



Potential anti-inflammatory diterpenes from *Premna obtusifolia*

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ABSTRACT

Eleven new diterpenoids: pimaranes (**1,2**), rosanes (**3**) abietanes (**4–7**), icetexanes (**8**), and rearranged icetexanes (**9–11**), together with fifteen known diterpenes (**12–26**) and two known sesquiterpenes (**27,28**) have been isolated from the roots and twigs of *Premna obtusifolia*. The structures of these compounds were elucidated on the basis of spectroscopic analysis and X-ray crystallographic data of **8** and **9** were also determined. The antibacterial and anti-inflammatory activities of the isolates were evaluated.

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1. Introduction

The genus *Premna* L. (Verbenaceae) contains about 200 species worldwide, which are mainly distributed in tropical and subtropical Asia, Africa, Australia, and Pacific Islands¹ and 30 species are found in Thailand.² Previous studies on the biological and chemical constituents of members of the genus *Premna* have resulted in the isolation of triterpenes,³ diterpenes,^{4–8} sesquiterpenes,⁹ iridoid glycosides,^{10,11} phenylethanoids,^{12,13} and xanthenes.¹⁴ Some of which have exhibited significant antibacterial,^{5,6,9,10} and cytotoxic activities.^{7,8} *Premna obtusifolia* is widely distributed in the southern part of Thailand and locally known as 'akkhithawan thale'.¹⁵ Crude hexane and CH₂Cl₂ extracts from the twigs and roots of this plant have displayed potent inhibitory activity against lipopolysaccharide (LPS)-induced NO production in RAW 264.7 cell lines from our preliminary test. We report herein eleven new diterpenes (**1–11**) and seventeen known compounds (**12–28**). The antibacterial and anti-inflammatory activities of selected compounds are also reported.

2. Results and discussion

Investigation of the roots and twigs of *P. obtusifolia* furnished eleven new compounds (**1–11**) (Fig. 1), along with seventeen known compounds (**12–28**) (Fig. 2). The known compounds were identified as lambertic acid (**12**),¹⁶ ferruginol (**13**),¹⁷ O-methyl ferruginol (**14**),¹⁸ sugiol (**15**), royleanone (**16**),¹⁹ horminone (**17**),²⁰ montbretrol (**18**),²¹ 14-deoxycoleon (**19**),²² taxodion (**20**),²³ arucadiol (**21**),²⁴ 12-hydroxy-6,7-secoabieta-8,11,13-triene-6,7-dial (**22**),¹⁸ salvicinaraldehyde (**23**),²⁵ 5,6-dihydro-6 α -hydroxy-salviasperanol (**24**), salviasperanol (**25**),²⁶ 11,12-dihydroxy-8,11,13-icetexatrien-1-one (**26**),²⁷ 4 β ,5 β -dihydroxy-10-*epi*-eudesmane (**27**),²⁸ 4 β ,10 β -dihydroxyaromadendrane (**28**),²⁹ by comparison of their physical, UV, IR, ¹H and ¹³C NMR data with those already published. We now report the characterization of **1–11**.

Compound **1** was obtained as a white amorphous solid, which exhibited a molecular ion [M]⁺ peak at 304.2407 (calcd 304.2402) in the HREIMS corresponding to a molecular formula C₂₀H₃₂O₂ with 5° of unsaturation. The IR spectrum showed strong absorption bands for hydroxyl at 3339 cm⁻¹ and double bond at 1637 cm⁻¹. The combined analysis of the ¹³C NMR (Table 2) and DEPT spectra revealed the presence of 20 carbon signals assigned to four methyls, six methylenes, six methines including those of two oxymethines, and two olefinic methine

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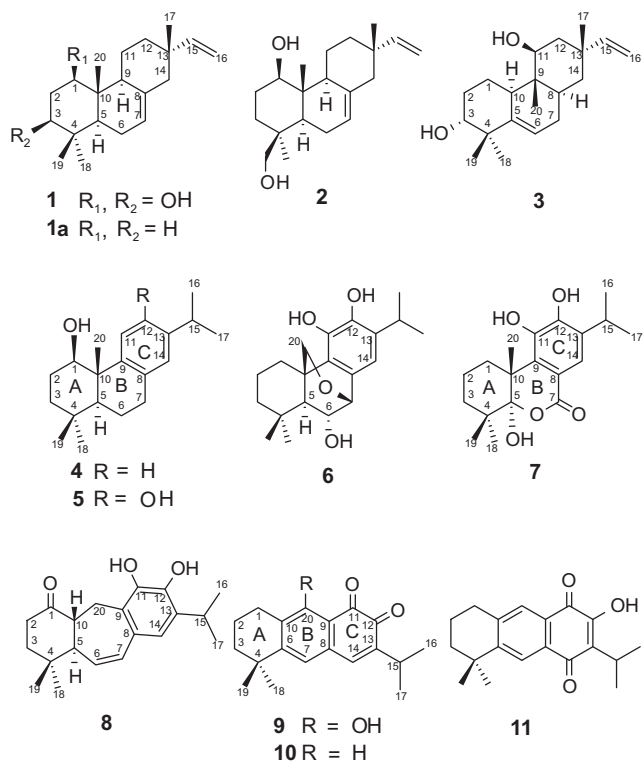


Fig. 1. Structures of compounds 1–11.

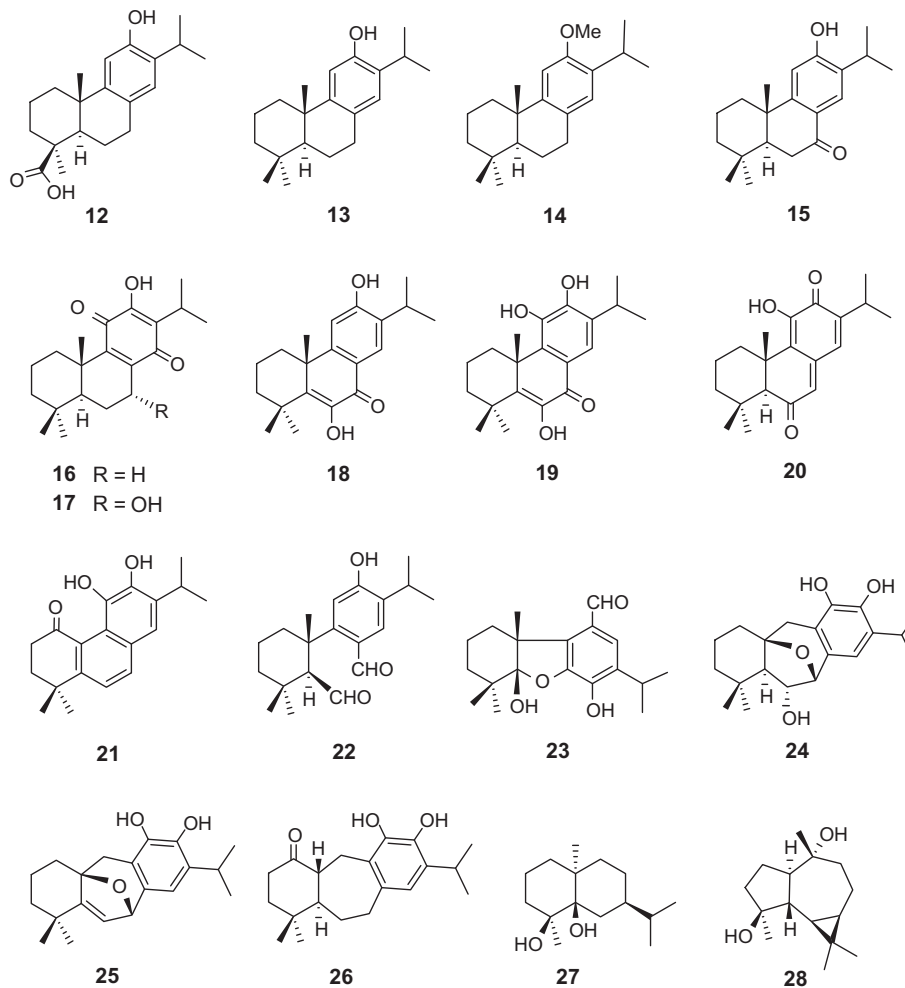


Fig. 2. Structures of compounds 12–28.

carbons and four quaternary carbons. The occurrence in the ^1H NMR spectrum (Table 1) of a vinyl group with three dd resonances at δ_{H} 5.80 ($J=17.4$ and 10.8 Hz, H-15), δ_{H} 4.92 ($J=17.4$ and 1.2 Hz, H_b-16), and δ_{H} 4.86 ($J=10.8$ and 1.2 Hz, H_a-16) and the presence of a broad doublet for an olefinic methine proton at δ_{H} 5.39 ($J=3.0$ Hz, H-7) and four methyl group singlets suggested a pimarane type skeleton.³⁰ Two oxymethine carbons resonated at δ_{C} 79.0 (C-1) and 76.1 (C-3). Their positions were deduced from the HMBC correlations (Fig. 3) observed between H-1 (δ_{H} 3.55) and C-2 (δ 37.9) and C-10 (δ 41.5) and between H-3 (δ_{H} 3.33) and C-2 (δ 37.9) and C-4 (δ 38.7), respectively. The oxymethine protons H-1 and H-3 both resonated as a doublet with large and small coupling constants of 12.0 and 4.5 Hz and 12.0 and 4.2 Hz, respectively. The magnitude of these coupling constants suggested the α axial position for both of these protons. These assignments were further supported from NOESY experiments (Fig. 3). An olefinic methine proton at δ 5.39 (H-7) showed HMBC correlations (Fig. 3) with the carbons at δ 46.7 (C-14), 48.7 (C-5), and 52.4 (C-9) and COSY cross-peak with the methylene protons H₂-6 at δ 1.97–2.04 and 2.03–2.11. The relative stereochemistry at C-13 was established by comparison of the ^{13}C NMR chemical shifts of C-12, C-13, C-14, C-15, C-16, and C-17 with those of isopimarane diterpenoids:^{30–33} isopimaradiene³⁰ (**1a**) establishing an axial position for the Me-17 and an equatorial position for the vinyl group. Compound **1** was therefore identified as isopimara-7,15-dien-1 β ,3 β -diol.

Compound **2** was also obtained as a white amorphous solid. The HREIMS showed a molecular ion $[\text{M}]^{+\bullet}$ peak at m/z 304.2403 (calcd

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