Available online at www.sciencedirect.com
ScienceDirect
Mendeleev Commun., 2008, **18**, 262–264
Mendeleev Communications

New products of the reaction of aldimines with dialkylphosphites

Mukattis B. Gazizov,*[a] Rafail A. Khairullin,[a] Anastasiya I. Alekhina,[a] Igor A. Litvinov,[b] Dmitrii B. Krivolapov,[b] Shamil K. Latypov,[b] Alsu A. Balandina,[b] Rashid Z. Musin[b] and Oleg G. Sinyashin[b]

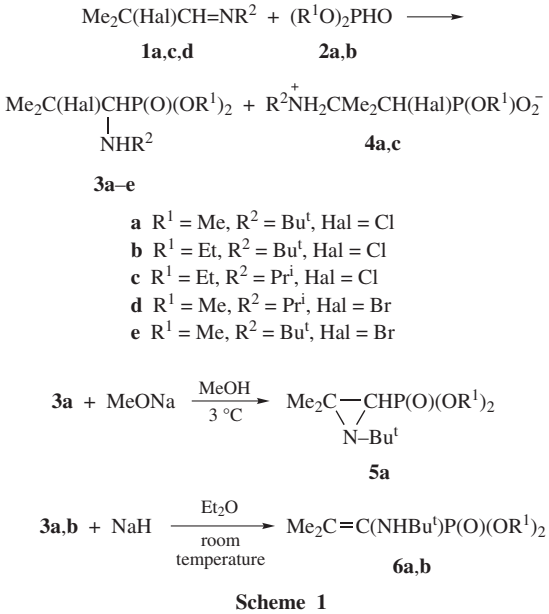
[a] *Kazan State Technological University, 420015 Kazan, Russian Federation. Fax: +7 843 231 4103; e-mail: mukattisg@mail.ru*
[b] *A. E. Arbuzov Institute of Organic and Physical Chemistry, Kazan Scientific Centre of the Russian Academy of Sciences, 420083 Kazan, Russian Federation. E-mail: oleg@iopc.knc.ru*

DOI: 10.1016/j.mencom.2008.09.012

Two types of new phosphobetaine compounds with aziridiniumyl and isomerised substituent at the phosphorus atom were synthesised by the reaction of 2-halogenaldimines with dialkylphosphites.

The interaction of 2-chloroaldimines **1** with trialkylphosphites and triphenylphosphine was described earlier.[1,2] However, no data on the reactions of compounds **1** with dialkylphosphites **2** are available, though the addition of the latter to halogen-unsubstituted imines is widely known.[3] We assumed that the presence of a halogen atom at the 2-position is responsible for new reaction paths and the formation of new types of organic compounds.

We found that the structure of the products of the interaction of 2-halogenaldimines **1**[4] with dialkylphosphites **2** depends strongly on the halogen atom. When Hal = Cl, the product of dialkylphosphite addition to the imine group **3b**† is formed at holding the mixture of corresponding compounds **1** and **2** for a short time (24 h) at room temperature. By keeping a mixture of **1a** and **2a** for a long-term period (14 days) at room temperature, a new product of a phosphobetaine structure with isomerised substituent at the phosphonate phosphorus atom **4a**† was obtained along with compound **3a** (Scheme 1). At a long-term interaction of reagents **1c** and **2a** at 25 °C, only compound **4c**† was isolated from the reaction mixture, though the formation of addition product **3c** (δ_P 29.4 ppm) was detected by ^{31}P NMR spectroscopy. The structures of products **3** and **4** were confirmed by ^{1}H and ^{31}P NMR spectra, and that of compound **4c**, by X-ray single crystal diffraction analysis‡ (Figure 1).



Scheme 1

† NMR spectra were recorded on a Bruker Avance-600 spectrometer [600 MHz (^{1}H), 150 MHz (^{13}C), 242.88 MHz (^{31}P) and 60 MHz (^{15}N)] in $CDCl_3$ at 30 °C. Chemical shifts for ^{1}H, ^{13}C were measured with reference to the residual protons of $CDCl_3$, for ^{31}P – with external reference to H_3PO_4, for ^{15}N – with reference to CD_3CN.

Mass spectra (EI) were obtained on a TRACE MS Finnigan MAT instrument, the energy of ionizing electrons was 70 eV. The temperature of the ion source was 200 °C. MALDI mass spectrum was obtained on a DYNAMO MALDI TOF (Thermo Bioanalysis Finnigan, USA) mass spectrometer and ion *m*/*z* 222 $[M + H]^+$ peak (compound **7e**) was maximum.

Reaction of N-*(2-chloro-2-methylpropylidene)*tert-*butylamine* **1c** *with dimethylphosphite* **2a**. Amine **1c** (6.53 g, 0.04 mol) was mixed with compound **2a** (4.4 g, 0.04 mol) under argon atmosphere. A slight warming-up of the reaction mixture was observed. The mixture was kept for 14 days at room temperature. The crystals precipitated after working-up of the reaction mixture with diethyl ether were filtered off, washed with Et_2O and recrystallised from acetonitrile. 1.23 g of *O*-methyl [3-(*tert*-butylammonio)-1-chloro-2-methylpropyl]phosphonate **4a** was obtained: yield 12%, mp 150–151 °C. ^{1}H NMR ($CDCl_3$) δ: 1.68 (s, 9H, CMe_3), 1.76 and 1.82 (s, 6H, CMe_2), 3.94 (d, 6H, POMe, *J* 11 Hz), 4.60 (d, PCH, *J* 8.5 Hz), 9.62 and 10.91 (br. d, 2H, $^+NH_2$, *J* 11 Hz).

After removing diethyl ether from mother solution *in vacuo*, 9.28 g of *O*,*O*-dimethyl [1-(*tert*-butylamino)-2-chloro-2-methylpropyl]phosphonate **3a** was obtained: yield 85%, mp 76–77 °C. ^{1}H NMR ($CDCl_3$) δ: 1.29 (s, 9H, CMe_3), 1.83 and 1.90 (s, 6H, CMe_2), 1.82–1.87 (br. s, 1H, NH), 3.37 (d, 1H, PCH, *J* 20 Hz), 3.93 and 3.94 (d, 6H, POMe, *J* 11 Hz). ^{31}P NMR ($CHCl_3$) δ: 29.3.

Reaction of **1c** *with diethylphosphite* **2b**. 6.8 g (0.04 mol) of **1c** and 5.52 g (0.04 mol) of **2b** gave 9.7 g of *O*,*O*-diethyl [1-(*tert*-butylamino)-2-chloro-2-methylpropyl]phosphonate **3b**: yield 81%. ^{1}H NMR ($CDCl_3$) δ: 1.08 (s, 9H, CMe_3), 1.28 (t, 6H, $POCH_2Me$, *J* 7 Hz), 1.28–1.44 (br. s, 1H, NH), 1.60 and 1.64 (s, 6H, CMe_2), 3.03 (d, 1H, PCH, *J* 19 Hz), 4.05 (quintet, 4H, $POCH_2$, *J* 7 Hz). ^{31}P NMR ($CHCl_3$) δ: 26.43.

Reaction of N-*(2-chloro-2-methylpropylidene)isopropylamine* **1d** *with diethylphosphite* **2b**. 5 g (0.03 mol) of **1d** and 4.14 g (0.03 mol) of **2b** gave 9.7 g of *O*-ethyl [3-(isopropylammonio)-1-chloro-2-methylpropyl]-phosphonate **4c**: yield 10%, mp 145–146 °C. ^{1}H NMR ($CDCl_3$) δ: 1.43 (t, 3H, $POCH_2Me$, *J* 7 Hz), 1.58 and 1.63 (d, 6H, $CHMe_2$, *J* 6.5 Hz), 1.67 (s, 6H, CMe_2), 3.36–3.64 (m, 1H, $CHMe_2$), 4.30 (quintet, 2H, OCH_2Me, *J* 7 Hz), 4.56 (d, 1H, PCH, *J* 9 Hz), 9.67 and 10.43 (br. d, 2H, $^+NH_2$, *J* 10.5 Hz).

© 2008 Mendeleev Communications. All rights reserved.

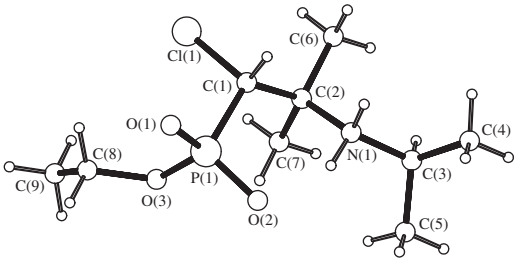
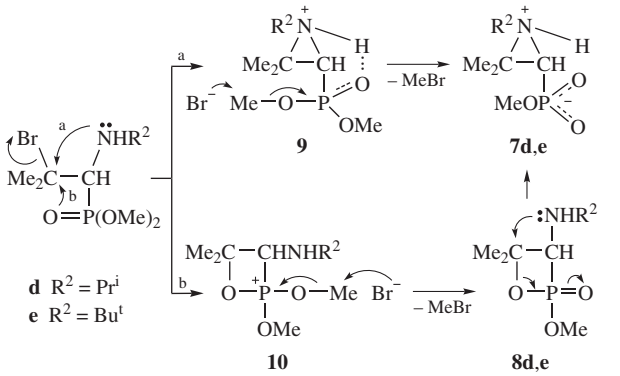


Figure 1 The structure of product **4c**. Selected bond lengths (Å): P(1)–O(1) 1.478(2), P(1)–O(2) 1.484(2), P(1)–O(3) 1.578(2), P(1)–C(1) 1.850(3), Cl(1)–C(1) 1.788(3), O(3)–C(8) 1.443(4), N(1)–C(2) 1.508(4), N(1)–C(3) 1.522(4), C(1)–C(2) 1.550(4), C(2)–C(7) 1.512(5), C(2)–C(6) 1.532(4), C(3)–C(5) 1.489(6), C(3)–C(4) 1.501(6), C(8)–C(9) 1.381(6).

Compounds **3** were transformed into aziridine-2-ylphosphonate **5a** and phosphorylated enamines **6a**,**b** (Scheme 1)§ by treatment with sodium methylate in methanol and sodium hydride in absolute diethyl ether, respectively.

When Hal = Br, bromimine **1d** appeared to be cosiderably more active than imine **1e**. In 10 h after mixing reagents **1d** and **2a**, two resonance signals at 26.14 and 1.17 ppm, corresponding to the phosphorus atoms in addition product **3d** and a new product, were detected in the ^{31}P NMR spectrum of the reaction mixture. In 30 h, the signal at 26.14 ppm vanished, and only one of the intense signals (at 1.17 ppm) was in the spectrum. New signals appeared in 25 h after mixing compounds **1e** and **2a**, and a signal at 26.50 ppm vanished completely in three days. Thus, in the case of bromimines **1d**,**e**, at first addition products **3d**,**e** were formed, which easily transformed into new compounds.¶ Unlike primary addition compounds **3d**,**e**, the new products did not contain bromine. A bromine atom leaves a reaction system as methyl bromide, as confirmed by the presence of an intense signal at 2.6 ppm in the ^{1}H NMR spectrum of the reaction mixture (Scheme 2).



Scheme 2

Two isomeric structures with either aziridiniumyl substituent at phosphonate phosphorus atom (**7d**,**e**) or with 1,2-oxaphosphetane cycle (**8d**,**e**) may correspond to the new isolated products, substituted at the nitrogen atom [R^2 = Pr^i ($C_8H_{18}NO_3P$) or Bu^t

‡ *X-Ray diffraction data for* **4c** were collected at 20 °C on an Enraf-Nonius CAD4 automatic diffractometer using graphite monochromated MoKα (0.71073 Å) radiation. Crystals of compound **4c** ($C_9H_{21}ClNO_3P$), monoclinic, a = 10.089(2), b = 11.865(2) and c = 11.688(4) Å, β = 102.12(2)°, V = 1367.9(6) Å^3, Z = 4, d_{calc} = 1.25 g cm^{-3}, space group $P2_1/n$.

X-Ray diffraction data for **7d** were collected on a Bruker Smart Apex II CCD diffractometer using graphite monochromated MoKα (0.71073 Å) radiation. Crystals of compound **7d** ($C_8H_{18}NO_3P{\cdot}2CHCl_3$), monoclinic, a = 15.452(7), b = 9.254(4) and c = 16.493(6) Å, β = 117.182(6)°, V = 2097.9(14) Å^3, Z = 4, d_{calc} = 1.41 g cm^{-3}, space group $P2_1/n$.

Cell parameters and intensities of 2768 (**4c**) and 4891 (**7d**) independent reflections, from which 1577 (**4c**) and 1929 (**7d**) with $I \geq 2\sigma$, were measured in the $\omega/2\theta$-scan mode, $\theta \leq 26.30°$ for **4c** and ω-scan, $\theta \leq 28.00°$ for **7d**. Absorption correction was not applied [μ(MoKα) = 3.86 cm^{-1} for **4c**], [μ(MoKα) = 9.0 cm^{-1} for **7d**]. The structures were solved by direct method using the SIR[8] program and refined by the full-matrix least-squares using the SHELXL-97[9] program. All non-hydrogen atoms were refined anisotropically. The hydrogen atoms were calculated and refined as riding atoms. All calculations were performed on a PC using the WinGX program.[10] The final residuals were R_{ob} = 0.046, wR_{ob} = 0.1272 (for **4c**) and R_{ob} = 0.062, wR_{ob} = 0.1419 (for **7d**). Data collection and reductions for **4c** were performed on an Alpha Station 200 computer using the MolEN program package.[11] For **7d** the images were indexed, integrated, and scaled using the APEX2 data reduction package.[12] All figures were made using the PLATON program.[13]

CCDC 696939 and 696940 contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre *via* www.ccdc.cam.ac.uk/data_request/cif. For details, see 'Notice to Authors', *Mendeleev Commun.*, Issue 1, 2008.

§ *Interaction of compound* **3a** *with alcoholic solution of sodium methylate*. To 6.08 g (0.02 mol) of **3a** in 35 ml of ice cooled methanol 2.55 g of sodium methylate (1.2 M solution in methanol) was added mantaining a reaction mixture temperature of 3 °C. The mixture was stirred during 24 h. Precipitated NaCl was filtered off, mother solution was steamed and the residue was distilled. 3 g of *O*,*O*-dimethyl (1-*tert*-butyl-3,3-dimethylaziridin-2-yl)phosphonate **5a** was obtained: yield 60%, bp 74 °C (0.1 Torr), n_D^{20} 1.4532. ^{1}H NMR ($CDCl_3$) δ: 0.97 (s, 9H, CMe_3), 1.15 (s, 1H, *Me*CMe) and 1.22 (d, 3H, MeC*Me*, *J* 2.3 Hz), 1.52 (d, 1H, CH, *J* 22 Hz), 3.5 (d, POMe, *J* 11 Hz). ^{13}C NMR ($CDCl_3$) δ: 22.32 and 26.56 (s, C*Me*$_2$), 30.17 (s, C*Me*$_3$), 37.91 (d, PCH, *J* 214.5 Hz), 44.14 (s, *C*Me$_2$), 53.17 (d, OMe, *J* 17 Hz), 55.75 (s, *C*Me$_3$).

Reaction of compound **3a** *with sodium hydride*. Under argon atmosphere by stirring to 4.32 g (0.18 mol) of NaH in 50 ml of dry diethyl ether 15.89 g (0.06 mol) of **3a** in 200 ml of dry Et_2O was dropped. The mixture was stirred at room temperature during 1 h and refluxed for 24 h. The cooled reaction mixture was washed with 50 ml of cold warter, extracted with diethyl ether (2×50 ml) and combined extracts were dried with $MgSO_4$. The solvent was removed and the residue was distilled. 9 g of *O*,*O*-dimethyl [1-(*tert*-butylamino)-2-methyl-1-propenyl]phosphonate **6a** was obtained: yield 81%, bp 66 °C (0.09 Torr), n_D^{20} 1.4655. ^{1}H NMR ($CDCl_3$) δ: 0.96 (s, 9H, CMe_3), 1.78 and 1.81 (d, 6H, CMe_2, *J* 1.2 Hz), 2.39 (br. s, 1H, NH), 3.52 (d, 6H, POMe, *J* 11 Hz). ^{13}C NMR ($CDCl_3$) δ: 22.49 and 24.63 (s, C*Me*$_2$), 31.78 (s, C*Me*$_3$), 53.30 (d, POMe, *J* 4.5 Hz), 56.96 (s, *C*Me$_3$), 128 (d, PC=, *J* 183.53 Hz), 149.28 (d, HNC=, *J* 30.35 Hz). ^{31}P NMR ($CHCl_3$) δ: 22.5.

O,O-*Diethyl [1-*(tert-*butylamino)-2-methyl-1-propenyl]phosphonate* **6b**. 11.19 g (0.037 mol) of **3b** and 2.66 g (0.111 mol) NaH gave 9.73 g of **6b**: yield 70%, bp 76 °C (0.09 Torr), n_D^{20} 1.4568. ^{1}H NMR ($CDCl_3$) δ: 1.07 (s, 9H, CMe_3), 1.28 (t, 6H, $POCH_2Me$, *J* 7 Hz), 1.89 and 1.92 (d, 6H, CMe_2), 2.56 (br. s, 1H, NH), 4.05 (quintet, 4H, $POCH_2$, *J* 7 Hz). ^{31}P NMR ($CHCl_3$) δ: 26.4.

¶ *Interaction of* N-*(2-bromo-2-methylpropylidene)isopropylamine* **1d** *with dimethylphosphite* **2a**. Under argon atmosphere 1.44 g (0.75 mmol) of **1d** was mixed with 0.83 g (0.75 mmol) of **2a**. The mixture was kept at room temperature for 33 h. The precipitated product was filtered off, washed with diethyl ether and acetone and dried with $MgSO_4$. 0.57 g of *O*-methyl (1-isopropyl-3,3-dimethylaziridinium-2-yl)phosphonate **7d** was obtained: yield 37%, mp 125–126 °C. ^{1}H NMR ($CDCl_3$) δ: 1.38 and 1.42 (d, 6H, CH*Me*$_2$, *J* 6.6 Hz), 1.47 and 1.69 (s, 6H, CMe_2), 2.17 (d, 1H, PCH, *J* 2.9 Hz), 2.66 (m, 1H, NCH), 3.59 (d, 3H, POMe, *J* 10.3 Hz), 9.90 (br. s, 1H, NH). ^{13}C NMR ($CDCl_3$) δ: 19.08 (s) and 19.24 (d) (C*Me*$_2$, *J* 3.0 Hz), 19.68 (d) and 19.96 (s) (CHMe$_2$, *J* 6.6 Hz), 47.7 (d, PCH, *J* 161.6 Hz), 50.0 (s, *C*Me$_2$), 54.3 (s, *C*HMe$_2$), 51.77 (d, POMe, *J* 6.0 Hz). ^{15}N NMR, δ: 46. ^{31}P NMR, δ: 1.17.

O-*Methyl (1-*tert-*butyl-3,3-dimethylaziridinium-2-yl)phosphonate* **7e**. 2.26 g (0.01 mol) of *N*-(2-bromo-2-methylpropylidene)*tert*-butylamine **1e** and 1.10 g (0.01 mol) of **2a** gave 1.23 g of compound **7e**: yield 56%, mp 132 °C. ^{1}H NMR ($CDCl_3$) δ: 1.39 (s, 9H, CMe_3), 1.57 and 1.73 (s, 6H, CMe_2), 2.59 (d, 1H, PCH, *J* 2.9 Hz), 3.58 (d, 3H, POMe, *J* 11 Hz), 8.40 (br. s, 1H, NH). ^{13}C NMR ($CDCl_3$) δ: 20.87 (s, C*Me*$_2$), 21.59 and 27.19 (s, C*Me*$_3$), 42.74 (d, PCH, *J* 162.8 Hz), 51.58 (d, POMe, *J* 5.4 Hz), 52.3 (s, *C*Me$_2$), 60.66 (s, *C*Me$_3$). ^{15}N NMR, δ: 52. ^{31}P NMR, δ: 1.19. MS, *m/z* (%): 222 (3.6) $[M + H]^+$, 221 (1.9) $[M]^+$, 206 (2.9) $[M - Me]^+$, 164 (11.4) $[M - Bu]^+$, 138 (100.0) $[C_4H_{11}O_3P]^+$, 127 (39) $[C_8H_{17}N]^+$, 112 (66.6) $[C_7H_{14}N]^+$, 96 (41.0) $[C_6H_{10}N]^+$, 94 (1.5) $[MeOPO_2]^+$, 70 (62.3) $[C_4H_8N]^+$, 57 (53.5) $[C_3H_7N]^+$, 41 (34.3) $[C_2H_3N]^+$.

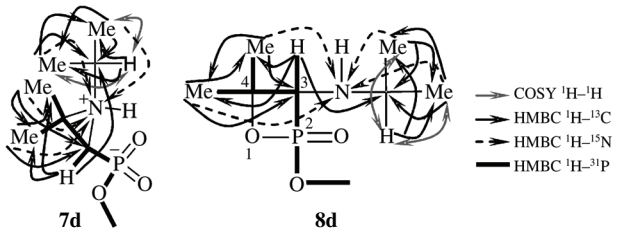


Figure 2 Principal COSY and HMBC correlations for **7d** and **8d** (from protons to carbons, nitrogen and phosphorus).

($C_9H_{20}NO_3P$)] (Scheme 2). The compounds structures were established by 2D NMR correlation methods.[5] The COSY and HMBC key correlations for a compound with $R^2 = Pr^i$ are summarized in Figure 2. Unfortunately, these HMBC correlations can correspond to both structural isomers, so they failed to distinguish these structures.

Since the chemical shifts of a number of atoms are expected to differ significantly in these structures [*e.g.*, P–C*H*, N*H*, P–*C*H and C(4)], the theoretical chemical shifts should be involved to establish the isomeric structures. The quantum-chemical gauge invariant atomic orbitals density functional theory (GIAO DFT) calculation of chemical shifts proved to be a useful method for establishing such spectrum–structure correlations.[6,7] For this purpose, non-empirical chemical shifts calculations were carried out [GIAO B3LYP/6-31G(d)//HF/6-31G][6(*e*)] for **7d** and **8d** and compared with the experimental data.

Thus, we compared a discrepancy between the experimental and calculated chemical shifts for the corresponding nuclei [*e.g.*, P–C*H*, N*H*, P–*C*H, C(4)] for both isomers (Figure 3). As can be seen in these diagrams, the difference in chemical shifts is minimal only for isomer **7d**, while for **8d** the deviations are obviously higher for the case of both proton and carbon.

Thus, upon the 2D NMR data and GIAO DFT chemical chifts calculations the new products of the reaction of **1d**,**e** and **2a** are proved to have the structure of **7d**,**e**.

The mass-spectroscopic data[§] also give evidence for the structure of **7d**,**e**. Finally, the structure of compound **7d** was confirmed by X-ray analysis (Figure 4).

The transformation of primary products **3d**,**e** with methyl bromide elimination can proceed in two ways (Scheme 2): (a) nucleophilic substitution of bromine atom under the influence of tricoordinated nitrogen atom with the formation of aziridiniumyl fragment in intermediates **9** and their demethylation with the formation of reaction products **7d**,**e**; (b) nucleophilic substitution of bromine atom under the influence of phosphoryl oxygen with the formation 1,2-oxaphosphetane fragment in intermediates **10** and their demethylation by the scheme of the second stage of the Arbuzov–Michaelis reaction with the formation of compounds **8d**,**e**, which can isomerize to products **7d**,**e** under the influence of tricoordinated nitrogen of the secondary amine group.

The NMR spectroscopy, mass spectrometry and elemental analysis data correspond to the structural formulae of the obtained compounds.[†]

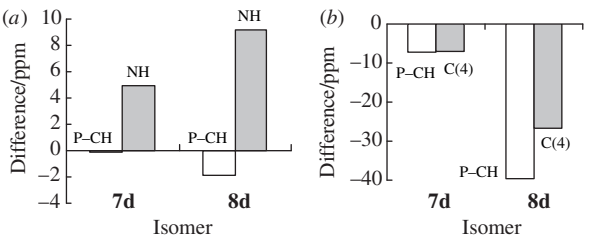


Figure 3 Difference of experimental and calculated (*a*) ^{1}H and (*b*) ^{13}C NMR chemical shifts for **7d** and **8d**.

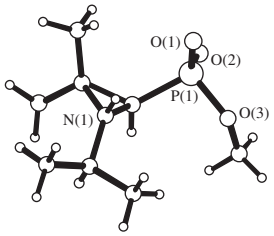


Figure 4 Molecular structure of **7d**, the disordered $CHCl_3$ molecules are not shown. Selected bond lengths (Å): O(2)–P(1) 1.467(2), O(3)–C(13) 1.443(5), O(3)–P(1) 1.600(3), O(1)–P(1) 1.464(3), N(1)–C(1) 1.480(4), N(1)–C(5) 1.489(5), N(1)–C(2) 1.501(5), C(1)–C(2) 1.496(5), C(1)–P(1) 1.813(4), C(5)–C(7) 1.497(6), C(5)–C(6) 1.502(5), C(2)–C(3) 1.496(6), C(2)–C(4) 1.503(6).

Sh.L. acknowledges the Russian Science Support Foundation for a doctoral grant. This investigation was carried out in NMR Department of the Federal Collective Spectral Analysis Center for Physical and Chemical Investigations of Structure, Properties and Composition of Matter and Materials (CKP SAC) and Federal CKP for Physical and Chemical Investigations of Matter and Materials (FCKP PCI) (state contracts of the Russian Federation Ministry of Education and Science nos. 02.451.11.7036 and 02.451.11.7019).

References

1 T. Kametani and O. Umezawa, *Chem. Pharm. Bull. (Tokyo)*, 1966, **14**, 369.
2 G. Gaudiano, R. Mondelli, P. P. Ponti, C. Ticozzi and A. Umani-Ronchi, *J. Org. Chem.*, 1968, **33**, 4431.
3 I. V. Konovalova and L. A. Burnaeva, *Reaktsiya Pudovika* (*The Pudovik Reaction*), Izd. KGU, Kazan, 1991, p. 146 (in Russian).
4 N. De Kimpe and N. Schamp, *Org. Prep. Proced. Int.*, 1979, **11**, 115.
5 (*a*) A. E. Derome, *Modern NMR Techniques for Chemistry Research*, Pergamon, Oxford, 1988; (*b*) Atta-ur-Rahman, *One and Two Dimensional NMR Spectroscopy*, Elsevier, Amsterdam, 1989, p. 579; (*c*) T. Parella, *Magn. Reson. Chem.*, 1998, **36**, 467; (*d*) M. Findeisen and S. Berger, *Magn. Reson. Chem.*, 2003, **41**, 431; (*e*) R. Crouch, R. D. Boyer, R. Johnson and K. Krishnamurthy, *Magn. Reson. Chem.*, 2004, **42**, 301; (*f*) T. Sprang and P. Bigler, *Magn. Reson. Chem.*, 2004, **42**, 55.
6 (*a*) A. De Dios, *Prog. Nucl. Magn. Reson. Spectrosc.*, 1996, **29**, 229; (*b*) G. Barone, L. Gomez-Paloma, D. Duca, A. Silvestri, R. Riccio and G. Bifulco, *Chem. Eur. J.*, 2002, **8**, 3233; (*c*) P. Cimino, L. Gomez-Paloma, D. Duca, R. Riccio and G. Bifulco, *Magn. Reson. Chem.*, 2004, **42**, 26; (*d*) I. Alkorta and J. Elguero, *Magn. Reson. Chem.*, 2004, **42**, 955; (*e*) A. A. Balandina, A. A. Kalinin, V. A. Mamedov, B. Figadere and Sh. K. Latypov, *Magn. Reson. Chem.*, 2005, **43**, 816; (*f*) A. Balandina, D. Saifina, V. Mamedov and Sh. Latypov, *J. Mol. Struct.*, 2006, **791**, 77; (*g*) G. Bifulco, P. Dambruoso, L. Gomez-Paloma and R. Riccio, *Chem. Rev.*, 2007, **107**, 3744.
7 (*a*) V. F. Mironov, A. A. Shtyrlina, A. T. Gubaidullin, A. V. Bogdanov, I. A. Litvinov, N. M. Azancheev, Sh. K. Latypov, R. Z. Musin and Yu. Ya. Efremov, *Izv. Akad. Nauk., Ser. Khim.*, 2004, **53**, 186 (*Russ. Chem. Bull., Int. Ed.*, 2004, **53**, 194); (*b*) V. F. Mironov, E. R. Zagidullina, A. B. Dobrynin, A. T. Gubaidullin, Sh. K. Latypov, R. Z. Musin, I. A. Litvinov, A. A. Balandina and I. V. Konovalova, *Arkivoc*, 2004, **22**, 95.
8 A. Altomare, G. Cascarano, C. Giacovazzo and D. Viterbo, *Acta Crystallogr., Sect. A*, 1991, **47**, 744.
9 G. M. Sheldrick, *SHELXL-97 Program for Crystal Structure Refinement*, University of Göttingen, Germany, 1997.
10 L. J. Farrugia, *J. Appl. Crystallogr.*, 1999, **32**, 837.
11 L. H. Straver and A. J. Schierbeek, *MolEN Structure Determination System*, Nonius B.V., 1994, **1**, 108.
12 *APEX2 (Version 2.1), SAINTPlus. Data Reduction and Correction Program (Version 7.31A)*, Bruker Advanced X-ray Solutions, Bruker AXS Inc., Madison, Wisconsin, USA, 2006.
13 A. L. Spek, *Acta Crystallogr., Sect. A*, 1990, **46**, 34.

Received: 5th March 2008; Com. 08/3093