Bisphosphorylated ligands based on isomeric N, N'-phenylenedibenzimidoyl dichlorides

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The Arbuzov reaction of N,N'-phenylenedibenzimidoyl dichlorides furnished new tetradentate ligands, *viz.*, N,N'-bis[(diphenylphosphoryl)phenylmethylidene]benzenediamines, which form complexes with lanthanides.

Key words: phenylenediamines, imidoyl chlorides, the Arbuzov reaction, phosphonates, phosphine oxides, complexes, lanthanides.

Imidoyl chlorides are useful as building blocks in the synthesis of imidates,¹ amidines,^{2–4} imidoyl cyanides,⁴ alkylated imines,⁵ ketimines,⁶ as well as various heterocyclic compounds possessing biological activity, since the chlorine atom in them can be easy enough substituted for the nitrogen or sulfur atom under the action of, for example, NH and SH nucleophiles. For *N*-arylimidoyl chlorides activated with a perfluoroalkyl substituent, the corresponding phosphonates (I_A) have been obtained by the Arbuzov reaction,⁷ which exhibit anticholinesterase activity.^{7a}



 $R^{F} = CF_{3}, C_{2}F_{5}, C_{3}H_{7}$ R = 4-Me, 3-Me, H, 4-Cl, 3-Cl, 3-CF₃

A detailed study of the reactivity of imidoyl chlorides in the Arbuzov reaction revealed⁸ that compounds with electron-withdrawing substituents at the carbon atom react more readily, with the products being able to undergo various elementotropic transformations.

Currently, a C=N bond as a functional group competes with a C=O bond in its importance in organic chemistry. In the era of metal complex catalysis, this is much due to the interesting coordinating and catalytic properties of the ligands containing an imine functional group (Schiff bases), where wide variation of the nature of sub-

* Institut für Anorganische und physikalische Chemie, Universität Bremen, Germany. gested that imidoyl chlorides can be used for the synthesis of ligands of various types too, including phosphorus-containing ones. Nevertheless, the only example for the synthesis of bidentate ligands (I_B) based on imidoyl chlorides has been described,² where palladium complexes proved to be efficient in the Suzuki and Heck reactions.

stituents in the azomethine fragment is possible. Such

compounds are characterized by diverse coordinating be-

havior allowing one to obtain mono-, bi-, and multinucle-

ar complexes of the chelate and molecular types, as well as metal-containing polymeric structures.⁹ It could be sug-



It should be noted that the most data on the reactivity of imidoyl chlorides obtained by now are related to their mono derivatives, whereas investigations of their bis-analogs comprise only the reactions with NH (see Refs 2–4) and OH nucleophiles,^{1,10} sodium azide,¹¹ and malonic ester dianion.¹²

Known methods for the synthesis of imidoyl chlorides are based on the reaction of the corresponding acid amides with various chlorinating agents, such as phosphorus pentachloride (in solvents,^{3,4} including thionyl chloride¹³), thionyl chloride,¹⁴ phosgene,¹⁵ and a triphenylphosphine—carbon tetrachloride system.¹⁶ Here, even in the case of mono derivatives, the target imidoyl chlo-

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Fig. 1. General view of the *Z*,*Z*-isomer of compound **2a**. Main bond distances (Å): Cl(1)-C(7) 1.783(2), Cl(2)-C(14) 1.778(1), N(1)-C(1) 1.418(2), N(1)-C(7) 1.254(2), N(2)-C(3) 1.417(2), N(2)-C(14) 1.255(2); main bond angles (deg): C(2)-C(1)-C(7)-N(1) 71.0(1), C(2)-C(3)-C(14)-N(2) 68.2(1).

rides frequently are not isolated in the individual state and are used in subsequent reactions without additional purification. Moreover, diimidoyl dichloride obtained from m-phenylenediamine is found to be unstable.³

It could be assumed that the Arbuzov reaction in the series of diimidoyl dichlorides, synthesized in turn from isomeric phenylenediamines, would lead to new types of oligodentate ligands containing the oxygen atom of the phosphoryl group and the sp²-hybridized nitrogen atom as the coordinating sites. Dibenzamides **1a**,**b**, obtained from p- and m-phenylenediamines, gave rise to diimidoyl dichlorides **2a**,**b** (Scheme 1). For the synthesis of compounds **2a**,**b**, we applied a modified procedure using phosphorus pentachloride as a chlorinating agent. It differs from the known versions by milder conditions, in particular, tolu-

Scheme 1



2a,b

Here and further: a, meta-isomer, b, para-isomer.

ene (reflux, 10 h) was used as a solvent. Diimidoyl dichlorides **2a,b** formed precipitate state the reaction mixtures after cooling and can be isolated in the individual form by simple filtration, though this is accompanied by the loss of the target compounds due to their partial solubility in toluene (the yield was ~75%). Evaporation of the volatile components *in vacuo* and washing of products **2a,b** with light petroleum seems an optimum method for the isolation of such compounds (the yields were 95 and 97% for **2a** and **2b**, respectively).

The structures of stable in the individual state diimidoyl dichlorides **2a**,**b** were confirmed not only by the spectral methods and elemental analysis, but also by a single crystal X-ray diffraction.* General views of the diimidoyl dichlorides molecules 2a and 2b are given in Figures 1 and 2, respectively. In the crystal, *m*-diimidoyl dichloride **2a** exists as the Z, Z-isomer (relative to the double bonds). *p*-Diimidoyl dichloride **2b** occupies a particular position, the center of symmetry, and is also characterized by the Z,Z-isomerism of the multiple bonds. The main geometrical parameters of structures 2a and 2b are close. Due to the steric reasons, the C=N groups are not coplanar with the central plane of the arene ring with the turn angles equal to ~ 70 and 50° in **2a** and **2b**, respectively. In turn, the absence of conjugation of the C=N bonds leads to the fact that their distances correspond to the values

^{*} It should be noted that the melting point for p-diimidoyl dichloride **2b** obtained by us, whose structure was confirmed by elemental analysis and physico-chemical methods, considerably differs from the data of other authors (see Experimental). This can be explained by the fact that earlier the authors obtained a mixture of diimidoyl dichloride, amide, and products of their resinification under significantly more severe reaction conditions.



Fig. 2. General view of the *Z*,*Z*-isomer of compound **2b**. Main bond distances (Å): Cl(1)-C(7) 1.777(2), N(1)-C(7) 1.257(2), N(1)-C(1) 1.417(2); the bond angle C(7)-N(1)-C(1)-C(3A) 54.2(1)°.

typical for of double bonds. In crystals, excluding a number of weak interactions C–H... π and C–H...Cl, the rest of contacts correspond to the Van der Waals interactions. Examination of structural homogeneity of these compounds by X-ray powder diffraction showed that the sample of *para*-isomer **2b** corresponds to the structure of a single crystal, whereas for the sample of *meta*-isomer **2a**, the presence of additional peaks is characteristic, which indicates the presence of other isomer (or polymorph).

Upon study of the Arbuzov reaction for diimidoyl dichlorides 2a,b with triethyl phosphite, it was found that, in contrast to the highly electrophilic perhaloalkyl-substituted imidoyl chlorides,^{7,8} under standard conditions, *i.e.* upon reflux of a stoichiometric mixture of reagents in benzene or toluene, the target products are formed in less than 15% yield even after prolonged (60 h) heating of the reaction mixture. When triethyl phosphite is used as the solvent, the content of the target product increases (Scheme 2), but a parallel formation of the phosphorylation products at both the nitrogen atom and the carbon atom, 4a,b and 5a,b, is observed. The use of nickel dichloride as a catalyst (5-7 mol.%) leads to the increase in the yield of all three compounds 3-5 (see Scheme 2). Note that phosphonates 3 undergo partial transformation into the products of phosphorotropic rearrangements 4 and 5 also in the process of chromatographic purification, which does not allow us to isolate these compounds in the individual states.

The mass spectra of the reaction mixtures exhibit peaks corresponding to the molecular ions of the compounds indicated. The illustrative mass spectrum of a mixture of compounds obtained after chromatographic purification of product **3b** are given in Fig. 3 $(m/z: 557 \text{ [M]}^+, \textbf{3b}; 695 \text{ [M']}^+, \textbf{4b}; 833 \text{ [M'']}^+, \textbf{5b}$). The ³¹P—{¹H} NMR spectrum of the reaction mixture obtained when the *meta*-isomer **2a** was used exhibits a broadened singlet signal in the region δ 6.7, which, apparently, corresponds to product **3a** and the phosphorus atom at the multiple bond in compound **4a**. The two doublet signals at δ 5.84 (P''N)

Scheme 2



and 21.44 (P'CH) with the spin-spin coupling constant of 22 Hz correspond to the bis-phosphorylated fragment in compounds **4a** and **5a**. Similar pattern is observed in the spectrum of the reaction mixture in the case when trieth-ylphosphite reacts with *para*-isomer **2b**.

At the same time, the Arbuzov reaction of diimidoyl dichlorides **2a,b** with ethyl diphenylphosphinite on reflux of a stoichiometric mixture of reagents in toluene leads to the corresponding phosphine oxides, though in not very high yields (Scheme 3). Taking into account that the ${}^{31}P{-}{^{1}H}$ NMR spectra of the reaction mixtures exhibit



Fig. 3. The illustrative mass spectrum of a mixture of compounds 3b, 4b, and 5b.

three singlet signal with different intensities (δ_P 24.18, 24.45, 24.50 with the ratio of intensities 75: 15: 10 for 6a and $\delta_{\rm P}$ 24.11, 24.31, 24.40 with the ratio of intensities 70: 20: 10 for **6b**), one can assume that in the course of E,E) of phosphine oxides **6** are formed with one of them, giving the upfield signal, being predominant (>70%). After purification by recrystallization (compounds partially decompose when purified by chromatography on silica gel), N,N'-bis[(diphenylphosphoryl)phenylmethylidene]benzenediamines **6a**,**b**, being yellow powders, were isolated as the individual stereoisomers, whose signals predominated in the reaction mixtures. Unfortunately, since we failed in growing crystals for such phosphine oxides suitable for X-ray diffraction analysis, the question about their exact configuration remains open.

Scheme 3



Upon study of complexation of m-bisphosphine dioxide **6a** with neodymium and praseodymium nitrates in acetone, it was found that, irrespective of the reagent ratios, the reaction results in the formation of stable complexes of the form $ML(NO_3)_3$. The coordination shift of the signal for the phosphorus atom in the ${}^{31}P - {}^{1}H$ NMR spectra (for the complexes with Pr and Nd in solutions, $\Delta \delta = 105$ and 117 ppm, respectively) and the shift of the absorption bands for the C=N and P=O bonds in the IR spectra of the complexes formed (Nujol) unambiguously testify the participation of the ligand phosphorus and nitrogen atoms in the coordination. In addition, the presence of the absorption bands in the region ~1480 cm⁻¹ (v(N=O)) and ~1302 cm⁻¹ ($v_{as}(NO_2)$) allowed us to suppose a bidentate coordination of the nitrate groups. The presence of a small shoulder in the IR spectrum in the region 1390 cm^{-1} does not exclude that one of the nitrate groups either is coordinated by only one oxygen atom or is in the external coordination sphere. It should be also noted that the presence of a sharp band at 1637 and 3368 cm⁻¹ allowed us to suggest the presence of water molecules in the coordination sphere of the metal. The entire identity of the IR spectra of the praseodymium and neodymium complexes indicates the similarity of their structures. It could be assumed that in the complexes ML, either the metal cation is included into the cavity formed by the dentate fragments and coordinated with the nitrogen atom of the fragment C=N and the oxygen atom of the phosphoryl group (A) or, which seems more probable to us, a polymer chain is formed with the similar type of the metal coordination (B).



 $M = Nd^{3+}$, Pr^{3+} ; nitrate groups are not shown.

Experimental

NMR spectra were recorded on a Bruker AMX-400 spectrometer in CDCl₃ or DMSO-d₆ using the signal for the residual protons of a deuterated solvent as a reference (¹H and ¹³C relatively to Me₄Si) and 85% aq. H₃PO₄ as an external standard (³¹P). IR spectra were recorded on a Magna-IR 750 Fourierspectrometer (Nicolet), resolution, 2 cm⁻¹, number of scans 128 (KBr pellets or a suspension in Nujol). Mass spectra were recorded on a Finnigan LCQ Advantage tandem mass spectrometer in the mode for detection of positively charged ions (the sensitivity limit, 2000 m/z).

Isomeric N, N'-(phenylene)dibenzamides **1a,b** were synthesized by the reaction of corresponding isomeric diamines with benzoyl chloride in the presence of pyridine according to the procedure described earlier,¹⁷ their physico-chemical constants are in agreement with the literature data.

N,N'-(*m*-Phenylene)dibenzimidoyl dichloride (2a). A mixture of *N,N'*-(*m*-phenylene)dibenzamide 1a (5 g, 16 mmol) and phosphorus pentachloride (6.6 g, 32 mmol) in anhydrous toluene (100 mL) was refluxed for 10 h. The solvent was evaporated *in vacuo*, the residue was washed with anhydrous light petroleum and dried *in vacuo* (70 °C, 1 Torr, 4 h). The yield was 5.3 g (95%), m.p. 89 °C (*cf.* Ref. 4: m.p. 86 °C). Found (%): C, 68.00; H, 3.94; N, 8.01. C₂₀H₁₄Cl₂N₂. Calculated (%): C, 68.00; H, 3.99; N, 7.93. ¹H NMR (CDCl₃), δ : 6.69 (br.s, 1 H, NC₆H₄N); 6.86, 6.88 (both d, 1 H each, NC₆H₄N, ³J_{H,H} = 2.0 Hz); 7.42 (s, 1 H, NC₆H₄N); 7.44–7.57 (m, 6 H, Ph); 8.17–8.19 (m, 4 H, Ph). IR (KBr), v/cm⁻¹: 1659 (C=N).

N,N'-(*p*-Phenylene)dibenzimidoyl dichloride (2b) was obtained similarly. The yield was 97%, m.p. 168 °C (*cf.* Ref.: m.p. 213 °C (see Ref. 1); 176 °C (see Ref. 4)). Found (%): C, 68.07; H, 3.94; N, 7.91. $C_{20}H_{14}Cl_2N_2$. Calculated (%): C, 68.00; H, 3.99; N, 7.93. ¹H NMR (CDCl₃), δ : 7.13 (s, 4 H, NC₆H₄N); 7.47–7.56 (m, 6 H, Ph); 8.18–8.21 (m, 4 H, Ph). IR (KBr), v/cm⁻¹: 1649 (C=N).

N, N'-Bis[(diphenylphosphoryl)phenylmethylidene]-1,3-benzenediamine (6a). A mixture of *m*-diimidoyl dichloride 2a (2.2 g, 0.006 mol) and ethyl diphenylphosphinite (2.9 g, 0.012 mol) in anhydrous toluene (30 mL) was refluxed for 3 h. A precipitate formed was filtered off and recrystallized from EtOH to obtain the target product (0.6 g, 14%) as a light yellow powder, m.p. 247 °C. A sample for elemental analysis was prepared by recrystallization from CH₂Cl₂. Found (%): C, 73.50; H, 4.85; N, 3.85; P, 8.52. C₄₄H₃₄N₂O₂P₂•0.5CH₂Cl₂. Calculated (%): C, 73.47; H, 4.52; N, 3.95; P, 8.81. ³¹P (CDCl₃): δ 23.08. ¹H NMR $(CDCl_3)$, δ : 6.10 (s, 1 H, NC₆H₄N); 6.23, 6.26 (both br.s, 1 H each, NC₆H₄N); 6.89 (t, 1 H, NC₆H₄N, ${}^{3}J_{H,H} = 7.8$ Hz); 7.05-7.07, 7.14-7.22, 7.44-7.55, 7.84-7.89 (all m, 4 H + 6 H + + 12 H + 8 H, 6 Ph). ¹³C NMR (CDCl₂), δ : 112.44 (s, C(2)); 116.49 (s, C(4), C(6)); 128.00 (s, p-PhP); 128.30 (d, m-C, PhP, ${}^{3}J_{PC} = 12.0 \text{ Hz}$; 128.84 (s, C(10), C(12), C(17), C(19)); 128.96 (s, C(5)); 129.43 (s, C(11), C(18)); 131.14 (d, ipso-C, PhP, ${}^{1}J_{P,C} = 102.0 \text{ Hz}$; 131.95 (d, C(9), C(13), C(16), C(20), ${}^{3}J_{P,C} =$ = 6.9 Hz); 132.01 (d, o-C, PhP, ${}^{2}J_{P,C}$ = 9.0 Hz); 133.98 (d, C(8), C(15), ${}^{2}J_{P,C} = 24.4 \text{ Hz}$; 149.76 (d, C(1), C(3), ${}^{3}J_{P,C} = 25.6 \text{ Hz}$); 172.72 (d, C(7), C(14), ${}^{1}J_{P,C} = 116.0 \text{ Hz}$). IR (KBr), v/cm⁻¹: 513, 566, 584, 695, 724, 1121, 1182 (P=O), 1437, 1588 (C=N), 3055, 3034 (br). MS (EI, 70 eV), m/z (I_{rel} (%)): 684 [M]⁺ (5), $484 [M - P(O)Ph_2]^+ (55), 483 [M - H - P(O)Ph_2]^+ (100), 283$ $[M - H - 2 P(O)Ph_2]^+$ (40), 201 $[P(O)Ph_2]^+$ (40), 179 [M - $-P(O)Ph_2 - NCPh - 2H^+(51), 77[Ph]^+(40), 76[C_6H_4]^+(58).$

N,N′-Bis[(diphenylphosphoryl)phenylmethylidene]-1,4-benzenediamine (6b) was obtained similarly. The yield was 26%, m.p. 268 °C. Found (%): C, 73.50; H, 4.85; N, 3.85; P, 8.52. C₄₄H₃₄N₂O₂P₂·0.5CH₂Cl₂. Calculated (%): C, 73.50; H, 4.57; N, 3.96; P, 8.28. ³¹P (CDCl₃): δ 23.15. ¹H NMR (CDCl₃), δ : 6.56 (s, 4 H, NC₆H₄N); 7.20–7.22, 7.29–7.32, 7.51–7.59, 7.92–7.96 (all m, 6 H + 5 H + 13 H + 6 H, 6 Ph). IR (KBr), v/cm⁻¹: 520, 543, 578, 696, 723, 774, 834, 1119, 1187 (P=O), 1438, 1489, 1591 (C=N), 1602, 1667, 3056, 3034 (br). MS (EI, 70 eV), *m/z* (*I*_{rel} (%)): 684 [M]⁺ (3), 485 [M + H – P(O)Ph₂]⁺ (40), 483 [M – H – P(O)Ph₂]⁺ (100), 283 [M – H – 2 P(O)Ph₂]⁺ (50), 201 [P(O)Ph₂]⁺ (40), 179 [M – P(O)Ph₂ – NCPh – 2 H]⁺ (60), 77 [Ph]⁺ (40), 76 [C₆H₄]⁺ (36). A sample for elemental analysis was prepared by recrystallization from CH₂Cl₂.

 ${N,N'-Bis[(diphenylphosphoryl)phenylmethylidene]-1,3$ $benzenediamine}praseodymium trinitrate (7a).$ *m*-Bisphosphine
 Table 1. Main crystallographic data and refining parameters for the structures 2a and 2b

Parameter	2a	2b
Molecular formula	$C_{20}H_{14}Cl_2N_2$	$C_{20}H_{14}Cl_2N_2$
Molecular weight	353.23	353.23
T/K	120	120
Crystal system	Monoclinic	Monoclinic
Space group	$P2_1/n$	$P2_1/c$
Z(Z')	4(1)	2 (0.5)
a/Å	8.941(2)	18.0005(13)
b/Å	8.5598(18)	5.9999(4)
c/Å	22.345(5)	7.5480(5)
β/deg	100.152(4)	93.505(2)
$V/Å^3$	1683.3(6)	813.67(10)
$d_{\rm calc}/{\rm g~cm^{-3}}$	1.394	1.442
μ/cm^{-1}	3.88	4.02
F(000)	728	364
$2\theta_{\rm max}/{\rm deg}$	60	58
Number of measured reflections	15199	5698
Number of independent reflections $[R_{int}]$	4881 [0.0248]	2147 [0.0274]
Number of reflections with $I > 2 \sigma(I)$	3852	1662
Number of refining parameters	273	137
R_1	0.0431	0.0427
wR_2	0.0968	0.0972
GOF	1.059	1.020
Residual electron	0.419/-0.296	0.537/-0.272
density/e Å ⁻³ (ρ_{min}/ρ_{max})		•

dioxide **6a** (50 mg, 0.07 mmol) was dissolved upon heating in acetone (5 mL) followed by addition of $Pr(NO_3)_3 \cdot 6H_2O$ (32 mg, 0.07 mmol). After 24 h, the solvent was half evaporated, a precipitate formed was filtered off. The yield was 38 mg (52%), m.p. 218 °C (decomp.). Found (%): C, 51.12; H, 4.17; N, 6.58. C₄₄H₃₄N₅O₁₁P₂Pr · (CH₃)₂CO · 2H₂O. Calculated (%): C, 51.05; H, 4.01; N, 6.33. ³¹P (DMSO-d₆): δ 113.31. IR (Nujol), v/cm⁻¹: 541, 709, 725, 1089, 1133 (P=O), 1170, 1306 (br), 1377, 1439, 1463, 1485, 1531, 1603 (C=N), 1634, 1705, 3062, 3368 (br).

{N,N'-Bis[(diphenylphosphoryl)phenylmethylidene]-1,3benzenediamine}neodymium trinitrate (7b) was obtained under similar conditions from compound 6a (40 mg, 0.06 mmol) and Nd(NO₃)₃·6H₂O (36 mg, 0.06 mmol). The yield was 40 mg (70%), m.p. 238 °C (decomp.). Found (%): C, 52.32; H, 4.44; N, 6.37. C₄₄H₃₄N₅NdO₁₁P₂·2(CH₃)₂CO·H₂O. Calculated (%): C, 52.26; H, 4.21; N, 6.09. ³¹P (DMSO-d₆): δ 137.83. IR (Nujol), v/cm⁻¹: 541, 708, 726, 1087, 1132 (P=O), 1169, 1302 (br), 1378, 1439, 1463, 1484, 1533, 1602 (C=N), 1621, 1708, 3061, 3351 (br).

X-ray diffraction study of compounds 2a,b was performed on a SMART 1000 CCD diffractometer (Mo-K α -irradiation, a graphite monochromator, ω -scanning). Crystals for the study were obtained from Et₂O—EtOH. Absorption was accounted *ab initio* on equivalent reflections using the Sadabs program. The structures were decoded by the direct method and refined by the least squares method in anisotropic full-matrix approximation on F_{hkl}^2 . Hydrogen atoms were localized from the differential Fourier-syntheses of electron density and refined in isotropic approximation. The main crystallographic data and refining parameters are given in Table 1. All the calculations were performed using SHELXTL PLUS program package.

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References

- 1. G. Bock, Chem. Ber., 1967, 100, 2870.
- K.-M. Wu, C.-A. Huang, K.-F. Peng, C.-T. Chen, *Tetrahe*dron, 2005, 61, 9679.
- 3. J. K. Hutchison, G. Srdanov, R. Hicks, H. Yu, F. Wudl, J. Am. Chem. Soc., 1998, **120**, 2989.
- 4. H. K. S. Rao, T. S. Wheeler, J. Chem. Soc., 1937, 1643.
- 5. H. Quast, R. Frank, A. Heublein, E. Schmitt, *Liebigs Ann. Chem.*, 1979, 83.
- M. Kosugi, M. Koshiba, A. Aton, H. Sano, T. Migita, Bull. Chem. Soc. Jpn, 1986, 59, 677.
- V. F. Shchegel'skii, V. V. Sokolov, G. A. Shataeva, V. I. Fetisov, I. V. Martynov, *Khim. Pharm. Zh.*, 1996, **11**, 26 [*Pharm. Chem. J. (Engl. Transl.)*, 1996, **11**]; H. Weisheng, Z. Yixin, Y. Chengye, *Phosphorus, Sulfur, Silicon*, 1995, **107**, 21.
- A. D. Sinitsa, V. S. Krishtal', V. I. Kal'chenko, L. N. Markovskii, *Zh. Obshch. Khim.*, 1980, **50**, 465 [*J. Gen. Chem. USSR (Engl. Transl.*), 1980, **50**, No. 2]; P. P. Onys'ko, E. I. Suvalova, T. I. Chudakova, A. D. Sinitsa, *Zh. Obshch. Khim.*,

1987, **57**, 1514 [*J. Gen. Chem. USSR (Engl. Transl.*), 1987, **57**, No. 7]; P. P. Onys'ko, T. V. Kolodka, A. A. Kudryavtsev, A. D. Sinitsa, *Zh. Obshch. Khim.*, 1993, **63**, 1562 [*J. Gen. Chem. USSR (Engl. Transl.*), 1993, **63**, No. 7].

- A. D. Garnovskii, I. S. Vasil'chenko, Usp. Khim., 2002, 71, 1064 [Russ. Chem. Rev. (Engl. Transl.), 2002, 71, 943]; A. D. Garnovskii, I. S. Vasil'chenko, Usp. Khim., 2005, 74, 211 [Russ. Chem. Rev. (Engl. Transl.), 2005, 74, 193].
- J. W. Schulenberg, S. Archer, J. Am. Chem. Soc., 1960, 82, 2035; J. E. Rowe, Synthesis, 1980, 114.
- T. V. Artmonova, A. B. Zhivich, M. Yu. Dubinskii, G. Koldobskii, *Synthesis*, 1996, **12**, 1428.
- 12. H. K. S. Rao, T. S. Wheeler, J. Chem. Soc., 1938, 476.
- G. I. Koldobskii, I. V. Nikonova, A. B. Zhivich, V. A. Ostrovskii, V. S. Poplavskii, *Zh. Obshch. Khim.*, 1992, **62**, 194
 [*J. Gen. Chem. USSR (Engl. Transl.)*, 1992, **62**, No. 2].
- 14. J. von Braun, W. Pinkernelle, *Chem. Ber.*, 1934, 67, 1218;
 A. R. Katritzky, A. T. Tods, *Heterocycles*, 1982, 18, 21.
- 15. H. von Eilingsfeld, M. Seefelder, H. Weidinger, *Angew. Chem.*, 1960, **22**, 70.
- R. Appel, K. Warning, K.-D. Ziehn, Chem. Ber., 1973, 106, 3450.
- V. V. Korshak, T. M. Frunze, L. V. Kozlov, *Izv. Akad. Nauk* SSSR, Ser. Khim., 1962, 2062 [Bull. Acad. Sci. USSR, Div. Chem. Sci. (Engl. Transl.), 1962, 11].

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