NATURAL FLAVONOID C-GLYCOSIDES

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The paper gives a review of flavonoid C-glycodes.

C-Glycosides is the name given compounds in which an aglycone is directly attached to the first carbon atom of a pyranose or furanose ring of a carbohydrate. However, C-glycosides cannot be considered as natural carbohydrates and it is more correct to regard them as anhydropolyols [1].

One of the first known natural C-glycosides — barbaloin — was isolated in 1851 from species of aloe. It is the 9-C-β-D-glucopyranoside of aloe-emodin, and its structure has been shown by independent synthesis [2]. Later, another anthrone C-glycoside — homonataloin (9-C-D-glucopyranosyl-4,6-dihydroxy-5-methoxy-2-methylanthrone) — was discovered in species of aloe, 4- or 5-O-glycosides of barbaloin in *Rhamnus purschiana*, and 10-C-D-glucosyl-1,8,10-trihydroxy-3-methyl-9(10)-anthracenome (cassialoin) in *Cassia garrettiana* [3, 4].

Carminic acid, which occurs in *Dactylopsis coccus* Costa, is an anthraquinone C-glycoside. Its structure has also been shown by independent synthesis and by NMR spectroscopy [2, 5]. An isocoumarin C-glycoside, bergenin, and a C-glycosidic derivative of pyrimidine, pseudouridine (5-C-β-D-ribofuranosyluracil), are also known [2]. Unfortunately, there is no more complete information on the C-glycosides of anthrone, anthraquinone, isocoumarin, and pyrimidine derivatives in the literature.

The most voluminous group is formed by the C-glycosides of flavonoids. The C-glycosides of flavones (glycoflavones), of flavonols, of flavonones, of chalcones, of isoflavones, and of xanthones are distinguished [6].

The first flavonoid C-glycosides isolated were scoparin (1851), vitexin (1898), and saponarin (1906) [7-9], but their structure could not be determined for a long time. An intensive study of flavonoid C-glycosides was begun at the end of the fifties and beginning of the sixties of this century after Horhammer and Wagner had isolated orientin (a luteolin C-glycoside) from *Polygonum orientale* [10].

At the present time, the flavone C-glycoside group is the best studied. Among them C-monoglycosides, C-diglycosides, C-acylglycosides, and C-O-di-, and triglycosides are distinguished. About 140 such compounds are known. The number of C-glycosides of other flavonoids is small [6, 11]. The method of detecting C-glycosides in plant raw material is based on their resistance to acid hydrolysis and on the use of chromatographic method of analysis followed by treatment of the chromatograms with chromogenic reagents (solutions of aluminum chloride, basic lead acetate, ferric chloride, etc.) [12]. For paper chromatography the following solvent systems are used: butan-1-ol—CH_3COOH—H_2O (4:1:5), butan-1-ol—ethanol—water (4:1:2.2), ethyl acetate—$HCOOH$—H_2O (10:2:3), isopropanol—$HCOOH$—H_2O (2:5:5), and aqueous solutions of acetic acid (5% and 15%) [6, 12].

For chromatography in thin layers of cellulose, in addition to the butan-1-ol—CH_3COOH—H_2O systems and acetic acid solutions, a mixture of butan-1-ol with 27% CH_3COOH (1:1) is used; in thin layers of silica gel, ethyl acetate—CH_3OH—H_2O (100:16.5:13.5), ethyl acetate—$HCOOH$—H_2O (100:30:20), ehtyl acetate—methanol—H_2O/toluene (100:16.5:13.5), and ethyl acetate—pyridine—H_2O—methanol (80:20:10:5); and, in thin layers of polyamide, methanol, CH_3OH—H_2O (6:4), water, and some other solvents [6, 13].

It has been established that on paper chromatography in the butan-1-ol—CH_3COOH—H_2O (4:1:5) system the 6,8-di-C-glycosides of apigenin have lower R_f values than the 6-C-glycosides of apigenin [6]. Chromatography in a thin layer of silica gel in the ethyl acetate—pyridine—H_2O—methanol (80:20:10:5) system shows that in this case it is easily possible to

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separate 6-C-glucosyl- and 6-C-arabinosylvitexins, but 6-C-glucosyl- and 6-C-galactosylvitexins are poorly separated. 6-C-Xylosyl- and 6-C-rhamnosylvitexins migrate together [6].

In the same system at a ratio of the components of 80:12:10:5, in terms of their R_f values on silica gel flavone C-glycosides form the following sequence: 8-C-xylosyl- > 8-C-glucosyl- > 6-C-xylosyl- > 6-C-glucosylflavone, and the di-C-glycosides form the following sequence; 6,8-di-C-glucosyl- > 6-C-glucosyl-8-C-xylosyl- > 6-C-xylosyl-8-C-glucosyl- > 6,8-di-C-xylosylflavone [6].

Table 1 gives the structures and R_f values of some C-glycosides in various systems.

On chromatograms (Table 2) the spots of flavonone C-glycosides have a yellow or dark yellow coloration in UV light. The spraying agents for chromatograms most frequently used are a solution of basic lead acetate, a 1% ethanolic solution of aluminum chloride, and ferric chloride (1% solution in 0.5 N HCl) [20-22].

The glycosidic nature of the glycoflavones can be determined by performing Bryant's reactions [19], whereupon a pink coloration of the aqueous layer arises because of the formation of C-glycosidated anthocyans. It is interesting to note that anthocyan C-glycosides have not so far been detected in nature.

To isolate C-glycosides from plant material, the same methods are used as for O-glycosides [23]. Their purification and separation is being successfully performed with the aid of column chromatography on cellulose, silica gel, or polyamide. Thus, for example, chromatography on polyamide using aqueous methanol of various concentrations as eluent has permitted the isolation of C-glycosides from *Rhyncosia minima* [24] and *Sarothamnus scoparius* [25]. K. Galle [20] initially adsorbed the C-glycosides of wheat shoots by Amberlite IRC-50, and then separated them on a column of cellulose using 15% CH_3COOH as eluent. The use of gel filtration on Sephadexes [26] and of gas-liquid chromatography has also been described [32].

The determination of the structures of the C-glycoside was originally a difficult task. Perkin isolated vitexin from *Vitex lucens* in 1898, but the structure of this compound was finally established only in 1966 [6]. At the present time, in the determination of structures wide use is made of spectral methods, of which PMR spectroscopy and mass spectrocopy are particularly promising, but here also great difficulties arise in determining the carbohydrate component because of the capacity of these compound for isomerization [6].

The UV spectral characteristics of the C-glycosides have been given widely in a number of reviews [11, 12, 27]. As a rule, the absorption maxima are close to those of their aglycones. Thus, in the UV spectra of apigenin C-glycosides there are two main absorption maxima in the 325-340 and 268-275 nm regions, and in luteolin C-glycosides they appear at 346-353 and 251-258 nm [11]. It has been reported that in the presence of freshly fused sodium acetate a larger bathochromic shift ($\Delta\lambda$ 21-25 nm) is observed for luteolin C-glycosides with a free hydroxy group at C_7 than for apigenin C-glycosides ($\Delta\lambda$ 8-11 nm) [27] (Table 3). In the case of the isoflavone C-glycosides, one of the maxima is better-defined than for the corresponding aglycones.

The study of the IR spectrum of C-glycosides is of interest only where it is possible to compare their IR spectra with the IR spectra of known compounds. In contrast to the O-glycosides, the C-glycosides have only two absorption bands in the 1000-1100 cm^{-1} region, at 1010 and 1035 cm^{-1}.

The methods of PMR spectroscopy and mass spectroscopy are particularly promising for establishing the structure of C-glycosides. As can be seen from Table 4, PMR spectroscopy readily permits 6- and 8-C-glycosides to be distinguished from one another. In mass spectroscopy, the products of the fragmentation of C-glycosides differ from the fragmentation products of O-glycosides and have been described in detail in the literature [34, 35].

N. K. Kochetkov and O. S. Chizhov [36] have established that with the aid of mass spectroscopy it is possible to distinguish disaccharides with a 1→6 bond from disaccharides with 1→2, 1→3, or 1→4 bonds. This fact has been used to establish the structure of the carbohydrate component of adonivernitol [20, 37]. However, it is difficult on the basis of mass spectra to distinguish a C-galactoside from a C-glucoside residue, or a C-xyloside from a C-arabinoside residue [6].

TABLE 1. Structures and R_f Values of Various C-Glycosides [6, 12-17]

C-Glycoside	R_f values in the following systems				
	1	2	3	4	5
	A		B	C	
Apigenin C-glycosides					
8-C-Glucopyranosyl (vitexin)	0,63	0,71	0,19—0,24		0,14
8-C-Galoctopyranosyl		0,59			
8-C-Xylopyranosyl		0,73			
8-C-Arabinopyranosyl		0,68			
8-C-Rhamnopyranosyl		0,72		0,54	
6-C-Glucopyranosyl (saponaretin)	0,46	0,59	0,50	0,33	0,42
6-C-Galactopyranosyl		0,37			
6-C-Xylopyranosyl (katchimoside)		0,72	0,68		
6-C-Arabinopyranosyl (isomollupentin)		0,61			
6-C-Rhamnopyranosyl		0,78			
6,8-Di-C-glucopyranosyl (vicenin-I)		0,13	0,42		
6-C-Glucopyranosyl-6-C-xylopyranosyl (vic-I)		0,26			
6-C-Galactopyranosyl-8-C-glucopyranosyl		0,11			
6-C-Arabinopyranosyl-8-C-glucopyranosyl		0,15	0,35		
8-C-Glucopyranosyl-6-C-rhamnopyranosyl (isoviolanthin)		0,26			
8-C-α-Arabinopyranosyl-6-C-glucopyranosyl (schaftoside)			0,46		
C-Glycosides of luteolin and its methyl derivatives					
8-C-Glucopyranosyl (orientin)	0,56		0,13	0,03	0,10
6-C-Glucopyranosyl (homoorientin)	0,39		0,36	0,08	0,19
8-C-β-Glucopyranosyl-1 → 6-β-xylosyl (adonivernitol)			0,53		
6-C-β-Glucopyranosyl-7-O-methyl (swertiajaponin)			0,33		
6-C-Glucopyranosyl-4'-O-methyl (diosmetin 6-C-glucoside)					
8-C-Glucopyranosyl-3'-O-methyl (scoparin)			0,18	0,08	
Isoflavone C-glycosides					
3',5-Dihydroxy-8-C-rhamnopyranosyl-4',7-dimethoxy (volubilin)	0,65				
3',5-Dihydroxy-6-C-rhamnosyl-4',7-dimethoxy (isovolubilin)	0,61				
8-C-Glucopyranosyl-5,7-dihydroxy-4',6-dimethoxy (volubidinin)	0,71				
6,8-Di-C-glucopyranosyl-4',5,7-trihydroxy (paniculatin)	0,31				
8-C-Glucosyl-4',7-dihydroxy (puerarin)	0,71				
8-C-Glucopyranosyl-4',5-dihydroxy-7-methoxy (prunetin 8-C-glucoside)	0,52				
8-C-Glucopyranosyl-4',5,7-trihydroxy-3'-m methoxy (dalpanitin)			0,18		0,08

Note. Systems: 1) ethyl acetate—CH_3OH—H_2O/toluene (100:15.5:13.5), 2) ethyl acetate—pyridine—H_2O—CH_3OH (80:20:10:5)- 3) H_2O; 4) 5% solution of CH_3COOH, 5) 15% CH_3COOH, A — in a thin layer of silica gel, B — in a thin layer of cellulose, C — on a paper chromatogram.

TABLE 2. Color Reactions of Some C-Glycosides on Chromatograms

C-Glycoside	Color of the spots on treatment		
	UV light	ethanolic solution of $FeCl_3$	solution of basic lead acetate
Vitexin	Yellow	Violet	Yellow
Saponaretin	Yellow	Violet-red	Yellow
Orientin	Yellowish blue	Violet	Orange
Homoorientin	Dark yellow	Violet	Orange
Violanthin	Dark yellow		Yellow
Adonivernitol	Dark yellow	Gray-green	Orange
Katchimoside	Dark yellow	Gray-green	
Puerarin	Blue-violet Yellow		
Volubilin	Yellow	Violet	
Isovolubilin	Yellowish blue	Green-brown	
Paniculatin	Yellow	Green	
Prunetin 8-C-glucoside	Yellowish blue	Violet-blue	

TABLE 3. UV Spectral Characteristics of Some G-Glycosides

C-Glycoside	λ, nm	
	CH_3OH	CH_3ONa
	Apigenin C-glycosides	
Vitexin	270, 302 sh. 336	279, 329, 395
Saponaretin	271, 336	278, 329, 398,
Violanthin	274, 311 sh., 335	281, 333, 398
	C-Glycosides of luteolin and its methyl derivatives	
Orientin	255, 267, 293 sh., 346	268, 278 sh., 334 sh., 405
Homoorientin	242 sh., 255, 271, 349	267, 278 sh., 280, sh., 406
Lucenin-I	257, 272, 349	240 sh., 266, 280, 344 sh.,408
Scoparin	251, 270, 345	265, 277, 334 sh., 406
	Isoflavone C-glycosides	
Paniculatin	265	284
Volubilin	269, 340	
Volubilinin	268, 336	
Prunetin 8-C-glycoside	216, 287 sh.,	268, 294 sh.

In recent years, CD spectra have been used successfully to distinguish C-glycosides from O-glycosides and the 6- and 8-glycosylflavones [38, 39].

In view of the complexity of determining the structures of C-glycosides by chemical methods (resistance to acid hydrolysis, isomerization), methods of independent synthesis have been developed. Thus, in determining the structures of vitexin and orientin it was established that C-glycosylation takes place as the result of an attack on the aglycone by the active sugar in the form of a carbonium ion.

The reaction of 2,3,4-triacetyl-α-D-xylopyranosyl bromide with chrysin, acacetin, apigenin, and luteolin leads to 6-C-β-D-xylopyranosyl flavones [6, 40]. Similarly, the reactions of 2,3,4,6-tetraacetyl-α-D-galactopyranosyl bromide with apigenin and acacetin and of 2,3,4-triacetyl-β-L-arabinosyl bromide with apigenin give, respectively, 6-C-β-D-galactopyranosyl-, 6-C-D-arabinopyranosyl-, and 6-C-α-L-rhamnopyranosylflavones [41].

5,7-Dihydroxyflavone C-diglycosides can be obtained from natural 8-C-glucosyl-5,7-dihydroxyflavones. Thus, the reaction of 2,3,4,6-tetraacetyl-α-D-glucopyranosyl bromide with cytisoside and vitexin leads to the 6,8-di-C-β-glucosyl derivatives of acacetin and of api-

TABLE 4. Chemical Shifts of the Protons of Flavonoid C-Glycosides and Their Acetates [16, 20, 27, 31]

C-Glycoside	Chemical shifts, δ scale, ppm, J, Hz							
	H-2',6'	H-3',5	H-6	H-8	H-3	H-2	OCH_3	sugar protons
Vitexin	7,94	6,98	6,31		6,30			
Vitexin metaacetate	8,15	7,42	6,83		6,70			
Saponaretin	7,87	7,00		6,58	6,62			
Saponaretin hexaacetate	7,96	7,42		6,94	6,69			
Adonivernatol decaacetate	7,57	7,43	6,93		6,69d (J=4)			5,5—3,5
Swertiajaponin heptaacetate	7,70	7,46		6,90	6,62		4,06	4,0—6,0
Schaftoside	8,13 (J=8)	6,99 d (J=8,5)			6,82			3,23—4,2
Schaftoside decaacetate	8,08d (J=9)	7,34 d (J=9)			6,60			3,8—5,8
Isoshaftoside	7.94 d	6,91 d			6,85			3,31—4,75
Isoswertisin	7.94 d (J=9)	6,77	6,39		6,70		3,77	3,0—5,0 m
Molludistin	6,1 d J=8	6,77	6 36		6,70		3,75	3,0—5,9
Bayin hexaacetate	8,25	7,36	7·15		6,81			
Puerarin	7,36	6,98	7,00			8,05		

C-Glycoside	Chemical shifts, δ scale, ppm, J, Hz								Solvent
	OAc in position							OH at C_5	
	5	7	4'	2''	3''	4''	6''		
Vitexin								13,13	$CDCl_3$, DMSO-d_6
Vitexin metaacetate	2,42		2,37	1,72	2,10	2,02	1,92		$CDCl_3$
Saponaretin		2,47						13,60	$CDCl_3$/ DMSO-d_6
Saponaretin hexaacetate		2,47	2,36	1,85	2,10	2,10	2,05		$CDCl_3$
Adonivernatol decaacetate		2,42—2,33			1,97	2,07	1,92		$CDCl_3$
Swertiajaponin heptaacetate	2,49			1,81	2,09		2,03		$CDCl_3$
Schaftoside									DMSO-d_6
Schaftoside decaacetate				1,60—2,21 m					$CDCl_3$
Isoshaftoside									DMSO-d_6
Isoswertsin									DMSO-d_6
Molludistin									DMSO-d_6
Bayin hexaacetate		2,45	2,37	1,72	2,10	2,02	1,90		$CDCl_3$
Puerarin									DMSO-d_6

genin, respectively [42]. These compounds proved to be identical with natural compounds isolated from *Citrus lemon*.

Lucenin and isoviolanthin have been synthesized in the same way [6]. A method of obtaining 4',7-di-O-methylbayin by condensing 2,6-dimethoxyphenylmagnesium bromide with α-acetochloroglucose has been described. In this case, a mixture of α- and β-D-glucopyranosyl-2,6-dimethoxybenzenes was obtained initially. On heating, the α-isomer was converted into the β isomer. The tetraacetate of the latter was condensed with anisaldehyde and the resulting chalcone was oxidized with selenium dioxide to 4',7-di-O-methylbayin [6]. However, in all cases the yield of C-glycosides was low (∿ 1%), and therefore it is extremely promising to perform additional investigations in this direction.

At the present time, natural flavonoid C-glycosides have been detected in 300 species of higher and lower plants and have been isolated from algae, mosses, ferns, and monocotyledonous plants. They are found particularly frequently in species of the family Leguminosae (*Arachis*, *Castanospermum*, *Cytisus*, *Hedysarum*, *Lespedeza*, *Pueraria*, *Sarothamnus*, *Sophora*, *Spartium*, *Tamarindus*, *Trigonella*, and others) [1].

TABLE 5. Distribution of Flavonoid C-Glycosides in Plants

Type, class, family	Genus	What it contains	Literature
ALGAE			
Characeae	Nitella Ag.	Flavone C-diglucosides	46
Rhodophyceae	Phyllophora Grev.	Glycoflavones	20
BRYOPSIDA			
Conocephalaceae	Conocephalum Weber	Vicenin-2, lucenin-2	48
Hepaticae	Hemenophytum	Schaftoside, isoschaftoside	47, 49
	Marchantia L.	Vicenin-2	50
Jungermanniaceae	Plagiochila Dum.	Tricetin 6,8-diglucosides, lucenin-2	51
	Reboulia	Acacetin 8-C-glycoside	52
Musci	Mnium Hedw.	Apigenin 8-C-glucoside, vicenin-2 Vicenin O-glucoside, scoparin, schaftoside, isoschaftoside	53, 61
Porellaceae	Porella L. (-Madotheca)	Apigenin 6,8-di-C-glycoside, isovitexin, saponarin	54, 55
SPHENOPSIDA			
Equisetaceae	Equisetum L.	Saponaretin	56
PTERIDOPSIDA			
Psilotaceae	Psilotum	Apigenin C-glycosides	57
	Tmesipteris	" "	
Cyantheaceae	Cyantea	Vitexin, isovitexin	58
Meratiales	Angiopteris		59
Aspidiaceae	Ctenitis	Violanthin, isoviolanthin	60
Athyriaceae, Dryopteridaceae	(6 genera have been investigated)	1,3,6,7-Tetrahydroxyanthone 2-C-glucoside (mangiferin), isomangiferin (the 4-C-glucoside)	62
GYMNOSPERMAE			
Cycadaceae	Cycas L.	Vitexin and derivatives of it	63
(Zamiaceae)	Dioon	Orientin, apigenin 8-C-glucoside	64
Pinaceae	Larix Mill.	Vitexin xyloside, apigenin 8-C-xylosidoglucoside, vitexin rhamnoside	65—69, 72
		Vitexin and derivatives of it	63
Taxodiaceae	Sequoia	Vitexin	63
Ephedraceae	Ephedra L.	Isovitexin, 7-O-glucosylisovitexin, vicenin-2, swertisin, x"-O-glucoswertisin. isoswertisin, swertiajaponin, isoswertiajaponin	70
Gnetaceae	Gnetum		
ANGIOSPERMAE DICOTYLEDONEAE			
Aceraceae	Acer L.	Vitexin, saponaretin, orientin, homoorientin	71
Aizoaceae	Mollugo L.	Isoswertisin, 8-C-α-L-arabinopyranosylgenkwanin (molludistin), isoswertisin 2"-rhamnoside, molludistin 2"-rhamnoside	72

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
Apiaceae	Anethum L.	Vicenin	73, 74
	Astrantia L.	6-C-Glucopyranosylluteolin	75
	Gryptotaenia D. C.	Glycoflavones	76
	Daucus L.	Vicenin	77
	Heptaptera Marg. et Reut	Mangiferin	76
	Laretia	Glycoflavones	76
	Onopanax	Glycoflavones	75
	Smyrnium L.	6,8-Di-C-rhamnosylapigenin	74
Asteraceae	Ajania Poljak.	Apigenin 6,8-di-C-β-D-glucopyranoside	78
	Artemisia L.	Apigenin di-C-β-D-diglycoside, 6-C-glucopyranosyl-pyranosylapeginin	79
	Carlina L.	Lucenin-2, orientin, homoorientin, vitexin, isoshaftoside, corymboside	80, 81
	Catananche L.	Shaftoside, isoschaftoside, 3-hydroxyschaftoside, 3'-hydroxyisoschaftoside	82
	Centaurea L.	Vitexin 7-methyl ether swertisin	83, 84
	Eupatorium L.	Vicenin-2	85
	Flourensia DC.	Vicenin-1, isoschaftoside, schaftoside, neoschaftoside	86
	Gaillardia Fourder	Swertisin, orientin, 3',4,5-trihydroxy-7-methoxyflavone 6-C-glucoarabinoside, 6-C-hexosyl-8-C-rhamnosyl-apigenin η-glucoarabinoside, luteolin glucoxylosido-6-8-di-C-glucoside, isovitexin, isoorientin, a C-glycoside of the vicenin type, saponaretin	87—90
	Hazardia	Apigenin 8-C-glycoside, 6,8-C-diglucosides.	91
	Helenium L.	Saponaretin, orientin, isoorientin, vitexin, swertisin	92, 93
	Liatris Schreb.	Vicenin-1, vicenin-2, vicenin-4	94
	Senecio L.		
	Tragopogon L.	x"-O-Xylosylvitexin, vicenin-2, lucenin-1, swertiajaponin, swertisin	95
Brassicaceae	Alliaria DC.	Alliaroside (7-O-glucosyl-C-glycosylapiginin)	96
Cannabaceae	Cannabis Tourn.	2"-O-glucopyranosylorientin, 2"-glucopyranosylvitexin, 2"-glucopyranosylvitexin, orientin, vitexin, isovitexin, glucoarabinoside, O-rhamnoglucosylisoorientin, orientin 7-O-glucoside.	26, 47
Caryophyllaceae	Humulus L.	Vitexin	98
	Arenaria L.	Vitexin, orientin, isovitexin, homoorientin, and their 8β, 6α, and 6β isomers, vicenin isomers, O-glycosides, C-monosides	99

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
Caryophyllaceae	Cerastium L.	Vitexin, saponaretin, orientin, homoorientin, di-, tri-, and tetraglycosides of C-monosides of apigenin and luteolin	99, 100
	Coranaria L.	Apigenin C-monoglucosides	101, 102
	Cucubalis L.	Saponaretin, vitexin, orientin, homoorientin, isosaponaretin, 6-O-galactosylsaponaretin	101
	Dianthus L.	Vitexin, saponaretin, homoorientin, orientin, neoavroside, isoneoavroside	103
	Gypsophila L.	Vitexin, saponaretin, orientin, homoorientin, adenivernitol, saponaretin $4'$-β-glucoside, katchimoside, a C-diglycoside of the vicenin type, O-mono-di-glycosides	104, 105
	Lychnis L.	Vicenin, orientin, isoorientin, vitexin, isovitexin	102
	Melandrium Roehl.	Vitexin, isovitexin, orientin, isoorientin, isovitexin-$3'$-O-xyloside, x''-arabinosyl-6-C-glucosylapigenin, x''-glucosylisovitexin, O-glucosylsaponarin, arabinosylsaponarin, 7-O-xylosyl-6-C-glucoarabinosylapigenin $3'$-or $4''$ -O-glucosyl-7-O-galactoside, $2''$-O-rhamnosylisovitexin, isovitexin 7-O-galactoside $2''$-O-glucoside, isovitexin 7-O-galactoside (neosaponarin)	44, 106, 107, 108
	Otites Adans.	Vicenin and its isomers, isovitexin, orientin, homoorientin, 8α, 6α, 6β isomers	109
	Petrocoma Rupr. (Silene)	Avroside, orientin, homoorientin, petrocoside	110
	Saponaria L.	Saponarin	6
	Silene L.	Vicenin and its isomers, isovitexin, homoorientin, schaftoside	102, 109, 111, 112, 113
	Spergularia J. et C. Presl.	Lucenin-2, vicenin-2, isomollupentin	114—116
	Stellaria L.	Saponaretin, vitexin, saponaretin F-O-glucoside	115, 116
	Telefium L.	Orientin, isoorientin, vitexin, isovitexin	102
	Vaccaria Medic.	Vacarin, isocaponaretin	117, 118
	Viscaria Roehl.	Saponaretin, vitexin, orientin, homoorientin, $6''$-O-galactosylsaponaretin	101

TABLE 5 (continued)

Type, class, family	Genue	What it contains	Literature
Caesalpiniaceae	Gleditschia L.	Vitexin, saponaretin, homoorientin, orientin	121—123
	Parkinsonia L.	Epiorientin (parkinsonin A) and O-methylepiorientin (parkinsonin B)	124
	Tamarindus	Vitexin, homoorientin, isovitexin, orientin	125, 126
Chenopodiaceae	Beta L.	Vitexin derivatives	127
Combretaceae	Combretum L.	Vitexin, saponaretin	128
Cucurbitaceae	Bryonia L.	Saponarin, bryonoside	129
	Cucumis L.	Meloside A, x'-O-caffeylfeylmeloside L	44
Dipsacaceae	Cephalaria Schrad.	Swetisin, leucanthoside (swertiajaponin), orientin, vitexin, isoorientin, isovitexin	20, 130, 132 133, 134
	Dipsacus L.	Saponarin	20, 131
	Knautia L.	Orientin, isoorientin, vitexin, knautinoside	133, 135
	Pterocephalus Vaill.	Swetiajaponin, swertisin	131
	Scabiosa L.	Saponarin, swertisin, orientin, isoorientin, vitexin, isovitexin	131, 134, 135
Euphorbiaceae	Croton L.	Vicenin-2, vitexin, saponaretin orientin, isoorientin	136
	Jatropha L.	Vitexin	137
Fabaceae	Arachis L.	Vitexin and its derivatives	138
	Aspalanthus L.	Aspalanthin, orientin, isoorientin	139, 140
	Calycotome Link.	Glycoflavones	141, 142
	Chamaecytisus Link.	Glycoflavones	141, 142
	Coronilla L.	Isoorientin, isoorientin 2"-rhamnoside, isoorientin 4'" glucoside	141
	Cladrastis Rafin	Isoliquiritigenin C-glycoside, bayin, cladrastin C-glycoside, 8-C-β-D-glucosyl-4',5,7-trihydroxy-6-C-β-D-xylosylflavone	145, 146
	Crotalaria L.	Isovitexin, orientin, isoorientin, vitexin 4'-xyloside	141, 143, 144
	Cytisus L.	Vitexin, isovitexin, orientin 3'-methyl ether, scoparin, cytisoside	6, 141
	Dalbergia L.	Paniculatin, prunetin 8-C-glucoside, dalpanitin, volubilin, volubin	147, 151
	Desmodium Desv.	Vitexin, saponaretin, homoorientin, luteolin C-diglucoside, vicenin, genkwanin C-glucoside	6, 152

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
	Genista L.	Vitexin, isovitexin	141
	Glycyrrhyza L.	Saponaretin, vitexin	153, 154
	Hedysarum L.	Mangiferin, isomangiferin	155, 156
	Lespedeza Medix.	Orientin. homoorientin, vitexin, saponaretin	157, 160
	Lupinus L.	Orientin, homoorientin, vitexin, cytisoside, genistein 8-C-glucoside	161, 163
	Lygos Adans	Glycoflavones	142
	Melilotus Juss.	Vicenin, orientin, homoorientin	164
	Meristotropis Fisch. et C. A. Mey	Saponaretin	165
	Psoralea L.	Scoparin, 6,8-di-C-glycosyl chrysoeriol, vicenins	6, 141
	Pueraria DC	Puerarin, C-glucosylisoliquiritigenin, daizein 8-C-glucoside, di-O-acetylpuerarin, puerarin D-xyloside	166, 168
	Sarothamnus Wimm	Orientin, scoparin, vitexin	6, 170
	Sophora L.	Lucenin-2, vicenin-2, rhamnosylvitexin, rhamnosylisovitexin	141, 171
	Spartium L.	Orientin	172
	Teline Medic.	Glycoflavones	141
	Trigonella L.	Vitexin, saponaretin, vicenin-1, vicenin-2, vitexin 7-glucoside, vitexin 2"-O-p-coumarate	11, 141, 173
	Ulex L.	Glycoflavones	141
Fabaceae	Castanospermum A. Cunn	Bayin	11
	Nothofagus Blume	Nothofagin (a C-glycosyl-2', 4,4' 6'-tetrahydroxy-dihydrochalcone), Konnanin (a C-glycosyl-2', 3,4,4', 6'-pentahydroxy-	6
Gentianaceae	Gentiana L.	Vitexin, vitexin 4'-O-glucoside, isoorientin 4'-O-glucoside, 2'-trans-caffeylisoorientin and its 4'-β-D-glucoside, swertisin, isoorientin 3'-O-glucoside, saponaretin, isopyrenin (6-C-β-D-glucosyltricin), 7-O-β-D-glucopyranosylisopyrenin, isoscoparin 7-O-glucopyranoside acacetin 7-O-β-D-glucopyranoside, saponarin	6, 174, 186
	Swertia L.	Isovitexin, homoorientin, isovitexin x"-arabinoside, swertiajaponin, swertisin	187—189

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
Gesneriaceae	Cyrtandra J. R. et G. Forst.	Glycoflavones	20
Geraniaceae	Geranium L.	Homoorientin, orientin, vicenin-2, vitexin	190
Hypericaceae	Hypericum L.	Orientin, homoorientin, $2''$-O-acetylorientin	191, 192
	Ochrocarpus Thou	Vitexin	193
Lauraceae	Beilschmiedia Nees	Orientin, isoorientin, vitexin, isovitexin	194
	Persea Mill.	Orientin, isoorientin, vitexin, isovitexin	195
Linaceae	Linum L.	Linosides A and B	196
Lythraceae	Lythrum L.	Glycoflavones	20
Malvaceae	Hibiscus L.	Glycoflavones	129
Martyniaceae	Martynia L.	Luteolin C-glycosides	197
Myrtaceae	Eucalyptus L. Herit.	Hemiphloin (6-C-β-D-glucopyranosylnaringenin), isohemiphloin (8-C-β-D-glucopyranosylnaringenin)	6
Nymphaeaceae	Nymphaea L.	Orientin	198
Ochnaceae	Brackenridgea	Vitexin, isoorientin, $2''$-O-acetyl-7-O-methylvitexin	199
	Sauvagesia	Vitexin, orientin, isoorientin, vicenin-2	200
Onagraceae	Circaea L.	Vitexin, isovitexin, vicenin-1, vicenin-2	201
Oxalidaceae	Oxalis L.	Orientin, $2''$-glucosylorientin	202
Passifloraceae	Passiflora L.	Vitexin, saponaretin, saponarin, homoorientin	203, 207
Plumbaginaceae	Limonium Mill.	Vitexin, saponaretin, saponarin, issorientin	207
Polemoniaceae	Phlox L.	Vicenin, 6-C-xylosyl-O-rhamnosylapigenin, 6-C-glucosyl-O-rhamnosylapigenin, 6-C-glucosyl-O-glucosylapigenin, lucenin, 6-C-xylosyl-O-rhamnosyl luteolin, 6-C-glucosyldi-O-xylosylluteolin, 6-C-glucosylluteolin, saponaretin, 6-C-xylosylluteolin	5, 208
Polygonaceae	Fagopyrum Gaertn.	Orientin, vitexin, homoorientin, saponaretin	209
	Polygonum L.	Orientin, isoorientin	6, 210, 211
	Rumex L.	Vitexin, saponaretin	11
Ranunculaceae	Adonis L.	Adonivernitol, orientin, homoorientin, vitexin, homoadonivernitol, saponaretin	11, 212, 20
	Ranunculus L.	Vitexin, saponaretin, orientin, homoorientin, adonivernitol, luteolin 8-C-β-D-glucopyranosyl-6-O-β-D-xyloside	11, 207
	Thalictrum L.	Vitexin, saponaretin, isoorientin, isovitexin 5-rhamnoside	11

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
	Trollius L.	Vitexin, orientin,	11
Rosaceae	Crataegus L.	Saponaretin, vitexin,orientin, monoacetylhomoorientin, rhamnosylorientin, vitexin 4′-rhamnoside, acetyl-4″-rhamnosylvitexin, 2″-O-rhamnosylvitexin (8-C-neohesperidosylvitexin), x″-O-rhamnosylorientin, cratenacin, deacetylcratenacin	6, 11, 213′ 214
Rutaceae	Citrus L.	2″-xylosylvitexin, 6,8-di-C-β-D-glucopyranosylapigenin, 8-C-β-D-glucopyranosyl-diosmin, 2″-O-β-D-xylosyl vitexin, 6, 8-di-C-glucosyl apigenin, 6,8-di-C-glucosyl diosmetin	6, 11, 215—217
	Fortunella Swingl.	Margaritin (8-C-, isomargaritin (6-C-neohesperido sylacacetin)	44
	Teclea Delile	Vitexin, saponaretin, homo-orientin, orientin	218
Simarubiaceae	Ailanthus Desf	Glycoflavones •	
Scrophulariaceae	Gratiola L.	Saponaretin, vitexin, avroside, isoavroside, neoavroside, isoneoavroside	20 219, 220
Theaceae	Camellia L.	Apigenin 6,8-di-C-glucoside, saponarin	6, 11
Tiliaceae	Colona	4′-Glucosylvitexin, orientin O-glucoside	221
Ulmaceae	Trema	Cytisoside, 8-C-β-D-glucosyl-4′-O-methylapigenin 7-O-β-D-glucoside, tremasperin (a mixture of mono- and di-O-acetyl-7-O-glucosylcytiso-sides	6,222
	Zelkova Spach.	Keyakinin B (6-C-glycosyl-quercetin), keyakinin (7-O-methyl-kaempferol 6-C-glucoside)	6
Verbenaceae	Vitex L.	Saponaretin, vicenin 1, vicenin-2, vicenin-3 vicenin-4, lucenin-1, lucenin-2, lucenin-3, lucenin-4, 2″-O-β-D-xylopyranosylvitexin, 2″-O-p-hydroxybenzoylvitexin, 2(?)-O-xylosylorientin, vitexin	6
Violaceae	Viola L.	Violanthin, 6,8-di-C-β-glucopyranosylapigenin	223, 224
Vitaceae	Vitis L.	Vitexin, saponaretin, orientin, homoorientin	225
MONOCOTYLEDONEAE			
Alismataceae	Alisma L. Echonodorus, Sagittaria	Glycoflavones	20
	Acorus	Lucenin	
	Arum	Saponarin	6
	Biarum	Saponarin	6

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
	Chamaedorea Chamaerops Howea Oreodoxa	Glycoflavones	20
	Phoenix	Isovitexin 7-sulfate, orientin 7-sulfate, orientin 7-glucoside sulfate, isoorientin 7-sulfate	226
Butomaceae	Butomus Limnocharis	Glycoflavones	20
Commelinaceae	Commelina	Swetisin, flavocommelinin (4′-glucosylswertisin)	6
Cyperaceae	Carex L. Cladium Cyperus L. Rhynchospora Scirpus Eriophorum	Glycoflavones	20
Iridaceae	Crocus L.	8-C-rhamnosidedoglucosidoglucosylchrysoeriol	227
	Iris L.	Vicenin, lucenins, flavoanamenin, x″-O-xylosylswertisin, embigenin (6-C-D-glucopyranosyl-4′, 7-di-O-methylapigenin, embinin (x″-O-rhamnosylembigenin), swetiajaponin, isoorientin, vitexin	6, 11, 228, 229
Lemnaceae	Lemna L.	Vicenins, isoscoparin, lutonarin, isovitexin, isosaponarin, adonivernitol, lucenin	6, 11
	Spirodella Schleid.	Saponarin, isosaponarin, lutonarin, orientin	230—232
	Wolffia	Vicenins	6, 11
Liliaceae	Asphodelus	Homoorientin	233
	Convallaria L.	Glycoflavones	234
	Ornitogallum	Saponaretin, saponarin	235
	Polygomatum Mill.	Vitexin xyloside, 6-C-β-D-glucosyl rhamnosyl-7-O-glucosyl, 4′, 5-dihydroxyl flavone, vitexin 2″-O-sophoroside, saponarin, 6-C-galactosyl-8-C-arabinosylapigenin, 8-C-galactosylapigenin	236—238
	Ruscus	4′-O-Glucosylisovitexin, isovitexin	239
	Urginea Steinh.	Vitexin, isovitexin, xylosylvitexin, vicenin-2, orientin, isoorientin, scoparin, isoscoparin	240, 241
Orchidaceae	Tribes of Epidendroideae and Vandoideae	Vitexin, isovitexin, and their 7-O-glucosides and C-diglucosides	242
Poaceae	Agropyron	Violanthin	243
	Agrostis	Glycoflavones	20

TABLE 5 (continued)

Type, class, family	Genus	What it contains	Literature
	Avena L.	Saponaretin, x″-arabinosylisovitexin, x″-O-rhamnosylisovitexin, 8-C-rhamnosylglucosylapigenin, 8-C-glucosidoglucosylapignin, O-rhamnosylisoswertisin, 6-C-glucosyl-O-arabinosylluteolin, 6-C-glucosylorientin, 8-C-glucosyl-O-rhamnosyl-7-methoxyapigenin, vitexin 2-rhamnoside, vicenin-2, isovitexin 2″-arabinoside	6, 244—246
	Brachypodium	Isoorientin, isovitexin	243
	Bromus	Glycoflavones	20
	Briza L.	Vitexin, isovitexin, orientin, isoorientin 4′-O-glucoside, 8-C-galactosylapigenin, 8-C-galactosylluteolin	247, 248
	Cathesticum J. Presl.	Vicenin	6
	Dactylis L.	Isovitexin, isoorientin	243
	Digitaria	Isoorientin	243
	Eleusine	Vitexin, orientin, isovitexin, violanthin, lucenin-1, isoorientin	243, 249
	Eregrostis	Violanthin	243
	Hordeum L.	Saponaretin, orientin, lutonarin 3-methyl ether, isoscoparin, vitexin 7-rhamnoside, vitexin 7-glucobioside, isovitexin 7-glucoside, isovitexin 7-glucobioside, isovitexin x-arabinoglucoside	6, 250, 251
	Leersia	Isovitexin, isoorientin, violanthin	243
	Microstegium	Violanthin	243
	Oryza L.	Violanthin	243
	Setaria	Isoorientin	243
	Sorghum L.	C-Glycosides of luteolin and apigenin	252
	Stipa L.	Isoorientin and its glycosides	253
	Triticum L.	Vicenin-2, lucenin-1, lucenin-3, saponarin, ester of 6,8-di-C-glycosylapigenin with sinapic acid, 4′-O-glucosylisoswertisin, lutonarin, viomin (7-O-rutinosylorientin)	6, 11, 254
	Zea L.	2″-O-α-L-Rhamnosyl-6-C-(6-deoxy-xylo-hexose-4-ulosyl) luteolin	255

Table 5 lists the plants in which C-glycosides have been detected. As the carbohydrate components in the C-glycosides have been found galactose, rhamnose, xylose, glucose, and arabinose in positions 6 or 8. C,O-Diglycosides are frequently found which have the second sugar residue in position 7 or 4'(O-glycosidic bond). In recent years, series of acylated C-glycosides have been found. These include as acid residues those of ferulic, caffeic, and acetic acids. Examples of such compounds are the glycosides isolated from *Linum usitatissimum* (one of them is 6"-O-acetyl-3',4',7-trimethyl-2"-O- α-L-rhamnopyranosylisoorientin) [44] and from *Gentiana burseri* (2"-O-trans-feruloylorientin, 2"-O-trans-feruloylisoorientin 4'-O-glucoside, and 2"-O-trans-caffeoylisoorientin 4'-O-glucoside) [45].

The biogenesis of the C-glycosides has been studied inadequately, but it is already possible to state with confidence that C-glycosidation takes place in the initial fragments, while O-glycosidation may be observed after the biosynthesis of the aglycone. This fact has made it possible for some authors to asert that C-glycosides are characteristic for the most primitive families, less evolutionarily advanced. However, the fact that C-glycosides are present in flower pigments, in the form of complexes with anthocyans and with metal ions and are responsible for the bright colors of flowers [6],and their detection in the most evolutionarily advanced families (see Table 5) show the invalidity of this hypothesis.

The study of flavonoid C-glycosides is of interest not only from the point of view of chemotaxononomy, but also for their use in medicine. It has been established that flavone C-glycosides, like other flavonoids, possess a blood-vessel strengthening action [256]. They are practically free from toxicity and, depending on their structure, exhibit a more or less pronounced diuretic, cholagogic, spasmolytic, anti-inflammatory, and antifungicidal action. An antisclerotic action has been established for some of them and also their capacity for lowering the total nitrogen content of the blood [256]. At the present time, medicinal preparations have been proposed in the form of total flavonoids, including C-glycosides. One of such preparations is lesphenephril [257].

In recent years, xanthone C-glycosides have been studied intensively. Xanthones are characteristic for a small number of families — Gentianaceae *(Gentiana, Swertia)*, Fabaceae *(Hedysarum)*, Guttiferae *(Gardenia, Hypericum)*, Polygonaceae *(Polygonum)*, and Moraceae [11]. The presence of C-glycosides in lichens has been reported [258]. The most widespread C-glycoside is mangiferin (1,3,6,7-tetrahydroxyxanthone 2-C-β-D-glucopyranoside). It has been established that in the biosynthesis of mangiferin C-glycosylation takes place in the stage of inclusion of the corresponding pentahydroxybenzophenone [259].

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NOMENCLATURE OF BENZYLISOQUINOLINE AND RELATED ALKALOIDS

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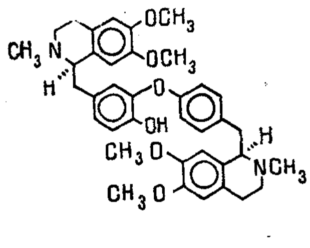
UDC 547.944/945

A classification of the bisbenzylisoquinoline, benzylisoquinoline, and related alkaloids is proposed which is based on the biogenetic characteristics of these alkaloids.

The bisbenzylisoquinoline alkaloids form a large — more than 160 — group of plant bases found in plants of twelve families [1]. Characteristic of them is a common pathway of biosynthesis through the oxidative coupling of benzylisoquinoline fragments. In accordance with the method of coupling of these fragments, the alkaloids can be divided into thirty subgroups, including benzylisoquinoline-aporphine, benzylisoquinoline-proaporphine, and other alkaloids genetically related to them. M. Shamma included this biogenetic principle in the basis of the broad classification that he has proposed of 26 subgroups of bisbenzylisoquinoline alkaloids [2]. However, when this classification is subjected to careful consideration, it can be seen to have several defects; for example, not all the alkaloids mentioned above can be classified on this principle.

In order to broaden the possibility of numerical classification, we have introduced letter symbols for the type of substitution: o — OH, om — OCH$_3$, m — CH$_3$, omo — OCH$_2$O, d — dehydro, h — homo, : — additional bond, x — bond cleavage, R and S — symbols of absolute configuration. The figures relating to the right-hand half bear a prime. This permits intramolecular ether or carbon-carbon bonds in the aporphine, coumarin, and other alkaloids to be shown. The 1-benzyl-1,2,3,4-tetrahydroisoquinoline skeleton is taken as the basis in all cases. Let us give some examples.



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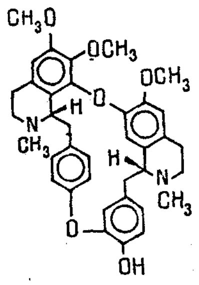

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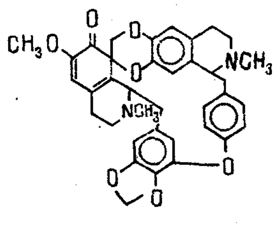

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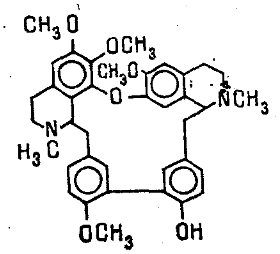
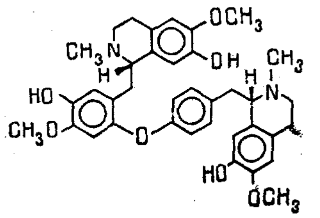


IV



V

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