

Composition and Nutritional Properties of Seeds and Oil From *Terminalia catappa* L.

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Abstract: The seeds of *Terminalia catappa* were analyzed to establish their chemical compositions and nutritional properties in order to investigate the possibility of using them for human and/or animal consumption. Proximate analyses showed that the seed contained 4.13% moisture, 23.78% crude protein, 4.27% ash, 4.94% crude fiber, 51.80% fat, 16.02% carbohydrate and 548.78 Kcal Calorific value. The seeds were found to be good sources of minerals. Potassium (9280 ± 0.14 mg/100g) was the highest, followed in descending order by Calcium (827.20 ± 2.18 mg/100g), Magnesium (798.6 ± 0.32 mg/100g) and Sodium (27.89 ± 0.42 mg/100g). The physical properties of the oil extracts showed the state to be liquid at room temperature. The oil was found to contain high levels of unsaturated fatty acids, especially oleic (up to 31.48%) and linoleic (up to 28.93%). *Terminalia catappa* oil can be classified in the oleic-linoleic acid group. The dominant saturated acids were palmitic (up to 35.96%) and stearic (up to 4.13%). The oil extracts exhibited good physicochemical properties and could be useful as edible oils and for industrial applications.

Key words: *Terminalia catappa*, oil yield, proximate composition, essential fatty acid, DSC and activation energy

INTRODUCTION

Terminalia catappa L. Believed to have originated in Malaysia, this tree is generally confined to mesic and wet coastal habitats and is distributed throughout the Old World tropics and tropical America (Morton, 1985). Reaching heights of 15 to 25 m, *T. catappa* shows strong salt-, drought- and wind-tolerance and produces fruit (5-10 cm long) with a thin flesh surrounding a large fibrous nut. While the fleshy fruit is the target of larval infestation, *T. catappa* leaf extracts have also been shown to preferentially attract female oriental fruit flies (Chen and Dong, 2000). Clarke *et al.*, (2001) found that *T. catappa* along with *Psidium guajava* L. constituted the major hosts for *B. dorsalis* in a survey of Thailand and Malaysia. In addition, *T. catappa* reared a particularly high number of larvae in proportion to the weight and number of fruit sampled, leading to the suggestion that it is a "primary native host" in the surveyed areas (Clarke *et al.*, 2001). Similar disproportionately high numbers of larvae from *T. catappa* have been documented in Hawaii (Maehler, 1949), and these observations drew our

attention to this, host. Stands of tropical almond appear to be somewhat isolated from other known hosts of oriental fruit fly, but the fruit infestation levels suggest that this tree may contain a powerful attractant for female oriental fruit flies.

MATERIALS AND METHODS

This study was led to the laboratory of engineering and biomolecule of the ENSAIA-INPL, Vandoeuvre – lès-Nancy (France) for the period of Jan. 5, 2009 to Feb. 27, 2009.

The fruits containing seeds of *Terminalia catappa* L. were obtained with the feet trees of *Terminalia catappa* L. which are in the centre town of Brazzaville (Congo). Then, they were further dried in our Laboratory at about 40°C and then crushed in Moulinex coffee blender (type Prep' line 850). Powdered *Terminalia catappa* L. seeds were kept at 5°C in polyethylene bag before analysis.

Proximate analysis of *Terminalia catappa* L. seed Moisture, crude protein (micro-Kjeldahl), crude fiber and oil (Soxhlet) contents were determined using the methods

described by Pearson (1976), whereas the ash content was determined using the method of Pomeranz *et al.*, (1994), and total carbohydrate was determined by difference. The sample calorific value was estimated (in Kcal) by multiplying the percentage crude protein, crude lipid and carbohydrate by the recommended factor (2.44, 8.37 and 3.57 respectively) used in vegetable analysis (Asibey-Berko and Tayie, 1999). All determinations were done in triplicate.

Oil extraction: Dried *Terminalia catappa* L seeds were ground in a Moulinex Model SeB PREP'LINE 850 (Moulinex coffee). For solvent extraction (soxhlet method), 50g of ground seeds were placed into a cellulose paper cone and extracted using light petroleum ether (b.p 40–60 °C) in a 5-l Soxhlet extractor for 8 h (Pena *et al.*, 1992). The oil was then recovered by evaporating of the solvent using rotary evaporator Model N-1 (Eyela, Tokyo Rikakikal Co., Ltd., Japan) and residual solvent was removed by drying in an oven at 60 °C for 1 h and flushing with 99.9% nitrogen. For methanol/chloroform extraction (Bligh and Dyer, 1959), 100g of the ground seeds were homogenised with a chloroform mixture methanol (1:1) and water. Two phases was obtained, aqueous layer (methanol-water) and organic layer (chloroform). Oil was recovered by evaporating of the solvent (chloroform) using rotary evaporator Model N-1 (Eyela, Tokyo Rikakikal Co., Ltd., Japan) and residual solvent was removed by drying in an oven at 60 °C for 1 h and flushing with 99.9% nitrogen. All experiments were done in triplicates and the mean and standard deviations were calculated.

Physical and chemical analysis of crude oil:

Thermal behaviour: The thermal property of the oil samples was investigated by differential scanning calorimetry using a Perkin–Elmer Diamond DSC (Norwalk, USA). The instrument was calibrated using indium and zinc. The purge gas used was 99.99% nitrogen with a flow rate of 100 ml/min and a pressure of 20 psi. Sample weights ranged from 5–7 mg and were subjected to the following temperature program: Frozen oil sample was heated at 50 °C in an oven until completely melted. Oil sample was placed in an aluminium volatile pan and was cooled to -50 °C and held for 2 min, it was then heated from -50 to 50 °C at the rate of 5 °C.min⁻¹ (normal rate) (Che Man *et al.*, 1995), and held -50 °C isothermally for 2 min and cooled from -50 to 50 °C at the rate of 5 °C per minute. The heating and cooling thermograms for the normal and the fast (hyper DSC) scan rates were recorded and the onset, peak, and offset temperatures were tabulated. These values provide information on the temperature at which the melting process starts, the temperature at which most of the TAG have melted, and the complete melting temperature of the oil, respectively.

Viscosity measurements: A rheometer as described by Nzikou *et al.* (2007) was used to measure the different oil viscosities. By this procedure, a concentric cylinder system is submerged in the oil and the force necessary to overcome the resistance of the viscosity to the rotation is measured. The viscosity value, in mPa.s, is automatically calculated on the basis of the speed and the geometry of the probe. Temperature (20 °C) was controlled with a water bath connected to the rheometer. The experiment was carried out by putting 3 ml of sample in a concentric cylinder system using 100 s⁻¹ as shear rate.

Chemical analysis: Determinations for peroxide, iodine, and saponification values, unsaponifiable matter and free fatty acid contents were carried out using Pena *et al.* (1992) standard analytical methods. The fatty acid composition was determined by conversion of oil to fatty acid methyl esters prepared by adding 950 µl of n-hexane 50 mg of oil followed by 50 µl of sodium methoxide using the method of Cocks *et al.* (1966). The mixtures were vortex for 5 s and allowed to settle for 5 min. The top layer (1 µl) was injected into a gas chromatograph (Model GC-14A, Shimadzu Corporation, Kyoto, Japan) equipped with a flame-ionisation detector and a polar capillary column (BPX70 0.25), 0.32 mm internal diameter, 60 m length and 0.25 µm film thickness (SGE Incorporated, USA) to obtain individual peaks of fatty acid methyl esters. The detector temperature was 240 °C and column temperature was 110 °C held for one minute and increased at the rate of 8 °C/min to 220 °C and held for one minute. The run time was 32 min. The fatty acid methyl esters peaks were identified by comparing their retention time with those of standards. Percent relative fatty acid was calculated based on the peak area of a fatty acid species to the total peak area of all the fatty acids in the oil sample. The minerals were determined by atomic absorption spectrophotometry. One gram samples, in triplicate, were dry ashed in a muffle furnace at 550 °C for 8 h until a white residue of constant weight was obtained. The minerals were extracted from ash by adding 20.0 ml of 2.5% HCl, heated in a steam bath to reduce the volume to about 7.0 ml, and this was transferred quantitatively to a 50 ml volumetric flask. It was diluted to volume (50 ml) with deionised water, stored in clean polyethylene bottles and mineral contents determined using an atomic absorption spectrophotometer (Perkin-Elmer, Model 2380, USA). These bottles and flasks were rinsed in dilute hydrochloric acid (0.10 M HCl) to arrest microbial action which may affect the concentrations of the anions and cations in the samples. The instrument was calibrated with standard solutions.

Statistical analysis: Values represented are the means and standard deviations for three replicates. Statistical analysis was carried out by Excel Version 8.0 software. Significance was defined at P < 0.05.

RESULTS AND DISCUSSION

Proximate analysis of *Terminalia catappa* L. seed oil:

Results obtained showed that the seeds contained 4.13% moisture, 51.80% crude oil, 23.78% crude proteins, 16.02% carbohydrate (by difference), 4.94% crude fibre, 4.27% ash and 548.78 Kcal calorific value (Table 1). The high percentage of oil makes this seed a distinct potential for the oil industry. According to Omeje *et al.* (2008) and Guillermo Arrázola *et al.* (2008). Variation in oil yield may be due to the differences in variety of plant, cultivation climate, ripening stage, the harvesting time of the seeds and the extraction method used.

Minerals: The *Terminalia catappa* L seeds contained significant amount of important minerals (Table 2). The Potassium concentration (9280.0±0.14 mg/100g dry mater) was the highest, followed in descending order by Calcium (827.20±2.18 mg/100g dry mater), Magnesium (798.6±0.32 mg/100g dry mater) and Sodium (27.89±0.42 mg/100g dry mater). Potassium is an essential nutrient and has an important role is the synthesis of amino acids and proteins (Malik, 1982). Calcium and Magnesium plays a significant role in photosynthesis, carbohydrate metabolism, nucleic acids and binding agents of cell walls (Russel, 1973). Calcium assists in tech development (Brody, 1994). Magnesium is essential mineral for enzyme activity, like calcium and chloride; magnesium also plays a role in regulating the acid-alkaline balance in the body. High magnesium levels in drinking water have been linked to resistance to heart disease (Fallon, 2001).

Oil extraction: Characteristics of the oil were compared with *Terminalia Catappa* L. varieties described by Dos Santos *et al.* (2008). The extracted oils were liquid at room temperature. The oil content of *Terminalia Catappa* L “Congo-Brazzaville” seeds for the two methods utilised and the level at which the differences are significant are shown in Table 3. The oil extraction with the Soxhlet method had the highest yield, due to the increased ability of the solvent to overcome forces that bind lipids within the sample matrix (Lumley *et al.*, 1991). The Blye and Dyer method, showed the low yield due to losses during the separation of the two phases, aqueous layer (methanol-water) and organic layer (chloroform). The results of the above authors agree with those of the present work.

Physical and chemical properties of oil:

Physical properties:

Differential Scanning Calorimetry (DSC): DSC is suitable to determine these physical properties. The results of thermal analysis of oils are presented in Table 4. The obtained peaks were asymmetries and may

Table 1: Proximate analysis of *Terminalia catappa* oil seed

Characteristic	Obtained values ^a (M±S.D.)	Reported values ^b	
		1	2
Moisture content (%)	4.13±0.24	1.54	4.5
Crude protein ^c (%)	23.78±0.15	26.30	24
Ether extract (%)	51.80±0.21	56.71	54
Crude fiber (%)	4.94±0.32	4.40	12
Ash content (%)	4.27±0.74	4.55	4.0
Total carbohydrate ^d (%)	16.02	10.9	13.5
Calorific value (Kcal/100g)	548.78	577.75	588.74

a M ± S.D. mean ± standard deviation.

^b (1) Omeje *et al.* (2008). (2) Guillermo Arrázola *et al.* (2008)

^c Crude protein = N (%) x 6.25

^d Carbohydrate obtained by difference

Table 2: Mineral elemental Composition of *Terminalia catappa* seeds

Mineral Elements	Composition (mg/100g) of Seed
Calcium, Ca	827.20±2.18
Magnesium, Mg	798.6±0.32
Potassium, K	9280.0±0.14
Sodium, Na	27.89±0.42

Values are mean ± S.D of triplicate determinations

Table 3: Physical and chemical properties of *Terminalia catappa* seed oil extracted using solvent process

Properties	Obtained values		Reported values ^a
	Blye & Dyer	Soxhlet	
Oil ^b (%)	47.30 ± 1.25 ^B	56.30 ± 2.35 ^A	49
PV	0.43 ± 0.24 ^A	0.51 ± 0.35 ^A	0.5
FFA (as % oleic acid)	3.02 ± 0.22 ^A	2.42 ± 0.27 ^B	ND
IV (w ijs)	80.89 ± 0.43 ^A	82.43 ± 1.10 ^A	83.92
Saponification value	196 ± 1.32 ^A	207 ± 0.13 ^A	ND
Unsaponifiable matter	0.47 ± 0.02 ^A	0.50 ± 0.07 ^B	ND
Content (%)			
Viscosity (mPa.s) at 38°C	43 ± 0.21 ^B	32.92 ± 0.17 ^B	39.8
E _a (KJ. mol ⁻¹)	7.34	10.23	ND

ND: not determined.

Means for the determined values in the same row followed by the same superscript letter are not significantly different (P < 0.05).

^a Dos Santos *et al.* (2008).

^b Oil = weight of extracted oil x 100/weight of seed.

Abbreviations: PV: Peroxide Value, FFA: Free Fatty Acid, IV: Iodine Value

Table 4: Melting behaviour of *Terminalia catappa* L. seed oil using different scan rates. Experimental conditions: temperature program set at -50 °C for 10 min, rising to 50 °C at rate of 5 °C.min⁻¹

Thermogram	5 °C.min ⁻¹	
	Blye and Dyer	Soxhlet
Peak 1 [°C]	-18.91	-19.75
?H _f [J.g ⁻¹]	+1.57	+1.47
Peak 2 [°C]	+3.56	+4.56
?H _f [J.g ⁻¹]	+21.02	+8.64

indicate the presence of two components in oil extracted from the two methods. The first peaks at low melting points appear at - 18.91 °C (H_f = +1.57 J.g⁻¹) for Blye and Dyer method and - 19.75 °C (H_f = +1.47 J.g⁻¹) for Soxhlet method. These peaks correspond to triglycerides formed by poly unsaturated acids (PUFA). The second melting points are at +3.56 °C (H_f = +21.02 J.g⁻¹) for Blye and Dyer method and +4.56 °C (H_f = +8.64 J.g⁻¹) for Soxhlet method. This is a characteristic of saturated acids (SFA).

Viscosity: Viscosity is a measure of resistance of a fluid to deform under shear stress. It is commonly perceived as thickness, or resistance to pouring. Viscosity describes a fluid's internal resistance to flow and may be thought of as a measure of fluid friction. In optics to know the rheological proprieties of these oils, we studied the influence of temperature on viscosity. Activation energies of the various classes of fatty acids contained in these oils were given Table 3. When the temperature increases, viscosity decreases exponentially (Fig. 1) some is the extraction method (Arslan *et al.*, 2005; Nzikou *et al.*, 2007). Viscosity varies between 58.51 and 31.50 mPa.s when temperature decreases of 50 to 5 °C by Soxhlet method. By Blye and Dyer method, the viscosity of oil decreases of 64.50 to 40.50 mPa.s (Table 5). The viscosity of the oil obtained by Blye and Dyer method was highest, possibly because of the water that was absorbed by the gums (phospholipids) during extraction. This calculator calculates the effect of temperature on reaction rates using the Arrhenius equation.

$$\eta = A \cdot \exp(-E_a / R \cdot T)$$

Where η is the viscosity, A is constant, E_a is the activation energy (in KJ mol⁻¹), R is the universal gas constant and T is the temperature (in degrees Kelvin). R has the value of 8.314×10^{-3} KJ mol⁻¹ K⁻¹. We should use this calculator to investigate the influence of temperature on viscosity. Linear regression analysis was applied to the logarithmic form of Arrhenius equation in order to determine the parameters of the relation (Fig. 2, Table 6). $\ln \eta$ against $1/T$, $-E_a/RT$ is the slope from which E_a was evaluated. Activation energies of oils are given in Table 3. The highest value of activation energy is obtained by Blye and Dyer method (7.34 KJ. mol⁻¹) and 10.23 KJ. mol⁻¹ by Soxhlet method. The higher the activation energy, the more stable the fatty acid is.

Chemical properties: The chemical properties of oil are amongst the most important properties that determines the present condition of the oil. Free fatty acid and peroxide values are valuable measures of oil quality. The iodine value is the measure of the degree of unsaturation of the oil. The free fatty acid and the unsaponifiable matter

Table 5: Oil viscosity at various temperature in degree celsius

T (°C)	η (mPa.s)	
	Blye and Dyer	Soxhlet
5	64.50	58.51
10	57.70	51.00
15	53.30	45.60
20	50.00	41.60
25	47.40	38.10
30	45.40	35.70
35	43.60	33.90
40	42.80	32.70
45	41.60	32.00
50	40.50	31.50

Table 6: Energie plot derived from the Arrhenius equation

1/T (K ⁻¹)	Ln η (mPa.s)	
	Blye and Dyer	Soxhlet
0.00359712	4.16666522	4.06919768
0.00353357	4.05525717	3.93182563
0.00347222	3.97593633	3.81990772
0.00341297	3.91202301	3.72810017
0.00335570	3.85862223	3.64021428
0.00330033	3.81551211	3.57515069
0.00324675	3.77505715	3.52341501
0.00319489	3.75653810	3.48737508
0.00314465	3.72810017	3.46573590
0.00309598	3.70130197	3.44998755

Table 7: Relative percent composition of fatty acid in *Terminalia catappa* seed oil

Fatty acid	Determined values		Reported values ^a
	Blye & Dyer	Soxhlet	
C16:0	36.12±1.37 ^A	35.81±1.41 ^A	35.0
C18:0	4.12±0.32 ^A	4.14±0.25 ^A	5.0
C18:1	31.30±0.35 ^A	31.65±0.48 ^A	32.0
C18:2	28.46±0.49 ^B	29.40±0.37 ^A	28.0
C18:3	-	-	-
Saturated	40.24	39.95	40.0
Unsaturated	59.76	60.05	60.0

ND: not determined., Means for the determined values in the same row followed by the same superscript letter are not significantly different (P < 0.05).

^a (1) Dos Santos *et al.* (2008).

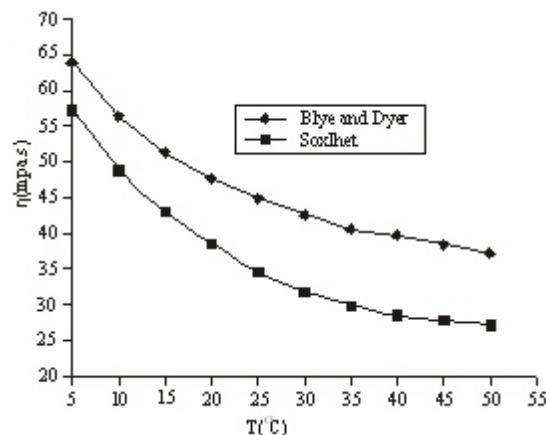


Fig 1: Effect of temperature on *terminalia catappa* L. seed oil viscosity

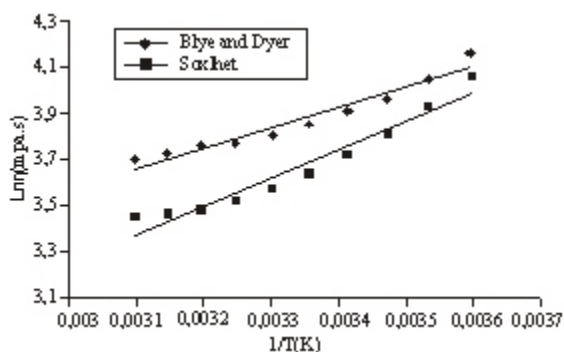


Fig. 2: Relationship between viscosity and temperature for *Terminalia catappa* L. seed oil extracted by Blye and Soxhlet. Solid line Arrhenius model

Table 8: Comparison of the profile in fatty vegetable oil acids

Huiles	C14 :0	C14 :1	C16 :0	C16 :1	C18 :0	C18 :1	C18 :2	C18 :3	C20 :0
Palm	1.0	-	44.5	0.2	4.6	38.7	10.5	0.3	0.3
Safou	-	-	45.5	-	2.8	28	24.9	1.24	-
Maize	-	-	10.5	-	2.5	28	58.5	1.0	0.5
Groundnut	-	-	10.0	-	2.0	46.0	31.0	-	-
Cotton	0.9	-	23.0	-	2.2	17.7	55.8	-	-
Hazel nut	-	-	7.0	0.1	2.0	74.5	16.5	-	-
Soybean	-	-	11.0	-	4.0	22.0	54.3	7.5	-
Jatropha	-	-	15.6	1.0	5.8	40.1	37.6	-	-
Terminalia	-	-	35.96	-	4.13	31.48	28.93	-	-

content of the Soxhlet method were significantly higher ($P < 0.05$) than those of the Blye and dyer method (Table 3). There was no significant difference in the iodine and saponification values, in the two extraction methods ($P > 0.05$). The slightly higher value of unsaponifiable matter in the Soxhlet method may be due to the ability of the solvent to extract other lipid associated substances like, sterols, fat soluble vitamins, hydrocarbons and pigments (Bastic *et al.*, 1978; Salunke *et al.*, 1992).

Fatty acid composition: The major saturated fatty acids in *Terminalia catappa* L. seed oil were palmitic (35.97%), stearic (4.13%) acids. The main unsaturated fatty acids are linoleic (28.93%) and oleic (31.48%) acids (Table 7). There was no significant difference ($P > 0.05$) in the amounts of the major fatty acids in the two oil samples. The two oil samples of *Terminalia catappa* L. contained saturated and unsaturated acids (40.10% and 59.90%) respectively. *Terminalia catappa* L. oil can be classified in the oleic-linoleic acid group. Linoleic acid which is one of the most important polyunsaturated fatty acids in human food because of its prevention of distinct heart vascular diseases (Boelhouwer, 1983). Apart from preventing cardiovascular disorders such as coronary heart diseases and atherosclerosis, linoleic acid also prevents high blood pressure. Also linoleic derivatives serve as structural components of the plasma membrane and as precursors of some metabolic regulatory compounds (Vles, 1989). *Terminalia capatta* oil is predominantly made up of palmitic (35.96%), oleic (31.48%) and linoleic (28.93%) acids respectively (Table 8). The results obtained are in agreement with those of the literature Dos santos *et al.* (2008). The comparison of the composition in fatty acids of *Terminalia catappa* seed oil with that of vegetable oils (Table 8) indicates that this plant is rich in acids oleic (C18:1), linoleic (C18:2) and palmitic (C16:0). This oil can about be simulated with the oil of safou (*Dacryodes edulis*).

CONCLUSION

This study showed that the *Terminalia catappa* L. seed is a good source rich in protein, minerals and oil. *Terminalia catappa* L. seed oil was obtained from the kernels with good yield (51.80%), allowing the possibility

of economical exploitation, and its fatty acid composition is comparable to that of some conventional oils. *Terminalia catappa* L. seed oil is of unsaturated type and contains mainly the fatty acids oleic C18:1(31.48%) and linoleic C18:2 (28.93%). The oil can be classified in the oleic-linoleic acid group. The oil extracts exhibited good physicochemical properties and could be useful for industrial applications.

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