17. R. T. C. Brownlee and R. D. Topsom, Tetrahedron Lett., No. 51, 5187 (1972).
18. G. P. Syrova and Yu. N. Sheinker, Khim. Geteortsikl. Soedin., No. 3, 345 (1972).
19. L. W. Deady, P. M. Harrison, and R. D. Topsom, Org. Magn. Resonance, 7, 41 (1975).
20. D. J. Brown and P. W. Ford, J. Chem. Soc., C, No. 7, 568 (1967).
21. M. Robba, Ann. Chim., 5, 351 (1960).
22. V. P. Mamaev and É. A. Gracheva, Khim. Geterotsikl. Soedin., No. 6, 1086 (1969).
23. V. A. Koptyug (editor), Atlas of the Spectra of Aromatic and Heterocyclic Compounds [in Russian], Nauka, Novosibirsk (1975), No. 8.

ANALOGS OF PYRIMIDINE MONO- AND POLYNUCLEOTIDES.

VI.* PHOSPHATES OF 1-(1,4-DIHYDROXY-2-PENTYL)THYMINE AND 1-(1,3-DIHYDROXY-2-PROPYL)URACIL

S. A. Giller,† L. A. Sherin',
D. É. Zarin', and R. A. Zhuk

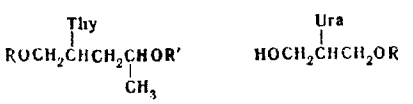
UDC 547.963.32'854.4

The corresponding monophosphates, cyclophosphates, and α,ω-diphosphates were obtained by phosphorylation of 1-(1,4-dihydroxy-2-pentyl)thymine, 1-(1,3-dihydroxy-2-propyl)uracil, and their derivatives with selective protection of one of the hydroxyl groups. 2-Cyanoethyl phosphate (CEP) in the presence of N,N'-dicyclohexylcarbodiimide (DCC), polyphosphoric acid, and pyrophosphoryl chloride were used as phosphorylating agents. The dependence of the yields of the products of phosphorylation of 1-(1,4-dihydroxy-2-pentyl)thymine with CEP on the ratio of the reacting substances and the reaction time was studied. The monophosphates were cyclized under the influence of DCC. In the case of 1-(1,3-dihydroxy-2-propyl)uracil 1-phosphate a dimeric phosphate was obtained in addition to a cyclophosphate. The acid hydrolysis of the cyclophosphates was investigated.

The phosphates of 1-(1,4-dihydroxy-2-butyl)thymine (I) that we synthesized in [3, 4] were used as monomers for the preparation of analogs of oligothymidylic acid, which displayed interesting biological properties [5]. However, these oligomers have the disadvantage that their structures are indeterminate, since a 1'—1, 1'—4', or 4'—4' bond may be realized during polycondensation.

Our research was directed toward a search for new dihydroxyalkyl derivatives of pyrimidine bases, on the bases of which the synthesis of oligomers with a more regular structure is possible.

The present paper is devoted to the synthesis of phosphates of 1-(1,4-dihydroxy-2-pentyl)thymine (IIa) [6] and 1-(1,3-dihydro-2-propyl)uracil (IIIa) [7]. The presence in diol IIa of primary and secondary hydroxyl groups facilitates the selective phosphorylation with the use of, respectively, protected derivatives IIb,c, and the symmetrical character of the dihydroxyalkyl group in IIIa makes it possible to obtain only one monophosphate (Vb).



II a-c III a,b

II a R=R'=H; b R=C(C6H5)3, R'=H; c R=H, R'=COCH3;
III a R=H; b R=C(C6H5)3

*See [1] for communication V; see [2] for a preliminary communication.
†Deceased.

Institute of Organic Synthesis, Academy of Sciences of the Latvian SSR, Riga 226006. Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 5, pp. 678-683, May, 1978. Original article submitted December 23, 1976; revision submitted October 26, 1977.

 0009-3122/78/1405-0556$07.50

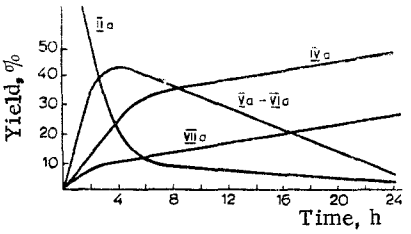


Fig. 1. Yields of IVa, Va + VIa, and VIIa and consumption of the starting IIa as a function of the reaction time for a IIa:CEP:DCC ratio of 1:2:3.

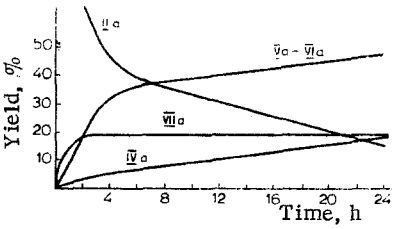


Fig. 2. Yields of IVa, Va + VIa, and VIIa and consumption of starting IIa as a function of the reaction time for a IIa:CEP:DCC ratio of 1:4:8.

The polycondensation of monophosphates Va,b and VIa may give oligomers with regular distances between the bases.

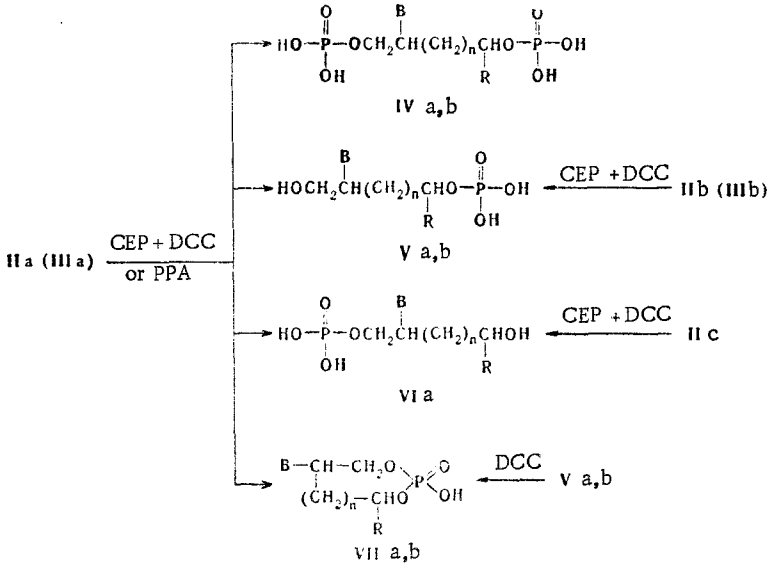
The yields of diphosphates IVa,b are considerably lower in the case of phosphorylation of diols IIa or IIIa with 2-cyanoethyl phosphate (CEP) in the presence of N,N'-dicyclohexylcarbodiimide (DCC) under conditions that are optimal for the formation of α,ω-diphosphates [3] than in the case of phosphorylation of dihydroxy derivative I (80%). The corresponding cyclophosphates (VIIa,b) and monophosphates (a mixture of Va and VIa or Vb) are formed in addition to diphosphates IVa or IVb.

Approximately the same ratio of phosphates (IVb:Vb:VIIb = 2.3:1:1.4) is retained in the phosphorylation of dihydroxy compound IIIa with polyphosphoric acid (PPA). The reduced yield of diphosphate IVb may be due to the closeness of the hydroxyl groups and the ease of formation of cyclophosphate VIIb.

The relatively low yield of diphosphate IVa in the reaction of DCC with dihydroxy derivative IIa can evidently be explained by the lower reactivity of the secondary hydroxyl group in the 4' position. The yields of monophosphates Va and VIa or cyclophosphate VIIa did not exceed 40-50% when the reagent ratio and reaction time were varied (Figs. 1 and 2).

The high yields (71-79%) of monophosphates obtained in the phosphorylation of 2,3-dihydroxy derivatives of nucleic bases [8] by this method are evidently explained by the presence of a vicinal diol grouping in the latter.

The phosphorylation of dihydroxy compound IIa with pyrophosphoryl chloride, which has been proposed as a selective phosphorylating agent that reacts selectively with the 5'-hydroxyl group of unprotected nucleosides [9, 10] did not give high yields of the monophosphate. A diphosphatewas obtained in 78% yield when the reaction was carried out at 0-5°C for



IV–VII a B = Thy, R = CH_3, n = 1; b B = Ura, R = H, n = 0

2 h with subsequent hydrolysis by water, and the monophosphate constituted only 6% of the reaction mixture. Consequently, we did not observe selectivity of the action of $P_2O_3Cl_4$; however, this method can be used for the preparation of diphosphates. Its application for the phosphorylation of diol IIIa gave the highest yield of IVb (62%). It is interesting to note that a decrease rather than an increase in the yield of diphosphate IVb is observed when the same reaction is carried out at room temperature for 6 h. The principal reaction products in this case after hydrolysis of the reaction mixture with water at the boiling point are monophosphate Vb (51%) and cyclophosphate VIIb (38%), and diphosphate IVb constitutes 4% of the mixture of products. The yield of monophosphate Vb is higher in this case than when CEP or PPA is used as the phosphorylating agent.

Because of the unsatisfactory results in the preparation of monophosphates from unsubstituted diols we developed a method for phosphorylation of their derivatives with selectively protected hydroxyl groups (IIb,c [6] and IIIb [7]).

The phosphorylation of 1-(1-O-trityl-1,3-dihydroxy-2-propyl)uracil (IIIb) was carried out with CEP in the presence of DCC at room temperature for 24 h and a IIIb:CEP:DCC ratio of 1:2:5. Monophosphate Vb was obtained in 88% yield after removal of the protective groups.

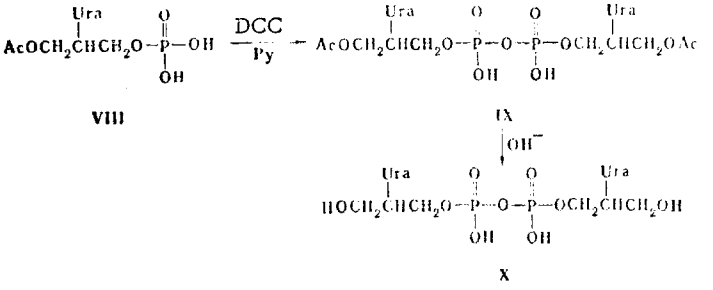
Monophosphorylation product VIa was formed in 60% yield (after removal of the protective acetyl group) in the phosphorylation under these conditions of 1-(4-O-acetyl-1,4-dihydroxy-2-pentyl)thymine (IIb), which has a free primary hydroxy group. In addition, the reaction mixture contains 20% starting diol IIc and about 20% unidentified side products.

Phosphorylation of 1-(1-O-trityl-1,4-dihydroxy-2-pentyl)thymine (IIb) with a free secondary hydroxyl group under the same conditions gives monophosphate Va in 43% yield.

The physical and chemical properties of the monophosphates differ appreciably as a function of the structure of the alkyl group. Thus, monophosphate Vb is virtually insoluble in anhydrous pyridine, whereas the monophosphates of diols I and II are quite soluble.

Differences were also observed in the cyclization of the monophosphates under the influence of DCC. The action of a sixfold excess of DCC on an 0.01 M solution of monophosphate Va in pyridine for 3 days leads to the formation of cyclophosphate VIIa in 80% yield. Similar results were obtained in the cyclization of the monophosphates of diol I. Dimeric pyrophosphate X is formed in 47% yield in the reaction of an 0.01 M solution of the pyridinium salt of phosphate Vb with DCC in dry dimethylformamide (DMF). The corresponding cyclophosphate (VIIb) was obtained in 76-77% yield by cyclization of an 0.06 M solution of phosphate Vb with DCC in dry DMF.

The electrophoretic mobilities of cyclophosphate VIIb and pyrophosphate X are very close. For identification of the reaction products, pyrophosphate X was synthesized by alternative synthesis from 1-(3-O-acetyl-1,3-dihydroxy-2-propyl)uracil 1-phosphate (VIII) in the presence of DCC.



Acid hydrolysis showed that pyrophosphate X undergoes complete cleavage to monophosphate Vb under the influence of 0.1 N HCl at 100°C in 2 h, whereas it undergoes 82% hydrolysis with acetic anhydride in pyridine at room temperature in 3 days.

The acid hydrolysis of cyclophosphates VIIa,b (0.1 N HCl, 100°C) shows that, in conformity with the data in [11], the seven-membered phosphate ring is somewhat more stable.

The characteristics of the phosphates obtained are given in Table 1. Small negative chemical shifts (from −0.12 to −3.54 ppm) are characteristic for the ^{31}P NMR spectra of monophosphates Va,b and VIa. The chemical shift of the phosphorus atom in the spectra of the cyclophosphates depends on the ring size: it is 4.15 ppm for cyclophosphate VIIb, in agree-

TABLE 1. Characteristics of the Phosphates of 1-(1,4-Dihydroxy-2-pentyl)thymine (IIa) and 1-(1,3-Dihydroxy-2-propyl)uracil (IIIa)

Compound	UV spectrum, λ_{max}, nm		R_f*		E_f†		^{31}P NMR spectra, δ, ppm (aqueous solution)
	pH 2	pH 12	A	B	pH 7	pH 3	
IVa	273	273	0,14	0,21	1,13	1,12	−1,37; 0,30
IVb	265	265	0,02	0,12	1,17	1,09	−1,80
Va	273	273	0,35	0,50	0,97	1,00	−0,12
Vb	266	266	0,16	0,27	1,03	1,01	−3,54
VIa	273	273	0,35	0,50	0,97	1,00	−1,17
VIIa	273	272	0,62	0,54	0,87	1,05	−2,59
VIIb	266	266	0,34	0,35	0,92	1,05	4,15
X	268	267	0,20	0,14	0,96	1,06	11,59

*On paper in isopropyl alcohol—concentrated NH_4OH–H_2O (7:1:2) (A) and isobutyric acid—concentrated NH_4OH–H_2O (66:1.5:33) (B) systems.
†The electrophoretic mobilities relative to uridine 5'-monophosphate.

ment with the values for other six-membered cyclophosphates [12], and −2.59 ppm in cyclophosphate VIIa with a seven-membered ring.

The synthesis of oligomers on the basis of the phosphate esters obtained will be reported in subsequent papers.

EXPERIMENTAL

The UV spectra of the compounds were obtained with a Specord spectrophotometer. The PMR spectra of solutions of the compounds in D_2O were recorded with a Perkin–Elmer R-12A spectrometer. The ^{31}P NMR spectra were recorded with an HX-90 spectrometer with Fourier transformation with a Bruker–Physik AG B–NC computer at 36.43 MHz. The chemical shifts (δ) are presented relative to 85% H_3PO_4 as the external standard.

Chromatography was accomplished by the ascending method on FN-1 paper. Electrophoresis was carried out on FN-12 paper in a phosphate buffer (pH 7) at 800 V or in a glycine buffer (pH 3) at 600 V.

The pyridinium salt of 2-cyanoethyl phosphate was prepared immediately prior to use by the method in [13]. Pyrophosphoryl chloride was synthesized from $POCl_3$ and PCl_5 [14].

1-(1,4-Dihydroxy-2-pentyl)thymine 1',4'-Diphosphate (IVa). A) A 1.84-g (8 mmole) sample of diol IIa was phosphorylated with 32 mmole of CEP in the presence of 64 mmole of DCC, as described in [3]. After purification on AGN charcoal to remove the inorganic phosphates, the mixture was chromatographed on QAE Sephadex A-25. Elution was accomplished with the stepwise gradient of an $(NH_4)_2CO_3$ buffer containing 10% ethanol; starting IIIa was eluted with an 0.01 N buffer, cyclophosphate VIIa was eluted with an 0.05 N buffer, a mixture of monophosphates Va and VIa was eluted with an 0.2 N buffer, and diphosphate IVa was eluted with an 0.3 N buffer. The fraction containing diphosphate IVa was evaporated repeatedly with water, and the residue was crystallized from absolute ethanol to give 1.5 g (47%) of the ammonium salt of diphosphate IVa with mp 156-160°C. PMR spectrum, δ: 1.27 (d, 5'-H), 1.87 (s, 5-CH_3), 2.01 (t, 3'-H), 3.50-4.80 (m, 1',2',4'-H), and 7.53 ppm (s, 6-H). Found, %: C 24.6; H 6.6; N 17.0; P 12.2. $C_{10}H_{30}N_6O_{10}P_2 \cdot 2H_2O$. Calculated, %: C 24.4; H 6.6; N 17.1; P 12.5.

B) An 0.07-ml (0.5 mmole) sample of pyrophosphoryl chloride was added at 0°C to a suspension of 0.023 g (0.1 mmole) of diol IIa in 1.5 ml of dry acetonitrile, and the mixture was maintained at 0-5°C for 2 h. It was then poured into 25 ml of water, and the acidic solution was neutralized with ammonia and evaporated to dryness. The residue was dissolved in water and subjected to separation on QAE-Sephadex A-25. The yield of phosphate IVa was 78%.

1-(1,3-Dihydroxy-2-propyl)uracil 1',3'-Diphosphate (IVb). This compound was similarly synthesized (method A) from 3.3 mmole of diol IIIa, 13 mmole of CEP, and 26 mmole of DCC. The yields of phosphates were: 41% IVb, 18% Vb, and 25% VIIb. PMR spectrum, δ: 4.32 (t, 1',3'-H), 5.00 (quintet, 2'-H), 5.90 (d, 5-H), and 7.79 ppm (d, 6-H).

B) This compound was similarly obtained (method B) in 62% yield from 1 mmole of diol IIIa and 5 mmole of pyrophosphoryl chloride. Phosphates Vb and VIIb were obtained in 15 and 14% yields, respectively. Found for the cyclohexylammonium salt: C 46.3; H 7.8; N 10.6; P 9.8%. $C_{25}H_{51}N_5O_{10}P_2$. Calculated: C 46.6; H 8.0; N 10.9; P 9.6%.

C) A 0.6-g (3.2 mmole) sample of diol IIIa was mixed with freshly distilled PPA (12.8 g of P_2O_5 + 9.6 ml of 85% H_3PO_4), and the mixture was heated at 50-60°C for 2 h and maintained at room temperature for 22 h. Water (50 ml) was then added, and the mixture was heated at 100°C for 15 min. The resulting solution was applied to a column filled with AGN charcoal, and the mixture was separated as in the phosphorylation with CEP to give IVb in 42% yield (phosphate Vb and cyclophosphate VIIb were obtained in 24 and 29% yields, respectively).

1-(1,4-Dihydroxy-2-pentyl)thymine 4'-Phosphate (Va). The reaction was carried out as in the synthesis of phosphate IVa (method A) starting from 3.8 g (8 mmole) of trityl derivative IIb, 16 mmole of CEP, and 8 g (40 mmole) of DCC. At the end of the reaction, the dicyclohexylurea was removed by filtration, the filtrate was evaporated, and the residue was treated with 50 ml of 80% acetic acid at 100°C for 30 min. The mixture was cooled, and the precipitated triphenylmethanol was removed by filtration. The filtrate was evaporated, and the residue was worked up as in the synthesis of phosphate IVa (method A). After separation on QAE-Sephadex A-25, workup of the fraction eluted with the 0.2 N buffer gave 1.3 g (43%) of the ammonium salt of Va with mp 128-130°C. PMR spectrum, δ: 1.26 (d, 5'-H), 1.87 (s, 5-CH_3), 1.95 (t, 3'-H), 3.73 (d, 1'-H) (quintet, 2'-H), 3.90-4.34 (m, 4'-H), and 7.52 ppm (s, 6-H). Found, %: C 30.4; H 7.2; N 14.0; P 7.5. $C_{10}H_{23}N_4O_7P \cdot 3H_2O$. Calculated, %: C 30.3; H 7.3; N 14.1; P 7.8.

1-(1,3-Dihydroxy-2-propyl)uracil 1'-Phosphate (Vb). A) The procedure used to prepare Va was employed to synthesize this compound in 88% yield from 2.75 mmole of IIIb, 5.5 mmole of CEP, and 13.8 mmole of DCC. PMR spectrum, δ: 3.95 (d, 3'-H), 4.22 (t, 1'-H), 4.82 (quintet, 2'-H), 5.87 (d, 5-H), and 7.77 ppm (d, 6-H). Found for the cyclohexylammonium salt: C 45.4; H 8.4; N 10.9; P 6.4%. $C_{19}H_{37}N_4O_7P \cdot 2H_2O$. Calculated: C 45.6; H 8.3; N 11.2; P 6.2%.

B) A 5-mmole sample of pyrophosphoryl chloride was added with stirring to a suspension of 1 mmole of dihydroxy derivative IIIa in 1.5 ml of anhydrous acetonitrile, and the mixture was maintained at room temperature for 6 h. It was then poured into 25 ml of water, and the acidic solution (pH ~ 1) was refluxed for 2 h, neutralized with ammonia, and separated on QAE-Sephadex A-25 to give Vb in 51% yield.

C) A 3.2-mmole sample of diol IIIa was mixed with freshly prepared PPA, and the mixture was heated at 50-60°C for 2 h. Water (50 ml) was added, and the mixture was worked up as in the synthesis of phosphate IVb (method C) to give phosphate Vb in 41% yield, IVb in 23% yield, and VIIb in 16% yield.

1-(1,4-Dihydroxy-2-pentyl)thymine 1'-Phosphate (VIa). This compound was synthesized by the method used to prepare phosphate IVa (method A) from 0.93 g (3.4 mmole) of hydroxy compound IIc, 6.8 mmole of CEP, and 3.5 g (17 mmole) of DCC. The protective acetyl group was removed by treatment of the reaction mixture with 20 ml of 1 N NaOH at room temperature for 30 min, after which the mixture was passed through Dowex-50 (H^+). The eluate was purified to remove the inorganic phosphate and separated on QAE-Sephadex A-25. Workup of the 0.2 N eluate gave 0.8 g (60%) of the ammonium salt of VIa with mp 131-134°C. PMR spectrum, δ: 1.18 (d, 5'-H), 1.88 (s, 5-CH_3), 1.92 (t, 3'-H), 3.67 (m, 4'-H), 3.99 (t, 1'-H), 4.60-4.80 (m, 2'-H), and 7.54 ppm (s, 6-H). Found, %: C 29.9; H 7.0; N 13.9; P 7.4. $C_{10}H_{23}N_4O_7P \cdot 3H_2O$. Calculated, %: C 30.3; H 7.3; N 14.1; P 7.8.

1-(1,4-Dihydroxy-2-pentyl)thymine 1',4'-Cyclophosphate (VIIa). An 0.1-g (0.25 mmole) sample of phosphate Va (H^+) was evaporated several times with absolute pyridine, and the residue was dissolved in 20 ml of pyridine. The solution was treated with 1.5 mmole of DCC, and the mixture was allowed to stand at room temperature for 3 days. Water (30 ml) was added, the dicyclohexylurea was separated, and the pyridine solution was chromatographed on QAE-Sephadex A-25 to give 0.07 g (90%) of the ammonium salt of VIIa with mp 214-216°C. PMR

spectrum, δ: 1.32 (d, 5'-H), 1.84 (s, 5-CH_3), 2.08 (t, 3'-H), 3.74-4.80 (m, 1'-, 2'-, 4'-H), and 7.44 ppm (s, 6-H). Found, %: C 37.1; H 6.2; N 12.8; P 9.3. $C_{10}H_{18}N_3O_6P \cdot H_2O$. Calculated, %: C 36.9; H 6.2; N 12.9; P 9.5.

1-(1,3-Dihydroxy-2-propyl)uracil 1',3'-Cyclophosphate (VIIb). A 0.45-mmole sample of phosphate Vb was evaporated three times with 8 ml of dry DMF, after which a solution of 0.4 g (1.9 mmole) of DCC in 8 ml of DMF was added to the residue, and the mixture was allowed to stand at room temperature for 20 min. Water (50 ml) was added, the dicyclohexylurea was removed by filtration, and the filtrate was separated on QAE-Sephadex A-25. Cyclophosphate VIIb was eluted with a 0.1 N buffer and was obtained in 76% yield. PMR spectrum, δ: 4.50 (m, 1'-, 2'-, 3'-H), 5.87 (d, 5-H), and 8.12 ppm (d, 6-H). Found, %: C 29.5; H 5.1; N 15.0; P 11.2. $C_7H_{12}N_3O_6P \cdot H_2O$. Calculated, %: C 29.7; H 5.0; N 14.8; P 10.9.

1-(3-O-Acetyl-1,3-dihydroxy-2-propyl)uracil 1'-Phosphate (VIII). A solution of 5.5 ml of acetic anhydride in 24 ml of anhydrous pyridine was added to 1 mmole of the pyridinium salt of Vb, and the mixture was allowed to stand at room temperature for 16 h. It was then cooled and treated with 90 ml of water, and the mixture was allowed to stand at room temperature for 3.5 h. It was evaporated repeatedly with water to remove the acetic acid. The residue was evaporated repeatedly with dry pyridine, and the saturated pyridine solution was added dropwise to 150 ml of absolute ether. The precipitated VIII was separated by centrifugation to give VIII with Ef 1.0 (pH 7) and Rf 0.45 (system B) in 80% yield. ^{31}P NMR spectrum (in pyridine), δ: −1.05 ppm.

P,P'-Bis[1-(1,3-dihydroxy-2-propyl)uracil] 1',1'-Pyrophosphate (X). A) An 0.06-mmole sample of the pyridinium salt of phosphate VIII was evaporated with dry pyridine, a solution of 0.05 g (0.24 mmole) of DCC in 1 ml of dry pyridine was added to the residue, and the mixture was allowed to stand at room temperature for 3 days. The dicyclohexylurea was separated, the filtrate was evaporated, and the residue was treated with 10 ml of 0.1 N NaOH at room temperature for 1 h. The reaction mixture was passed through Dowex-50 (H^+) and separated on QAE-Sephadex A-25. Pyrophosphate was eluted with an 0.3 N buffer and was obtained in 65% yield.

B) An 0.09 mmole sample of the pyridinium salt of phosphate Vb was evaporated with dry pyridine, after which a solution of 0.07 g (0.34 mmole) of DCC in 10 ml of anhydrous DMF was added, and the mixture was allowed to stand at room temperature for 3 days. Water (10 ml) was added, and the mixture was worked up as in the synthesis of phosphate VIIa. Separation of QAE-Sephadex A-25 gave pyrophosphate X in 47% yield (0.3 N buffer) and phosphate VIIb in 19% yield.

LITERATURE CITED

1. S. A. Giller, Z. A. Shomshtein, and T. A. Popova, Khim. Geterotsikl. Soedin., No. 4, 540 (1975).
2. S. A. Hiller, R. A. Zhuk, L. A. Sherinya, and D. E. Zarin, Nucleic Acids Res., Special Publication No. 1, 169 (1975).
3. S. A. Giller, L. A. Sherin', R. A. Zhuk, and A. E. Berzinya, Khim. Geterotsikl. Soedin., No. 12, 1671 (1974).
4. S. A. Giller, T. A. Popova, and Z. A. Shomshtein, Khim. Geterotsikl. Soedin., No. 3, 401 (1975).
5. S. A. Giller, E. Ya. Gren, Z. A. Shomshtein, R. A. Zhuk, V. M. Berzin', P. P. Pumpen, and G. F. Rozental', Dokl. Akad. Nauk SSSR, 221, 473 (1975).
6. R. A. Zhuk, L. A. Sherin', É. É. Liepin'sh, and S. A. Giller, Khim. Geterotsikl. Soedin., No. 12, 1666 (1974).
7. S. A. Hiller, D. E. Zarin, and R. A. Zhuk, Nucleic Acids Res., 3, 721 (1976).
8. A. M. Kritsyn, L. I. Kolobushkina, S. N. Mikhailov, and V. L. Florent'ev, Khim. Geterotsikl. Soedin., No. 1, 125 (1975).
9. K. Imai, S. Fujii, K. Takanohasi, Y. Furukawa, T. Masuda, and M. Hanjo, J. Org. Chem., 34, 1547 (1969).
10. N. G. Shinskii, N. N. Preobrazhenskaya, M. G. Ivanovskaya, Z. A. Shabarova, and M. A. Prokof'ev, Dokl. Akad. Nauk SSSR, 184, 622 (1969).
11. H. G. Khorana, G. M. Tener, R. S. Wright, and J. G. Moffatt, J. Am. Chem. Soc., 79, 430 (1957).
12. M. M. Crutchfield, C. H. Dungan, J. H. Letcher, V. Mark, and J. R. Van Wazer, P^{31} Nuclear Magnetic Resonance Topics in Phosphorus Chemistry, Vol. 5, Wiley and Sons, New York (1967), p. 340.
13. G. M. Tener, J. Am. Chem. Soc., 83, 159 (1961).
14. H. Grunze, Monatsh. DAW DDR, 5, 636 (1963).