N-GLYCOSIDES.
9.* REACTION OF 3,5-O-ISOPROPYLIDENEXYLO-FURANOSYLAMINE WITH β-ISOTHIOCYANATOALDEHYDES. SYNTHESIS OF NEW CYCLONUCLEOSIDES WITH A HEXAHYDROPYRIMIDINE AGLYCONE

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The reaction of 3,5-O-isopropylidenexylofuranosylamine p-toluenesulfonate with β-isothiocyanatoaldehydes in the presence of triethylamine gives 4,2′-anhydro-4-hydroxy-3-(3′,5′-O-isopropylidene-α-D-xylofuranosyl)hexahydropyrimidine-2-thiones. The structure of these compounds and their deblocking products was studied by IR, UV, PMR, and ORD spectroscopy and mass spectrometry.

Nucleosides with a partially or completely hydrogenated aglycone have not been studied extensively. Some such compounds display high biological activity. Thus, for example, tetrahydrouridine (4-hydroxy-1-β-D- ribofuranosylhexahydropyrimidin-2-one) and 1-β-D-ribofuranosylhexahydroxypyrimidin-2-one are effective cytidinedeaminase inhibitors [2, 3], 5,6-dihydro-5-azacytidine, 3-desazauridine, and 3-desazacytidine have anti-tumor activity [4], and 3-desazauridine triphosphate is an inhibitor of cytidine triphosphate synthetase [4].

A number of approaches have been reported for the synthesis of nucleosides with a hexahydropyrimidine aglycone. Thus, tetrahydrouridine was obtained by the reduction of uridine [5], while 1-β-D-ribofuranosylhexahydropyrimidin-2-one was synthesized using the condensation of 2,3,5-tri-O-benzoylribofuranosyl bromide with the trimethylsilyl derivative of hexahydropyrimidin-2-one in the presence of HgO—$HgBr_2$ [2]. Two alternative methods have also been reported for the preparation of 4-hydroxyhexahydropyrimidine-2-thiones N-glycosides based on the reaction of peracetyl glycosylisothiocyanates with β-aminocarbonyl compounds [6, 7] or reaction of glycosylamines with β-isothiocyanatocarbonyl compounds [8].

In previous work [9-11], we have shown that 4-hydroxy-3-ribofuranosylhexahydropyrimidine-2-thiones, which have a free 5′- or 2′-hydroxyl group in the sugar residue are capable of converting to the corresponding cyclonucleosides as result of intramolecular dehydration, 4,5′-anhydro-4-hydroxy-3-(β-D-ribofuranosyl)hexahydropyrimidine-2-thiones or 4,2′-anhydro-4-hydroxy-3-(α-D-ribofuranosyl)hexahydropyrimidine-2-thiones. In a continuation of our previous work [6-11] and in a search for methods for the preparation of new biologically active nucleosides with a hexahydropyrimidine aglycone, we undertook the synthesis of 4-hydroxy-3-xylosylhexahydropyrimidine-2-thiones and studied the conversion of these compounds in the corresponding cyclonucleosides.

The condensation of 3,5-O-isopropylidene-D-xylofuranosylamine, containing a free hydroxyl group at $C_{(2)}$, with β-isothiocyanatoaldehydes (IIa)-(IIc) was used to obtain these compounds [12]. The starting xylosylamine was generated *in situ* upon treating readily available 3,5-O-isopropylidene-D-xylofuranosylamine *p*-toluenesulfonate (II) [13] with bases. The optimal conditions for the reaction of I with isothiocyanatoaldehydes IIa-IIc in chloroform feature the presence of triethylamine at about 0°C. In this case, the corresponding 4,2′-anhydro-4-hydroxy-3-(3′,5′-O-isopropylidene-α-D-xylofuranosyl)hexahydropyrimidine-2-thiones (Va)-(Vc) are obtained as the major products in 43-59%. The yields of xylosides Va-Vc were much less when the reaction was carried out between I and IIa-IIc in methanol in the presence of sodium methylate or in pyridine.

*For Communication 8, see [1].

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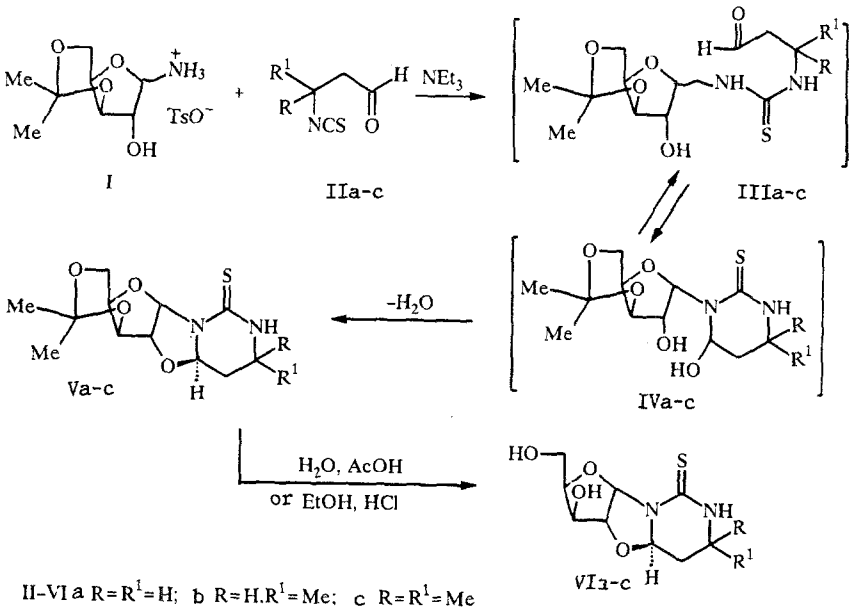

The proposed reaction pathway is shown in the above scheme and involves the intermediate formation of a mixture of anomers of N-(3,5-O-isopropylidene-D-xylofuranosyl)-N′-(3-oxopropyl)thioureas (IIIa)-(IIIc). Heterocyclization of the α-forms in IIIa-IIIc and subsequent intramolecular dehydration of 4-hydroxy-3-(3′,5′-O-isopropylidene-α-D-xylofuranosyl) hexahydropyrimidine-2-thiones (IVa)-(IVc), which is facilitated by the steric approximation of the 2′- and 4′-hydroxyl groups, lead to the major reaction products, namely, cyclonucleosides Va-Vc. The alternative reactions such as formation of β-forms of the N-glycosides and hydrolysis of the glycoside bonds account for the moderate yields of Va-Vc and the formation of a series of minor products, whose structure is now under study.

A new chiral site arises upon the formation of 4,2′-anhydroxylosides Va-Vc at $C_{(4)}$ such that Va and Vc may be obtained in two diastereomeric forms and Vb may be obtained as four diastereomeric forms.* PMR spectroscopy showed that Va and Vc are formed entirely diastereoselectively as a single isomer, while Vb is obtained as a 1:1 mixture of two diastereomers. One of the diastereomers of Vb [the (4R,6R)-isomer, see below] was isolated as a pure compound by crystallization from methanol. The isopropylidene protective group in xylosides Va-Vc is very labile and is readily removed upon treatment of these compounds by 25% aqueous acetic acid at 97°C for 5 min or ethanolic HCl at reflux for 5 min. This gives 4,2′-anhydro-4-hydroxy-3-(α-D-xylofuranosylhexahydropyrimidine-2-thiones (VIa)-(VIc) in 82-100% yield. Recrystallization of xyloside VIb from ethanol gave one of the diastereomers of this compound [the (4R,6R)-isomer, see below] as a pure compound.

The structures of Va-Vc and VIa—VIc were established by UV, IR, PMR, and ORD spectroscopy and mass spectrometry.

The presence of the hexahydropyrimidine-2-thione fragment in xylosides Va-Vc and VIa-VIc accounts for the strong UV bands at 206-208 (log ε 3.98-4.11) and 252-253 nm (log ε 4.12-4.23) for solutions of these compounds in methanol (Table 1), which are characteristic for the thioureide chromophore [14].

The IR spectra of Va-Vc and VIa-VIc in vaseline mull show a strong thioamide-II band at 1506-1545 cm^{-1}, assigned to the thioamide fragment [15]. Furthermore, the IR spectra of xylosides Va-Vc have a broad stretching band for the associated N—H group at 3000-3700 cm^{-1} (Table 1). The spectrum of a 10^{-3} M solution of Vc in CCl_4 (the low concentration excludes the formation of intermolecular hydrogen bonds) has only one narrow band at 3435 cm^{-1} (ηNH) and lacks stretching bands for free O—H groups, which supports our assignment of the structure of Va-Vc as 4,2′-anhydroglycosides. The IR spectra of crystalline samples of deblocked xylosides VIa-VIc have a number of broad bands at 3000-3700 cm^{-1} assigned to stretching vibrations of associated N—H and O—H groups.

*Racemic isothiocyanate IIb was used in the reactions.

TABLE 1. Characteristics of 4,2′-Anhydro-4-hydroxy-3-(α-D-xylofuranosyl)hexahydropyrimidine-2-thiones Va-Vc and VIa-VIc

Compound	Chemical formula	Mp, °C***	$[\alpha]_D^{20}$ (c, solvent***)	$[\alpha]_{436}^{20}$ (c, solvent***)	$[\alpha]_{365}^{20}$ (c, solvent***)	R_f**	UV spectrum (in methanol), λ_{max}, m (log ε)	Mass spectrum, m/z (I_{rel}, %)	IR spectrum (in Vaseline mull), cm^{-1}	Yield, %
Va	$C_{12}H_{18}N_2O_4S$	220...220,5(M)	+176,3 (0,66; M)	+356,1	+546,6	0,40	206 (3,98), 252 (4,12)	286 (22), 271 (7), 172 (57), 157 (7), 115 (100), 72 (9), 56 (9)	3255, 1520, 1301, 1281, 1198, 1091	58,7
			+155,6 (0,86; DMSO)	+302,9	+423,4					
Vb*	$C_{13}H_{20}N_2O_4S$	254,5...255 (dec.) (M)	+199,2 (0,67; M)	+401,4	+606,3	0,50	208 (4,11), 253 (4,23)	300 (19), 285 (6), 186 (47), 171 (4), 129 (100), 86 (20), 70 (10)	3325, 1506, 1321, 1290, 1199, 1059	44,2
Vc	$C_{14}H_{22}N_2O_4S$	270...271 (dec.) (M)	+179,4 (0,68; M)	+360,7	+546,9	0,55	208 (4,09), 253 (4,22)	314 (26), 299 (7), 200 (51), 185 (2), 143 (100), 100 (33), 84 (5)	3204, 1533, 1328, 1278, 1204, 1078	42,5
VIa	$C_9H_{14}N_2O_4S$	164...164,5 (E)	+199,9 (0,63; M)	+409,9	+644,4	0,33	206 (4,11), 252 (4,19)	246 (45), 172 (57), 157 (12), 115 (100)	3483, 3357, 3292, 1524, 1304, 1205, 1075, 991	81,8
			+159,7 (0,77; DMSO)	+307,8	+424,7					
VIb*	$C_{10}H_{16}N_2O_4S$	241...242 (dec.) (E)	+237,4 (0,71; M)	+484,5	+745,4	0,39	208 (4,05), 253 (4,20)	260 (25), 186 (39), 171 (6), 129 (100), 86 (34), 70 (20)	3370, 3283, 1545, 1315, 1238, 1093, 1041, 1012	100
VIc*	$C_{11}H_{18}N_2O_4S$	153,5...154,5 (E)	+195,9 (0,66; M)	+398,9	+617,3	0,43	208 (4,08), 253 (4,23)	—	3382, 1527, 1326, 1206, 1072, 1038	94,6

*For the (4R,6R) diastereomer.

**In 10:1 chloroform—methanol for Va-Vc and in 5:1 chloroform—methanol for VIa-VIc.

***M) methanol and E) ethanol.

TABLE 2. PMR Spectra of Va-Vc and VIa-VIc in DMSO-d_6

Compound	Chemical shift, δ, ppm (coupling constant, Hz)														
	Signals of saccharides residue protons									Aglycone proton signals					
	1'-H ($J_{1'2'}$)	2'-H ($J_{2'3'}$)	3'-H ($J_{3'4'}$)	4'-H ($J_{4'5'}$)	5'-H ($J_{4'5''}$)	5''-H ($J_{5'5''}$)	3'-OH ($J_{H,OH}$)	5'-OH ($J_{H,OH}$)	6-Me	4-H ($J_{4,5a}$; $J_{4,5e}$)	5-H_e ($J_{5e,5a}$)	5-H_a ($J_{5a,6}$)	6-H ($J_{5e,6}$)	6-Me ($J_{Me,H}$)	N-H ($J_{NH,H}$)
Va	6,45 (4,6)	4,46 (0)	4,39 (2,2)	3,85 (2,2)	4,02 (0)	3,79 (13,3)	—	—	1,40, 1,24	5,11 (9,3; 4,3)	2,19...2,28 m	1,34...1,50 m	3,09...3,16 m	—	8,65
Vb*	6,41 (4,6)	4,45 (0)	4,38 (2,3)	3,92 (2,3)	4,00 (0)	3,79 (13,2)	—	—	1,40, 1,25	5,14 (9,7; 4,3)	2,14 (12,2)	1,51 (5,8)	3,45 (~1,5)	1,10 (6,6)	8,75 (4,0)
Vc	6,55 (4,8)	4,53 (0)	4,46 (2,5)	3,99 (2,5)	4,07 (0)	3,86 (13,3)	—	—	1,42, 1,28	5,23 (9,8; 4,5)	2,25 (12,5)	1,37	—	1,24, 1,18	8,77
VIa	6,43 (4,6)	4,40 (0)	4,07 (3,2)	3,87 (4,7)	3,58 (6,5)	3,49 (11,2)	5,25 (5,2)	4,73 (5,5)	—	5,10 (9,5; 4,2)	2,17...2,27 m	1,33...1,49 m	3,08...3,16 m	—	8,59
VIb**	6,44 (4,5)	4,41 (0)	4,11 (3,4)	3,95	3,61	3,53 (11,2)	5,26 (5,2)	4,72 (5,3)	—	5,16 (9,9; 4,2)	2,15 (12,4)	1,51 (5,9)	3,35...3,54 m***(< 2,0)	1,11 (6,8)	8,78 (4,0)
				(Σ = 10,8)											
VIc	6,45 (4,7)	4,41 (0)	4,10 (3,2)	3,94 (4,7)	3,59 (6,5)	3,51 (11,4)	5,21 (5,3)	4,66 (5,5)	—	5,14 (9,9; 4,4)	2,19 (12,5)	1,33	—	1,20, 1,15	8,64

*For the (4R,6R)-diastereomer. Proton signals of the (4R,6S)-isomer: 8.55 (s, NH, $J_{NH,H}$ = 0 Hz), 6.43 (d, 1'-H, $J_{1'2'}$ = ~4.4 Hz), ~4.46 (d, 2'-H, $J_{2'3'}$ = ~0 Hz), 2.25 (d.q, 5-H_e, J_{5e5a} = ~12.5, J_{4a5e} ~ J_{5e6a} = ~3.8 Hz), 1.14 ppm (d, 6-Me, J = ~6.5 Hz).

**For the (4R,6R)-diastereomer. Proton signals of the (4R,6S)-isomer: 8.57 (s, NH, $J_{NH,H}$ = 0 Hz), 6.45 (d, 1'-H, $J_{1'2'}$ = ~4.0 Hz), ~4.42 (2'-H), 2.26 (d.q, 5-H_e, J_{5e5a} = ~12.0, J_{4a5e} ~ J_{5e6a} = ~3.5 Hz), 1.14 ppm (d, 6-Me).

***Signals overlap with signals for 5''-H and signal of the protons of HOD found in DMSO-d_6.

The PMR spectra of xylosides Va-Vc in DMSO-d_6 (Table 2) show signals for only one labile N—H group proton and lack signals for protons of 2′-OH or 4-OH groups. The lack of hydroxyl groups in Va-Vc in these molecules is also indicated by the nature of the multiplets for 2′-H (doublet) and 4-H (quartet). The PMR spectra of deblocked xylosides VIa-VIc have signals for protons of two O—H groups, one of which is bound to a primary carbon atom (CH_2OH, triplet) and the other is bound to a secondary carbon (CHOH, doublet) as well as signals for the protons of the deblocked N—H group. Analysis of the multiplets of the protons of the saccharide residue as well as of 4-H in the PMR spectra of VIa-VIc indicate that the hydroxyl groups are bound to $C_{(3')}$ and $C_{(5')}$, while $C_{(2')}$ and $C_{(4)}$ are linked by an ether bridge.

The existence of an oxazolidine ring in 4,2′-anhydroxylosides Va-Vc and VIa-VIc is possible only in the case of α-orientation of the glycoside bond. The α-anomeric configuration of the glycoside bond is supported by the ORD spectra of these compounds (Table 1). The ORD curves of Va-Vc and VIa-VIc (smooth positive curves at 365-589 nm) are similar to the ORD curves of 4,2′-anhydro-4-hydroxy-3-(α-D-ribofuranosyl)hexahydropyrimidine-2-thiones* and opposite in sign to the ORD curves for 4,5′-anhydro-4-hydroxy-3-(β-D-ribofuranosyl)hexahydropyrimidine-2-thiones.*

The absolute configuration of $C_{(4)}$ in Va-Vc and VIa-VIc was established by comparing the chemical shifts of the analogous protons in the PMR spectra of these compounds and 4-alkoxy-3-methylhexahydropyrimidine-2-thiones [16]. The most significant differences are observed for 4-H. Specifically, these protons are found in the spectra of Va-Vc and VIa-VIc 0.5-0.7 ppm downfield. Hence, the 4-H protons in Va, Vc, VIa, VIc, and both isomers of Vb and VIb are close to the oxygen atom of the tetrahydrofuran ring, which is possible in the case of (R)-configuration of $C_{(4)}$. Thus, both diastereomers of Vb and VIb apparently differ only in the configuration of chiral atom $C_{(6)}$.

The chemical shifts $J_{4,5e}$ (4.2-4.5 Hz) and $J_{4,5a}$ (9.3-9.9 Hz) in the PMR spectra of xylosides Va-Vc and VIa-VIc indicated that the hexahydropyrimidine ring in these compounds exists in a conformation with equatorial orientation of the 4-OCH group. One of the diastereomers of Vb and VIb was isolated as a pure compound by crystallization from methanol or ethanol and found to have *trans* arrangement of the 6-Me and 4-OCH groups and axial orientation of the 6-Me group ($J_{5a,6e}$ = 5.8-5.9, $J_{NH,6e}$ = 4.0 Hz), i.e., (4R,6R)-configuration, while the other diastereomer of Vb and VIb has *cis* orientation of the 6-Me and 4-OCH groups and equatorial orientation of the 6-Me group ($J_{NH,6a}$ = 0 Hz), i.e., (4R,6S)-configuration.

The structures of these products were supported by mass spectrometry (Table 1). The mass spectra of Va-Vc, VIa, and VIb have molecular ion peaks of rearranged ions $[(B - 18) + 1]^+$ (B is the corresponding 4-hydroxyhexahydropyrimidine-2-thione). Furthermore, the spectra of Va-Vc have strong peaks for $[M - 114]^{+\cdot}$ ions due to cleavage of a $C_6H_{10}O_2$ fragment from the molecular ions, probably as a result of dissociation of the $C_{(2')}$—$C_{(3')}$ and $C_{(4')}$—$O_{(1')}$ bonds. An analogous pathway for fragmentation of the molecular ions leads to $[M - 74]^+$ ions in the mass spectra of xylosides VIa and VIb (loss of a $C_3H_6O_2$ fragment). The mass spectra of blocked xylosides Va-Vc also have $[M - 15]^+$ ions related to loss of a methyl group from the molecular ions.

EXPERIMENTAL

The specific rotations were determined on Perkin—Elmer 241 and 241MC polarimeters for solutions in methanol and DMSO. The IR spectra were taken on Specord IR-75 and Shimadzu IR-435 spectrometers as vaseline mulls and in CCl_4 solution. The UV spectra were taken on a Beckmann DU-B spectrophotometer in methanol. The PMR spectra were taken on Bruker MSL-200 (200 MHz) or WM-250 (250 MHz) spectrometers for solutions in DMSO-d_6 or $CDCl_3$ with HMDS as the internal standard. The mass spectra were taken on a Varian MAT-112 spectrometer with direct sample inlet. The ionizing electron energy was 70 eV. The ionization chamber temperature was 180°C. Thin-layer chromatography was carried out on Merck Kieselgel 60 F_{254} plates in 10:1 or 5:1 chloroform—methanol. The spots were observed in UV light or developed with iodine vapor. The R_f values were determined under standard chromatography conditions according to Stahl [17]. Column chromatography was carried out on L 40/100μ silica gel manufactured in Czechoslovakia.

The elemental analysis data for C, H, N, and S for the previously unreported products corresponded to the calculated values.

*Unpublished data.

4,2′-Anhydro-4-hydroxy-3-(3′,5′-O-isopropylidene-α-D-xylofuranosyl)hexahydropyrimidine-2-thione (Va). A solution of 0.73 ml (5.26 mmoles) dry triethylamine in 8 ml chloroform cooled to −15°C was added to a suspension of 1.85 g (5.11 mmoles) I in 6 ml dry chloroform cooled to −15°C. Then, a solution of 765 mg (6.64 mmoles) freshly distilled isothiocyanate IIa in 8 ml chloroform cooled to −15°C was added. The solution obtained was maintained at 5°C for 16 h and evaporated in vacuum. The residue was dissolved in a minimal amount of chloroform. The solution was deposited on a 3.7×12-cm column packed with 70 g silica gel and eluted with 1:1 → 2:1 chloroform—ether and then chloroform to give 859 mg (58.7%) Va, which was recrystallized from methanol.

4,2′-Anhydro-4-hydroxy-3-(3′,5′-O-isopropylidene-α-D-xylofuranosyl)-6-methylhexahydropyrimidine-2-thione (Vb) was obtained analogously to Va from 1.783 g (4.93 mmoles) I, 0.68 ml (4.90 mmoles) dry triethylamine, and 637 mg (4.93 mmoles) freshly distilled isothiocyanate IIb in 20 ml dry chloroform. After evaporation of the reaction mixture, 2 ml cold water and 2 ml ether were added to the residue. The mixture was stirred and cooled to −15°C. The precipitate formed was filtered off, washed once with ice water, several times with ether cooled to −15°C, and once with ethanol cooled to −15°C to give 344 mg (23.2%) Vb. The combined mother liquors were extracted with chloroform. The extract was washed with water and dried over $MgSO_4$. The solvent was distilled off and the residue was dissolved in chloroform. The solution was deposited on a column packed with 25 g silica gel and eluted with chloroform to give an additional 311 mg Vb. The combined yield of Vb was 655 mg (44.2%).

Xyloside Vb, which is a 1:1 mixture of two diastereomers, was recrystallized several times from methanol to give the pure (4R,6R)-diastereomer.

Analogously, xyloside Bc was obtained from I and isothiocyanate IIc in 42.5%.

4,2′-Anhydro-4-hydroxy-3-(α-D-xylofuranosyl)hexahydropyrimidine-2-thione (IVa). A. A solution of 110 mg (0.385 mmoles) xyloside Va in 2 ml 25% acetic acid was heated for 5 min at 97°C and evaporated in vacuum to dryness. A sample of 1 ml absolute ethanol was added to the residue and the mixture was again evaporated. The solid foam formed was triturated with 0.5 ml dry acetone and cooled to −15°C. The precipitate was filtered off, washed with cold acetone, and dried to give 78 mg (81.8%) VIa, which was recrystallized from absolute ethanol.

(4R,46)-VIb and VIc were synthesized analogously from (4R,6R)-Vb and Vc and crystallized from methanol or ethanol.

(4R,6R)-VIb was obtained analogously by deblocking a mixture of the diastereomers of xyloside Vb with two or three recrystallizations of the isomer mixture of VIb formed from ethanol.

B. A solution of 157 mg (0.549 mmoles) xyloside Va in 3 ml 96% ethanol containing two drops of concentrated hydrochloric acid was heated at reflux for 5 min and the mixture was evaporated to dryness. A sample of 1 ml absolute ethanol was added to the residue and the mixture was again evaporated. The solid foam formed was triturated with dry ether. The crystals were filtered off, washed with ether, and dried to give 132 mg (97.7%) VIa.

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