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A new flavone from *Excoecaria agallocha L*

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ABSTRACT

Isolation of a new flavone [8-hydroxy-2(3-hydroxy-4-methoxy phenyl) 4-oxo-3-propoxy-4H-chromen-7yl-propionate] from Excoecaria agallocha was confirmed through spectroscopic analysis.

Key words*: Excoecaria agallocha*; 8-hydroxy-2(3-hydroxy-4-methoxy phenyl) 4-oxo-3-propoxy-4H-chromen-7yl-propionate.

INTRODUCTION

In this investigation the widespread tropical genus *Excoecaria* is known for the production of toxic metabolites [1]. It is distributed on seashores and edge mangroves sometimes cultivated for wind and sea-breaks in tropical Africa and East Asia. The leaves and milky fluid obtained from damaged branches have been used as a fish poison in New Caledonia and in Okinawa, the resinous wood including latex, the so-called "Okinawa-Jinko", has also been used as a substitute for the incense of agalwood[2-3]. The bark and wood of this tree have been used in traditional medicines for flatulence in Thailand [4-5]. Anjaneyulu and LakshmanaRao 2000 reported a large number of new diterpenoids from the n-hexane extract and from the ethyl acetate solubles of the CH_3OH:CH_2Cl_2 (1:1) extract of the roots of Indian mangrove plant *Excoecaria agallocha L.* (family: Euphorbiaceae) [6-7]. The piscicidal constituent of the twigs and bark of *E.agallocha* native to Okinawa has been characterized as the daphnanediterpene ester excoecariatoxin. This diterpene ester and some related compounds have also been obtained from the latex of *E.agallocha* in Thailand [8].

Being one of non-viviparous true mangrove species *E.agallocha* is naturally distributed on wetland along the coastlines in china which has been used as a dart poison and fish poison in south-eastern Asia [9]. In Traditional Thai medicine, the bark and wood of the plant are used to compact flatulence. Several skin irritant daphnane and tiglianediterpene esters have been isolated from the latex of *E.agallocha*[10].*E.agallocha*is herb medicine traditionally used for the treatment of ulcers, rheumatism, leprosy, and paralysis in the coast regions of south china. While the latex was used as a purgative and abortifacient [11-15].

A new Flavone 8-hydroxy-2(3-hydroxy-4-methoxy phenyl) 4-oxo-3-propoxy-4H-chromen-7yl-propionate and their structural elucidation.

MATERIALS AND METHODS

Melting points were determined on a VEB-Analytic Dreader HMK hot plate and are uncorrected. IR spectra were reported on a Perkin-Elmar 841 IR Spectrometer in $CHCl_3$ solution. UV spectra were recorded on a Milton Roy Spectronic 1201 spectrometer in $CHCl_3$. ^{1}H NMR spectra were measured on a Bruker Advance DRX 600 and JEOL JNM EX-90 spectrometers. ^{13}C NMR spectra were measured on a Bruker Advance DRX 600 spectrophotometer at

400 MHz and JEOL JNM EX-90 spectrophotometer at 22.5 MHz using $CDCl_3$ as a solvent and tetra-methylsilane as an internal reference. Mass spectra were obtained on a JEOL JMS-300 spectrometer.

Plant Material

The roots of E.agallocha were quiet from Corangi Mangrove forest near Bhiravapalem of Godavari Estuary (16^0 58' N latitude and 82^0 15' E longitudes) in March 2002 and was established by Prof.B.KondalaRao, Dept.of Marine Living Sources, Andhra University, and Visakhapatnam. Voucher specimens (Code: AU 1/60) have been deposited at the Marine Museums of School of Chemistry, Andhra University and National Institute of Oceanography, Goa.

Extraction, Isolation, and Characterization

The air-dried and powdered plant material (6 kg) was exhaustively extracted with n-hexane. Removal of the solvent from the combined n-hexane extracts under reduced pressure gave a residue (45 g). This residue was subjected to column chromatography over a column of silica gel (Acme brand, 100-200 mesh, and 450 g) using solvents of increasing polarity from n-hexane through EtoAc. In all 200 Fractions (500 ml) were collected. The fractions displaying similar spots in TLC were combined and the residues from therein were subjected to re-chromatography over silica gel column to yield one pure compound Fig.I In the form of an off-white solid.

RESULTS AND DISCUSSION

The crude product obtained by isolation was subjected to exhaustive column chromatography by using silica-gel (100-200 mesh). The chromatographic fractions were collected using n-hexane through ethyl-acetate. The TLC of all the fractions was checked with n-hexane and ethyl-acetate as eluent. The products were identified in the iodine chamber by using sulphuric acid spray. The fractions containing to the pure product as per TLC was evaporated by distillation followed by drying the product. The product thus obtained was triturated with n-hexane to furnish the pure product(HPLC purify 95%). The compound was then subjected to spectroscopic studies carried out. Like UV, IR, ^{1}H NMR, ^{13}NMR, MASS. Which Compound is further conformed by 2DNMR like COSY, NOESY, HMBC, HMQC, HSQC were obtained for the compound.

UV ($CHCl_3$) λ_{max} 270nm.It indicates conjugated-enone. Melting Point is 110^0c.

IR $\nu KBr_{max}cm^{-1}$ 3534, 2918,2850,1710,1513,1464,1267,1174,755.

1) The Peakat 3534 cm^{-1} indicates the presence of alcoholic group
2) The peak at 1710cm^{-1} indicates the presence ofcarbonyl group
3) The Peak at 2850cm^{-1} indicates the presence of saturated hydro carbon group
4) The Peaks at 1513 cm^{-1}, 1464cm^{-1}, 1267 cm^{-1}, 755cm^{-1} indicates the presence of aromatic group
5) The Peakat 1174cm^{-1} indicates aliphatic cyclic chain

^{1}HNMR (400 MHz, $CDCl_3$)

δ 7.6(d, 1H, J=16Hz), 7.1(dd,1H,J=3.6 Hz), 7.0(dd, 1H, J=1.6 Hz), 6.9 (d, 1H, J=8 Hz), 6.3(d, 1H, J=16 Hz), 4.2(t, 2H, J=6.8 Hz), 3.9(s,3H), 2.4-2.3(t, 3H, 7.6 Hz), 1.7-1.5(m), 1.4-1.2(m), 0.9(t,9H,6.4 Hz).

The Presence of two doublets, two doublets of doublets and singlet indicate the presence of aromatic rings and has flavone structure. Similarly a singlet at δ3.9 indicates the presence of methoxy, a triplet at δ 4.2 indicates the presence ofCH_2 attached to oxygen. A triplet observed at δ2.4 indicates the presence of CH_2 attached to carbonyl. The presence large number of protons in the form of a broad & high peak at δ 1.1 indicates the presence of either aliphatic side chain or cyclic aliphatic chain.

^{13}CNMR (400 MHz, $CDCl_3$)

δ 178.0, 167.4,148,146.8, 144.6, 127.1, 123, 115.8, 114.7, 109.4, 77.3, 77, 76.7, 64.6, 63.1, 56, 33, 32, 31.9, 29.7, 29.6, 29.5, 29.4, 29.3, 29.2, 29.1, 28.8, 26, 25.7, 24.7.

1) In this investigation the peak at 178& 167 corresponds to carbonyl group.
2) The peaks in between 150-110 indicated the presence of aromatic ring.
3) The Peaks at 63 & 56 indicates the presence of carbon attached to oxygen.
4) The peaks observed between 20&15 indicated long aliphatic side chain which could be probably derived from esterification with fatty-acid.

M+1ion is 639.Further conformed by the proposed structure.

Based on the above spectral data the following tentative structure has been proposed. Molecular Formula is $C_{38}H_{54}O_8$.

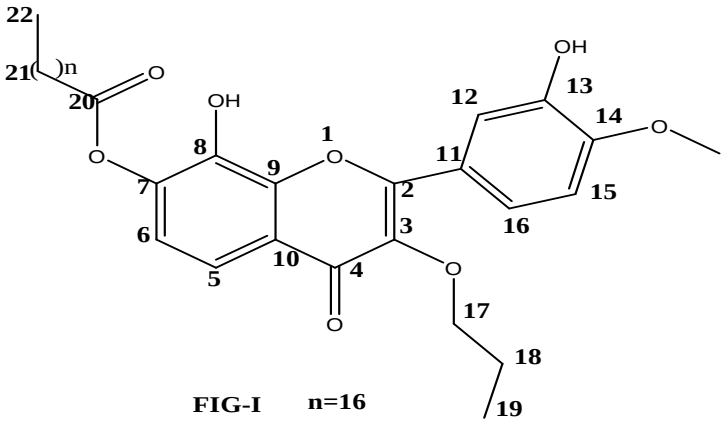
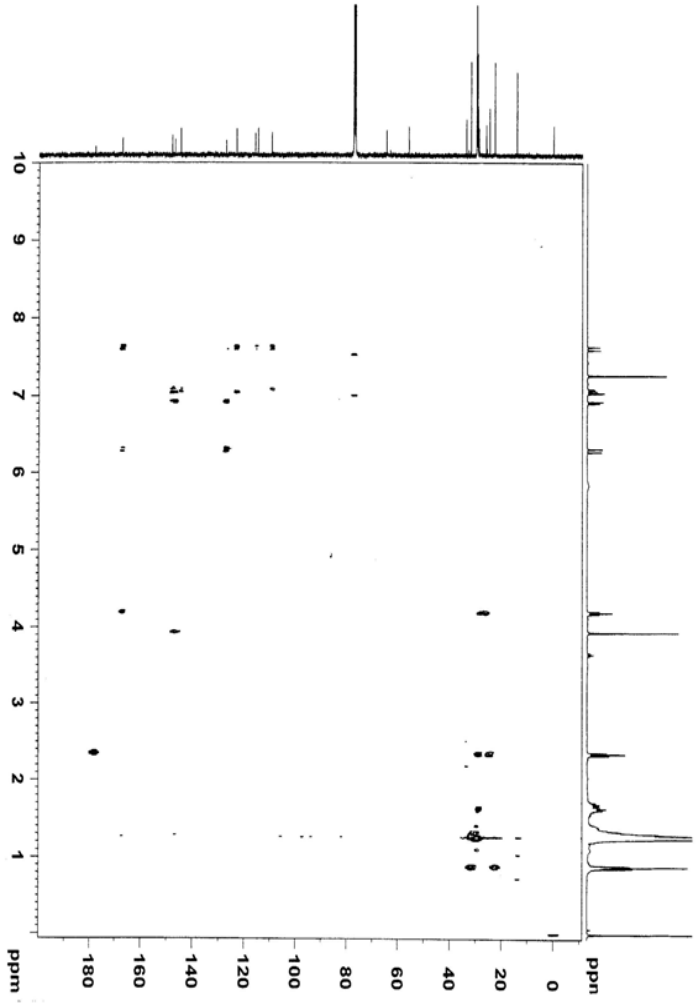


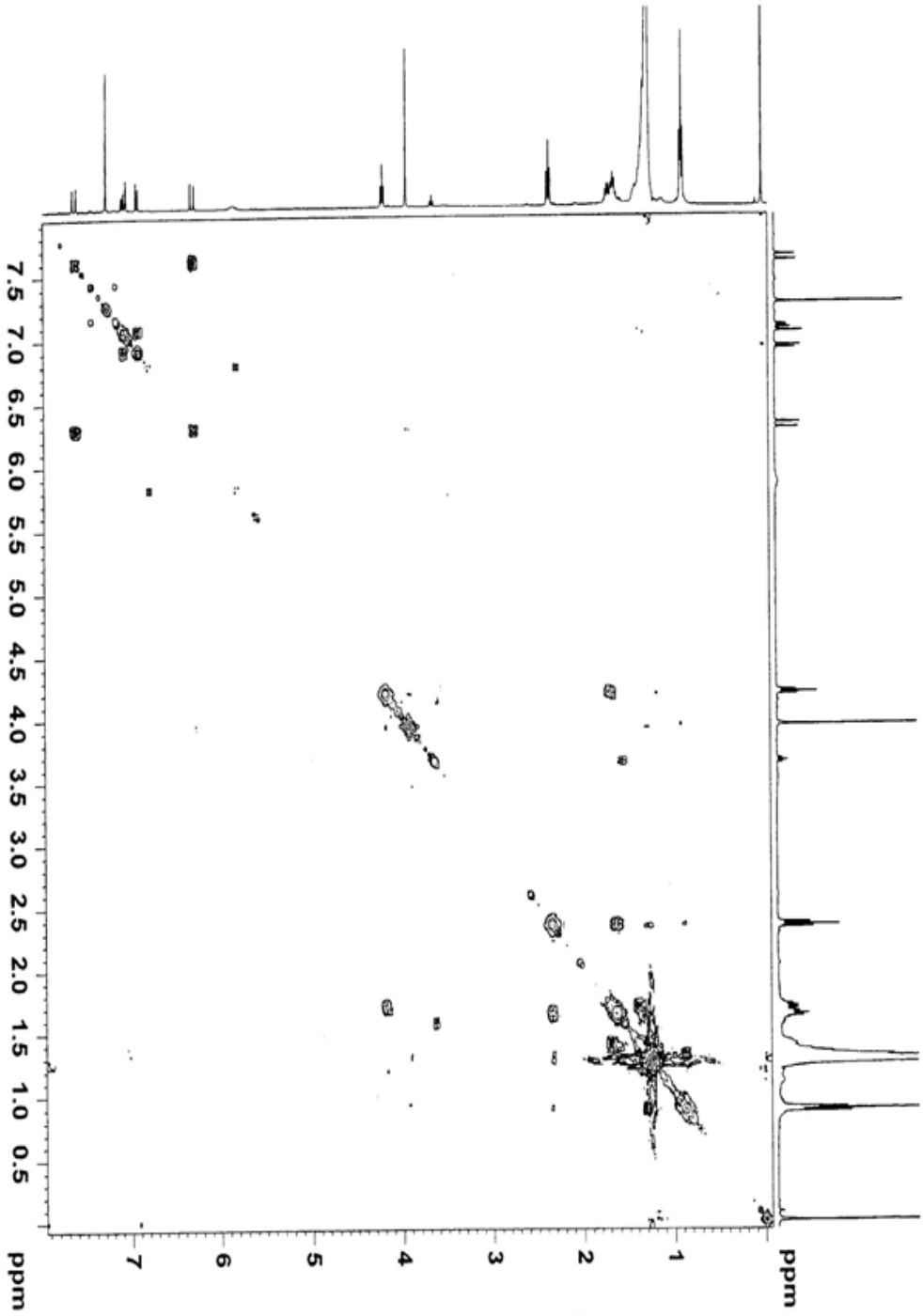
FIG-I n=16

For further conformation advanced NMR-data such as 2D-COSY, NOESY, HMBC etc. has been recorded which support the structure

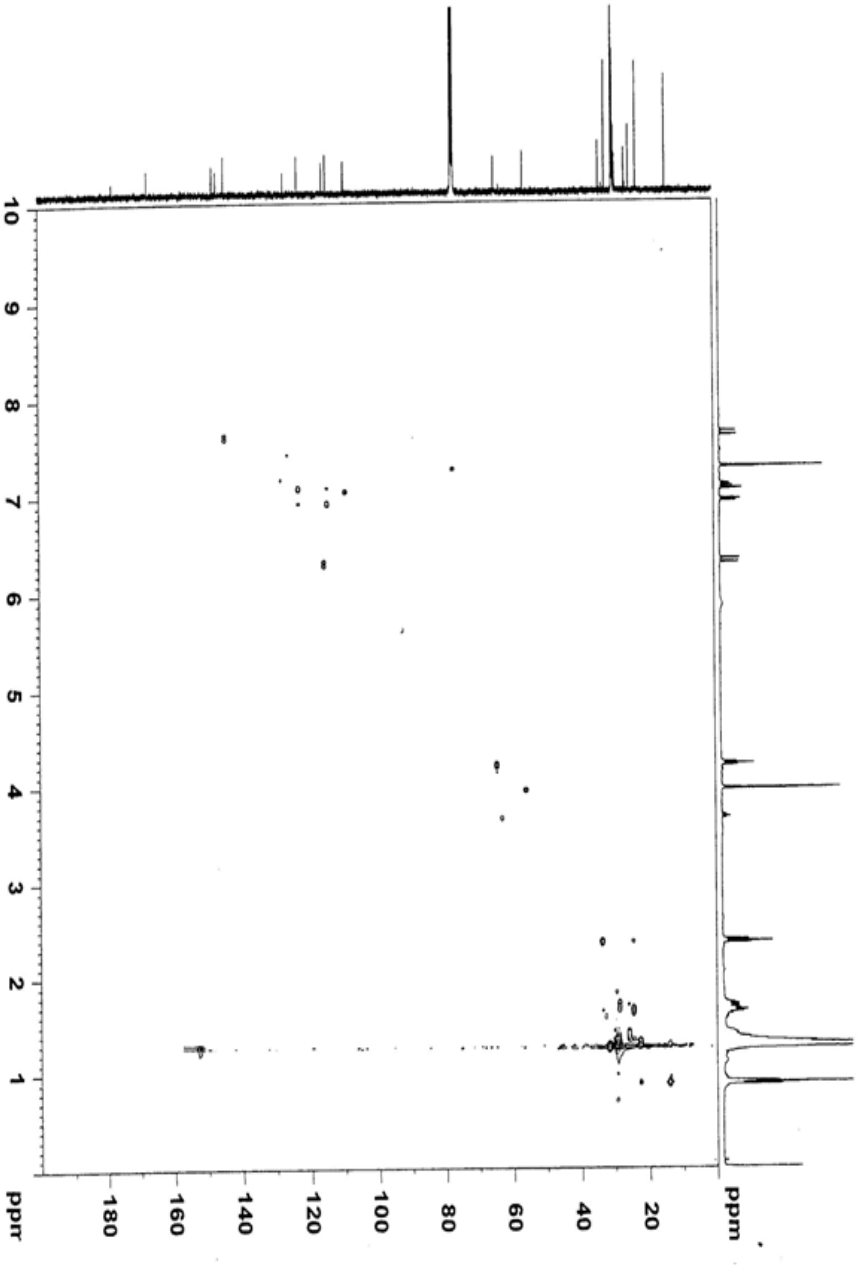
Excoecaria Agallocha **HMBC ($CDCl_3$) -SPECTRA**



Excoecaria Agallocha **COSY ($CDCl_3$) -SPECTRA**



Excoecaria Agallocha **HSQC ($CDCl_3$) -SPECTRA**



All the above spectral studies has supported to Basic Skeleton proposed for the Compound.

CONCLUSION

The compound isolated from the root of *Excoecaria agallocha* was given chemical structure **FIG-I** based on the spectral analysis. Further work is in progress to obtain suitable derivatives and followed by Single Crystal X-RAY analysis.

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