004 |
| Diesel index | 43.10±0.00 | 33.65±5.60 | 37.60±2.64 | *****NF |
| Acid value (mgKOHg$^{-1}$) | 0.34±0.00 | 0.52±0.06 | 1.24±0.16 | 0.6±0.22 |

*source: [22, 23] and [24]. *****NF=Not Found

### *Composition and Characteristics of Mono-Alkyl Esters Produced*

Kinematic viscosity is a property that represents the flow characteristics of fuel. One reason why biodiesel is used as an alternative fuel instead of pure vegetable oils is due to its reduced viscosity which enhances fuel flow property. Viscosity is a key fuel property which persuades the atomization of a fuel upon injection into the engine ignition chamber and ultimately, the formation of engine deposits [30]. Kinematic viscosity increases with fatty acid chain length and with increasing degree of saturation of either the fatty acid or alcohol moiety in the fatty ester [31]. Viscosity of transesterified oils in the present study are lower than that of their parent oils. Mono-alkyl esters of adansonia, lagenaria and citrullus had kinematic viscosities as presented in Table 2. The values obtained fall within the ASTM standard specification and that of Jatropha (control) as shown in the study.

Flash point (FP) is the temperature at which a fuel ignite when exposed to a flame, FP is used to assess the flammability hazard of fuels while transporting or storage. Flash points of mono-alkyl esters under investigation agree with the ASTM specification for biodiesel (Table 2). The results suggest that, there is less tendency of the produced alkyl esters to inflame accidentally and could be handled safely. FP values obtained are slightly less than that of Jatropha curcas but are appreciably high enough to confirm to the prescribed limits in American and European biodiesel standards. Sulfur level in the analyzed mono-alkyl esters is slightly above the ASTM specification limits. Presence of sulfur in biodiesel could be from the vegetable oil's phospholipids which are available in all vegetable oils. This implies that, the alkyl esters produced may emit sulfur oxide gases when burned.

Acids value indicates the level of free fatty acids or processing acids present in an alkyl ester and is a measure of the quality of ester. The acid values of biodiesels studied are shown in Table 2, with citrullus methyl esters having the highest value followed by Jatropha curcas. A high acid value could have a strong solvency effect on rubber seals and hoses in the engine which could lead to engine failure. It may cause deposits that can clog the fuel filter. High acid value causes corrosion of engine parts. The results suggests that, citrullus methyl ester may cause wear in the storage tanks.

Diesel index (DI) indicates the ignition quality of a fuel. DI of a fuel is related to the ignition delay time, i.e. the time required for injection of the fuel into the cylinder and onset of ignition. The shorter the ignition delay time, the higher the index value and vice versa [32]. Just like Cetane number, diesel index decreases with decreasing chain length and increasing branching. The higher the value, the better would be the ignition quality of diesel fuel. It was reported elsewhere [33] that, standard ASTM D 675 for biodiesel prescribes a minimum diesel index of 45.0—55.0.It is determined by calculation from the specific gravity and aniline point of sample. DI values for the mono-alkyl ester samples under study are in the order of 33.65±5.60, 37.60±2.64 and 43.10±0.00 for lagenaria, citrullus and adansonia respectively (Table 2). Lower DI values suggests a possibility of smoky exhaust. This parameter could be enhanced by addition of cetane improver which can enhance the ignition quality of the alkyl esters [16].

The profile of seed oils presented in Figure 1, indicates fatty acid composition of four seeds oil (*Adansonia, lagenaria, Citrullus* and *Jatropha*). The analyses showed that, major fatty acid compounds in the oils are; Palmitic acid ($C_{16}$:$_0$), Stearic acid ($C_{18}$:$_0$), Oleic acid ($C_{18}$:$_1$), Linoleic acid ($C_{18}$:$_2$), and Eicosanoic acid ($C_{20}$:$_0$); with oleic and linoleic acids as most dominant in all the oils. Of the five major fatty acids obtained, oleic acid has the highest value followed by linoleic acid. The profile of *Adansonia* oil indicates presence of palmitic, stearic, oleic and linoleic acids (Figure 1). *Adansonia* seed oil has high unsaturation level compared to its saturated fatty acids content.

The unsaturation level of *Adansonia* oil is very close to *Jatropha* oil as shown in Figure 1. Presence of oleic and linoleic acids can influence the structure of fatty acid ester; with double bonds in the molecules producing a high level of reactivity with oxygen that can affect the oxidative stability of its derived biodiesel. This effect manifests on prolong storage of mono-alkyl esters and is responsible for autoxidation. Since exhaust emissions increase with increasing unsaturation level, *Adansonia* biodiesel will have less and better emissions than sunflower, soybean and rapeseed oils as they contained higher unsaturation levels; even though the latter would have better cold flow property performance [31, 33].

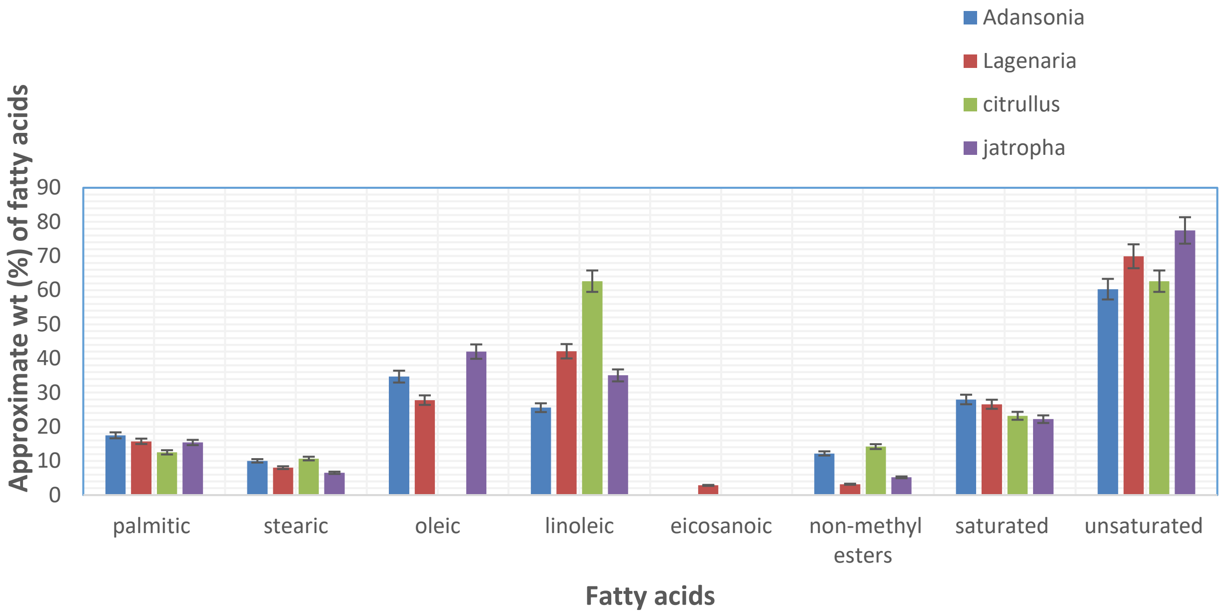


**Figure 1: Fatty acid composition of the studied vegetable oils with corresponding values of ****Jatropha curcas***

*Jatropha data source, [18, 19, 20, 23, 25] and [26]. Wt(%).- **percentage weight.**

Only three of the four major long chain fatty acids are observed in *Citrullus* seed oil (Figure 1). These include, palmitic, stearic and linoleic acids. The unsaturation in *Citrullus lanatus* seed oil was as a result of the presence linoleic acid only which is polyunsaturated fatty acid observed in the oil. This possibly is reflected in the oil of Citrullus showing low values of pour point (18$^{o}$F) and viscosity (26.41cSt) compared to *Adansonia*, *Lagenaria* and *Jatropha*. The oil (citrullus) composition indicated a low saturation level, which is still higher compared to soybean, sunflower, and rape seed oils but less than *Adansonia* and palm oils as reported elsewhere [34]. Despite absence of oleic acid in the oil, its unsaturation level is very high almost close to that of *Jatropha* seed oil. However, absence of oleic acid in citrullus oil would imply that, biodiesel derived from the oil would be more stable and would experience low autoxidation due to less number of double bonds compared to sunflower,

soybean, safflower and Jatropha oils; all of which contain both oleic and linoleic acids [35]. Considering what was reported for Linoleic acid levels in soybean, rapeseed and sunflower oils (54.10%, 13.10%, and 71.40%) respectively, the result for *Citrullus lanatus* (Figure 1) is within what is required for biodiesel feedstock.

The profile of fatty acids in *lagenaria* oil shown in Figure 1, indicates linoleic as the dominant fatty acid, followed by oleic acid. Each of these compounds has a certain amount of unsaturation which may make them susceptible to peroxidation. This implies that, the alkyl esters produced from these feedstocks could deteriorate on prolong storage due to the attack of labile oxygen. Other fatty acids observed in lagenaria oil profile include palmitic, stearic and eicosanoic acids. However, the instability anticipated due to high levels of unsaturation could be corrected by the presence of the mentioned saturated compounds found in the oil (palmitic, stearic and eicosanoic acids). These compounds are saturated and less susceptible to peroxidation. Linoleic acid was reported [16] as the predominant fatty acid in sunflower, rape and soybean seeds oil. Thus, composition of lagenaria seed oil in this study compares favourably with those in the reported literature. *Jatropha* is the major source of oil currently recommended for biodiesel industry. *Jatroph*a seed oil contains palmitic, stearic, oleic and linoleic acids as reported by [17]. The results of this study showed (Figure 1), *Jatropha* seed oil contains four major long chain fatty acids. The seed oil contains more than 76% unsaturation level with only 22.20% saturation, which is reflected in the oil having low pour point (28$^{o}$F) and viscosity value of 31.10cSt. Fatty acid composition of *Jatropha curcas* oil is dominated by linoleic and oleic acids as indicated in this work and reported literature. The maturity stage of the fruits at the period of collection tends to influence the fatty acid composition of the oil [36]. It is important to state that, oil quality of different seeds is dependent on the interaction of environment where they are sourced and genetics [37]. Figure 1 shows the fatty acids profile for Jatropha curcas oil as reported elsewhere, [18, 20, 25] and [36]. All the seeds oil investigated contained some amount of non-fatty acids compounds in small proportion. Each of the oils exhibited high proportion of unsaturation than saturation levels which is closely comparable to the Jatropha seed oil reported (Figure 1) as control measure.

## CONCLUSION

The oils extracted from seeds of *Adansonia d.*, *Lagenaria s*. and *Citrullus l.* could be a viable source of triglycerides for the manufacture of mono-alkyl esters by transesterification. The results showed that, plant species investigated contained appreciable oil and the analyses of their derived mono-alkyl esters indicates that; they possessed physicochemical characteristics acceptable within the ASTM standard specification for biodiesel. The fatty acids profile obtained also compared favourably well with what is obtainable in sunflower, soybean, rape and safflower alkyl esters as internationally accepted oil seeds. Thus, the studied seeds could make a good source of feedstock for alkyl ester production.

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