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Dietylpentaerythritol Diacetals in the Synthesis of Phosphorus Containing Macroheterocycles

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Abstract — Synthesis of phosphorus-containing macroheterocycles based on the symmetrical dipentaerytritol diacetals and trivalent phosphorus reagent is studied. Effective peracetalization of dimethoxymethane with dipentaerytritol is performed.

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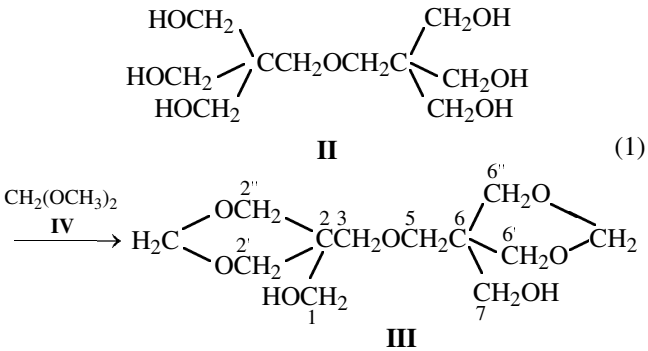
Design of new phosphorus-containing macroheterocycles is a fruitful and intensively developing area of chemistry [1–3]. Recently we have proposed to use accessible dipentaerythritol derivatives, a mixture of configuration isomers of 1,3;1',3'-dibenzylidenedipentaeritritol (**I**), as initial molecular platforms for creation of new 10- and 20-membered phosphocyclanes [4]. Synthesis of compounds **I** was achieved in reaction of dipentaerytritiol **II** with benzaldehyde catalyzed by hydrochloric or phosphotungstenic acids [5]. The reporting work is aimed at the development of this method to the application of a stereo individual reagents dimethylenedipentaerytritiol **III** and dibenzylidenedipentaerytritiol (**I**), as well as these diacetals combined together.

For the synthesis of dimethyleneacetal **III** we applied a method of peracetalization, which is in wide use for fine organic syntheses [6–8].



For the dual methylenization of hexaol **II** used boron trifluoride etherate (mode 1) or *p*-toluensulfonic acid (mode 2) as catalysts. In the mode 1, reaction was conducted at 100–110°C, in mode 2 at 120–125°C. The target compound **III** was isolated by column chromatography. The yield of chromatographically pure diacetal **III** achieves 30% (mode 1) and 59% (mode 2). The lower yield in the firs case is connected with accumulation of side compounds in the process of reaction.

The diacetal **III** prepared alongside the mode 1 just after removing of solvents appeared as a crystalline compound (colorless plates), while by mode 2 an oily substance was obtained which then crystallizes in a few days. ^{1}H NMR spectrum of compound **III** contains a signal of hydroxyl protons (δ 2.78 ppm), a singlet of methylene protons of bridgehead CH_2OCH_2 group (δ 3.14 ppm), doublets of methylene protons of 1,3-dioxane rings (δ 3.32 and 3.61 ppm), a doublet of methylene protons hydroxyl groups (δ 3.43 ppm), and two singlets of magnetically nonequivalent methylene protons of the two protecting acetal groups (δ 4.11 and 4.66 ppm). This assignment was confirmed by the data of molecular modeling[1] of diacetal **III** molecule in optimal conformation (Fig. 1). Both the 1,3-dioxane rings of compound **III** are in chair conformation, one has CH_2 protecting group and another acetal oxygen atom in the apex.

Note that dimethylenedipentaerytritiol **III** preparation described earlier included 14-h reaction of dipentaerytritiol **II** with 37% aqueous formaldehyde in the presence of substantial amount of hydrochloric acid (volume ratio of formaldehyde solution to hydro-

[1] Hereinafter the molecular modeling of the studied compounds was performed by MM2 method using Chem 3D Ultra 7.0 program [9].

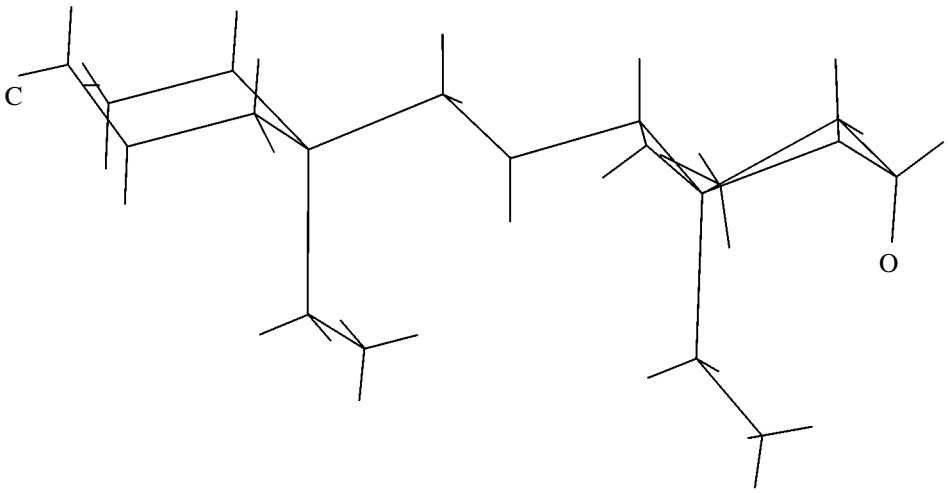
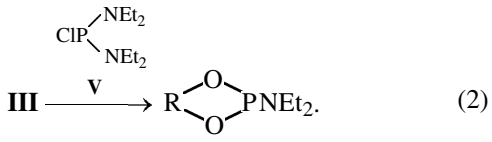


Fig. 1. Optimal conformation of 1,3;1',3'-dimethylenedipentaerytritiol **III**.

chloric acid is 1 : 1). The acetal **III** yield was 40.6% only [10].

The next step of our work is denoted to cyclophosphorylation of dimethylenedipentaerytritiol **III** with the use of tetraethyldiamidophosphoerous chloride (**V**).



(2)

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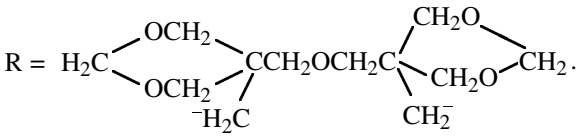
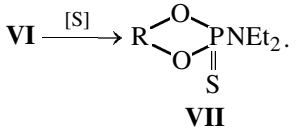


The phosphorylation was conducted in anhydrous dioxane at 0°C in the presence of pyridine. Upon the reaction progress, in the ^{31}P NMR spectrum of reaction mixture disappeared signal of diamidophosphorous chloride **V** (δ_P 157.3 ppm) and grew a signal at δ_P 147.92 ppm (br.s) which corresponds to the region of monoamidocyclophosphites. Note that in the above described reaction the dimethylenedipentaerytritiol **III** is more reactive compound than dibenzylidenedipentaerytritiols **I** [4].

The amidocyclophosphite **VI** without isolation was easily transformed into corresponding 10-membered cycloamidothionophosphate **VII**.

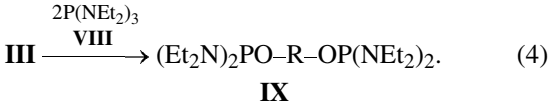


VII (3)

The thionophosphate **VII** was isolated by chromatography on a column with silica gel, yield in two steps was 55%. The compound was isolated as an oil which is stable on keeping at room temperature. Its individuality and structure were proven using elemental analysis, TLC, and ^{1}H and ^{31}P NMR spectroscopy. Molecular mass of **VII** was confirmed by the data of MALDI-TOF mass spectrometry.

The ^{31}P NMR spectrum of **VII** contains broad signal at δ_P 77.78 ppm. Its ^{1}H NMR spectrum shows a singlet and a doublet (δ 4.47 and 4.65 ppm, respectively) of the methylene protons of the acetal protecting group. Appearance of the doublet is a result of trans-annular interaction of the methylene protons of one acetal group with the methylene protons of neighboring 1,3-dioxane ring. Besdes, resonance of *N*-etyl group protons occurs as a typical triplet and a quadruplet at δ 0.99 and 3.01 ppm, and the molecyle backbone methylene protons give signals at δ 3.17–4.08 ppm.

Further, we studied a reaction of dimethylenedipentaerytritiol **III** with phosphorous hexamethyltriamide **VIII** in 1 : 2 molar ratio, leading to formation of symmetrical bisdiamidophosphite **IX**.



IX (4)

The phosphorylation was conducted in anhydrous dioxane at room temperature (25°C) for 48 h; the diethylamine formed in the reaction process was not removed. In ^{31}P NMR spectrum of crude amidophosphite **IX** we observed a broad signal at δ_P 135.40 ppm typical of diamidophosphites.

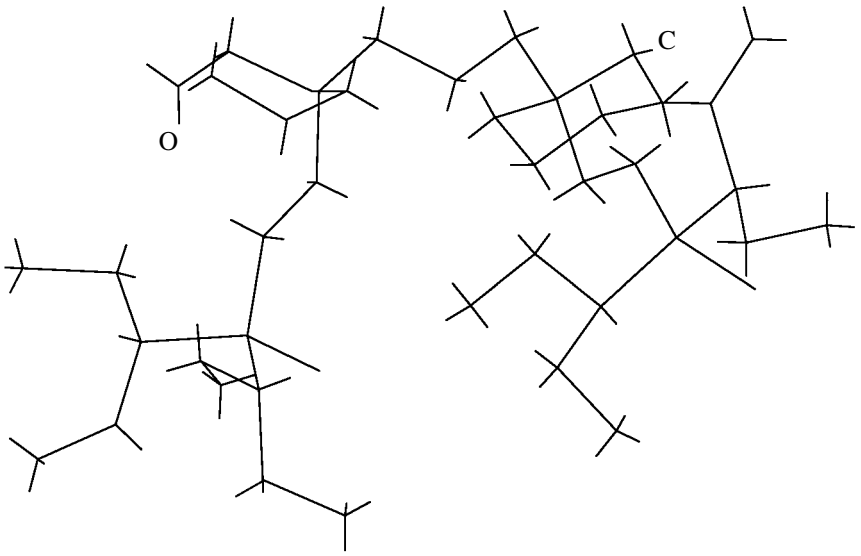


Fig. 2. Optimal conformation of the bisdiamidothionophosphate **X** molecule.

To confirm the structure of compound **IX** the latter was without additional purification transformed into bisamidothionophosphate **X**.

$$\mathbf{III} \xrightarrow{[S]} (Et_2N)_2P(S)O\text{–}R\text{–}O P(S)(NEt_2)_2. \quad (5)$$

X

Yield of chromatographically pure phosphachalcogenide **X** achieves 60% on two steps.

Structure of compound **X** is evidenced by ^{1}H and ^{31}P NMR spectroscopy. In the ^{31}P NMR spectrum was observed a broad singlet signal at δ_P 79.55 ppm. A feature of ^{1}H NMR spectrum of compound **X** is the presence in the spectrum of a singlet and a doublet of methylene protons of acetal protecting group, like the case of compound **VI**. The bridgehead methylene protons give two separate signals at δ 3.16 and 3.50 ppm. These spectral features are connected with steric organization of compound **X** molecules. This can be illustrated by the data of computer modeling of the molecule X (Fig. 2). The spectra also contained the signals of other protons belonging to this structure (see Experimental).

The bisdiamidophosphite **IX** obtained was explored as a basic compound for creation of the macrocyclic systems with two thionophosphoric fragments in their structure by the method of molecular assembling. Synthesis of such type dimeric phosphocyclanes which was proposed by us in [4] includes three steps: preparation of bisphosphorylated substituted diols, cyclophosphorylation by these compounds of second equivalent of the chosen diol and further stabilization by sulfurization of the biscyclophosphites obtained.

In correspondence with this synthetic scheme, the first step consisted of addition of bismethylated hexaol **III** to a solution of crude amidophosphite **IX** in dioxane.

$$\mathbf{IX} \xrightarrow[-HNEt_2]{\mathbf{III}} Et_2NP\begin{matrix}O\text{–}R\text{–}O\\O\text{–}R\text{–}O\end{matrix}PNEt_2\,. \quad (6)$$

XI

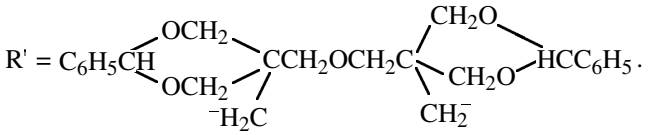
The ^{31}P NMR spectrum of the products of this reaction shows significant structural changes at the heating to 60–65°C even in 2 h. The ^{31}P NMR spectrum of the reaction mixture reflects accumulation of side compounds and only a weak signal occurs at δ_P 147.68 ppm (~5%) of the target compound **XI**.

Similar result (compound **XII**) occured with 1,3;1',3'-dibenzylidenedipentaerytritols **I** as a ring forming agents.

$$\mathbf{IX} \xrightarrow{\mathbf{I}} Et_2NP\begin{matrix}O\text{–}R\text{–}O\\O\text{–}R\text{–}O\end{matrix}PNEt_2. \quad (7)$$

XII

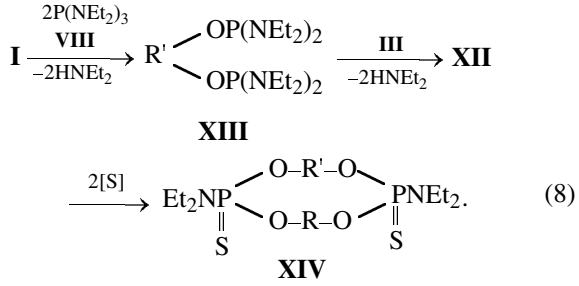
Here and hereinafter



These experiments show that dimethylenedipentaerytritiol diaidophosphite **IX** is not a convenient compound for preparation of macrocycles due to its termal instability.

Accounting for the foregoing, we performed synthesis of mixed macrophosphacycle **XII** with biscyclophosphites **XIII** obtained by us earlier from a mixture of the dibenzylidenepentaerythritols **I** geometric isomers [5] as initial reagents.

To the bisdiamidophosphites **XIII** prepared from dibenzylidenepentaerytritols **I** and hexaethyltriamidophsphite **VIII** alongside the procedure in [5] we added 1 equivalent of dimethylenedipentaerytritiol **III**. In the ^{31}P NMR spectrum appeared broad singlet of cyclophosphite **XII** at δ_P 147.68 ppm, which corresponds to chemical shift of the above described monophosphoric monomeric ring **VI**.



(8)

The amidocyclophosphite **XII** being not isolated readily reacted with sulfur to transform into corresponding 20-membered dithionidiphosphocyclane **XIV** containing fragments of two different acetals, dimethylene and dibenzylidene. Cyclothionophosphate **XIV** was isolated by column chromatography, yield up to 52%. Just after isolation the compound **XIV** was an oil which crystallized in two weeks. Its individuality was confirmed by the data of TLC and MALDI-TOF mass spectrometry. Its structure was also evidenced by elemental analysis and ^{31}P NMR spectroscopy. The ^{31}P NMR spectrum of cyclothionophosphate **XIV** contains a broad singlet signal at δ_P 76.64 ppm, in its ^1H NMR spectrum were fixed all proton groups characteristic of the above shown structure of **XIV** molecule. Besides, the signals of acetal methylene protons of the protected hexaol **III** were observed the proton signals of benzylidene groups: a broad singlet of methine proton in CHC_6H_5 (δ 5.41 ppm) and two signals at δ 7.37, 7.40 ppm and broad singlet at δ 7.47 of *m*-, *p*- and *o*-protons in the system of two aromatic rings.

Ordinary drying of 20-membered mixed dithionodiphosphocyclane **XIV** containing similtaneously dibenzylidene- (**I**) and dimethylenedipentaerytritiol (**III**) residues in its structure led to its anhydrous form, as follows from physicochemical study. Earlier described monotonic 20-membered dithionodiphosphocyclan **XV** synthesized from dibenzylidenedipentaerytritiols only, after similar drying contains one molecule of water [4]. The difference of compounds **XIV** and **XV** in the ability of retaining water results from the fact that dithiophosphocyclane **XV** molecule according to the data of molecular modeling (with accounting for atomic Van der Vaals radii) has a cavity which sterically can include water molecule [4]. But in dithionodiphosphocyclane **XIV** molecule there is no such a cavity: it is closed by one of 1,3-dioxane rings of methylene protecting group. These features of the molecule are clearly seen in the graphical representation of the molecule **XIV** optimal conformation (Fig. 3).

Thus, on the basis of amidophosphite chemistry we proposed a general method for the synthesis of the phosphorus containing macroheterocycles with regular coupling of phosphorus functional groups and structural blocks of dipentaerytritiol acetal derivatives.

EXPERIMENTAL

The ^1H NMR spectra were registered on a Bruker WM-250 instrument (250 MHz), chemical shifts are given relatively to internal HMDS. Assignment of proton signals was made on the basis of the data of double magnetic resonance. The $^{31}P–\{^1H\}$ NMR spectra were registered on a Bruker WP-80SY spectrometer with operating frequency 32.4 MHz, external reference 85% phosphoric acid.

Monoisotopic (^{12}C) molecular mass of compounds were measured on a Bruker UltraFlex mass spectrometer (Bruker Daltomics, Germany) for positive ions in the reflection mode using a N_2 laser (λ 337 nm) with accelerating voltage 25 kV. All the syntheses with trivalent phosphorus compounds were performed under dry argon atmosphere. Adsorption chromatography was performed on a 15 mm column through silica gel L 100–250 μk; the R_f values are determined by TLC on Silufol UV-254 plates using benzene–dioxane 3 : 1 (A), hexane–dioxane 3 : 1 (B), and ethyl acetate–hexane 4 : 1 (C) systems.

Melting points were measured in sealed capillary tubes, rate of temperature raise 1 deg min^{-1}.

Tetraethyldiamidophosphorous chloride (**V**) and phosphorous hexaethyltriamide (**VIII**) were prepared according to [11], dimethoxymethane (methinal) by [12], *cis–cis*, *cis–trans* and *trans–trans* isomeric mixture of 1,3;1',3'-dibenzylidenedipentaerytritiol (**I**) was obtained by method of [5], the products constants correspond to published data. Dipentaerytritiol (**II**) (Acros Organics), exactly 2',2'',6',6''-tetra(hydroxymethyl)-4-oxaheptane-1,7-diol prior to application was recrystallized from water (mp 222–225°C).

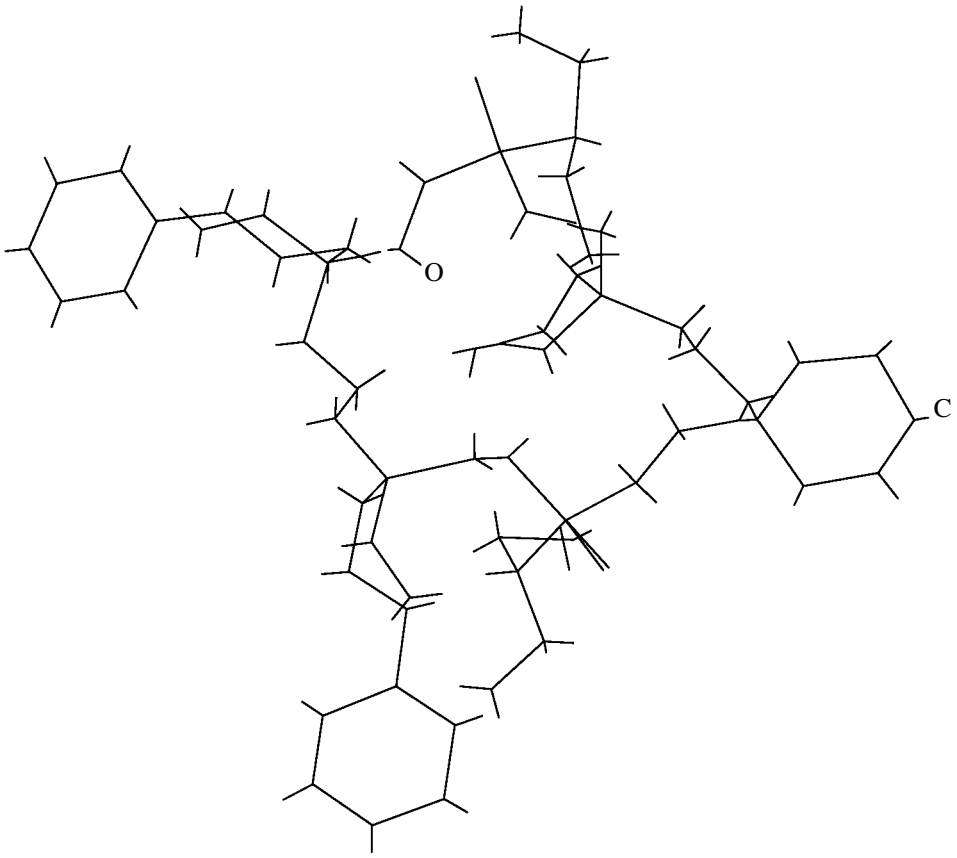


Fig. 3. Optimal conformation of the diamidocyclodithionodiphosphate **XIV** molecule.

2',2'':6',6''-Di-O-methylene-2',2'',6',6''-tetra(hydroxymethyl)-4-oxaheptan-1,7-diol (III). *a*. 0.5 g of dipentaerytritiol (**I**) was mixed with 3 ml of methinal **IV**, 0.01 g of boron trifluoride etherate was added and the mixture was kept in a sealed ampule at 100–110°C to complete dissolving of the initial compound **II**, 2 h (the reaction mixture darkened). The methinal excess was removed in a vacuum and dimethyleneacetal **III** was purified on a column with silica gel (10 g) filled with benzene. Compound **III** was elude with 35 ml of 1 : 1 benzene–dioxane mixture. The solvents were removed in a vacuum and residue was kept 2 h at 50°C (1 mm Hg). Chromatographic isolation yielded diacetal **III** 0.33 g (30%), mp 94–95°C (published [10]: mp 95–96°C), R_f 0.53 (A), 0.17 (B), 0.33 (C) [published [10]: R_f 0.34 (C)]. ^{1}H NMR spectrum (C_6D_6), δ, ppm: 2.78 br.s (2H, OH), 3.14 s (4H, CH_2OCH_2), 3.32 d (4H^a) and 3.61 d (4H^e) (CH_2OC, $^2J_{H^aH^e}$) 11.28 Hz), 3.43 d (4H, CH_2OH, $^3J_{HH}$ 6.41 Hz), 4.41 s and 4.66 s (4H, OCH_2O, $^2J_{HH}$ 6.1 Hz). Found, %: C 51.82; H 8.01. $C_{12}H_{22}O_7$. Calculated, %: C 51.79; H 7.97. *M* 278.30.

b. 1 g of dipentaerytritiol (**I**) was mixed with 5 ml of methinal **IV**, 0.01 g of *p*-toluensulfuric acid was added and the mixture was heated in a sealed ampoule at 120–125°C for 5 h. Diacetal **III** was isolated by column chromatography (see *a*). Yield 0.65 g (59%), n_D^{20} 1.5930.

10-Membered amidocyclothionophosphate VII. To a solution of 0.2 g of dimethylenedipentaerytritiol **III** and 0.8 g pyridine in 10 ml of anhydrous dioxane at stirring and cooling to 0°C was added dropwise for 2 min 0.15 g of chloride **V** (molar ratio 1 : 1) in 3 ml of the same solvent, then temperature was increased to 25°C and the reaction mixture was kept at this temperature for 1.5 h. Formation of cyclophosphite **VI** was monitored by the method of ^{31}P NMR spectroscopy (dioxane), δ_P, ppm: 147.9 br.s. Then to the reaction mixture was added 0.08 g of sulfur and mixture was kept at room temperature for 4 h. The sulfur excess was filtered off and solvents were removed in a vacuum. The cycloamidothionophosphate **VII** was purified on a column with silica gel (5 g) filled with hexane. Compound **VII** was elued with 25 ml of hexane–dioxane 5 : 1 mixture. The solvents were removed in a vacuum and residue was kept for 2 h at 80°C (1 mm Hg). Yield of compound **VII** 0.16 g (55%), n_D^{20} 1.5583, R_f 0.80 (A), 0.50 (B). ^{1}H NMR

spectrum (C_6D_6), δ, ppm: 1.99 t (6H, NCH_2CH_3, $^3J_{HH}$ 7.31 Hz), 3.01 q (4H, NCH_2CH_3, $^3J_{HP}$ 13.68 Hz), 3.17 s (4H, CH_2OCH_2), 3.39 d ($4H^a$) and 4.08 d ($4H^e$) [CH_2OC, $^2J_{H^aH^e}$ 5.57 Hz], 3.68 d.d (4H, CH_2OP, $^3J_{HP}$ 12.79 Hz), 4.47 s and 4.65 d.d (4H, OCH_2O, $^2J_{HH}$ 6.32 Hz, $^4J_{HH}$ 1.87 Hz). ^{31}P NMR spectrum (dioxane), δ_P, ppm: 77.78 br.s. Found, %: C 46.82; H 7.40; P 7.61. $C_{16}H_{30}NO_7PS$. Calculated, %: C 46.70; H 7.35; P 7.53. *M* 411.45. Found, *M* (^{12}C): 411.62. Calculated, *M* (^{12}C): 411.29.

1,7-Bis(tetraethyldiamido-2',2'';6',6''-di-*O*-methylene-2',2'';6',6''-tetra(hydroxymethyl)-4-oxaheptan-1,7-diol (X). 0.2 g dimethylenedipentaerytritiol (**III**) and 0.36 g of phosphorous hexaethyltriamide **VIII** (molar ratio 1 : 2) in 3 ml of anhydrous dioxane was stirred for 48 h at 25°C. Formation of amidphosphite **IX** was monitored by means of ^{31}P NMR spectroscopy. ^{31}P NMR spectrum (dioxane), δ_P, ppm: 135.40 br.s. To the reaction mixture was then added 0.07 g of sulfur at room temperature and mixture was kept at 40°C for 3 h. Sulfur excess was filtered off, dioxane was removed in a vacuum. Diamidodithionodiphosphate **X** was purified on a column with silica gel (5 g) filled with hexane. Compound **X** was elued with 30 ml of hexane–dioxane 3:1 mixture. Solvents were then removed in a vacuum and residue was kept for 2 h at 40°C (1 mm Hg). Yield of compound **X** 0.3 g (60%), n_D^{20} 1.5284, R_f 0.80 (A), 0.55 (B). ^1H NMR spectrum (C_6D_6), δ, ppm: 0.98 t (24H, $NCH_2 \cdot CH_3$, $^3J_{HH}$ 6.94 Hz), 2.99 q (16H, NCH_2CH_3, $^3J_{HP}$ 12.06 Hz), 3.16 s, 3.50 s (4H, CH_2OCH_2), 3.37 d ($4H^a$) and 4.05 d ($4H^e$) [CH_2OC, $^2J_{H^aH^e}$ 6.99 Hz], 3.68 d.d (4H, CH_2OP, $^3J_{HP}$ 11.69 Hz), 4.46 s and 4.67 d.d (4H, OCH_2O, $^2J_{HH}$ 6.22 Hz, $^4J_{HH}$ 1.46 Hz). ^{31}P NMR spectrum (dioxane), δ_P, ppm: 79.55 br.s. Found, %: C 48.81; H 8.90; P 9.03. $C_{28}H_{60}N_4O_7P_2S_2$. Calculated, %: C 48.67; H 8.75; P 8.97. *M* 690.87.

20-Membered diamidocyclodithionodiphosphate (XIV). 0.3 g of mixture of dibenzylidenedipentaerytritiols **I** and 0.35 g of phosphorous hexaethyltriamide **VIII** (molar ratio 1 : 2) in 5 ml of anhydrous dioxane was stirred for 12 h at 25°C. Formation of bisdiamidophosphite **XIII** was monitored by means of ^{31}P NMR spectroscopy. ^{31}P NMR spectrum (dioxane), δ_P, ppm: 135.20 br.s. Then to the reaction mixture was added equimolar amount (0.19 g) of dimethylenedipentaerytritiol **III** and the mixture was kept at 85°C for 40 h. Formation of cyclophosphite **XII** was monitored by ^{31}P NMR spectroscopy: δ_P 147.68 ppm, br.s. Then to the mixture was added 0.13 g of sulfur at room temperature and the mixture was kept at 40°C for 1 h. The sulfur excess was filtered off and dioxane was removed in a vacuum. Compound **XIV** was purified on a column with silica gel (5 g) willed with benzene. Compound **XIV** was elued with 40 ml of benzene. The solvent was removed in a vacuum and residue was kept for 5 h at 80°C (1 mm Hg). After 2 weeks keeping at 25°C compound has crystallized. Yield of **XIV** 0.13 g (52%), n_D^{20} 1.5693, mp 159–161°C, R_f 0.72 (A), 0.44 (B). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.15 t (12H, NCH_2CH_3, $^3J_{HH}$ 7.15 Hz), 3.17 q (8H, NCH_2CH_3, $^3J_{HP}$ 13.79 Hz), 3.50 m ($4H^a$) and 4.12 m ($4H^e$) (8H, CH_2OC), 3.69 s and 3.81 s (8H, CH_2OCH_2), 3.75 m ($4H^a$) and 4.33 m ($4H^e$) (CH_2OCH), 4.19 m (8H, CH_2OP), 4.57 d and 4.76 s (4H, OCH_2O, $^2J_{HH}$ 6.72 Hz), 5.41 br.s (2H, CHC_6H_5), 7.37 br.s (6H, *m*-, *p*-) and 7.47 br.s (4H, *o*-) (C_6H_5). ^{31}P NMR spectrum (dioxane), δ_P, ppm: 76.64 br.s. Found, %: C 54.23; H 7.18; P 6.41. $C_{44}H_{68}N_2O_{14}P_2S_2$. Calculated, %: C 54.19; H 7.03; P 6.35. *M* 975.08. Found, *M* (^{12}C): 974.24. Calculated *M* (^{12}C): 974.64.

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REFERENCES

1. Nifant'ev, E.E., Maslennikova, V.I., and Rasadkina, E.N., *Zh. Obshch, Khim.,* 1999, vol. 69, no. 11, p. 1813.
2. Bauer, I., Khabikher, V.D., Antipin, I.S., and Sinyashin, O.G., *Izv. Ross. Akad. Nauk, Ser. Khim.,* 2004, no. 7, p. 1348.
3. Uroguchi, D. and Terada, M., *J. Am. Chem. Soc.,* 2004, vol. 126, no. 17, p. 5356.
4. Predvoditelev, D.A., Malenkovskaya, M.A., and Nifant'ev, E.E., *Zh. Obshch. Khim.,* 2006, vol. 76, no. 8, p. 1254.
5. Predvoditelev, D.A., Malenkovskaya, M.A., Strebkova, E.V., and Nifant'ev, E.E., *Zh. Obshch. Khim.,* 2006, vol. 76, no. 7, p. 1095.
6. Yanovskaya, L.A., Yufit, S.S., and Kucherov, V.F., *Khimiya acetalei* (Cheistry of Acetals), Moscow: Nauka, 1973.
7. Kukharev, B.F., Stakevich, V.K., and Kukhareva, V.A., *Zh. Org. Khim.,* 2003, vol. 39, no. 5, p. 668.

8. Cabasso, I. and Sahni, S., *J. Polym. Sci., Part A: Polym. Chem.,* 1988, vol. 26, no. 11, p. 2997.
9. Allinger, H.L., *J. Am. Chem. Soc.,* 1977, vol. 99, no. 25, p. 8127.
10. Al-Mughaid, H., Crindley, T.B., Robertson, K.N., and Cameron, T.S., *Canad. J. Chem.,* 2003, vol. 81, p. 505.
11. Zwierzak, A., *Bull. Acad. Polon. Sci., Ser. Sci. Chim.,* 1965, vol. 13, no. 6, p. 609.
12. Veigand-Hilgetag, *Methoden in Organishe Chemie:* Moscow: Khimiya, 1969.