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Exploring *Gmelina arborea* Leaves for Biofuels and Petrochemical and Pharmaceutical Feedstocks

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Authors' contributions

This work was carried in collaboration among the authors. Authors HI, KON and SA interpreted results, compiled the write up and supervised the work. Authors ASZ, OBA and DCN cleaned, pulverized, hydrolyzed the leaves. All authors read and approved the final manuscript.

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ABSTRACT

An investigation was carried on the chemical constituents of *Gmelina arborea* leaves by hydrolyzing the pulverized dry leaves with 3% sulphuric acid solution at 100°C for 30 minutes. The extract on analysis with GCMS yielded 31 compounds. The major components of the extract include; 5-methul furfural 13.024%, 5-hydroxyl methyl furfural 7.548%, 2-furan methanol 22.728%, 6-methyl-3-pyridazinone 3.256%, o-hydroxyphenol 2.704%, p-hydoxyphenol 4.4%, m-hydroxyphenol 3.048%, levoglucosenone 7.576%, levoglucosan 3.26%, 4-Altrosan 1.63%, 1,6-Anhydro-.beta.-d-talopyranose 1.63% and D-Allose 1.63%. Biorefinary processes of *Gmelina arborea* will be capable of replacing petroleum products in future. The plant leaves have feedstocks for pharmaceutical, agro-industries and others.

Keywords: Biofuels; explore; Gmelina arborea; leaves; petrochemical; pharmaceutical.

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1. INTRODUCTION

Gmelina arborea is a very important medicinal plant belongs to the family of Verbenaceae [1]. It is a fast growing deciduous tree with a wide spread canopy [2] and straight trunk [3]. It is commonly known as white teak and Kashmir [4]. It grows from 3 to 30 m tall [2:4], thrives better at 22-34°C, survive from 16-46°C but get killed at 1°C and below [2]. It has life span of 40 years [2] borne flowers 3-4 years after planting [3]. The leaves are 10-23 cm long 6.3 cm broad used as demulcent to threat gonorrhea and cough and also applied to wounds and ulcer [2]. The flowers are abundant scented reddish brown or yellow terminals and axillary 1-3 flowered cymes on the panicle branches about 8-40 cm long [3]. The flowers are said to bisexual in nature [1] and have been used to treat leprosy and blood diseases [2]. The fruits are edible sweet taste and its flowers can be used with rice to make delicious cake like festive dish [2]. The roots and barks contain oil, resins, alkaloids, benzoic acid, butyric acid and tartaric acid used for stomachic, laxative and anthelmintic good for remedy of pile, improve appetite and inflammation [4:2]. The root also used for blood purifier and antidote for snake bite and scorpion sting [2:4]. The fruits and barks have medicine for bilious fever [2]. Both the wood ash and fruits yield very persistent yellow dye [2].

The wood is light, soft to hard, very susceptible to fungi, dry wood borer and termites. The natural durability is about 15 years [2]. The wood is good for furniture, plywood cores stock, mine prop, matches and timber for construction of dug canoes, musical instruments and carving images [2:3]. The wood produces good quality pulp suitable for cardboard and low grade writing papers.

Adisa and Olurunsogu [5] reported to have isolated para-hydroxyphenol from leaves of *Cnestis ferruginea* (D.C). Chakraborty et al. [6], have extracted furfural from leaves of decorative plants and also Ibrahim et al., [7], have extracted furfural, 2-furan methanol and hosts of other useful chemicals from the leaves of ear leaf acacia using 3% sulphuric acid. In this work, we delight to investigate the chemical components of *Gmelina* leaves by hydrolyzing it with 3% sulphuric acid for 30 minutes at 100°C. The chemical components of the resulting extract were analyzed with GCMS.

2. MATERIALS AND METHODS

The materials used in this study include; pulverized *Gmelina arborea* leaves, GC-MS and Hot plate magnetic stirrer. The following laboratory equipment were also used; mortar and pestle, 1000 ml conical flask, 1000 ml beaker, filter paper, separating funnel, burette and thermometer. The chemical used include; sulphuric acid and analytical grade methanol. Dried leaves of *Gmelina arborea* were collected from around the premises of NARICT, Zaria. These leaves were pulverized using mortar and pestle. 3% sulphuric acid solution was prepared and 500 ml of it was mixed with 50 g of the pulverized leaves and heated to 100°C for 30 minutes [7]. The hydrolyzed mixture was filtered and part of it was neutralized with hydrated lime to pH of 7. The neutral filtrate was analyzed with GC-MS to determine its chemical constituents.

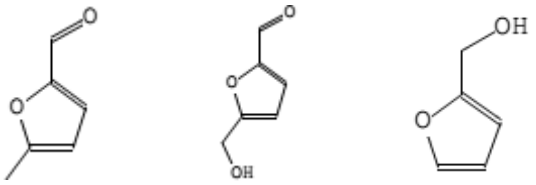
3. RESULTS AND DISCUSSION

The hydrolyzed product of *Gmelina arborea* leaves contain was found to 31 compounds as presented in Table 1. It has 20.576% furfural derivatives, 22.728% furan methanol, 3.256% 6-methyl-3-pyridazinone, 7.58% Levoglucosenone, 3.26% Levoglucosan, 15.022% benzene derivatives, 7.548% borane halides, 1.228% alkynes and 17.958% others as presented in Table 1. The benzene derivatives found in the product including; toluenes, phenols, styrene and amines. Hydroxyphenols consist of 10.152% which include ortho-hydroxyphenol, para-hydroxyphenol amd meta- hydroxyphenol. It had only 7.576% fatty acid which was 2, 4-pentadienoic acid.

3.1 Furfural Derivatives

Fig. 1 depicted the structural formula of three furfural derivatives found in the hydrolysis product which were; 5-methyl furfural 13.024%, 5-hydroxymethyl furfural 7.548% and 2-furan methanol 22.728% as presented in Table 1. 1. 5-methyl furfural is a very expensive compound. 98% concentration of this compound is selling at the rate of 23.50 euro for 25.0 g [8]. It boils at 186-188°C, relative density of 1.106, refractive index 1.5310, flash point 72°C soluble in alcohols and water. It is used in perfuming agent and a smoking essence [8]. 5-Methylfurfural is a volatile compound present in *Lavandula stoechas*, *Lavandula angustifolia*m and *Lavandula angustifolia* x *latifolia* unifloral honeys and formed

during the photo exposition of ranitidine hydrochloride. It is employed as potential age marker for Madeira wine [9]. 5-Hydroxylmethyl furfural abbreviated as HMF is known to be produced from hexose and cellulose [10:11]. It is used for production of polyurethane [10] and nylon 6,6 monomers [11]. It has been reported by Rosatella et al. [12] that HMF can be useful in the production of important molecules such as levulinic acid, 2,5-furandicarboxylic acid (FDA), 2,5-diformylfuran (DFF), dihydroxymethyl furan and 5-hydroxy-4-keto-2-pentenoic acid. 2-furan methanol has been found useful in Furan polymers, in making Sealants and Cements, Urea-formadehyde and Phenolic Resins, as a Solvent, Foundry cores, Flavorings [7].



5-methyl furfural **5-Hydroxymethyl furfural** **2-Furan methanol**

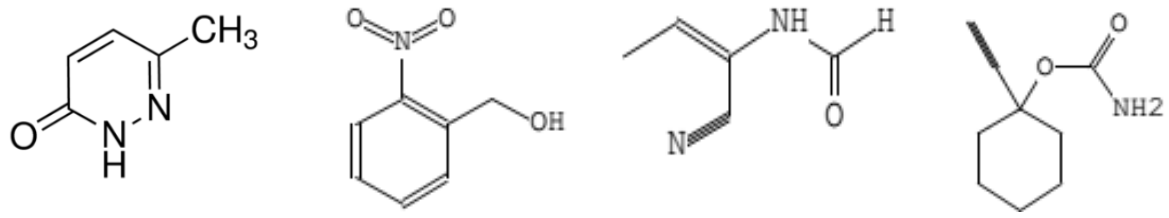
Fig. 1. Structural formulae of furfural derivatives extracted from *Gmelina arborea* leaves

3.2 Nitrogen Compounds

The structural formula of some nitrogen compounds extracted from *Gmelina arborea* is depicted in Fig. 2. They include; 6-Methyl-3-pyridazinone 3.256%, o-Nitrobenzylalcohol 0.912%, N-(1-Cyanopropenyl) formamide 0.848%, Ethinamat (Valamin) 0.614% among others as presented in Table 1. 6-methyl-3-pyridazinone is a derivative of pyridazine which has been found to be active in antimicrobial, antitubercular, analgesic and anti-inflammatory, antipyretic, antifeedant, herbicidal, antiplatelet, anticancer, cardiovascular and neurological disorder and other anticipated biological and pharmacological activities [7]. According to Phamacodes [13], Valamin is a very important pharmaceutical product for treatment for ascariasis caused by *Ascaris lumbricoides* (roundworm) and enterobiasis (oxyuriasis) caused by *Enterobius vermicularis* (pinworm). It is also used to treat partial intestinal obstruction by the common roundworm, a condition primarily occurring in children. Unfortunately, there was no information on uses of the other two compounds, o-Nitrobenzyl alcoholand N-(1-Cyanopropenyl) formamide.

3.3 Hydroxyphenols

Fig. 3 depicted the structural formula of three major benzene derivatives extracted from *Gmelina arborea* leaves. The hydrolytic product had a lot of benzene derivatives, but hydroxyphenols in Fig. 3 have higher concentration than others. They include; o-hydroxyphenol (pyrocatechol) 2.704%, p-hydroxyphenol (hydroquinone) 4.4% and m-hydroxyphenol (resorcinol) 3.048%. According to GPS-2011 [14] pyrocatechol or simply catechol is used as precursor to various flavorings such as vanillin or eugenol, used in food industry, perfumery, home and personal product. Adisa and Olurunsogu [5] reported that p-hydroxyphenol possesses very powerful antioxidant property. According to Drugs.com [15], the hydroquinone cream is used in lightening freckles, age spots and other skin discoloration associated with pregnancy, skin trauma, birth control pills or hormone. It has been reported by Xueyanghu [16] that resorcinol is used in antiseptic, disinfectant, as ointment in the treatment of skin diseases such as eczema and psoriasis and also antidandruff.

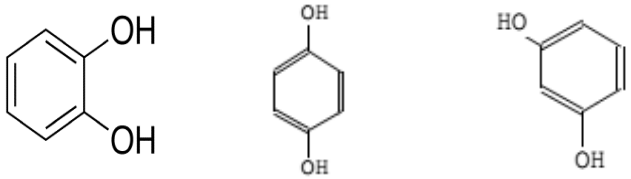


6-Methyl-3-pyridazinone **o-Nitrobenzylalcohol** **N-(1-Cyanopropenyl) formamide** **Valamin**

Fig. 2. Structural formulae of nitrogen compounds extracted from *Gmelina arborea* leaves

Table 1. Product components of *Gmelina arborea* leaves extract

	Compound	MF	% composition	Group composition%
1	5-methyl-Furfural	$C_6H_6O_2$	13.024	Furfural derivatives
2	5-Hydrxoymethylfurfural	$C_6H_6O_3$	7.548	
3	2-Furanmethanol	$C_5H_6O_2$	22.728	43.300
4	6-Methyl-3-pyridazinone	$C_5H_6N_2O$	3.256	Methyl pyridazinone 3.256
5	2,4-Pentadienoic acid	$C_5H_6O_2$	7.576	Fatty acid 7.576
6	3,4-Altrosan	$C_6H_{10}O_5$	1.63	
7	.beta.-D-Allose	$C_6H_{12}O_6$	1.63	
8	1,6-Anhydro-.beta.-d-talopyranose	$C_6H_{10}O_5$	1.63	
9	Levoglucosan	$C_6H_{10}O_5$	3.26	Sugar
10	Levoglucosenone	$C_6H_6O_3$	7.576	15.726
11	o-Hydroxyphenol	$C_6H_6O_2$	2.704	Benzene derivatives
12	p-Hydroxyphenol	$C_6H_6O_2$	4.4	15.022
13	m-Hydroxyphenol	$C_6H_6O_2$	3.048	
14	2-Hydroxy-4-methylphenol	$C_7H_8O_2$	0.456	
15	p-(Hydroxymethyl)phenol	$C_7H_8O_2$	0.912	
16	3-Nitro-4-methylaniline	$C_7H_8N_2O_2$	0.494	
17	(2-Nitrophenyl)methanol	$C_7H_7NO_3$	0.494	
18	1,2,3,6-Tetrahydrobenzaldehyde	$C_7H_{10}O$	0.614	
19	Toluene-2,6-diol	$C_7H_8O_2$	0.456	
20	2,6,2',6'-Tetramethylazobenzene	$C_{16}H_{18}N_2O_2$	0.494	
21	2,3-Dihydroxytoluene	$C_7H_8O_2$	0.456	
22	4-Hydroxy-3-methoxystyrene	$C_9H_{10}O_2$	0.494	
23	N-(1-Cyanopropenyl)formamide	$C_5H_6N_2O$	0.848	0.848
24	-4-Methyl-4-hepten-3-one	$C_8H_{14}O$	3.774	
25	Isopropyl(dipropyl)borane	$C_9H_{21}B$	3.774	Borane halides
26	Tripropylborane	$C_9H_{21}B$	3.774	7.548
27	2,7-Dioxa-tricyclo[4.4.0.0(3,8)]deca-4,9-diene	$C_8H_8O_2$	0.494	
28	3a,6-Methano-3aH-inden-4-ol, octahydro-, (3a.alpha.,4.alpha.,6.alpha.,7a.beta.)-	$C_{10}H_{16}O$	0.614	
29	Ethinamat (Valamin)	$C_9H_{13}NO_2$	0.614	
30	1-Undecen-3-yne	$C_{11}H_{18}$	0.614	Alkynes
31	1-Decen-3-yne	$C_{10}H_{16}$	0.614	1.228
	Total		**100**	

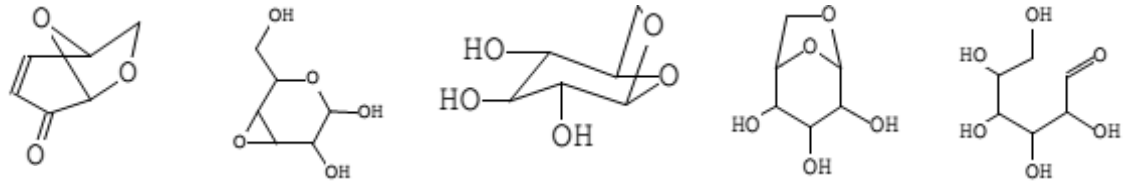


o-Hydroxyphenol p-Hydroxyphenol m-Hydroxyphenol

Fig. 3. Structural formula of Hydroxyphenols extracted from *Gmelina arborea* leaves

3.4 Sugars

Five simple sugars were identified from the hydrolytic extraction of *Gmelina arborea* leaves which are; levoglucosenone 7.576%, Levoglucosan, 3.26% 3,4-Altrosan, 1.63%, 1,6-Anhydro-.beta.-d-talopyranose, 1.63%, and D-Allose (or Hexose) 1.63%. The structural formulas of the five sugars are shown in Fig. 4. Levoglucosenone is produced from cellulose



Levoglucosenone **3,4-Altrosan** **Levoglucosan** **1,6-Anhydro-.beta.-d-talopyranose** **D-Allose (or Hexose)**

Fig. 4. Structural formulae of simple sugars extracted from *Gmelina arborea* leaves

which has been used for the synthesis of natural products such as tetrodotoxin [17]. According to Perez [18], levoglucosan can be hydrolyzed and converted to a multitude of chemical species, including alcohols and lipids and that the use of levoglucosan for the production of polymers was extensively studied in the 1980s and early 1990s. 3,4-Altrosan is a Bacteriostat and fungicide [19:20]. 1,6-Anhydro-.beta.-d-talopyranose also called 1,6-anhydro-beta-D-mannopyranose is found used as a marker of toasted oak wood used for ageing wines and distillates [21]. Cayman [22] described it as a major organic tracer used to evaluate in atmospheric samples of burning of wood. It is reported that β-D-Allose a rare sugar member of the aldohexose family and a C3 of glucose is used as an inhibitor of fruiting body formation and sporulation in myxococcus xanthus [23].

The extract of *Gmelina arborea* leaves is rich in pharmaceutical, biofuel and plastic feedstocks. 2-furan methanol (furfuryl alcohol) which has the highest quantity in the extract is described as the most important furfural derivative due to its numerous applications. It has been used in reinforced carbon-carbon composite materials, developed to protect the shuttle around its nose and wing leading edge from extremely high and cold temperatures (-121-1,649°C) encountered during there-entry of shuttles into space [24]. It has ingredient for wood modification processes that convert soft woods to products that like and have properties that are similar to tropical hard woods [24]. It was reported by Elbert [25] that the blend of hydroxymethyl furfural fossil gave smooth engine performance with significant reduction in sooth emissions from vehicles. Therefore furfural and its derivatives are transportation fuels for the future. Eseyin and Steele [24] also reported that hydrocarbon fuels can also be produced from furfural and its derivatives by their hydrogenolysis. Hence, biorefinary can be established with *Gmelina arborea* leaves as feedstock for the production of biofuels. The benzene derivatives have wide range of applications in petrochemicals, fuels, pharmaceutical, agrochemicals and food. Among the nitrogen compounds found in the extract methyl pyridazinone is most useful with wide range of applications. The sugar content of the extract all have important applications, this makes the leaves of the *Gmelina arborea* as important as other parts of it. The exploration of *Gmelina arborea* will provide good alternative for petroleum base products for industrial applications. *Gmelina arborea* leaves is renewable and sustainable and that can have the capacity to replace fossil fuel for transportation fuels and plastic chemicals.

4. CONCLUSION

Extraction of *Gmelina arborea* leaves extract with 3% sulphuric acid yielded 43.3% furfural derivatives made up of 13.028, 7.548 and 22.728% 5-methyl furfural, 5-hydroxyl methyl furfural and 2-furan methanol. Also 3.256% 6-methyl-3-pyridazinone, 15.726% sugars and 15.022% benzene derivatives were found in the extract. The sugars comprises of Levoglucosenone, 3,4-Altrosan, Levoglucosan, 1,6-Anhydro-.Beta.-D-talopyranose and D-Allose with compositions of 7.576, 1.63, 3.26, 1.63 and 1.63% respectively. Pyrocactechol (o-hydroxyphenol), hydroquinone (p-hydroxyphenol) and resorcinol (m-hydrxyphenol) had 2.704, 4.4 and 3.048% respectively were the major benzene derivatives in the extract. Exploration of *Gmelina arborea* as a renewable source, transportation fuels, chemicals for petrochemical, plastic, pharmaceutical and food industries can be found in abundance to replace fossil fuel sources.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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