ALKALOIDS OF *Liriodendron tulipifera*

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UDC 547.944/945

The alkaloids of the tulip tree *Liriodendron tulipifera* L., family Magnoliaceae, are considered. More then 20 alkaloids have been isolated during different vegetation periods from various organs of the plant growing in Uzbekistan, and these have been assigned to the aporphine alkaloids and their dehydro, oxo, and 7-hydroxy derivatives; only two alkaloids proved to be derivatives of proaphorphine and of tetrahydroberberine. On the basis of the results of a comparative study of the NMR spectra of aporphines unsubstituted in ring D and some chemical transformations, the structure and configuration of the (R)-3-hydroxy-1,2-dimethoxyaporphine have been proposed for the new alkaloid lirinine. The absolute configurations, possible biogenetic interconnections, and mutual transitions of the alkaloids of *L. tylipifera* that are derivatives of aporphine, oxoaporphine, and dehydroaporphine are discussed. A summery table is given which includes 41 alkaloids found in this plant.

The tulip tree *Liriodendron tulipifera* (in the country of its origin — the USA — it is also called the yellow poplar) is one of the most interesting representatives of the Magnoliaceae family. At the present time it is being widely cultivated in Central Asia, including Uzbekestan.

The biology, anatomy, morphology, and methods of propagating *Tulipifera* have been studied by F. M. Murzova [1]. In the dendrarium of the Botanical Garden of the Uzbek SSR Academy of Sciences there are tulip trees aged 27-30 years 30-60 m high with trunks 50-60 cm in diameter covered with grey finely fissured cork. Depending on the meteorological conditions in winter and spring, the flowering of the tulip tree takes place either in the second half of April and throughout May or at the beginning of May and the first half of June and lasts 45-48 days. The leaf buds open in April, and complete leaf formation is complete by the end of April, or the beginning of the autumn and winter. Mass leaf-fall of the tulip tree begins in October-November, The tree is propogated by seeds.

The presence of alkaloids in the leaves of *L. tulipifera* was first determined in 1935 by A. P. Orekhov [2]. In 1960, M. A. Buchanan and E. E. Dickey [12] and, somewhat later, M. Taylor [13] isolated from the wood of this species glaucine (XXV), and also liriodenine (XXX) and O-methyllatheroline (XXXV), which are representatives of the 7-oxodibenzo[de, g]quinoline (7-oxoaporphine) group.

A more detailed chemical investigation of the tulip tree was begun in 1973 by workers at the Institute of the Chemistry of Plant Substances of the Uzbek SSR Academy of Sciences [6-9, 16-23]. Alkaloids were detected on all the organs of the plant. More than 20 alkaloids were isolated from various organs of *L. tulipifera* in different periods of vegetation (Table 1).

Later, abroad, the alkaloids of this plant were studied by A. Chen and coworkers [29, 32, 37-50]. More than 40 alkaloids were isolated from *L. tulipifera* including 22 new ones (Table 2).

The alkaloids of *L. tupilifera* are aporphine alkaloids or their dehydro, oxo, or 7-hydroxy derivatives, only two bases (N-methylcrotsparine and isocorypalmine) having proved to be derivatives of proaporphine and of tetrahydroprotoberberine, respectively. The larger number of alkaloids isolated belongs to the aporphine group, and of the 20 alkaloids of this group 17 proved to be new. The structures and absolute configurations of the new alkaloids have been established on the basis of the results of a study of UV, IR, NMR, and mass spectra, in combination with chemical transformations.

Institute of the Chemistry of Plant Substances of the Uzbek SSR, Academy of Sciences, Tashkent. Tashkent Agricultural Institute. Translated from Khimiya Prirodnykh Soedenii, No. 5, pp. 628-638, September-October, 1987. Original article submitted January 28, 1987.

0009-3130/87/2305-0521$12.50

TABLE 1. Alkaloids Present in *Liriodendron tulipifera*

Plant organ	Vegetation period, date of collection	Total amt. of alkaloids, %	Alkaloids isolated
1	2	3	4
Leaves	Beginning of flowering, May 5, 1972	0,32	
Leaves	Flowering, May, 1978	0,28	Remerine, lirinidine, anonaine, remerine N-oxide, isocorypalmine, N-methylcrotsparine, lirinine, N-methyllaurotetanine
Leaves	Fruit-bearing, September 28, 1972	0,22	Remerine, d-nornuciferine, lirinine, d-caaverine, lirinidine, O-methyllirinine, lirinine N-oxide, lanuginosine, isolaureline
Leaves	Withering, November 3, 1972	0,11	Remerine, liriodenine, O-methylmoschatoline (liridine), lysicamine, d-caaverine, lirinidine, d-isolaureline, N-methylcrotsparine
Wood	End of flowering, May 30, 1972	0,15	Liriodenine, lysicamine, O-methylatheroline, glaucine, remerine, dehydroremerine
Bark	The same	0,11	Liriodenine, lysicamine, lanuginosine, remerine
Young flowers	Flowering, May 10, 1974	0,12	Lirinidine, caavarine, predicentrine, asimilobine, liriodenine, dehydroremerine, dehydroisolaureline
Petals	End of flowering, May 30, 1972	0,10	
Seeds	Withering, Ocotber 29, 1972	0,08	

TABLE 2. Alkaloids of *Liriodendron tulipifera*

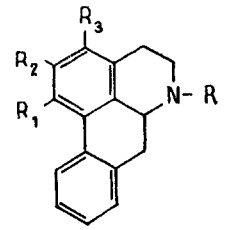
Alkaloid	Structure	Configuration	Literature
1	2	3	4
	I. Aporphines		
I. Anonaine $C_{17}H_{15}NO_2$, mp 120-122° (decomp.) $[\alpha]_D$ −60° (ethanol)	 $R=R_3=H$; $R_{1,2}=CH_2O_2$		3, 23, 33
II. N-Acetylanonaine $C_{13}H_{17}NO_3$, mp 229-230° (ethanol) $[\alpha]_D$ −356° (chloroform)	$R=COCH_3$; $R_{1,2}=CH_2O_2$; $R_3=H$	R	24, 45
III. Remerine $C_{18}H_{17}NO_2$, mp. 263-264° (hydrochloride (decomp.) $[\alpha]_D$ +68° (ethanol)	$R=CH_3$; $R_{1,2}=CH_2O_2$; $R=CH_3$	S	3, 6, 26
IV. Remerine N-oxide $C_{18}H_{17}NO_3$, mp. 164-165° (acetone)		S	23
V. Caaverine $C_{17}H_{17}NO_2$, mp. 207-209° (acetone), $[\alpha]_D$ +95° (methanol)	$R=R_3=H$; $R_1=OH$; $R_2=OCH_3$	S	7,27
VI Asimilobine $C_{17}H_{17}NO_2$, mp. 175-176° (acetone), $[\alpha]_D$ −210° (chloroform)	$R=R_3=H$; $R_1=OCH_3$; $R_2=OH$	R	21, 22,28
VII. N-Acetylasimilobine $C_{19}H_{19}NO_3$, mp. 280-281° (decomp.), $[\alpha]_D$ −405° (pyridine)	$R=COCH_3$; $R_3=H$; $R_1=OCH_3$; $R_2=OH$	R	24
VIII. Nornuciferine $C_{18}H_{19}NO_2$, mp. 128-129° (acetone) $[\alpha]_D$ +140° (ethanol), $[\alpha]_D$ −145° (ethanol)	$R=R_3=H$; $R_1=R_2=OCH_3$	S, R	15, 16, 29, 30
IX. N-Acetylnornuciferine $C_{20}H_{21}NO_2$, mp. 229-230° (acetone) $[\alpha]_D$ −406° (chloroform)	$R_1=R_2=OCH_3$; $R_3=H$; $R=COCH_3$	R	24, 32
X. Nuciferine $C_{19}H_{21}NO_3$, mp. 165-166°, $[\alpha]_D$ −152° (ethanol)	$R=CH_3$; $R_3=H$ $R_1=R_2=OCH_3$	R	29, 33
XI. Lirinidine $C_{18}H_{19}NO_2$, mp. 237-239° (hydrochloride (decomp.)) $[\alpha]_D$ +79° (chloroform)	$R=CH_3$; $R_1=OH$ $R_2=OCH_3$; $R_3=H$	S	7, 34, 63
XII. Lirine $C_{19}H_{21}NO_3$, mp. 152-154° (ethanol), $[\alpha]_D$ −55° (chloroform)	$R=CH_3$; $R_1=R_2=OCH_3$, $R_3=OH$	R	9, 37, 64
XIII. Lirinine N-oxide $C_{18}H_{21}NO_4$, mp. 162-164° (acetone), $[\alpha]_D$ −49° (methanol)		R	3, 14, 37, 64
XIV. O-Methylirinine $C_{20}H_{23}NO_3$, mp. 105-106° $[\alpha]_D$ −52° (chloroform)	$R=CH_3$; $R_1=R_2=R_3=OCH_3$	R	14, 38, 39
XV. Liridine $C_{19}H_{21}NO_3$, mp. 140-142° (acetone) $[\alpha]_D$ −38° (chloroform)	$R_1=R_3=OCH_3$; $R_2=OH$; $R=CH_3$	R	17, 37
XVI. O-Methylnorlirinine $C_{19}H_{21}NO_3$	$R=H$ $R_1=R_2=R_3=OCH_3$	R	37
XVII. Tuliferoline $C_{21}H_{23}NO_4$, mp. 145-146° (ethanol–hexane), $[\alpha]_D$ −330° (chloroform)	$R=COCH_3$; $R_1=R_2=R_3=OCH_3$	R	24, 40

TABLE 2. (Continued.)

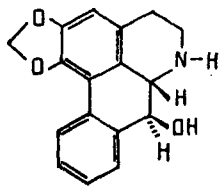
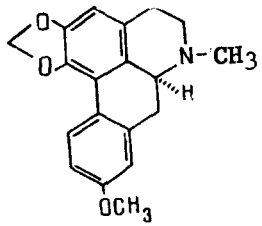
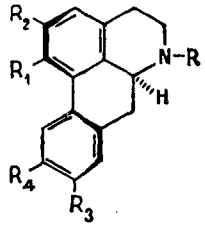
1	2	3	4
XVIII. Norushinsunine $C_{17}H_{15}NO_3$, mp. 204-205°, $[\alpha]_D$ −105° (chloroform)			32, 35, 36
XIX. Isolaureline $C_{19}H_{29}NO_3$, mp. 244-245° (hydrochloride), $[\alpha]_D$ + 35° (water)		S	3, 18, 19
			
XX. Thalicmidine $C_{20}H_{23}NO_4$, mp. 192-193° (methanol) $[\alpha]_D$ + 44° (ethanol)	$R=CH_3$; $R_1=OH$ $R_2=R_3=R_4=OCH_3$	S	3, 29, 39
XXI. Predicentrine $C_{20}H_{23}NO_4$, mp. 208-210° (hydrochloride) $[\alpha]_D$ + 97° (ethanol)	$R=CH_3$; $R_2=OH$ $R_1=R_3=R_4=OCH_3$	S	22, 29, 41
XXII. N-Methyllaurotetanine $C_{20}H_{23}NO_4$, mp. 237-238° (hydrobromide), $[\alpha]_D$ + 88° (chloroform)	$R=CH_3$; $R_3=OH$ $R_1=R_2=R_4=OCH_3$	S	29, 42, 31, 39
XXIII. Lirioferine $C_{20}H_{23}NO_4$, mp. 173-174° (ethyl acetate) $[\alpha]_D$ + 128° (chloroform)	$R=CH_3$; $R_4=OH$ $R_1=R_2=R_3=OCH_3$	S	29
XXIV. Norglaucine $C_{20}H_{23}NO_4$, mp. 221-222° (decomp.), $[\alpha]_D$ + 102° (methanol)	$R=H$ $R_1=R_2=R_3=R_4=OCH_3$	S	29, 44, 45
XXV. Glaucine $C_{21}H_{25}NO_4$, mp. 117-119° (ethyl acetate), $[\alpha]_D$ + 84° (ethanol)	$R=CH_3$ $R_1=R_2=R_3=R_4=OCH_3$	S	3, 10, 29, 32
XXVI. Boldine $C_{21}H_{21}NO_4$, mp. 161-161° (acetone), $[\alpha]_D$ + 111° (ethanol)	$R=CH_3$ $R_1=R_4=OH$; $R_2=R_3=OCH_3$	S	29, 30
XXVII. N-Acetylnormantenine $C_{21}H_{21}NO_5$, mp. 283-284 (ethanol), $[\alpha]_D$ + 340° (chloroform)	$R=COCH_3$ $R_1=R_2=OCH_3$; $R_{3,4}=O_2CH_2$	S	24, 46, 47
XXVIII. Liriotulipiferine $C_{19}H_{21}NO_4$, mp. 184-185° (ethanol), $[\alpha]_D$ + 184° (CCl_4)	$R=CH_3$ $R_1=R_3=OCH_3$; $R_2=R_4=OH$	S	29

TABLE 2 (Continued.)

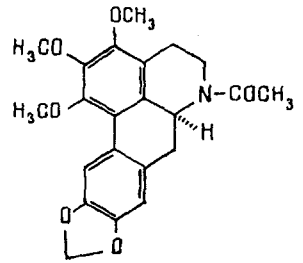
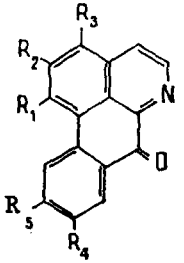
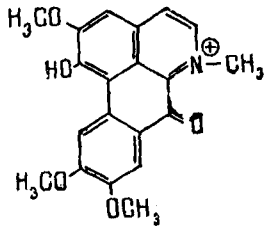
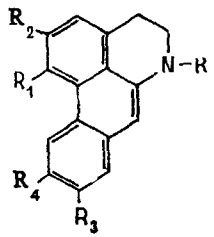
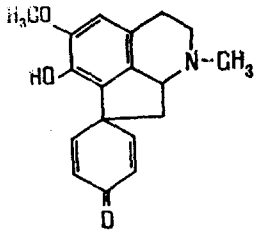
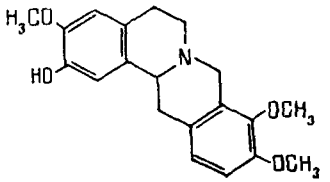
1	2	3	4
XXIX. N-Acetyl-3-methoxynornantenine $C_{22}H_{23}NO_6$, mp. 216-217° (ethanol), $[\alpha]_D$ + 271° (chloroform)		S	29, 46, 48
	II. Oxoaporphines		
			
XXX. Liriodenine $C_{17}H_9NO_3$, mp. 273-275° (decomp.)	$R_{1,2}=O_2CH_2$ $R_3=R_4=R_5=H$		3, 6, 10
XXXI. Lysicamine $C_{18}H_{13}NO_3$, mp. 208-210 (ethanol; decomp.)	$R_1=R_2=OCH_3$ $R_3=R_4=R_5=H$		3,18, 49
XXXII. Liriodendronine $C_{16}H_9NO_3$, mp. 265-270° (chloroform; decomp.)	$R_1=R_2=OH$ $R_3=R_4=R_5=H$		50
XXXIII. N-Methylmoschatoline (liridine) $C_{19}H_5NO_4$, mp. 161-163° (chloroform)	$R_1=R_2=R_3=OCH_3$ $R_4=R_5=H$		8, 51—53
XXXIV. Lanuginosine $C_{18}H_{11}NO_5$, mp. 319-321° (ethanol; decomp.)	$R_{1,2}=CH_2O_2$; $R_4=OCH_3$; $R_3=R_5=H$		17, 30, 54, 55
XXXV. N-Methylatheroline $C_{20}H_{17}NO_5$, mp. 220-222° (chloroform)	$R_3=H$ $R_1=R_2=R_4=R_5=OCH_3$		12, 16, 19
XXXVI. Corunnine $C_{20}H_{17}NO_5$, mp. 255-257° (ethanol)			29, 30, 56

TABLE 2. (Continued.)

1	2	3	4
	III. Dehydroaporphines		
			
XXXVII. Dehydroremerine $C_{18}H_{15}NO_2$, mp. 88-89° (benzene)	$R=CH_3$; $R_{1,2}=O_2CH_2$ $R_3=R_4=H$		22, 33
XXXVIII. Dehydroisolaurenine $C_{19}H_{17}NO_3$, mp. 143-145 (benzene)	$R=CH_3$; $R_{1,2}=O_2CH_2$ $R_3=OCH_3$; $R_4=H$		22
XXXIX. Dehydroglaucine $C_{21}H_{25}NO_4$, mp. 133-134° (benzene)	$R=CH_3$ $R_1=R_2=R_3=R_4=OCH_3$		29, 47
	IV. Proaporphine		
XL. N-Methylcrotsparine $C_{18}H_{19}NO_3$, mp. 222-224° (acetone) $[\alpha]_D$ − 39° (chloroform)			16, 58
	Tetrahydroprotoberberine alkaloid		
XLI. Isocorypalamine $C_{20}H_{23}NO_4$, mp. 220-222° (methanol)			43, 65, 66

The aporphine alkaloids were isolated mainly from the green leaves of the tulip tree collected in the flowering and fruit-bearing stages. The aporphines isolated are representatives of this group unsubstituted or monosubstituted in ring D and 1,2,9,10-tetrasubstituted. However, the 1,2,10,11-tetrasubstituted aporphines of the corydeine group have not yet been found in this plant.

It is known that aporphines unsubstituted and monosubstituted in ring D are usually levorotatory with the (R) absolute configuration [67]. In addition to the levorotatory aporphines of this type, caaverine (V), lirinidine (XI), nornuciferine (VIII), and remerine (III), which are dextrorotatory, with the (S) absolute configuration, have also been isolated from *L. tulipifera*. All the 1,2,9-10- tetrasubstituted aporphines found are dextrarotatory and, consequently, have the (S) absolute configuration.

Together with aporphine alkaloids (III, V, XI, XII, XIX), the leaves contain oxoaphorphine bases corresponding to them — liriodenine (XXX), lysicamine (XXXI), O-methylmoschatoline (XXXIII), and lanuginosine (XXXIV), and also a proaporphine alkaloid — N-methylcrotsparine (LX).

Oxo- and dehydroaporphine alkaloids have been found in the wood and the bark. It must be mentioned that in the green leaves the main alkaloid is remerine (III), while yellowed leaves contain a smaller amount of remerine than liriodenine.

Passages have been performed from the aporphines (III, VIII, IX, XIV) to the oxoaporphines (XXX, XXXI, XXXIII, and XXXIV), the dehydroaporphines (XXXVII, XXXVIII), and the proaporphine (LX), and from the oxoaporphine alkaloids (XXX, XXXI) to the aporphine alkaloids (III, VIII), and so on [16, 17, 23].

The detection in *L. tulipifera* of the aporphine alkaloids and the oxo, dehydro, and hydroxy derivatives corresponding to them show that redox processes take place in the plant organism between which there is a definite relationship, and that there are mutual transitions that depend on the vegetation period and the organ of the plant [4, 5, 22].

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