THE OCCURRENCE OF IRIDOIDS IN PLANTS

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ABSTRACT

The occurrence of iridoids (iridoid glucosides, secoiridoids, valtrates, iridoids Plumeria) in the taxa of plants and their taxonomic significance are referred.

KEY WORDS: iridoids, taxonomy.

INTRODUCTION

Iridoids (formerly called pseudoindicanes or aucubinoides) are naturally occurring monoterpenic products of plants. Their characteristic feature is the presence in their structure of an α -pyran ring. The first iridoid – verbenalin (cornin) (I) was isolated as early as 1835 from *Cornus* (*Cornaceae*). However, the detailed structure of these compounds had not been determined until 1963. It was found out then, that the structure of iridoids resembled that of iridodial (II) – a compound isolated from ants living in Argentina, belonging to the genus *Iridomyrmex*. Iridodial was later detected also in the Australian plants belonging to the genus *Myoporum* (*Myoporaceae*). Two other iridoids were also isolated from the above mentioned ants: iridomyrmecin (III) and isoiridomyrmecin (IV). Some butterflies contain the iridoid glycosides, which are obtained from iridoid-containing plants via larval feeding (Stermitz et al. 1994). However the iridoids occur mainly in the plant kingdom.

The considerable interest in iridoids has been caused by their relatively wide spectrum of biological activity, discussed by the author in another paper (Andrzejewska-Golec – in press), as well as by their use as markers in the taxonomy of plants (Rimpler & Sauerbier 1986; Jensen et al. 1988; Inouye et al. 1988; Grabias & Świątek 1987; Readdy et al. 1990; Nicoletti 1991).

The best known works among the monographies discussing this group of compounds are those by El-Naggar and Beal of 1980 as well as a supplementary monography by Boros and Stermitz of 1990. They include the structural formulae, the properties allowing for the identification, families or even genera (and sporadically species) containing the particular iridoids, as well as the lists of references.

The presence of iridoids has not been hitherto observed in spore plants, gymnosperms and monocotyledons. According Damtoft et al. (1983) these compounds are characteristic of 6-8% dicotyledons.

The occurrence of iridoids in plant was the subject of works by Hegnauer (1969, 1986) who pointed out the significance of this group of compounds in plant taxonomy, but only in the cases of taxonomic units of a range higher than that of the species.

The isolation and the structure elucidation of the iridoids have been described many authors, for example Andrzejewska-Golec et al. 1993, Damtoft et al. 1994, Handjieva et al. 1993, Nicoletti 1989, Świątek et al. 1981.

The occurrence of iridoids has been discussed in detail (i.a.) in the following families: *Rubiaceae* (Inouye et al. 1988) *Lamiaceae* (Kooiman 1972), *Scrophulariaceae* (Kooiman 1970; Świątek 1973), *Acanthaceae* (Jensen 1988), *Plantaginaceae* (Andrzejewska-Golec & Świątek 1984; Andrzejewska-Golec 1994). Up to the present, over 300 iridoid glycosides, 60 secoiridoids, and 90 nonglycoside iridoids have been described (Junior 1990).

IRIDOID GLYCOSIDES

They contain a cyclopentan- α -pyran ring. The majority of them are α -D-glucosides. Nicoletti (1989) reported their presence in 107 genera belonging to 39 families. The iridoid glycosides occur mainly in the representatives of the orders: *Scrophulariales*, *Lamiales*, *Gentianales*, *Dipsacales*, *Ericales*, *Saxifragales*, *Hamamelidales*.

Aucubin (V) is the best known and most widespread compound belonging to this group. The name of the compound has been derived from that of the plant *Aucuba japonica* Thbg. (*Cornaceae*) which has been used up to the present as the source of this iridoid. Aucubin was previously called rhinanthin – from the genus *Rhinanthus* (*Scrophulariaceae*) where it was first detected by Ludwig in 1968. El-Naggar and Beal (1986) report its occurrence in 13 families in dicotyledons. It is particularly common in such families as *Scrophulariaceae* and *Plantaginaceae*. Among 40 species of

STRUCTURAL FORMULAE OF IRIDOIDS

I.
cornin

II.
iridodial

III.
iridomyrmecin

IV.
isoiridomyrmecin

V.
aucubin

VI.
bartsioside

VII. catalpol

VIII. ajugol

IX. R = H harpagide
XXIII. R = t - cinnamoyl harpagoside

X.
asperuloside

STRUCTURAL FORMULAE OF IRIDOIDS

XI.
melittoside

XII.
geniposidic acid

XIII.
plantarenaloside

XIV.
oleoeuropin

XV.
gentiopicroside

XVI.
swertiamarin

XVII.
loganine

XVIII. gardenoside

XIX. geniposide

XX. ixoroside

XXI. valtrate

XXII. plumericin

Scrophulariaceae only in 4 aucubin has not been detected (Świątek 1973). Among 38 investigated species of the genus *Plantago*, the presence of this iridoid has not been detected only in 1 species (Andrzejewska-Golec & Świątek 1984). In the case of the *Scrophulariaceae* family, the presence of aucubin is the significant characteristics of the representatives of the genera *Verbascum* and *Scrophularia*, and its absence of the taxa of the genus *Digitalis*. The presence of cardenolide glycosides in the genus *Digitalis* excludes that of iridoids.

Bartsioside-6-deoxyaucubin (VI) had been known up to recent times as occurring only in *Scrophulariaceae* family. This iridoid was detected for the first time in *Bartsia trixago* L. (Bianco et al. 1976), then in *B. viscosa* L., *Lathrea squamaria* L., two species of the genus *Penstemon*, two species of the genus *Castilleja* and in species *Monochasma savatieri* Franchet. et Maxim (Bianco et al. 1984; Junior 1984; Arslanian 1985; Roby & Stermitz 1986; Yahara et al. 1986; Belofsky & Stermitz 1988). Recently it has been determined that bartsioside also occurs in the representatives of the *Plantaginaceae* family (Andrzejewska-Golec et al. 1993), which seems to have originated from the *Scrophulariaceae* family (Hegnauer 1969, Takhtadzjan 1987).

Subgenus *Plantago* one of the subgenera of *Plantaginaceae* family – is characterized by the presence of aucubin, while the other one – subgenus *Psyllium* (Miller) Harms contains bartsioside. Thus, a marked biochemical differentiation can be observed here.

An epoxide derivative of aucubin – catalpol (VII) occurs in the taxa of the orders *Scrophulariales*, *Lamiales*, *Gentianales* which are regarded by Takhtadzjan (1987) as related ones. Catalpol was detected for the first time by Duff et al. in 1965 in the leaves of *Plantago lanceolata* L. (*Plantaginaceae*). Its name has been derived from the plant *Catalpa bignonioides* Walt. (*Bignoniaceae*) in whose fruit a catalpol ester – catalposide – was detected in the second half of the 19th century. The presence of catalpol has been described in over 30 genera of the *Scrophulariaceae* family. Its also characteristic some sections of subgenus *Plantago* of the genus *Plantago* (Andrzejewska-Golec & Świątek 1984).

The iridoid glucosides common in both *Scrophulariaceae* and *Lamiaceae* families are ajugol (VIII) and harpagide (IX) (Kooiman 1970, 1972; Świątek 1973; Grabias & Świątek 1987).

Asperuloside (X) isolated as early as that second half of the 19th century is particularly well known for the presence in the *Rubiaceae* family. It has been formed so because it was detected for the first time in the genus *Asperula*. It also occurs in the *Globulariaceae* and *Ericaceae* families. Bianco et al. (1984) isolated it from the flowers of the representatives of *Plantaginaceae* family: *Plantago major* L. and *P. lanceolata* L.

Melittoside (XI) can sure as an example of iridoid diglycosides. It was isolated for the first time from *Melittis melissophyllum* L. (*Lamiaceae*) in 1967. It also occurs in the *Plantaginaceae* family (Świątek et al. 1981; Andrzejewska-Golec & Świątek 1984).

Geniposidic acid (XII) is characteristic of the genera *Genipa* and *Ixora* of the *Rubiaceae* family. It has been also known for the presence in the taxa of the *Scrophulariaceae*, *Verbenaceae* and *Bignoniaceae* families. It has also been detected in the species of the *Plantaginaceae* family occurring in East Asia: *Plantago japonica* Franchet et Savat. and *P. asiatica* L. (Tsumura Jutendo Co 1981).

Plantarenalosite (yuheinoside) (XIII) was first isolated by Popov in 1978 from the herb *Plantago arenaria* Waldst. et Kit. (*Plantaginaceae*). So far its presence has been described

in ten species of the *Scrophulariaceae* family, five of the *Bignoniaceae* family, three of the *Verbenaceae* family and four of the *Plantaginaceae* family (Ozaki et al. 1979; Bianco et al. 1981, 1985; Popov et al. 1981; Sticher & Salama 1981; Jacke & Rimpler 1983, Andrzejewska-Golec & Świątek 1984; Berg et al. 1985; Garbarino & Gambara 1985; Roby & Stermitz 1986; Stenzel et al. 1986; Lira-Rocha et al. 1987; Takeda 1988; Davioud et al. 1989; Debrauwer et al. 1989; Junior 1989; Afifi et al. 1990). All the four families are described by Dahlgren (1980) as belonging to the subordo *Lamiiflorae* and by Ehrendorfer (1983) as belonging to the subordo *Scrophularianae*.

Similarly as bartsioside, plantarenalosite does not occur in the taxa of the subgenus *Plantago* (Andrzejewska-Golec & Świątek 1984).

SECOIRIDOIDS

This group of iridoids contains, beside a α -pyran ring also a lactone ring. Secoiridoids are characteristic among others the following orders: *Dipsacales* (*Dipsacaceae*, *Caprifoliaceae*), *Oleales* (*Oleaceae*), *Gentianales* (*Gentianaceae*, *Menyanthaceae*, *Apocynaceae*, *Loganiaceae*), *Cornales* (*Cornaceae*).

Nicoletti (1989) describes their occurrence in 30 genera of 17 families. The best known secoiridoids are: oleoeuropein (XIV) obtained from the leaves and fruit of *Olea europea* L. (*Oleaceae*), as well as gentiopicroside (XV) – the main iridoid present in the roots of plants belonging to the genus *Gentiana* (*Gentianaceae*) (Do et al. 1987). The main iridoid found in *Erythraea centaurium* (L.) Pers. (*Gentianaceae*) is swertiamarin (XVI) (Do et al. 1987). Loganine (XVII) occurs in the families: *Apocynaceae*, *Caprifoliaceae*, *Cornaceae*, *Loganiaceae* and *Menyanthaceae*.

In the *Rubiaceae* family it has been found that the presence of secoiridoids is characteristic of particular subfamilies (*Cinchonoideae*, *Antirrhoeoideae*) while others contain iridoid glucosides [*Ixoroideae*: gardenoside (XVIII), geniposide (XIX) and ixoroside (XX); *Rubioideae*: asperuloside (X)] (Inouye et al. 1988).

VALEPOTRATES

Valepotrates (XXI) are a group of nonglycoside iridoids. They have the structure of triesters. They are characteristic of the *Valerianaceae* family – ordo *Dipsacales* (Bach et al. 1993).

IRIDOIDS PLUMERIA

They are also a group of non-glycosidic iridoids. The best known representative of this group is plumericin (XXII) found in *Plumeria* (*Apocynaceae*-*Gentianales*).

CONTENT OF IRIDOIDS IN VARIOUS PARTS OF THE PLANTS

It should be noted that various parts of the plants may differ quantitatively and qualitatively as far as the content of iridoids is concerned. Thus according to Świątek et al. (1984), the presence of harpagoside (XXIII) is characteristic for the roots

of plants belonging to the genus *Verbascum* of *Scrophulariaceae* family, while aucuboside (V) and catalpol (VII) occur in all the organs of the plant, the former mainly in vegetative and the latter in generative ones. The seeds contain only small quantities of iridoids.

Noro et al. (1990) observed that the overground parts of *Plantago asiatica* L. (*Plantaginaceae*) contained more aucubin (V) than the underground ones, and young leaves more than the older ones. According to Bowers and Stamp (1992) the contents of aucubin (V) and catalpol (VIII) in *Plantago lanceolata* is determined genetically and it also depends on the age of the leaves.

Moreover, according to Świątek et al. (1984) the annual leaves of plants *Verbascum phlomoides* L. and *V. thapsiformae* Schrad. (*Scrophulariaceae*) are richer in iridoid than biannual leaves.

In the cases of biocenotic parasite host interactions, the transmission of iridoids from the roots of the host plant to the parasite can be observed. The transmission of the main iridoid of *Penstemon teucrioides* – aucubin (V) to *Castilleja integra* has been observed by Stermitz et al. (1993).

BIOSYNTHESIS OF IRIDOIDS IN PLANTS

It has been described in numerous monographic publications (Jensen et al. 1975; Buzogany & Cucu 1983; Hegnauer 1986; Nicoletti 1989).

Damtoft and collaborators (Damtoft 1983, Damtoft et al. 1983) think that aucubin in *Plantago major* L. (*Plantaginaceae*) as well as in *Scrophularia racemosa* and *Antirrhinum majus* L. (*Scrophulariaceae*) is synthesized from 8-epiloganic acid, which is, according to these authors, characteristic of the plants belonging to the subordo *Lamiiflorae* whereas the iridoids present in the plants belonging to *Corniflorae*, *Gentianiflorae* and *Loasiflorae*, such as asperuloside, secoiridoids and iridoid alkaloids are synthesized from deoxyloganic acid.

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ROZPOWSZECHNIENIE IRYDOIDÓW W ROŚLINACH

STRESZCZENIE

Opisano występowanie w roślinach różnych grup irydoidów zwracając uwagę na ich taksonomiczne znaczenie, a także jakościowe i ilościowe różnice w zawartości tego typu związków w poszczególnych organach roślinnych oraz ich biosyntezę.

SŁOWA KLUCZOWE: irydoidy, taksonomia.