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CHEMICAL CONSTITUENTS FROM THE LEAVES OF *TERMINALIA CATAPPA* L. (COMBRETACEAE)

Ho Dac Hung*, Doan Duy Tien, Nguyen Thi Ngoan, Ba Thi Duong, Do Quoc Viet, Pham Gia Dien, Bui Kim Anh*

Institute of Chemistry, Vietnam Academy of Science and Technology, 18 Hoang Quoc Viet., Cau Giay, Ha Noi, Viet Nam

*Email: *hungvhh@gmail.com, anhvhh@gmail.com*

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Abstract. *Terminalia catappa* L. (cây Bàng) belongs to the family Bàng (Combretaceae), is a large tree species living mainly in the tropics. The phytochemical studies of this plant have only focused on the volatile fraction and tannins. The main components in the plant include terpenes, flavonoids, tannins, saponins and phytosterols. *T. catappa* species has been recognized as an important medicinal plant. In traditional medicine, *T. catappa* leaf, bark and fruit are used in treating dysentery, rheumatism, cough and asthma. The fruit is also helpful in the treatment of leprosy and headaches and the leaves are specifically used in getting rid of intestinal parasites, treatment of eye problems, wounds, and liver problems. Due to its rich medicinal ingredients, the leaves and bark are used in various herbal medicines for different purposes. This study aims to characterize the phyto-chemical constituents present in the leaf extracts of *Terminalia catappa* using chromatographic and spectroscopicmethods. Four flavonoids, tectochrysin (**1**), luteolin (**2**), kaempferol 3,7,4′-trimethyl ether (**4**), and kaempferol (**5**), one phenolic, gallic acid (**3**), were isolated from the ethylacetate fraction of the leaves of *Terminalia catappa* L.. Their structures were elucidated on the basis of spectroscopic analysis and comparisons with known related compounds. Compounds **1** and **4** were reported for the first time from the leaves of *Terminalia catappa*.

Keywords: Terminalia catappa, Combretaceae, tectochrysin, luteolin, kaempferol 3,7,4′-trimethyl ether.

Classification numbers: 1.1.1, 1.3.1, 1.5.2.

1. INTRODUCTION

Terminalia genus (Combretaceae) comprises about 250 species, which are widely distributed in tropical and subtropical regions of Asia, Australia and Africa [1]. *Terminalia catappa* L. is a tall deciduous and erect tree reaching 15 - 25 m, trunk 1 - 1.5 m in diameter, wide- spreading branches and a broad disk-shaped crown, bark grey-brown, rough with age. Leaves alternate obovate with short petioles, spirally clustered at the branch tips, 15 - 36 cm long, 8 - 24 cm wide, dark green above, paler beneath. Before falling, the leaves turn into red color or brown -yellow, due to pigments such as violaxanthin, lutein, and zeaxanthin. The tree usually sheds all its leaves twice a year in January-February and August [2]. The leaves contain

flavonoids, tannins, saponins and phytosterols [3, 4]. The leaves and the barks are used in herbal medicines for treatement of liver diseases [3]. Pharmacological studies proved that extracts and compounds from *T. catappa* exhibited a broad range of biological properties such as anti-inflammatory, anti-viral, anti-oxidant, and anti-diabetic activities [5, 6, 7]. In the folk medicine, the leaves or barks of *Terminalia* species are used for the treatment of skin diseases, and hepatitis. However, there have been no reports on chemical constituents of the leaves of *Terminalia catappa* L. in Viet Nam. In this paper, we reported the isolation and structural elucidation of four flavonoids, tectochrysin (**1**), luteolin (**2**), kaempferol 3,7,4′-trimethyl ether (**4**), and kaempferol (**5**), one phenolic, gallic acid (**3**), and three sterols, *β*-sitosterol (**6**), stigmasterol (**7**), and daucosterol (**8**) from the leaves of *T. catappa*.

2. MATERIALS AND METHODS

2.1. Instruments and chemicals

Nuclear magnetic resonance (NMR) spectra were recorded on a Bruker AM 500 FT-NMR spectrometer using tetramethylsilane as an internal standard. Mass spectra (ESI-MS) were obtained from an AGILENT 1100 LC-MSD Trap spectrometer. Melting points were measured on a Mikroskopheiztisch PHMK-50, VEB Waegetechnik Rapido, Germany. FT-IR spectra were recorded on an IMPACT-410FT-IR spectrometer (CARL ZEISS JENA). Thin layer chromatography (TLC) was conducted using silica gel 60 F_{254} (Merck). Column chromatography (CC) was performed using silica gel 60 (40 - 63 μm, Merck). Gel permeation chromatography was conducted, using Sephadex LH-20 in methanol. Organic solvents were pure for analysis or redistilled.

2.2. Plant materials

The leaves of *Terminalia catappa* L. were collected in Hai Ba Trung District, Ha Noi, Viet Nam in January 2019 and identified by Mrs. Nguyen Kim Dao, Institute of Ecology and Biological Resources, *Vietnam Academy of Science and Technology*. A voucher specimen (TC 01.19) was deposited at the Institute of Chemistry, *Vietnam Academy of Science and Technology*.

2.3. Extraction and isolation

The dried leaves of *T. catappa* (2.35 kg) were ground and extracted with methanol at room temperature (4 times, 1 day/time). The combined extracts were evaporated under reduced pressure at 45 ^{o}C to obtain 450 g of MeOH extract. The MeOH extract was suspended in H_2O and then fractionated into n-hexane, ethyl acetate and n-butanol (400 mL each), successively. The organic solvents were evaporated to yield the corresponding fractions of n-hexane (42 g), ethyl acetate (118 g) and n-butanol (62.5 g).

The ethyl acetate fraction (65 g) was subjected to a silica gel column (column diameter 5 cm, high 70 cm and 1 kg silica gel) with gradient solvents of $CHCl_3$-MeOH (100 % $CHCl_3$, 9:1, 8:2, 7:3, 6:4, 5:5, 4:6, 3:7, 2:8 and 1:9) to give 8 fractions. Fraction 2 (2.0 g) was purified on a silica gel column ($CHCl_3$-MeOH, 27:3, v/v) to yield compound **1** (28 mg). Fraction 4 (2.0 g) was purified on a silica gel column ($CHCl_3$-MeOH, 14:6, v/v) and then on a sephadex LH-20 column (MeOH as eluent solvent) to give compound **4** (12 mg). By repeated column chromatography on a silica gel ($CHCl_3$-MeOH, 3:7, v/v) and sephadex LH-20 (MeOH as eluent solvent) together

with recrystallization, compound **2** (15 mg) and compound **5** (18 mg) were obtained from fraction 5 (2.5 g). Fraction 1 (850 mg) was repeatedly chromatographed on a silica gel column ($CHCl_3$-MeOH, 99:1, v/v) to afford compounds **6** (15 mg) and **7** (11 mg). Fraction 6 (1.5 g) was repeatedly chromatographed on a silica gel column (CH_2Cl_2-MeOH-H_2O, 8:2:0.1, v/v) to afford compound **3** (18 mg). Fraction 7 (1 g) was chromatographed on a silica gel column (CH_2Cl_2-MeOH, 6:4, v/v) to afford compound **8** (18 mg).

Tectochrysin (**1**): Light yellow crystals, mp. 164 - 165 ^oC. IR υ_{max} (KBr, cm^{-1}): 3441 (OH), 1640 (C=O), 1615, 1595 (Ar). ESI-MS *m/z*: 269 $[M+H]^+$ ($C_{16}H_{12}O_4$).

^{1}H-NMR (500 MHz, $CDCl_3$), δ_H (ppm): 3.87 (3H, s, 7-OCH_3), 6.37 (1H, d, *J* = 2.0 Hz, H-8), 6.49 (1H, d, *J* = 2.0 Hz, H-6), 6.65 (1H, s, H-3), 7.54 (3H, m, H-3′, H-4′ and H-5′), 7.88 (2H, dd, *J* = 2.0 Hz and 7.0 Hz, H-2′ and H-6′), 12.71 (1H, s, 5-OH).

^{13}C-NMR (125 MHz, $CDCl_3$), δ_C (ppm): 182.4 (C-4), 165.6 (C-9), 163.9 (C-7), 162.2 (C-5), 157.8 (C-2), 131.8 (C-1′), 131.3 (C-4′), 129.0 (C-3′/C-5′), 126.2 (C-2′/C-6′), 105.8 (C-10), 105.7 (C-3), 98.2 (C-8), 92.6 (C-6), 55.8 (7-OCH_3).

Luteolin (**2**): Yellow crystal. IR υ_{max} (KBr, cm^{-1}): 3420 (OH); 1652 (carbonyl), 1265 and 1164 (C-O-C). ESI-MS *m/z*: 285 $[M-H]^-$ ($C_{15}H_{10}O_6$).

^{1}H-NMR (500 MHz, CD_3OD), δ_H (ppm): 6.21 (1H, d, *J* = 1.8 Hz, H-6), 6.43 (1H, d, *J* = 1.8 Hz, H-8), 6.54 (1H, s, H-3), 6.91 (1H, d, *J* = 8.2 Hz, H-5′), 7.38 (1H, d, *J* = 2.0 Hz, H-2′), 7.39 (1H, dd, *J* = 8.2 Hz, 2.0 Hz, H-6′).

^{13}C-NMR (125 MHz, CD_3OD), δ_C (ppm): 183.7 (C-4), 166.2 (C-7), 166.1 (C-2), 163.2 (C-5), 159.4 (C-9), 151.0 (C-4′), 120.3 (C-6′), 147.0 (C-3′), 123.7 (C-1′), 116.8 (C-5′), 114.1 (C-2′), 105.3 (C-10), 103.7 (C-3), 100.1 (C-6), 95.0 (C-8).

Gallic acid (**3**): White solid, mp 248 - 250 ^oC. ESI-MS *m/z*: 169 $[M-H]^-$ ($C_7H_6O_5$).

^{1}H-NMR (DMSO-d_6), δ_H (ppm): 7.12 (2H, s, H-2, 6).

^{13}C-NMR (125 MHz- CD_3OD), δ_C (ppm): 121.9 (C-1), 108.5 (C-2, 6), 146.2 (C-3, 5), 138.5 (C-4), 170.1 (-COOH).

Kaempferol 3, 7, 4′-trimethyl ether (**4**): Light yellow crystal, mp. 144 - 145 ^0C. ESI-MS *m/z*: 329 $[M-H]^-$($C_{18}H_{16}O_6$).

^{1}H-NMR ($CDCl_3$), δ_H (ppm): 3.84 (3H, s, 3-OCH_3), 3.86 (3H, s, 4′-OCH_3), 3.88 (3H, s, 7-OCH_3), 6.34 (1H, d, *J* = 2.0 Hz, H-6), 6.44 (1H, d, *J* = 2.0 Hz, H-8), 7.01 (2H, d, J= 9.0 Hz, H-3′ and H-5′), 8.06 (2H, d, *J* = 9.0 Hz, H-2′ and H-6′), 12.63 (1H, s, 5-OH-5).

^{13}C-NMR ($CDCl_3$), δ_C (ppm): 55.3 (7-OCH_3), 55.9 (4′-OCH_3), 59.9 (3-OCH_3), 92.1 (C-8), 97.7 (C-6), 106,1 (C-10), 114.0 (C-3′ and C-5′), 122.8 (C-1′), 130.1 (C-2′ and C-6′), 138.9 (C-3), 156.0 (C-2), 156.7 (C-9), 161.7 (C-5), 162.0 (C-4′), 165.4 (C-7) 178.8 (C-4).

Kaempferol (**5**): Light yellow solids, mp. 275 - 277 ^0C, ESI-MS *m/z*: 285 $[M-H]^-$ ($C_{15}H_{10}O_6$), UV (CH_3OH) λ_{max} nm: 310, 380; IR (KBr) υ_{max} cm^{-1}: 3324 (free OH), 1660 (C=O) 1614 (C=C aromatic).

^{1}H-NMR (500 MHz, CD_3OD), δ_H (ppm): 8.07 (2H, dd, *J* = 8.9, 2.0 Hz, H-2′, H-6′), 6.89 (2H, dd, *J* = 8.9, 2.0 Hz, H-3′, H-5′), 6.38 (1H, d, *J* = 2.0 Hz, H-8), 6.18 (1H, d, *J* = 2.0 Hz, H-6).

^{13}C-NMR (125MHz , CD_3OD), δ_C (ppm): 175.9 (C=O), 164.1 (C-7), 161.1 (C-5), 159.1 (C- 4′), 156.3 (C-9), 147.6 (C-2), 136.7 (C-3), 129.8 (C-2′, C-6′), 122.7 (C-1′), 115.8 (C-3′, C-5′), 104.4 (C- 10), 98.9 (C-6), 93.1 (C-8).

3. RESULTS AND DISCUSSION

Compound **1** was isolated as a light yellow crystal, mp. 164 - 165 ^{o}C. The ESI-MS spectrum showed a molecular ion peak at *m/z* 269 $[M+H]^+$ corresponding to the molecular formula of $C_{16}H_{12}O_4$. The ^{1}H-NMR of **1** showed two doublet protons at δ_H 6.49 (1H, *J* = 2.0 Hz, H-6) and 6.37 (1H, *J* = 2.0 Hz, H-8) located at *meta* positions. A singlet at δ_H 6.65 was assigned to H-3 while two multiplets signals at δ_H 7.54 (3H, *m*, H-3′, H-4′, H-5′) and δ_H 7.88 (2H, *J* = 2.0 Hz and 7.0 Hz, H-2′, H-6′) belonged to protons at monosubstituted ring B. A singlet indicated as a chelated OH was observed at δ_H 12.71 (1H, *s*, 5-OH) at a very downfield region due to the formation of hydrogen bond between protons from the hydroxyl group with the carbonyl group (C=O) in the heterocyclic ring C. A singlet at δ_H 3.97 was due to the presence of a methoxy group. The ^{13}C-NMR spectrum showed a signal of methoxy carbon at δ_C 55.8 and six aromatic methine carbons at δ_C 105.7, 98.2, 92.6, 126.2, 129.0 and 131.3. A signal at a very low region (δ_C 182.4) is definitive of carbonyl carbon of the flavone structure [8]. Four oxygenated carbons were observed at δ_C 163.9, 165.6, 165.9, and 165.2 and two quaternary carbons gave signals at δ_C 131.4 and δ_C 103.1. The HMBC correlation between δ_H 3.91 and δ_C 163.9 confirmed the attachment of the methoxy group to C-7. Comparison of the available value with the literature [9] confirmed that compound **1** was tectochrysin. This compound was reported from *Pranus cerasus* [10].

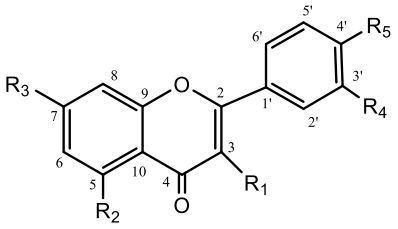
Compound **2** was isolated as a yellow crystal. The ESI-MS spectrum showed a molecular ion peak at *m/z* 285 $[M-H]^-$ corresponding to the molecular formula of $C_{15}H_{10}O_6$. The skeleton of **2** was confirmed of three ABX-type aromatic protons at δ_H 7.38 (1H, d, *J* = 2.0 Hz, H-2′), 7.39 (1H, dd, *J* = 8.2, 2.0 Hz, H-6′) and 6.91 (1H, d, *J* = 8.2 Hz, H-5′) in ^{1}H-NMR. The ^{13}C-NMR spectrum of **2** showed fifteen carbons including one carbon carbonyl at δ_C 183.7 (C-4) and the others from δ_C 95.0 to 166.2. Thus, the structure of **2** was identified as luteolin by comparing these data with those previously reported [10]. Luteolin has been reported to possess antioxidant, anti-inflammatory, anticancer, cardio-and antidiabetes activities [11].

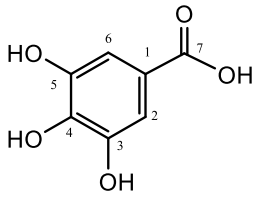
Compound **3** was isolated as a white amorphous solid. The ESI-MS spectrum showed a molecular ion peak at *m/z* 169 $[M-H]^-$. Compound **3** was identified as gallic acid, which was deduced from its singlet proton signal at δ_H 7.12 (2H, s, galloyl-H) and three broad hydroxyl signals at 12.2 (1H, s, -COOH), 9.21 (2H, s, -OH) and 8.86 (1H, s, -OH) in the ^{1}H-NMR spectra, seven carbon signals in the low field (δ_C 170.1 (-COOH), 146.8 (C-4), 139.4 (C-3,5), 121.9 (C-1), 110.2 (C-2,6)) in the ^{13}C-NMR spectrum, and further confirmed by comparison with the literature [12].

Compound **4** was obtained as a yellow crystal. The ^{1}H-NMR spectrum of **4** showed the presence of three methoxy groups at δ_H 3.84 (3H, s), 3.86 (3H, s), and 3.88 (3H, s) and one hydroxyl group at δ_H 12.63. Furthermore, a *meta* coupling system of two aromatic protons at δ_H 6.34 (1H, d, *J* = 2.0 Hz) and 6.44 (1H, d, *J* = 2.0 Hz), and an AB coupling system of four protons in a *para*-substituted benzene ring at δ_H 7.01 (2H, d, *J* = 9.0 Hz) and 8.06 (2H, d, *J* = 9.0 Hz). The ^{13}C-NMR spectrum showed signals of 14 aromatic carbons and one carbonyl group at δ_C 178.8 that was clearly approved of flavonol skeleton for **4**. Three carbons of methoxy groups were resonated at δ_C 60.1, 55.8 and 55.4. The signal of the chelated proton at δ_H 12.63 indicated that C-5 was linked to a hydroxyl group and **4** could be kaempferol 3,7,4′-trimethyl ether. The obtained results for compound **4** were in agreement with the spectral data of kaempferol 3,7,4′-trimethyl ether previously reported in the literature [13]. Thus, compound **4** was identified as kaempferol 3,7,4′-trimethyl ether, which was isolated from *T. catappa* for the first time.

Compound **5** was isolated as a yellow solid. Its mass spectrum showed a molecular ion peak at *m/z* 285 $[M-H]^-$ which corresponds to the molecular formula $C_{15}H_{10}O_6$. The ^{1}H-NMR spectrum showed proton signals of a typical flavonol-type ABX system (δ_H 6 - 8 ppm): In the B-ring system, proton signals appeared as a pair of doublet of doublets at δ_H 6.89 (2H, dd, J = 7.0 Hz, 2.0 Hz, H-3′, H-5′) at δ_H 8.07 (2H, dd, J = 7.0 Hz, 2.0 Hz, H-2′, H-6′). For the A-ring protons, two doublet peaks observed at δ_H 6.18 (1H, d, J = 2.0 Hz) and 6.38 (1H, d, J = 2.0 Hz) were assigned as H-6 and H-8, respectively. Both of these protons were meta-coupled as indicated by small coupling constant, J = 2.0 Hz. The ^{13}C-NMR spectrum exhibited signals of 15 carbons. A downfield signal at δ_C 175.9 was attributed to carbonyl C-4. In addition, four oxygenated aromatic carbons observed at δ_C 136.7, 159.1, 161.1, and 164.1 were attributed to C-3, C-4′, C-5, and C-7, respectively. Two quaternary carbons were observed at δ_C 104.4 (C-10), and 122.7 (C-1′) and two oxygenated aromatic carbons at δ_C 147.6, and 156.3 were observed. The remaining carbon signals observed at δ_C 93.1 (C-8), 98.9 (C-6), 115.8 (C-3′, C-5′), and 129.8 (C-2′, C-6′) were assigned to methine carbons. On the basis of spectral data and comparison with the literature [12], compound **5** was identified as kaempferol. This compound was found in *Terminalia* species [14].

The structures of compounds **6**, **7**, and **8** were identified as *β*-sitosterol, stigmasterol, and daucosterol by comparing physicochemical and spectroscopic data [15, 16].





1. tectochrysin, R_1= R_4= R_5=H, R_2= OH, R_3=OCH_3

2. luteolin, R_1=H, R_2= R_3= R_4=R_5=OH

4. kaempferol 3, 7, 4'- trimethyl ether, R_1=R_3=R_5=OCH_3, R_2= OH, R_4=H

5. kaempferol, R_1= R_2= R_3=R_5=OH, R_4= H

3. gallic acid

The structures of isolated compounds (**1**-**5**) from the leaves of *Terminalia catappa* L.

4. CONCLUSIONS

From the ethyl acetate extract of the leaves of *Terminalia catappa* L., eight compounds were isolated and indentified as tectochrysin (**1**), luteolin (**2**), gallic acid (**3**), kaempferol 3,7,4′-trimethyl ether (**4**), kaempferol (**5**). In which, tectochrysin (**1**) and kaempferol 3,7,4′-trimethyl ether (**4**) were isolated from the leaves of *Terminalia catappa* L. for the first time.

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CRediT authorship contribution statement. This study was conceived and designed by B.K.A and H.D.H., B.K.A., DDT and P.G.D. contributed reagents/materials/analysis tools. The experiments were conducted by H.D.H., N.T.N., B.T.D., D.Q.V., B.K.A., DDT and H.D.H. analyzed the data. The manuscript was drafted by H.D.H., and B.K.A. B.K.A. finalized the manuscript. All authors have read and agreed to the published version of the manuscript.

Declaration of competing interest. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

REFERENCES

1. Cock I. E. - The medicinal properties and phytochemistry of plants of the genus *Terminalia* (Combretaceae), Inflammo Pharmacology **23** (5) (2015) 203-229.
2. Arumugam Vijaya Anand, Natarajan Divya1, Pannerselvam Punniya Kotti. - An updated review of *Terminalia catappa*, Pharmacogn Rev. **9** (18) (2015) 93-98.
3. Fan Y. M., Xu L. Z., Gao J., Wang Y., Tang X. H., Zhao X. N., Zhang Z. X. - Phytochemical and anti inflammatory studies on *Terminalia catappa*, J. Ethnopharmacol **75** (3-4) (2004) 253-60.
4. Jain S., Yadav P. P., Gill V., Vasudeva N., Singla N. - *Terminalia arjuna* a sacred medicinal plant: phytochemical and pharmacological profile, Phytochemistry Reviews **8** (2009) 491.
5. Hanaa Mohamed El-Rafie and Manal Abdel-Aziz Hamed - Antioxidant and anti-inflammatory activities of silver nanoparticles biosynthesized from aqueous leaves extracts of four *Terminalia* species, Adv. Nat. Sci. Nanosci. Nanotechnol **5** (2014) 11.
6. Divya N., Vijaya Anand A. - Phytochemical investigation and *in vitro* anti-diabetic activity of *Terminalia catappa* leaves, Int J. of Phyto Pharm. **4** (2014) 132-4.
7. Lin C. C., Chen J. M., Lin J. M., Ujiie T. - Evaluation of the Antioxidant and Hepatoprotective Activity of *Terminalia catappa*, Am. J. Chin. Med. **25** (1997) 153-61.
8. Nagarajan G. R., Parmar V. S. - Flavonoids from *Pranus cerasus*, Planta Medica **32** (1977) 50-53.
9. Nesma M. Salah, Ahmed M. A. Souleman, Kamel H. Shaker, Seham El Hawary, Zeinab A. El-Shahid, Faten K. Abd El-Hady - Acetylcholinesterase, Alpha-Glucosidase and Tyrosinase Inhibitors from Egyptian Propolis, IJPPR **9** (4) (2017) 528-536.
10. McNulty J., Nair J. J., Bollareddy E., Keskar K., Thorat A., Crankshaw D. J., Holloway A. C., Khan G., Wright G. D., Ejim L. - Isolation of flavonoids from the heartwood and resin of *Pranus avium* and some preliminary biological investigations, Phytochemistry **70** (2009) 2040 -2046.
11. Mohamed Ali A. Alwahsh, Melati Khairuddean, and Wong Keng Chong - Chemical Constituents and Antioxidant Activity of *Teucrium barbeyanum* Aschers, Rec. Nat. Prod. **9** (1) (2015) 159-163.
12. Jie yin, Chuan-ling Si, and Myeong- Hyeon Wang - Antioxidant activity of flavonoids and glucosids from *Sonchus oleraceus* L., J. Appl. Bio. Chem. **51** (2) (2008) 57-60.
13. Eldahshan O. A. - Isolation and Structure Elucidation of Phenolic Compounds of Carob Leaves Grown in Egypt, Current Research Journal of Biological Sciences **3** (1) (2011) 52-55.
14. Mohale D. S., Dewani A. P., Chandewar A. V., Khadse C. D., Tripathi A. S., Agrawal S. S. - Brief review on medicinal potential of Terminalia catappa, Journal of Herbal Medicine and Toxicology **3** (1) (2009) 7-11.
15. Çitoglu G., Tanker M., and Seve B. - Flavonoid aglycones from *Ballota saxatilis* subsp. Saxatilis, Pharmaceutical Biology **37** (2) (1999) 158-160.
16. Khatun M., Billah M., and Quader M. D. A. - Sterols and Sterol Glucoside from *Phyllanthus* Species, Dhaka Univ. J. Sci. **60** (1) (2012) 5-10.