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< Review >

Chemical Compositions of the Genus *Hovenia*

Bao-jun Xu[1], Yu-qiu Deng[2], Jeong-hyun Lee, Eun-kyoung Mo, and Chang-keun Sung*

Department of Food Science and Technology, Chungnam National University, Taejon 305-764, Korea
[1]The Pharmaceutical Institute, Dalian University, Dalian 116622, China
[2]Department of Horticulture, Chungnam National University, Taejon 305-764, Korea

Abstract – Though a great deal of secondary metabolites and their bioactivities were discovered from the plants of the genus *Hovenia*, the structure-activity relationship was not well defined. To facilitate further pharmacological research and development in this area, we have summarized the progress of phytochemical and pharmacological research on the plants of the genus *Hovenia*.
Keywords – *Hovenia dulcis, H. acerba, H. trichocarpa*, triterpenoids saponin, flavonoids, alkaloids

Introduction

The plant genus *Hovenia* (Rhamnaceae) includes three species: *Hovenia dulcis*, *Hovenia acerba*, *Hovenia trichocarpa*, and two variations: *Hovenia dulcis* var. *tomentella* Makino and *Hovenia dulcis* var. *Koreana* Nakai (Choo *et al.*, 1993), major inhabitation of the plant being China, Korea, Japan and the Himalayan areas (Gupta and Safui, 1984). Among them, *H. dulcis* Thunb., *H. acerba* Lindl., and *H. dulcis* var. *tomentella* are used as medicinal plants.

According to the traditional Chinese medicine, *H. dulcis* is neutral in nature, sweet and sour in flavor, and has biological effects like defervescence, diuresis, and alcohol detoxification. It was traditionally known to have the lingering intoxication alleviation effect, treating thirst, emesis, urinal disorder, and constipation. *H. trichocarpa* is an indigenous plant of southern Japan, the bark of which is used as a remedy for crapulence and as an underarm deodorant.

Recent research showed that *Hovenia* contained extensive pharmaceutically active compositions, such as triterpenoids, flavonoids, alkaloids, polysaccharides, fructose, glucose, potassium nitrate, organic acids, and peroxidase. It was proved that the extract of *Hovenia* or its complex formulas hasten detoxification of alcoholic poisoning. In addition, it was reported that the extract of *Hovenia* had other various physiological activities including anti-mutation, anti-tumor, anti-hypertension, and anti-oxidation (Kennedy *et al.*, 1988; Lee *et al.*, 1999).

*Author for correspondence
Fax: +82-42-822-2287, E-mail: kchsung@cnu.ac.kr

In recent years, novel active compounds with remarkable pharmacological activities were discovered from genus *Hovenia*. In this review article, we have summarized and briefly discussed about these compounds, in order to facilitate the further pharmacological research and development.

Chemical compositions – By utilizing modern spectral and chemical techniques, various chemical compositions were isolated and elucidated from the leaves, roots, barks, xylems, fruits, and seeds of the genus *Hovenia*.

Terpenoids

Triterpenoids Saponins – Triterpenoids saponins were obtained firstly from the root, bark and leaves of *H. dulcis* (Kawai *et al.*, 1974; Ogihara *et al.*, 1976; Inoue *et al.*, 1978). Some compounds were reported as sweetness inhibitors from the root, bark and leaves of *H. dulcis*, including hovenoside I (**1**) (Inoue, *et al.*, 1978), saponin C_2 (**2**), D (**3**), G (**4**) (Kimura *et al.*, 1981), and saponins E (**5**) and H (**6**) (Kobayashi *et al.*, 1982). Similarly, several dammarane-type triterpene saponins such as hoduloside I (**8**), II (**9**), III (**10**), IV (**11**), and V (**12**) were reported as anti-sweet principles from the leaves of *H. dulcis*, along with the known saponins hovenoside I, saponins C_2, E,H and jujuboside B (**13**) (Yoshikawa *et al.*, 1992). In succession, five new dammarane glycosides named hodulosides VI~X (**14~18**) were isolated from the fresh leaves of *H. dulcis* Thunb. var. *tomentella* Makino (Yoshikawa *et al.*, 1993a). Their structures were determined on the basis of chemical and spectral evidence. Compound **5** VII-X showed anti-sweet activities.

From the leaves of *H. acerba* Lindl, two dammarane-type triterpene saponins were isolated for the first time and identified as hovenia saponin C_2 (**2**) and jujubogenin-

3-O-[α-L-rhamnopyranosyl(1→2)][α-D-galactopyranosyl (1→2)-α-D-glucopyranosyl (1→3)]-α-L-arabinopyranoside (**19**), named as hovacerboside A_1 (Liang *et al.*, 1996a). Similarly, from the seeds and fruits of *H. dulcis* Thunb., four bioactive methyl-migrated 16, 17-seco-dammarane type triterpene glycosides, hovenidulciosides A_1 (**20**), A_2 (**21**), B_1 (**22**), and B_2 (**23**) were isolated together with hoduloside III (**10**) and (+)-gallocatechin (**48**) (Yoshikawa *et al.*, 1996). The absolute stereostructures of hovenidulciosides A_1 and A_2 with a migrated 16, 17-seco-dammarane skeleton were determined on the basis of chemical and physicochemical evidence that included X-ray crystallographic analysis (Yoshikawa *et al.*, 1995). All were found to inhibit the histamine release from rat peritoneal exudate cells induced by compound 48/80.

Sapogenin and terpenoids – In the course of study on hovenia saponins, some sapogenins were obtained through chromatographic separation and chemical manners. Hovenolactone (7) was the genin of saponins E (5) and H (6) as well as of hoduloside I (8) and II (9), and its structure was confirmed by X-ray analysis (Kobayashi *et al.*, 1982). Sapogenin jujubogenin (25) and prosaponin 20-O-α-L-rhamnopyranosyljujubogenin (26) were obtained from hovenia saponin D (3) by mild alkaline degradation (Ogihara *et al.*, 1987). In addition, hovenidulcigenin (24) and hovenilactone were obtained in the course of the structure elucidation of hovenidulciosides A_1, A_2, B_1 and B_2 (Yoshikawa *et al.*, 1996).

Several aroma compositions were reported from the fresh leaves of *H. dulcis* var. *tomentella* Makino. These include monoterpenoids kenposide A (27), B (28) and sesquitepenoids icariside C_1 (29) (Yoshikawa *et al.*, 1993).

The information about the terpenoid compounds isolated from genus *Hovenia* is listed in Table 1, while their

Table 1. The information of compounds from genus *Hovenia*

No.	Compounds	Compound type	Plant materials	Species	References
1	Hovenoside I	dammarane-type triterpene saponin	root, barks	H. dulcis	Kawai *et al.*, 1974; Ogihara *et al.*, 1976; Inoue *et al.*, 1978
2~4	Saponin C_2, D, G	dammarane-type triterpene saponin	leaves	H. dulcis	Kimura *et al.*,1981
5, 6	Saponin E, H	dammarane-type triterpene saponin	leaves	H. dulcis	Kobayashi *et al.*, 1982
7	Hovenolactone	triterpene			
8~13	Hoduloside I, II, III, IV, and V, Jujuboside B	dammarane-type triterpene saponin	leaves	H. dulcis	Yoshikawa *et al.*, 1992
14~18	Hoduloside VI, VII, VIII, IX, and X	dammarane-type triterpene saponin	fresh leaves	*H. dulcis.* var. *tomentella*	Yoshikawa *et al.*, 1993a
2	Saponin C_2	dammarane-type triterpene saponin	leaves	H. acerba	Liang *et al.*, 1996a
19	Hovacerboside A_1				
20~ 23	Hovenidulcioside A1, A2, B1, and B2	dammarane-type triterpene saponin	seeds and fruits	H. dulcis	Yoshikawa *et al.*, 1996 Yoshikawa *et al.*, 1995
10	Hoduloside III	dammarane-type triterpene saponin			
24	Hovenidulcigenin	triterpene			
25	Jujubogenin	triterpene	Alkaline degradation from saponin D	H. dulcis	Ogihara *et al.*, 1987
26	20-O-α-L-rhamno pyranosyljujubogenin				
27, 28	Kenposide A, B	monoterpenoid	fresh leaves	*H. dulcis* var.*tomentella*	Yoshikawa *et al.*, 1993b
29	Icariside C_1	sesquiterpenoid			
30	Kaempferol	flavonol	leaves	H. acerba	Liang *et al.*, 1996b
31	Quercetin				
32	Iso-quercetin				
33	Kaempferol-3-O-α-L-rhamnopyranosyl(16)-α-D-galactopyranoside				
34	Kaempferol-3-O-rutinoside				
35	Quercetin 3-O-α-L-rhamnopyranosyl(1 → 6)-β-D-galactopyranoside				

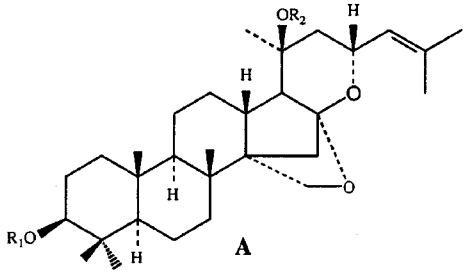
Table 1. The information of compounds from genus *Hovenia*

No.	Compounds	Compound type	Plant materials	Species	References
36	Rutin				
37	Dihydrokaempferol	dihydroflavonol	seeds	H. dulcis	Ding *et al.*, 1997
31	Quercetin	flavonol			
38	(+)-3,3',5',5,7-pentahydroflavanone	dihydroflavonol			
39	(+)-dihydromyricetin	dihydroflavonol			
30	Kaempferol	flavonol	seeds	H. acerba	Sha and Ding, 2001
40	Apigenin	flavone			
41	4',5,7-trihydroxy-3',5'-dimethoxy flavanone	flavone			
31	Quercetin	flavonol			
42	Myricetin	flavonol			
39	Dihydromyricetin	dihydroflavonol			
43	(+)-ampelopsin	dihydroflavonol	fruits	H. dulcis	Hase. *et al.*, 1997
44~46	Hovenitins I, II, and III	dihydroflavonol	seeds and fruits	H. dulcis	Yoshikawa *et al.*, 1997
43	(+)-ampelopsin,	dihydroflavonol			
47	Laricetrin	flavonol			
42	Myricetin	flavonol			
48	(+)-gallocatechin	flavanol			
49	Hovenodulinol	dihydroflavonol	fruits	H. dulcis.	Park *et al.*, 2002
50	Frangulanine	cyclopeptide alkaloid	root barks	H. dulcis	Makoto *et al.*, 1973 Kawai *et al.*, 1977
51	Hovenine A				
52	Perlolyrine	alkaloid	seeds	*H. dulcis.*	Park *et al.*, 1990
53	3-O-coumaroylquinic acid	organic acid	leaves	*H. acerba*	Liang *et al.*, 1997
54	4- hydroxyl-N-methylproline	amino acid			
55	Vanillic acid	organic acid		*H. dulcis*	Cho *et al.*, 2000
56	Ferulic acid				
55	Vanillic	aromatic acid	seeds	*H. acerba*	Sha and Ding, 2001
57	Emodin	anthraquinone			
58	Hovenic acid	triterpene	fresh barks	H. trichocarea	Yoshikawa *et al.*, 1998a
59	Hovetrichoside H	saponin			
60	Ceanothetric acid	triterpene			
61	(+)-lyoniresinol-3a-O-β-D-glucopyranoside	Phenolic glycosides			
62	(-)-lyoniresinol-3a-O-β-D-glucopyranoside	Phenolic glycosides			
63	Citrusin B	flavonoid			
64, 65	Hovetricoside A, B	Phenolic glycosides	barks	H. trichocarea	Yoshikawa *et al.*, 1998b
66~70	Hovetricoside C, D, E, F, G	Phenolic glycosides	barks	H. trichocarea	Yoshikawa *et al.*, 1998c
71	Acanthoside B	lignan			
72	Kelampayoside A	Phenolic glucoside			
73	Shashenoside I	Phenolic glucoside			
74	3,4,5-trimethoxyphenol-1-*O*-D-xylopyranosyl-(1 → 6)-D-glucopyranoside	Phenolic glucoside			

structures are shown in Fig. 1.

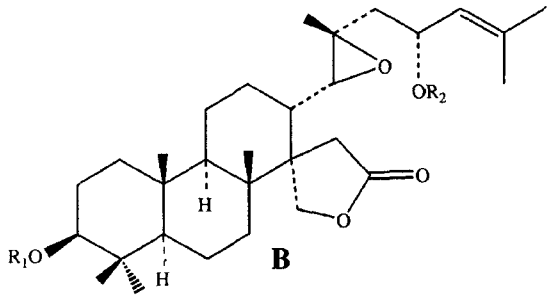
Flavonoids – Seven flavonoids were isolated from the leaves of *H. acerba* Lindl (Liang *et al.*, 1996b). They were identified as kaempferol (30), quercetin (31), iso-quercetin (32), kaempferol-3-O-L-rhamnopyranosyl (1 → 6)-α-D-galactopyranoside (33), kaempferol-3-O-rutinoside (34), quercetin3-O-α-L-rhamnopyranosyl (1 → 6)-α-D-galactopyranoside (35), and rutin (36). All of these were isolated from the genus *Hovenia* for the first time.

Four flavanoids were isolated from the seeds of *H. dulcis* Thunb (Ding *et al.*, 1997). Their structures were identified as dihydrokaempferol (37), quercetin (31), (+)-3,3',5',5,7-pentahydroflavanone (38) and (+)-dihydromyricetin (39) on the basis of physicochemical properties and



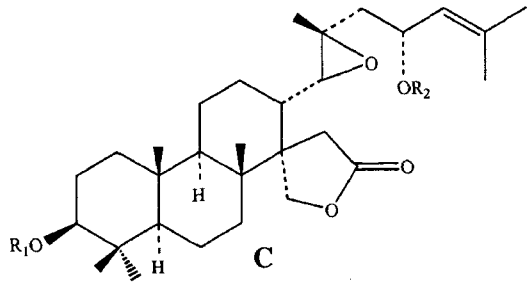
A

Compounds		R_1	R_2	References
Hovenoside I	(1)	2 α-ara-β-xyl 3 | β-glc	-Me	Inoue *et al.*, 1978 Yoshikawa *et al.*, 1992
Saponin C_2	(2)	2 α-ara-α-rha 3 | β-glc	-H	Kimura *et al.*, 1981 Yoshikawa *et al.*, 1992
Saponin D	(3)	2 -glc-rha	-Rha	Kimura *et al.*, 1981
Saponin G	(4)	β-glc	-Rha	Kimura *et al.*, 1981
Jujubogenin	(25)	-H	-H	Ogihara *et al.*, 1987
20-O-α-L-rhamnopyranosyl-		-H	-Rha	Ogihara *et al.*, 1987
Jujubogenin	(26)			
Hoduloside III	(10)	2 α-ara-β-qui 3 | β-glc	-H	Yoshikawa *et al.*, 1992
Hoduloside IV	(11)	2 α-ara-β-glc 3 | β-glc	-H	Yoshikawa *et al.*, 1992
Hoduloside V	(12)	2 β-glc-α-rha 3 | β-glc	-H	Yoshikawa *et al.*, 1992
Jujuboside B	(13)	3 2 α-ara-β-glc-β-xyl 2 | α-rha	-H	Yoshikawa *et al.*, 1992
Hovacerboside A_1	(19)	3 2 α-ara-β-glc-β-gal 2 | α-rha	-H	Liang *et al.*, 1996a



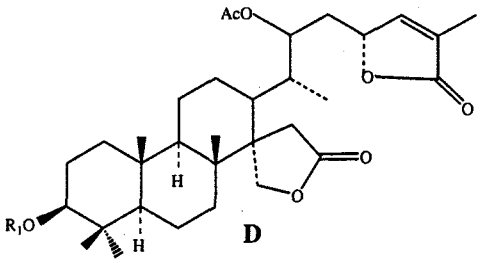
B

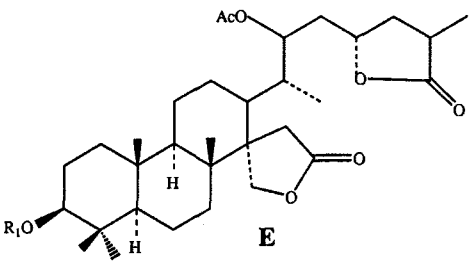
Compounds		R_1	R_2	References
Hoduloside VI	(**14**)	α-ara	β-glc	Yoshikawa *et al.*, 1993a
Hoduloside VII	(**15**)	2 α-ara-α-rha	β-glc	Yoshikawa *et al.*, 1993a
Hoduloside VIII	(**16**)	α-ara	6 β-glc-β-xyl	Yoshikawa *et al.*, 1993a
Hoduloside IX	(**17**)	2 α-ara-α-rha	6 β-glc-β-xyl	Yoshikawa *et al.*, 1993a
Hoduloside X	(**18**)	2 α-ara-α-rha 3 β-glc	β-glc	Yoshikawa *et al.*, 1993a



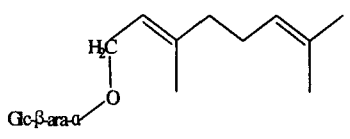
C

		R_1	R_2	References
Saponin E	(**5**)	2 β-glc-α-rha	-H	Kobayashi *et al.*, 1982
Saponin H	(**6**)	-β-glc	-H	Kobayashi *et al.*, 1982
Hovenolactone	(**7**)	-H	-H	Yoshikawa *et al.*, 1992
Hoduloside I	(**8**)	2 β-glc-α-rha	β-glc	Yoshikawa *et al.*, 1993
Hoduloside II	(**9**)	2 β-glc-α-rha 3 β-glc	-H	Yoshikawa *et al.*, 1993

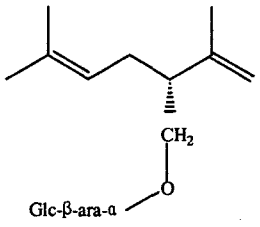




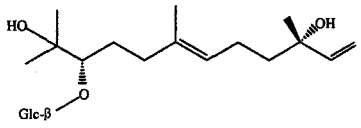
		R_1		References
Hovenidulcioside A_1	(**20**)	D-R_1	2 β-glc-α-rha	Yoshikawa *et al.*, 1996 Yoshikawa *et al.*, 1995
Hovenidulcioside A_2	(**21**)	D-R_1	β-glc	Yoshikawa *et al.*, 1996 Yoshikawa *et al.*, 1995
Hovenidulcigenin	(**24**)	D-R_1	H	Yoshikawa *et al.*, 1995
Hovenidulcioside B_1	(**22**)	E-R_1	2 β-glc-α-rha	Yoshikawa *et al.*, 1996 Yoshikawa *et al.*, 1995
Hovenidulcioside B_2	(**23**)	E-R_1	β-glc	Yoshikawa *et al.*, 1996 Yoshikawa *et al.*, 1995



Kenposide A (**27**)



Kenposide B (**28**)



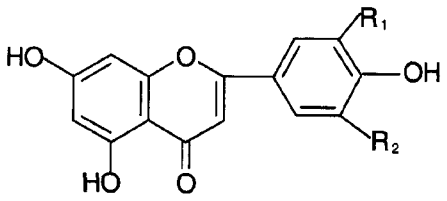
Icariside C_1 (**29**)

Fig. 1. Structures of terpenoids compounds from genus *Hovenia*.

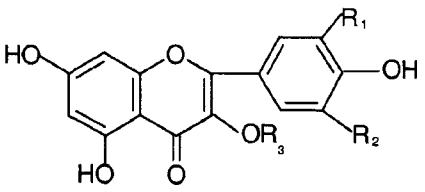
spectroscopic analyses. Among them, (**38**) was a new compound, while (**37**) and (**39**) were isolated from the genus *Hovenia* for the first time. Six flavanone compounds were isolated from the seeds of *H. acerba* Lindl. (Sha and Ding, 2001). These include kaemferol (**30**), apigenin (**40**), 4',5,7-trihydroxy-3',5'-dimethoxy flavanone (**41**), myricetin (**42**), quercetin (**31**), and dihydromyricetin (**39**). Compounds (**30**, **40**, **41**) were isolated from the seeds of *H. acerba* for the first time.

The hepatoprotective effects of the fruits of *H. dulcis* Thunb on chemically induced experimental liver injury models were investigated (Hase *et al.*, 1997). An active flavonoids constituent, (+)-ampelopsin (**43**) was obtained from the methanolic extract of the fruits. In the same year, the methanol-soluble fraction from the seeds and fruits of *H. dulcis* Thunb was found to show an inhibitory effect on the alcohol-induced muscular relaxation and a protective activity on the D-galactosamine-induced liver injury. Through bioassay-guided separation, three new dihydrofravonols named as hovenitins I (**44**), II (**45**), and III (**46**) were isolated together with four known flavonoids, (+)-ampelopsin (**43**), laricetrin (**47**), myricetin (**42**), and (+)-gallocatechin (**48**). The absolute stereostructures of hovenitins I, II, and III were determined on the basis of chemical and physicochemical evidence (Yoshikawa *et al.*, 1997). A new active flavonoid, hovenodulinol (**49**), was isolated from the fruits of *H. dulcis* Thunb (Park *et al.*, 2002). Its chemical structure was similar to hovenitins (**44-46**). Experiments on animals and clinical tests showed that hovenodulinol had excellent *in vivo* effects of alcohol and aldehyde decomposition, and showed excellent effect on alleviating lingering intoxication.

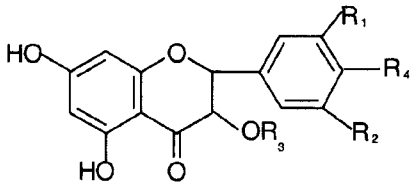
The information about the flavonoid compounds isolated



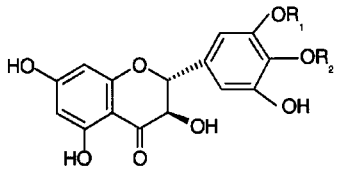
		R_1	R_2	References
Apigenin	(**40**)	-H	-H	Sha and Ding, 2001
4`,5,7-trihydroxy-3`,5`-dimethoxy flavanone	(**41**)	$-OCH_3$	$-OCH_3$	Sha and Ding, 2001



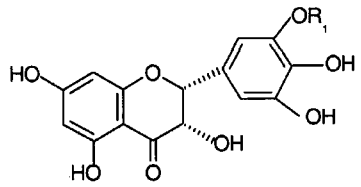
		R_1	R_2	R_3	References
Kaempferol	(**30**)	-H	-H	-H	Liang *et al.*, 1996b
Kaempferol-3-O-α-L-rhamnopyranosyl (1→6)-β-D-galactopyranoside	(**33**)	-H	-H	- α-rha-β-gal	Liang *et al.*, 1996b
Kaempferol-3-O-rutinoside	(**34**)	-H	-H	-Rutinose	Liang *et al.*, 1996b
Quercetin	(**31**)	-OH	-H	-H	Liang *et al.*, 1996b Sha and Ding, 2001
Iso-quercetin	(**32**)	-OH	-H	-Glucose	Liang *et al.*, 1996b
Quercetin-3-O-α-L-rhamnopyranosyl (1→6)-β-D-galactopyranoside	(**35**)	-OH	-H	- α-rha-β-gal	Liang *et al.*, 1996b
Rutin	(**36**)	-OH	-H	-Rutinose	Liang *et al.*, 1996b
Myricetin	(**42**)	-OH	-OH	-OH	Sha and Ding, 2001
Laricetrin	(**47**)	$-OCH_3$	-OH	-OH	Yoshikawa *et al.*, 1997



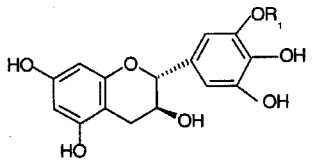
		R_1	R_2	R_3	R_4	References
Dihydrokaempferol	(**37**)	-H	-H	-H	-OH	Ding *et al.*, 1997
Dihydromyricetin	(**39**)	-OH	-OH	-H	-OH	Sha and Ding, 2001
(+)-3,3',5',5,7-pentahydroflavanone	(**38**)	-OH	-OH	-H	-H	Ding *et al.*, 1997



		R_1	R_2	References
(+) –ampelopsin	(**43**)	-H	-H	Hase *et al.*, 1997 Yoshikawa *et al.*, 1997
Hovenitin I	(**44**)	$-CH_3$	-H	Yoshikawa *et al.*, 1997
Hovenodulinol	(**49**)	$-CH_3$	$-CH_3$	Park *et al.*, 2002



		R_1	References
Hovenitin II	(**45**)	$-CH_3$	Yoshikawa *et al.*, 1997
Hovenitin III	(**46**)	- H	Yoshikawa *et al.*, 1997



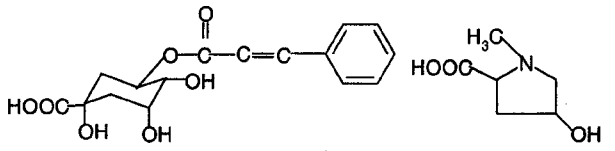
		R_1	References
(+)-gallocatechin	(**48**)	- H	Yoshikawa *et al.*, 1997

Fig. 2. Structures of flavonoids compounds from genus *Hovenia*.

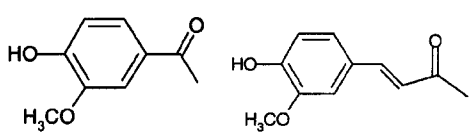
from genus *Hovenia* is listed in Table 1, while their structures are shown in Fig. 2.

Alkaloids and organic acids – Two cyclopeptide alkaloids, frangulanine (**50**) and hovenine A (**51**) were isolated from the root and bark of *H. dulcis* Thunb. and *H. dulcis* var. *tomentella* Makino (Makoto *et al.*, 1973). Alkaloid frangulanine showed ion selectivity on the induction of mitochondrial swelling (Kawai *et al.*, 1977). Another new alkaloid perlolyrine (**52**) was isolated from the seeds of *H. dulcis* Thunb (Park *et al.*, 1990).

An organic acid compound and an amino acid were isolated from the dry leaves of *H. acerba* Lind. By chemical and spectral analyses, they were identified as 3-O-coumaroylquinic acid (**53**) and 4-hydroxyl-N-methylproline (**54**), which were isolated from Rhamnaceae plant for the first time (Liang *et al.*, 1997). Two antioxidative and antimicrobial compounds were isolated and identified as vanillic acid (**55**) and ferulic acid (**56**) from the hot water extracts of *H. dulcis* Thunb (Cho *et al.*, 2000). More recently, an aromatic acid, vanillic acid (**55**) was also isolated from the seeds of *H. acerba* Lindl. for the first time (Sha and Ding, 2001).



3-O-coumaroylquinic acid (**53**) 4-hydroxyl-N-methylproline (**54**)



Vanillic acid (**55**) Ferulic acid (**56**)

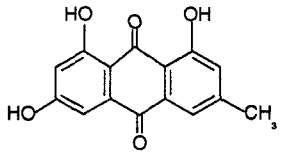
Others – An anthraquinone compound, emodin (**57**) was

isolated from the seeds of *H. acerba* Lindl (Sha and Ding, 2001).

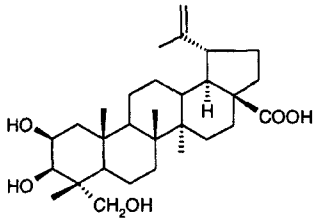
From the fresh bark of *H. trichocarea*, some new compounds including a new lupane-triterpene, hovenic acid (**58**) and an abeolupane glucoside, hovetrichoside H (**59**) were isolated together with ceanothetric acid (**60**), (+)-lyoniresinol-3a-O-β-D-glucopyranoside (**61**), (−)-lyoniresinol-3a-O-β-D-glucopyranoside (**62**) and citrusin B (**63**). The structures of these new compounds were established by extensive NMR experiments and chemical methods (Yoshikawa *et al.*, 1998a). In addition, Hovetrichosides A (**64**) and B (**65**), two 2-hydroxy-2-benzylcoumaranone glycosides, two neolignan glycosides and a phenylpropanoid glycoside were isolated from the bark of *H. trichocarea* (Yoshikawa *et al.*, 1998b). Compounds were named as hovetricosides C-G, (**66~70**) respectively, and their structures were established by extensive NMR experiments and chemical methods. This investigation also includes the known compounds acanthoside B (**71**), kelampayoside A (**72**), shashenoside I (**73**), and 3,4,5-trimethoxyphenol-1-*O*-β-D-xylopyranosyl-(1→6)-β-D-glucopyranoside(**74**) (Yoshikawa *et al.*, 1998c).

Perspective

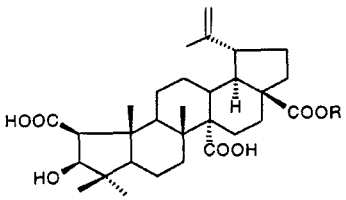
In summary, numerous chemical compositions were isolated and identified from various medicinal plants of the genus *Hovenia*, providing a sound base for further pharmacological development. The further isolation of



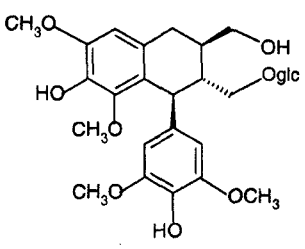
Emodin (**57**)



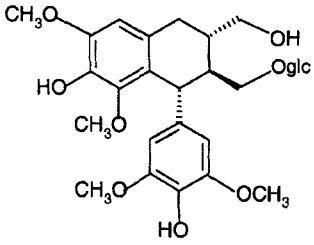
Hovenic acid (**58**)



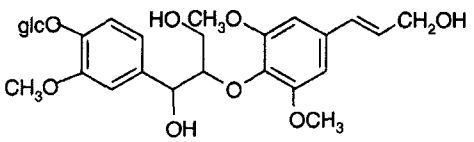
Hovetrichoside H (**59**): R -glc
Ceanothetric acid (**60**): R -H



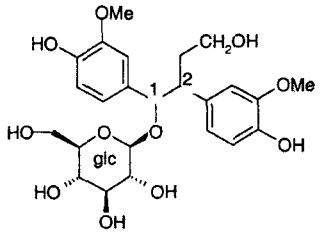
(**61**)

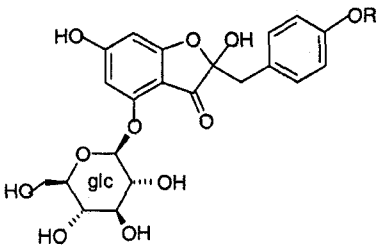


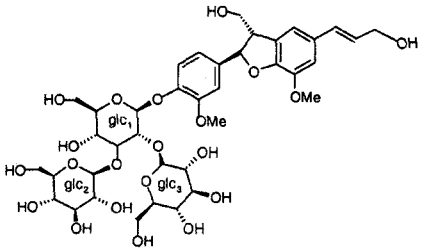
(**62**)



Citrusin (**63**)



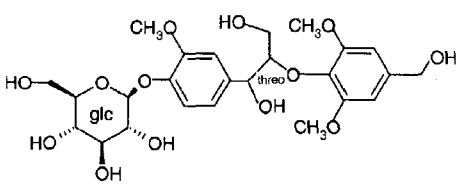




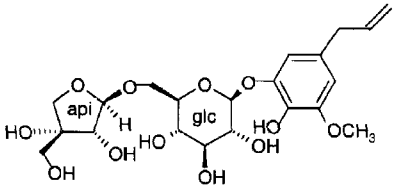
Hovetrichoside A (**64**) : 1R, 2S
Hovetrichoside B (**65**) : 1S, 2R

Hovetrichoside C (**66**) :R-H
Hovetrichoside D (**67**) :R-rha

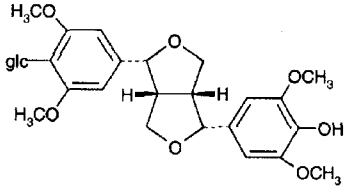
Hovetrichoside E (**68**)



Hovetrichoside F (**69**)



Hovetrichoside G (**70**)



Acanthoside B (**71**)

new active chemical compositions may prove to be a useful tool in alcoholic detoxification, hepatoprotective activity or other pharmacological activities.

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