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Phytochemical studies on roots of *Gmelina asiatica* Linn.

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ABSTRACT - A bioguided extraction and fractionation of the alcoholic extract of the roots of *Gmelina asiatica* afforded a new flavone derivative named ovalifolin [3-(3-methyl-1-butenyl)-6-methoxy-5, 7, 4'-trihydroxy flavone] has been isolated and identified together with the known compounds (+)Sesamin, Sakuranetin, (-)Piperitol, (+)Pinoresinol. The structures of these compounds have been established on the basis of chemical, chromatographic and spectral methods.

KEYWORDS - *Gmelina asiatica*; Verbenaceae; ovalifolin, flavonoid.

INTRODUCTION

Gmelina asiatica Linn. (syn. *Gmelina parvifolia*) (Family-Verbenaceae) is commonly known as 'Nilakkumil' in Tamil and 'Gopabhandra' in Sanskrit. The root of the plant is used in gonorrhoea, catarrh of the bladder, rheumatism and as a blood purifier.[1)] The plant is claimed to be useful in rheumatism, and also possess anti-inflammatory effect.[2)] Previous study reported on hypoglycemic and antihyperglycemic activity of alcoholic extract on roots of *G. asiatica*.[3)] Previous investigations of *G. asiatica* have isolated quercetagetin, glycosides of kaempferol, apigenin and luteolin[4,5)]. We report herein upon the isolation and structural elucidation of a new flavone derivative named ovalifolin (1). Ovalifolin, [3-(3-methyl-1-butenyl)-6-methoxy-5, 7, 4'-trihydroxy flavone] has been isolated and identified together with the known compounds (+)Sesamin (2), Sakuranetin (3), Piperitol (4), (+)Pinoresinol (5).

MATERIAL AND METHODS

Plant material

The roots of *Gmelina asiatica* L. were collected from Annavaram hills of East Godavari district, Andhra Pradesh, South India. Dr. K Hemadri, taxonamist, Regional Research Institute, Botanical Survey of India, Vijayawada, identified the herb. A voucher specimen (G-0567) was deposited in the herbarium of our department.

Extraction and isolation procedure

Freshly collected roots of *G. asiatica* were cut into small pieces and shade dried. The dried roots were powdered in Wiley mill. The powdered root (1500 gms) was extracted with ethyl alcohol (95.0% v/v) by process of continuous extraction (Soxhlation). The crude extract was evaporated to dryness in a rotary film evaporator (Roteava, Equitron, Medica instrument, India). 1 g of ethanolic extract equivalent to 21.3 g of crude drug was obtained. The alcoholic extract was fractioned with hexane (10x250 ml), ether (5x100 ml), chloroform (7x150 ml), ethylacetate (4x150 ml) and methanol (15x200 ml). All solubles were concentrated and the percentage yields were: hexane 9%, ether 21%, chloroform 15%, methanol 8% and ethylacetate 6% (w/w) in terms of dry plant material. Among all the solubles, the ether and chloroform fractions showed three and two distinct spots in thin layer chromatography over silica gel in different solvent systems respectively. The ether solubles were chromatographed over silica gel (finer than 200#, ACME) column and eluted taking 1000 ml fractions, starting with $CHCl_3$/hexane, 10:90 (frs. 10-23) to afford compound 1 (yield 0.011%), $CHCl_3$/hexane, 20:80 (frs. 32-46) to afford compound 2 (yield 0.016%), $CHCl_3$/hexane, 30:70 (frs 59-67) to afford compound 3 (yield 0.012%). The chloroform solubles were chromatographed over silica gel (finer than 200#, ACME) column and eluted taking 1000 ml fractions, starting with $CHCl_3$ 100% (frs. 51-60) to afford compound 4 (yield 0.011%), $CHCl_3/CH_3OH$, 95:05 (frs. 71-80) to afford compound 5 (yield 0.016%).

RESULTS AND DSUSSION

Compound 1 was isolated as colourless needles, m.p

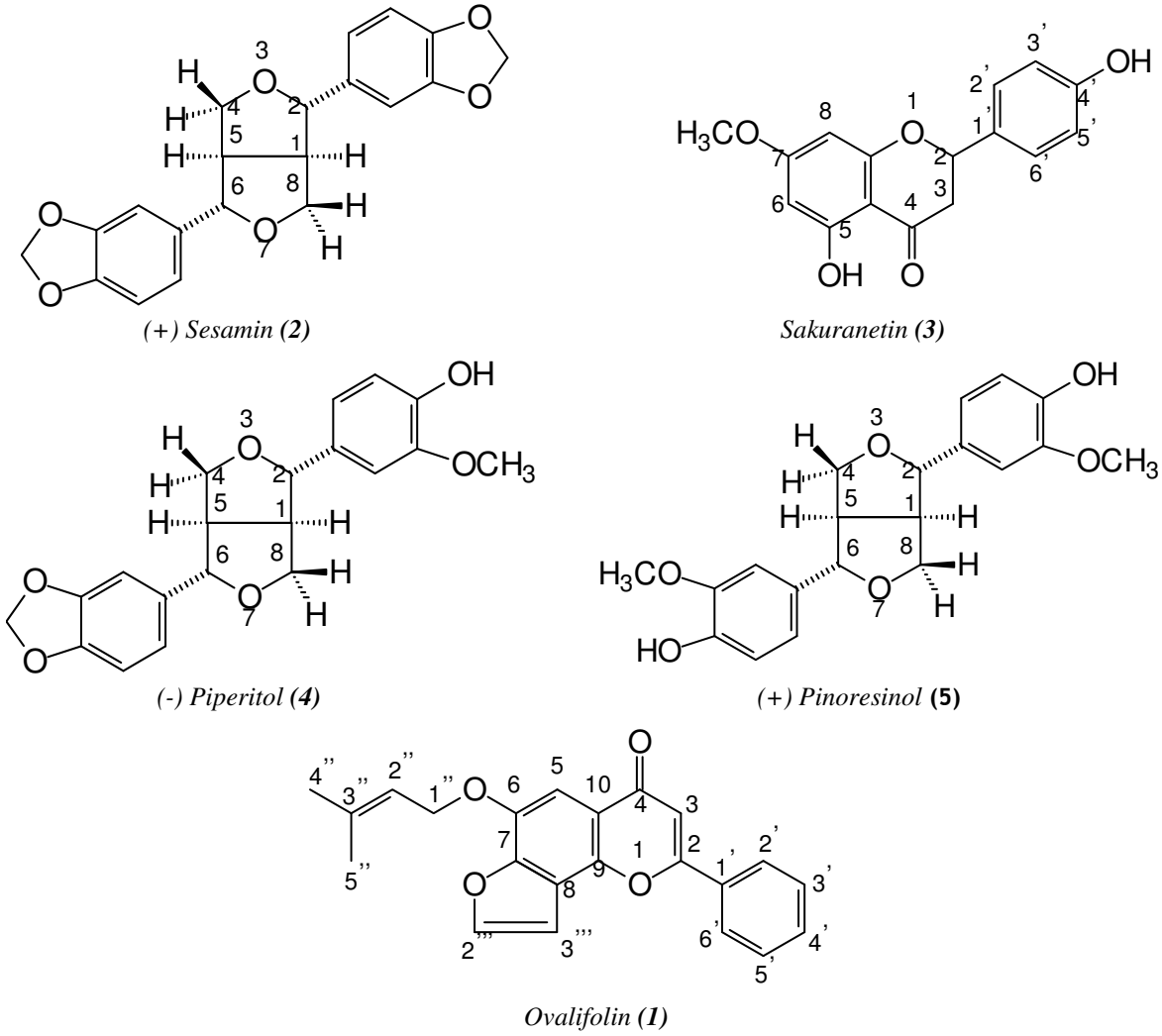

Fig.1. Structures of the compounds 1 - 5.

134-135ºC, its molecular formula, $C_{22}H_{18}O_4$ was established by HR-MS. Its IR spectrum showed the absorption for a flavonoid carbonyl group (1640 cm^{-1}), aromatic double bond stretchings (1620 and 1595 cm^{-1}) and stretchings of aromatic ethers, symmetric and asymmetric (1215, 1085, 1295 and 1060 cm^{-1}) respectively. ^1H NMR spectrum had resonances at δ 6.85 (1H, s, 3-H) indicating the compound is flavone in nature and furan ring was evidenced at δ 7.76(1H, d, J=2.1Hz, H-2‴) and δ 7.19(1H, d, J=2.1Hz, H-3‴). Unsubstituted aromatic ring was evidenced at δ 7.93(2H) and δ 7.53(4H) and the presence of γ-dimethylallyloxy side chain at δ 4.79(2H, d, J=6.8Hz, H-1˝), δ5.61(1H, t, J=13.7Hz, H-2˝), δ 1.82(3h, s, CH_3-4˝), δ 1.85 (3H, s, CH_3-5˝).

The ^{13}C NMR spectrum of compound 1 revealed the presence of flavone nature by resonances at δ 162.5(C-2), δ 107.6(C-3) and furan ring at δ 146.1(C-2‴) and δ 104.8(C-3‴). Unsubstituted aromatic ring was evidenced at δ 126.3(C-2′, 6′), δ 129.2(C-3′,5′), δ 131.5(C-4′) and the presence of γ-dimethylallyloxy side chine at δ 66.3 (C-1˝), δ 118.9 (C-2˝), δ 139.5 (C-3˝), δ 26.0 (CH_3-4˝) and δ 18.5 (CH_3-5˝).

Compound 2 was isolated as colourless crystalline compound from CH_3OH m.p./ 122-124ºC $[\alpha]^{25}_D$ + 59.5^0. Its molecular formula was determined as $C_{20}H_{18}O_6$ by HR-MS. Its IR spectrum, the bands between 1480 and 1610 cm^{-1} indicated its aromatic nature and bands between 1025 and 1100 cm^{-1} indicated the presence of ether linkages. Further a strong peak at 937 cm^{-1} and Labats colour reactions supports the presence of methylenedioxy group. The ^1H as well as ^{13}C NMR- data

were found to be identical with spectrum of those already reported earlier for (+) Sesamin.[6,7]

Compound 3 was isolated as colourless crystalline compound from aqueous CH_3OH m.p. 150-152°C, $[\alpha]^{25}_D$ - 9.2^0, (c 0.30, $CHCl_3$). Its molecular formula was determined as $C_{16}H_{14}O_5$ by HR-MS. Positive results of alcoholic $FeCl_3$ reaction and Shinoda's test indicating the flavonoids nature. IR V_{max} KBr cm^{-1}: 3580, 3050, 1665, 1610, 1460, 820, UV λ_{max} nm: 287. The ^1H as well as ^{13}C NMR- data was found to be identical with spectrum of those already reported earlier for Sakuranetin.[8,9]

Compound 4 was isolated as colurless gummy solid from CH_3OH, $[\alpha]^{25}_D$ - 75.2°C (c 1.2, $CHCl_3$). Its molecular formula was determined as $C_{20}H_{20}O_6$. Positive results of concentrated H_2SO_4 and Labats test indicating the presence of methylenedioxy group. IR spectra indicate the presence of hydroxyl (3550 cm^{-1}), aromatic nature (1610 cm^{-1}) functions. IR V_{max} $CHCl_3$ cm^{-1}: 3590, 2940, 2060, 1610, 1490, 1440, 1370, 1285, 1040, 930, 850. UV λ_{max} nm: 282. The ^1H as well as ^{13}C NMR- data was found to be identical with spectrum of those already reported earlier for (-) Piperitol.[10-12]

Compound 5: Colourless amorphous powder from acetone: ether, m.p.120-121°C, $[\alpha]^{25}_D$ + 62.76, (c 0.76, $CHCl_3$). Its molecular formula was determined as $C_{20}H_{22}O_6$ by HR-MS. The ^1H as well as ^{13}C NMR- data were found to be identical with spectrum of those already reported earlier for (+) Pinoresinol.[13]

NMR data

Compound 1: ^1H NMR (400 MHz, $CDCl_3$), δ 6.85 (1H, s, H-3), 7.53(4H, m, H-5, 3', 4', 5'), 7.93(2H, m, H-21, 61), δ 4.79 (2H, d, J=6.8Hz, H-1''), δ 5.61(1H, t, J=13.7Hz, H-2''), δ 1.82 (3h, s, CH_3-4''), δ 1.85 (3H, s, CH_3-5''), δ 7.76(1H, d, J=2.1Hz, H-2''') and δ 7.19(1H, d, J=2.1Hz, H-3'''). The ^{13}C NMR (100 MHz, $CDCl_3$), δ 162.5(C-2), δ 107.6(C-3), 178.3(C-4), 101.1(C-5), 148.6(C-6), 145.8(C-7), 120.2(C-8), 143.8(C-9), 119.1(C-10), 132.1(C-1'), 66.3 (C-1''), δ 118.9 (C-2''), δ 139.5 (C-3''), δ 26.0 (CH_3-4'') and δ 18.5 (CH_3-5''), 146.1(C-2''') and δ 104.8(C-3''').

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