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Flavone glycosides from *Uraria crinita*

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Abstract:

A common edible herb found in all regions of Vietnam is Uraria crinita (L.) DC. (Leguminosae). It has been used as a vegetable or in folk medicine to treat lung disorders, sprains, bruises, diarrhea, and rheumatism. Pharmacological studies have shown that U. crinita possesses a range of medicinal qualities, including as antioxidant, anti-inflammatory, and osteogenic activity, as well as the ability to lessen ulcers brought on by stress. Numerous substances, including flavonoids, triterpenes, megastigmanes, and nucleosides, have been identified as this plant's active ingredients. Four flavone glycosides including apigenin 7-*O*- *β*-glucoside (1), chrysoeriol 7-*O*-*β*-glucoside (2), rhoifolin (3), and 3'-methoxyapiin (4) were isolated from the n-butanol extract of the whole plant *Uraria crinita* collected in the Phu Tho province, Vietnam. Their structures were elucidated by 1D- and 2D-NMR spectra and compared with those reported in the literature. The structures of compounds 1-4 were determined by spectroscopic methods and comparison with published data. Compounds 1-3 were obtained from the genus *Uraria* for the first time. This is the first report of these compounds from the genus *Uraria*.

Keywords: **apigenin 7-*O*-*β*-glucoside, chrysoeriol 7-*O*-*β*-glucoside, flavone glycosides, rhoifolin, *Uraria crinita*.**

Classification number: **2.2**

1. Introduction

Uraria crinita (L.) DC. (Leguminosae) is an edible herb that is widely distributed across all regions of Vietnam. It has been consumed as a vegetable or used in folk medicine for the treatment of rheumatism, diarrhoea, sprains, injuries, and lung diseases [1]. Pharmacological investigations have demonstrated that *U. crinita* has various therapeutic properties including antioxidant, anti-inflammatory, and osteogenic activities as well as reduce stress-induced ulcers [2, 3]. Several compounds including flavonoids, triterpenes, megastigmanes, and nucleosides have been reported to be active components in this plant [3-6]. Our previous studies on *U. crinita* has led us to isolate four new phenolics, (3*S*)-5,7-dihydroxy-2′,3′,4′-trimethoxy-6,5′-diprenylisoflavanone, 3,5,7,2',4'-pentahydroxyisoflavanone, 3,4-dimethoxyphenyl 1-*O*-(6′-*O*-acetyl)-*β*-D-glucopyranoside, and 3,4,5-trimethoxyphenyl 1-*O*-(6′-*O*-acetyl)-*β*-D-glucopyranoside, along with eleven known compounds [7, 8]. Here, we report the isolation and structural determination of four flavone glycosides **1-4** from the n-butanol extract of the whole plant *U. crinita*.

2. Materials and methods

2.1. General experimental procedures

Nuclear magnetic resonance (NMR) spectra were taken on a Bruker Avance III 500 spectrometer (Bruker, Fällanden, Switzerland) and electrospray ionisation mass spectrometry (ESI-MS) was conducted on an Agilent LC-MSD-Trap-SL spectrometer (Varian, USA). Infrared (IR) spectra were performed on Perkin Elmer Spectrum Two IR spectrometer (Perkin Elmer, Waltham, MA, USA). Column chromatography was carried out on silica gel 60 (0.040-0.063 mm, Merck, Darmstadt, Germany), Sephadex LH-20 (Amersham Pharmacia Biotech, Tokyo, Japan), and Rp-18 (30-50 µm, Fuji Silysia Chemical Ltd, Aichi, Japan). Thin layer chromatography was performed on silica gel 60F254 (0.25 mm, Merck, Darmstadt, Germany). The spots on the plates were observed under UV light and by spraying with a solution of vanillin/sulfuric acid and 5 min of heating.

2.2. Plant material

The whole plant *U. crinita* was collected in the Phu Tho province, Vietnam, in May 2019. The botanical identification

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was made by D.H. Thu, Institute of Ecology and Biological Resources, Vietnam Academy of Science and Technology (VAST). A voucher specimen (UC05/2019) was deposited at the Laboratory of Natural Products Research, Institute of Chemistry, VAST, Hanoi, Vietnam.

2.3. Extraction and isolation

The dried whole plant *U. crinita* (12 kg) was ground and extraction was performed three times with methanol-water (95:5, v/v) at room temperature. The extracted solution was concentrated under reduced pressure in a rotary evaporator at 45°C to remove the methanol. The obtained crude extract was suspended in water and successively partitioned with n-hexane, ethyl acetate, and n-butanol, respectively. The organic solvents were evaporated to yield extracts of 138.6, 81.2, and 117.3 g, respectively.

The n-butanol extract (115 g) was subjected to silica gel chromatography eluted with a CH_2Cl_2-MeOH-H_2O system (from 10:0:0 to 7:3:1, v/v) to give 10 fractions. Fraction 3 was repeatedly chromatographed on a silica gel column (CH_2Cl_2-MeOH-H_2O, 4:1:0.1, v/v) and then on a Sephadex LH-20 column (MeOH) to give compounds **1** (6 mg) and **2** (4 mg). Fraction 6 was purified on a silica gel column (CH_2Cl_2-MeOH-H_2O, 3:1:0.1, v/v) and then on an Rp-18 column (MeOH-H_2O, 2.5:2, v/v) to yield compound **3** (7 mg). Fraction 7 was measured using silica gel chromatography, eluted with CH_2Cl_2-MeOH-H_2O (3:1:0.1, v/v), and then on Sephadex LH-20 column (MeOH) to give compound **4** (4 mg).

Apigenin 7-*O*-*β*-glucoside (1):

Yellow solid. ^{1}H-NMR (500 MHz, DMSO-d_6): δ_H 12.96 (1H, s, 5-OH), 7.96 (2H, d, *J*=9.0 Hz, H-2′, H-6′), 6.94 (2H, d, *J*=9.0 Hz, H-3′, H-5′), 6.86 (1H, s, H-3), 6.83 (1H, d, *J*=1.5 Hz, H-8), 6.45 (1H, d, *J*=1.5 Hz, H-6), 5.39 (1H, br s, OH), 5.12 (1H, br s, OH), 5.06 (1H, d, *J*=7.0 Hz, H-1″), 5.06 (1H, br s, OH), 4.61 (1H, m, OH), 3.72 (1H, d, *J*=10.5 Hz, H-6a″), 3.49-3.17 (5H, m, H-2″-H-6b″). ^{13}C-NMR (125 MHz, CD_3OD): δ_C 182.03 (C-4), 164.25 (C-2), 162.96 (C-7), 161.35 (C-4′), 161.08 (C-5), 156.90 (C-9), 128.59 (C-2′, C-6′), 120.99 (C-1′), 115.98 (C-3′, C-5′), 105.33 (C-10), 103.11 (C-3), 99.88 (C-1″), 99.49 (C-6), 94.88 (C-8), 77.15 (C-5″), 76.44 (C-3″), 73.10 (C-2″), 69.58 (C-4″), 60.60 (C-6″). ESI-MS *m/z* 433.3 $[M + H]^+$, $C_{21}H_{20}O_{10}$. IR (KBr) ν_{max}: 3390 (O-H), 2923 (C-H), 1635 (C=O), 1600, 1510, 1462 (C=C), 1205, 1143 (C-O) cm^{-1}.

Chrysoeriol 7-*O*-*β*-glucoside (2):

Yellow solid. ^{1}H-NMR (500 MHz, DMSO-d_6): δ_H 12.96 (1H, s, 5-OH), 7.60 (1H, dd, *J*=8.5, 1.5 Hz, H-6′), 7.59 (1H, d, *J*=1.5 Hz, H-2′), 6.98 (1H, s, H-3), 6.95 (1H, d, *J*=8.5 Hz, H-5′), 6.86 (1H, d, *J*=2.0 Hz, H-8), 6.45 (1H, d, *J*=2.0 Hz, H-6), 5.39 (1H, br s, OH), 5.12 (1H, br s, OH), 5.06 (1H, d, *J*=7.0 Hz, H-1″), 5.06 (1H, br s, OH), 4.61 (1H, m, OH), 3.89 (3H, s, 3′-OCH_3), 3.72 (1H, d, *J*=10.5 Hz, H-6a″), 3.49-3.17 (5H, m, H-2″-H-6b″). ^{13}C-NMR (125 MHz, CD_3OD): δ_C 182.03 (C-4), 164.14 (C-2), 162.96 (C-7), 161.08 (C-5), 156.90 (C-9), 150.93 (C-4′), 148.04 (C-3′), 121.31 (C-1′), 120.49 (C-6′), 115.76 (C-5′), 110.30 (C-2′), 105.33 (C-10), 103.43 (C-3), 99.99 (C-1″), 99.49 (C-6), 95.01 (C-8), 77.23 (C-5″), 76.44 (C-3″), 73.10 (C-2″), 69.58 (C-4″), 60.60 (C-6″), 55.96 (3′-OCH_3). ESI-MS *m/z* 463.4 $[M + H]^+$, $C_{22}H_{22}O_{11}$. IR (KBr) ν_{max}: 3405 (O-H), 2906 (C-H), 1648 (C=O), 1612, 1506, 1453 (C=C), 1185, 1126, 1087 (C-O) cm^{-1}.

Rhoifolin (3):

White solid. ^{1}H-NMR (500 MHz, CD_3OD): δ_H 7.90 (2H, d, *J*=9.0 Hz, H-2′, H-6′), 6.96 (2H, d, *J*=9.0 Hz, H-3′, H-5′), 6.81 (1H, d, *J* = 2.0 Hz, H-8), 6.68 (1H, s, H-3), 6.48 (1H, d, *J*=2.0 Hz, H-6), 5.31 (1H, d, *J*=2.0 Hz, H-1‴), 5.22 (1H, d, *J*=8.0 Hz, H-1″), 3.97-3.41 (C<u>H</u>-OH, C<u>H$_2$</u>-OH sugar), 1.35 (1H, d, *J*=6.0 Hz, H-6‴). ^{13}C-NMR (125 MHz, CD_3OD): δ_C 184.06 (C-4), 166.82 (C-2), 164.43 (C-7), 162.96 (C-4′, C-5), 159.01 (C-9), 129.65 (C-2′, C-6′), 123.07 (C-1′), 117.08 (C-3′, C-5′), 107.14 (C-10), 104.15 (C-3), 102.54 (C-1‴), 101.08 (C-6), 99.94 (C-1″), 95.95 (C-8), 79.11 (C-2″), 79.02 (C-5″), 78.36 (C-3″), 73.96 (C-4‴), 72.22 (C-4″, C-3‴), 71.41 (C-2‴), 70.02 (C-5‴), 62.44 (C-6″), 18.23 (C-6‴). ESI-MS *m/z* 579.2 $[M + H]^+$, $C_{27}H_{30}O_{14}$. IR (KBr) ν_{max}: 3458 (O-H), 2908 (C-H), 1653(C=O), 1600, 1452 (C=C), 1210, 1148 (C-O) cm^{-1}.

3′-Methoxyapiin (4):

Yellow solid. ^{1}H-NMR (500 MHz, CD_3OD): δ_H 7.56 (1H, d, *J*=8.5 Hz, H-6′), 7.52 (1H, s, H-2′), 6.95 (1H, d, *J*=8.5, H-5′), 6.83 (1H, brs, H-8), 6.69 (1H, H-3), 6.48 (1H, d, *J*=1.5 Hz, H-6), 5.48 (1H, d, *J*=1.5 Hz, H-1‴), 5.17 (1H, d, *J*=7.5 Hz, H-1″), 4.07-3.41 (C<u>H</u>-OH, C<u>H$_2$</u>-OH sugar). ^{13}C-NMR (125 MHz, CD_3OD): δ_C 184.05 (C-4), 166.67 (C-2), 164.66 (C-7), 162.91 (C-5), 158.98 (C-9), 152.36 (C-4′), 149.55 (C-3′), 123.51 (C-1′), 122.00 (C-6′), 116.83 (C-5′), 110.94 (C-1‴), 110.83 (C-2′), 107.11 (C-10), 104.50 (C-3), 101.07 (C-6), 100.29 (C-1″), 96.11 (C-8), 80.71 (C-3‴), 78.82 (C-2″), 78.46 (C-3″), 78.36 (C-5″), 78.19 (C-2‴), 75.45 (C-4‴), 71.34 (C-4″), 65.91 (C-5‴), 62.49 (C-6″), 56.75 (O<u>CH$_3$</u>). ESI-MS *m/z* 595.4 $[M + H]^+$, $C_{27}H_{30}O_{15}$. IR (KBr) ν_{max}: 3445 (O-H), 2895 (C-H), 1640 (C=O), 1585, 1426 (C=C), 1175, 1050 (C-O) cm^{-1}.

3. Results and discussion

Compound **1** was assigned to have a molecular formula of $C_{21}H_{20}O_{10}$ by a combination of its NMR data and ESI-MS *pseudo*-molecular ion peak at *m*/z 433.3 $[M+H]^+$. The

presence of intramolecularly hydrogen-bonded proton signal [δ_H 12.96 (1H, s)] suggested that this hydroxyl group was at C-5. The ^{1}H-NMR spectrum showed signals due to a *para*-disubstituted B-ring [δ_H 7.96 (2H, d, *J*=9.0 Hz, H-2′, H-6′), 6.94 (2H, d, *J*=9.0 Hz, H-3′, H-5′)], a *meta*-disubstituted A-ring [δ_H 6.83 (1H, d, *J*=1.5 Hz, H-8), 6.45 (1H, d, *J*=1.5 Hz, H-6)], and a one-proton singlet of the C-ring [δ_H 6.86 (1H, s, H-3)] on the flavone skeleton. In addition, signals of a glucopyranosyl unit [δ_H 5.06 (1H, d, *J*=7.0 Hz, H-1″), and 3.72-3.17 (6H, m, H-2″, H-3″, H-4″, H-5″, H-6″)] were also observed. The large coupling constant (*J*=7.0 Hz) of the anomeric proton indicated the *β*-configuration of the glucopyranosyl moiety [9]. The ^{13}C-NMR and heteronuclear single quantum coherence (HSQC) spectra revealed 15 carbon signals due to a flavone skeleton comprising one carbonyl carbon, seven sp^2 methine, and seven sp^2 quaternary carbons; in addition to six carbon signals due to a glucopyranosyl moiety. The heteronuclear multiple bond correlation (HMBC) from the anomeric proton H-1″ (δ_H 5.06) and two protons H-6 (δ_H 6.45) and H-8 (δ_H 6.83) to the carbon of A-ring at δ_C 162.96 confirmed that the glucosyl unit was at C-7. From the above spectral data, the structure of **1** was determined to be apigenin 7-*O*-*β*-glucoside. The ^{13}C-NMR data of **1** were in good agreement with those of apigenin 7-*O*-*β*-glucoside in the literature [10].

Compound **2** was isolated as a yellow solid. Its molecular formula was established as $C_{22}H_{22}O_{11}$ on the basis of an ion peak $[M + H]^+$ at *m/z* 463.4 in ESI-MS. ^{1}H- and ^{13}C-NMR spectral data of compound **2** were similar with those of **1** except for the presence of three aromatic protons of an ABX spin system of ring B at δ_H 7.60 (1H, dd, *J*=8.5, 1.5 Hz, H-6′), 7.59 (1H, d, *J*=1.5 Hz, H-2′), and 6.95 (1H, d, *J*=8.5 Hz, H-5′); and an additional methoxyl group at δ_H 3.89 (3H, s). The position of the methoxyl group was at C-3′ based on the nuclear Overhauser effect spectroscopy (NOESY) correlations of H-2′ (δ_H 7.59) with 3′-OCH$_3$ (δ_H 3.89) and the HMBC correlations between the protons of 3′-OCH$_3$ (δ_H 3.89), H-2′ (δ_H 7.59), H-6′ (δ_H 7.60), and carbon C-3′ (δ_C 148.04). Moreover, the carbon chemical shifts of C-1′-C-6′ in **2** were very similar to those reported for chrysoeriol [11]. On the basis of the above evidence, the structure of **2** was elucidated to be chrysoeriol 7-*O*-*β*-glucoside [12].

The ^{13}C-NMR and HSQC spectra of **3** confirmed the presence of 27 carbons that were attributed to 15 carbons of a flavone skeleton and 12 carbons of two sugar residues. The ^{1}H-NMR spectrum showed resonance for two *meta*-coupled aromatic protons at δ_H 6.81 (1H, d, *J*=2.0 Hz, H-8) and 6.48 (1H, d, *J*=2.0 Hz, H-6); four *ortho*-coupled aromatic protons of an AA′BB′ spin system at δ_H 7.90 (2H, d, *J*=9.0 Hz, H-2′, H-6′) and 6.96 (2H, d, *J*=9.0 Hz, H-3′, H-5′); and a one-proton singlet at δ_H 6.68 (1H, s, H-3). Thus, the flavonoid moiety of **3** was determined as apigenin. In addition, a series of sugar signals at *δ* 3.97-3.41, along with two signals at δ_H 5.31 (1H, d, *J*=2.0 Hz, H-1‴) and 5.22 (1H, d, *J*=8.0 Hz, H-1″) corresponding to anomeric protons of two sugar residues, were also observed in the ^{1}H-NMR spectrum. The coupling constants (*J*=8.0 Hz and 2.0 Hz) of the anomeric protons indicated the *β*- and *α*-configurations for glucopyranosyl and rhamnopyranosyl, respectively [9, 13]. The HMBC correlation from the anomeric proton H-1″ of glucose to C-7 (δ_C 164.43), H-1‴ (δ_H 5.31) with the carbon C-2″ (δ_C 79.11), and H-2″ (δ_H 3.74) with the carbon C-1‴ (δ_C 102.54) indicated 7-*O*-glycosidation and the interglycosidic linkage in **3** as -*O*-*α*-rhamnopyranosyl(1→2)-*O*-*β*-glucopyranoside. On the basis of the above evidence, the structure of **3** was assigned to be rhoifolin [14].

Compound **4** was obtained as a yellow solid. Its molecular formula was established as $C_{27}H_{30}O_{15}$ on the basis of an ion peak $[M+H]^+$ at *m/z* 595.4 in ESI-MS. The ^{1}H- and ^{13}C-NMR spectra of **4** were similar to those of **2** except for the appearance of an additional *α*-arabinofuranosyl moiety [δ_H 5.48 (1H, d, *J*=1.5 Hz, H-1‴); δ_C 110.94 (C-1‴), 80.71 (C-3‴), 78.19 (C-2‴), 75.45 (C-4‴), 65.91 (C-5‴)]. The *α*-arabinofuranosyl moiety was attached to C-2″ of glucopyranosyl based on the HMBC correlations between the anomeric proton H-1‴ (δ_H 5.48) and carbon C-2″ (δ_C 78.82) and between proton H-2″ (δ_H 3.69) and carbon C-1‴ (δ_C 110.94). Based on the evidence above and in comparison with those reported in the literature [15], compound **4** was determined to be 3′-methoxyapiin (Fig. 1).

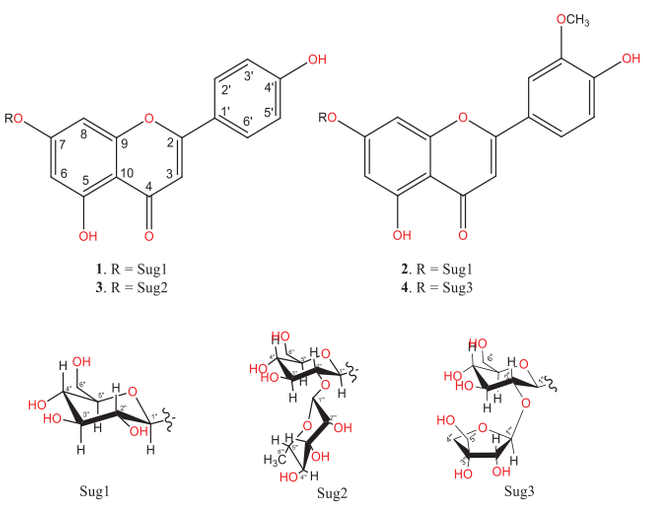

Fig. 1. Structure of compounds 1-4.

4. Conclusions

From the n-butanol extract of the whole plant *U. crinita*, using column chromatography, four flavone glycosides including apigenin 7-*O*-β-glucoside (**1**), chrysoeriol 7-*O*-β-glucoside (**2**), rhoifolin (**3**), and 3′-methoxyapiin (**4**) were isolated. The structures of compounds **1-4** were determined by spectroscopic methods and comparison with published data. This is the first report of these compounds from the genus *Uraria*.

CRediT author statement

Tran Duc Dai: Conceptualisation, Methodology, Software, Resources, Writing - Review and Editing; Dao Duc Thien: Data curation, Software, Writing - Original draft preparation; Nguyen Thanh Tam: Supervision, Software, Validation.

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COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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