A new method for the synthesis of *N*-(2-aminoethyl)azoles by alkylation of azoles with 2-alkyl-4,5-dihydrooxazoles

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Ring opening in 2-alkyl-4,5-dihydrooxazoles by the action of azoles gave intermediate *N*-(2-azolylethyl)alkanamides, whose hydrolysis afforded *N*-(2-aminoethyl)azoles.

Key words: azoles, oxazolines, amines, aminoalkylation.

N-(2-Aminoethyl)azoles are valuable starting materials for the preparation of antiaggregation,[1] hypotensive,[2] anti-HIV,[3] and antibacterial drugs.[4,5] They are mainly synthesized by the Gabriel method, which is known since a relatively long time. For instance, 2-(imidazol-1-yl)/(1,2,4-triazol-1-yl)ethylamines have been obtained[6] from azoles and 2-bromoethylphthalimide in the presence of NaH in DMF with subsequent elimination of the phthalimide protection with hydrazine hydrate (the total yield was 20—40%). Alkylation of azoles with 2-chloroethylamine under the conditions of phase-transfer catalysis leads to the desired amines in 60—70% yields;[7,8] the reaction produces highly toxic aziridine as a by-product. Substituted 1-(2-aminoethyl)benzimidazoles have been obtained in a two-step scheme. First, reactions of acrylamide or methacrylamide with substituted benzimidazoles give 3-(benzimidazol-1-yl)propanamides, which upon the Hoffman rearrangement produce 2-(benzimidazol-1-yl)ethylamines in 30—60% yields.[9] The same product has been obtained from *o*-fluoronitrobenzene in five steps.[10] Each of the above methods has substantial drawbacks.

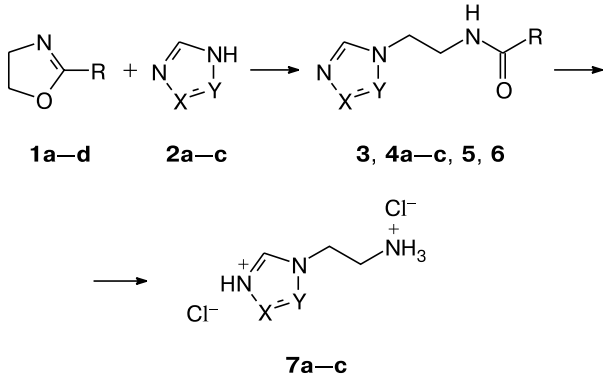
Here we proposed a new efficient general route to *N*-(2-aminoethyl)azoles that affords their high (85—95%) yields from inexpensive and accessible starting reagents. When developing this method, we verified three approaches.

According to the first approach, we tried to synthesize *N*-(2-aminoethyl)azoles through intermediate 2-azolylacetonitriles obtained in 70—80% yields by alkylation of 1,2,4-triazole and imidazole with chloroacetonitrile in acetonitrile in the presence of K_2CO_3.[11] The resulting nitriles were hydrogenated in an autoclave (70—90 °C, 100 atm, Raney nickel) in the presence of excess ammonia, by analogy with a known synthesis of azolylpropylamines.[6] However, final column chromatography gave only secondary *N*,*N*-bis(2-azolylethyl)amines.

The second approach involved the Gabriel reactions of 2-bromoethylphthalimide with azoles. Alkylation of sodium imidazolate in boiling methanol in the presence of catalytic amounts of KI gave 2-(imidazol-1-yl)ethylphthalimide in a higher yield (50%) than that provided by a related procedure.[6] In a reaction of 2-bromoethylphthalimide with melted imidazole used in a fourfold excess, the yield was increased to 75%. Under the same conditions, the yield of 2-(1,2,4-triazol-1-yl)ethylphthalimide was 62%. The phthalimide protection was eliminated with hydrazine hydrate (as proposed by Ing and Manske) to give 2-(imidazol-1-yl)ethylamine hydrochloride in 82% yield or its triazole analog in 52% yield. A reaction of benzimidazole with *N*-(2-bromoethyl)phthalimide in triethylamine gave rise to a difficult-to-separate mixture of alkylation products.

The third approach involved acid-catalyzed ring opening in 2-alkyl-4,5-dihydrooxazoles **1a—c** in the presence

Scheme 1



1a—d **2a—c** **3**, **4a—c**, **5**, **6** **7a—c**

R = Me (**1a**, **3**), Et (**1b**, **4a—c**), Pr (**1c**, **5**), Ph (**1d**, **6**);

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of azoles **2a—c**. Hydrolysis of intermediate *N*-(2-azolylethyl)alkanamides **3—6** gave the target *N*-(2-aminoethyl)azoles **7a—c** (Scheme 1). This method is based on the known[12] amine-assisted ring opening in 2-alkyl-4,5-dihydrooxazoles, which provides a convenient access to *N*,*N*-dialkylethylenediamines.

With $ZnCl_2$ as a catalyst, the yields of amides **3—6** varied from 40 to 87%, the highest yield being reached with commercial 2-ethyl-4,5-dihydrooxazole (**1b**).

Compound	**3**	**4a**	**4b**	**4c**	**5**	**6**
Yield (%)	65	80	85	87	50	40

Data on the effects of the catalyst nature and the conditions of the synthesis on the outcome of the reactions of oxazoline **1b** with azoles **2a—c** are given in Table 1. The yields of *N*-[2-(imidazol-1-yl)ethyl]propanamides were lower than the yields of analogous triazole and benzimidazole products.

Alkylation of azoles **2a—c** with oxazoline **1b** in a steel pressure vessel allowed us to elevate the reaction temperature and raise the conversion of the starting reagents. The yields of amides **4a—c** were increased to 75—86% versus 20—60% under atmospheric pressure. The highest yield (85%) of amide **4a** was reached with $BF_3 \cdot Et_2O$ as a catalyst. The use of $ZnCl_2$ somewhat lowered the yields; however, the resulting compounds were of higher purity. An increase in the catalyst amount from 1 to 10 mol.% decreased the yields of amides **4a—c** by half.

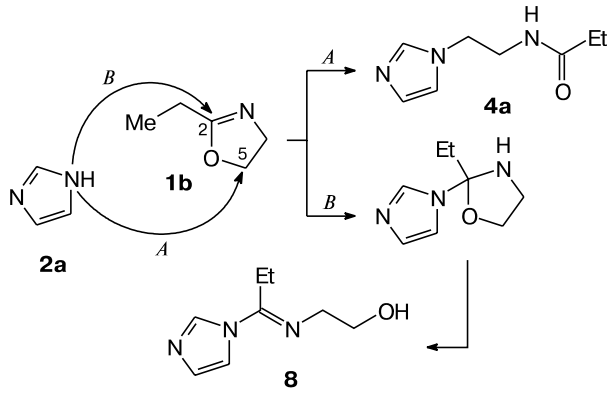
With a 50% excess of low-boiling oxazoline **1b**, we obtained a mixture of the target compound **4a** and by-product **8** in the ratio 2.3 : 1. According to the MS data, the molecular mass of compound **8** (*m/z* 168) equals that of amide **4a**. This suggests the formation of compound **8** *via* a nucleophilic attack of imidazole on position 2 of 2-ethyl-4,5-dihydrooxazole (Scheme 2, pathway *B*). It should be noted that reactions of oxazolines with amines also yield by-products due to the addition at position 2.[12]

The structure of compound **8** was proven by ^1H NMR spectroscopy for its mixture with compound **4a** (because of close boiling points and retention times, these products

Table 1. Conditions of the synthesis and the yields of *N*-(2-azolylethyl)propanamides **4a—c**

Compound	Catalyst	Yield (%)	
		1 atm, 138 °C	5 atm, 180 °C
4a	1 mol.% $Zn(OAc)_2 \cdot 2H_2O$	20	75
4a	2 mol.% $Zn(OAc)_2 \cdot 2H_2O$	8	50
4a	1 mol.% $ZnCl_2$	40	80
4a	10 mol.% $ZnCl_2$	—	40
4a	1 mol.% $BF_3 \cdot Et_2O$	40	85
4a	1 mol.% TsOH	40	75
4b	1 mol.% $ZnCl_2$	64	86
4c	1 mol.% $ZnCl_2$	—	87

Scheme 2



can be separated by neither distillation nor column chromatography).

Hydrolysis of amides **4a—c** gave amine hydrochlorides **7a—c** in 60—80% yields (see Scheme 1). The melting points of these hydrochlorides are not always the same as reported in the literature, because we did not obtain complete amine dihydrochlorides **7a—c**: the lower content of hydrogen chloride in the compounds we synthesized was determined by argentometric potentiometric titration.

The obvious advantages of the acid hydrolysis of amides **3—6** over the base one include the substantially higher hydrolysis rate, the formation of the product as a stable hydrochloride, and its higher yield. The total yields of the final amines **7a—c** were increased by 10—20% when intermediate amides **4a—c** were used in the second step without purification (Table 2).

Consideration for the alternative routes to amines **7a—c** as regards their isolation showed that alkylation of azoles with oxazoline **1b** is the most rational approach ensuring the highest total yield (87—95%). This method involves inexpensive and accessible reagents and shorter reaction times, appreciably lower waste outcome, and the higher atom efficiency.[13] This approach can be regarded as ecologically attractive for the synthesis of biologically active compounds.

Table 2. Conditions of the synthesis and the yields of *N*-(2-aminoethyl)azole hydrochlorides **7a—c**

Compound	*n**	Yield (%)		M.p. /°C
		with isolation of **4**	without isolation of **4**	
7a	1.83	80	—	214—216
	1.80	—	87	200—204
7b	1.86	85	95	170—175
7c	1.81	80	90	215—217

* The number of equivalents of HCl per molecule of compound **7**.

Experimental

IR spectra were recorded on a Specord M-80 instrument in Nujol (for solids) and a thin film (for liquids). ^{1}H NMR spectra were recorded on a Bruker AC-200 spectrometer (200 MHz) in $CDCl_3$ and $(CD_3)_2SO$ with Me_4Si as the internal standard. Mass spectra were recorded on a Surveoyr MSQ Thermo Finnigan instrument (ESI, positive ion detection, needle tip voltage 3 kV); 0.1% formic acid and acetonitrile were used as solvents. The chlorine content of the hydrochlorides obtained was determined on an Ekotest-2000 instrument (pH-meter, ionometer) with an ion-selective Cl electrode. The course of the reactions was monitored and the purity of the products was checked by TLC on Silufol UV-254 plates; spots were visualized under UV light and by treatment with Dragendorff modified reagent or Ehrlich reagent.[14]

N*-[2-(Imidazol-1-yl)ethyl]propanamide (4a).** ***A. A mixture of azole **2a** (3.4 g, 0.05 mol), dihydrooxazole **1b** (5.04 mL, 4.95 g, 0.05 mol), and $Zn(OAc)_2 \cdot 2H_2O$ (0.1 g, 0.5 mmol) was heated under nitrogen at 130 °C for 26 h. The reaction mixture was distilled *in vacuo* (oil pump) to collect a fraction with b.p. 151—154 °C (0.042 Torr). The yield of propanamide **4a** was 1.6 g (20%), n_D^{20} 1.5045. Found (%): C, 57.36; H, 7.82; N, 25.03. $C_8H_{13}N_3O$. Calculated (%): C, 57.46; H, 7.84; N, 25.13. IR, ν/cm^{-1}: 3280 (NH), 1670 (amide I), 1620 (amide II). ^{1}H NMR ($(CD_3)_2SO$), δ: 1.13 (t, 3 H, Me, J = 7.7 Hz); 2.21 (q, 2 H, C<u>H</u>$_2$Me, J = 5.7 Hz); 3.55 (q, 2 H, C<u>H</u>$_2$NH, J = 4.4 Hz); 4.11 (t, 2 H, CH_2N, J = 5.5 Hz); 6.72 (br.s, 1 H, NH); 6.89, 7.00, 7.38 (all s, 1 H each, CH of imidazole).

1-[1-(2-Hydroxyethylimino)propyl]imidazole (8) was obtained as a by-product (oil, b.p. 155—156 °C) for **2a** : **1b** = 1 : 1.5. ^{1}H NMR ($(CD_3)_2SO$), δ: 1.15 (t, 3 H, Me, J = 7.8 Hz); 2.23 (q, 2 H, C<u>H</u>$_2$Me, J = 7.5 Hz); 3.69 (t, 2 H, CH_2N, J = 5.3 Hz); 4.10 (t, 2 H, C<u>H</u>$_2$OH, J = 5.5 Hz); 6.76 (br.s, 1 H, OH); 6.91, 7.01, 7.42 (all s, 1 H each, CH of imidazole). MS (CI), m/z (I_{rel} (%)): 169 $[M + H]^+$ (42) (*cf.* **4a**: m/z 169 $[M + H]^+$ (100)).

B. A mixture of azole **2a** (3.4 g, 0.05 mol), dihydrooxazole **1b** (5.04 mL, 4.95 g, 0.05 mol), and $ZnCl_2$ (0.068 g, 0.5 mmol) was heated in a steel vessel at 180 °C for 26 h. The reaction mixture was distilled *in vacuo* (oil pump) to collect a fraction with b.p. 152—156 °C (0.035 Torr). The yield of amide **4a** was 6.7 g (80%).

Compounds **3**, **4b,c**, **5**, and **6** were obtained analogously.

***N*-[2-(Imidazol-1-yl)ethyl]ethanamide (3)**, an oil, b.p. 154—159 °C (0.07 Torr), n_D^{20} 1.5111. Found (%): C, 54.84; H, 7.16; N, 25.37. $C_7H_{11}N_3O$. Calculated (%): C, 54.89; H, 7.24; N, 27.43. IR, ν/cm^{-1}: 3305 (NH), 1640 (amide I), 1556 (amide II). ^{1}H NMR ($(CD_3)_2SO$), δ: 1.99 (s, 3 H, MeCO); 3.55 (q, 2 H, C<u>H</u>$_2$NH, J = 6.0 Hz); 4.10 (t, 2 H, CH_2N, J = 6.0 Hz); 6.73 (br.s, 1 H, NH); 6.90, 7.00, 7.35 (all s, 1 H each, CH of imidazole).

***N*-[2-(1,2,4-Triazol-1-yl)ethyl]propanamide (4b)**, an oil, b.p. 140—180 °C (0.03 Torr), n_D^{20} 1.5006. Found (%): C, 49.87; H, 7.23; N, 33.24. $C_7H_{12}N_4O$. Calculated (%): C, 49.99; H, 7.19; N, 33.31. IR, ν/cm^{-1}: 3300 (NH), 1640 (amide I), 1554 (amide II). ^{1}H NMR ($(CD_3)_2SO$), δ: 1.09 (t, 3 H, Me, J = 5.1 Hz); 2.15 (q, 2 H, C<u>H</u>$_2$Me, J = 4.7 Hz); 3.65 (q, 2 H, C<u>H</u>$_2$NH, J = 5.2 Hz); 4.35 (t, 2 H, CH_2N, J = 5.2 Hz); 6.30 (br.s, 1 H, NH); 7.90, 8.00 (both s, 1 H each, CH of triazole).

***N*-[2-(Benzimidazol-1-yl)ethyl]propanamide (4c)**, m.p. 55—58 °C (from EtOH), b.p. 180—185 °C (0.022 Torr). Found (%): C, 66.30; H, 6.87; N, 19.35. $C_{12}H_{15}N_3O$. Calculated (%): C, 66.34; H, 6.96; N, 19.34. IR, ν/cm^{-1}: 3290 (NH), 1635 (amide I), 1551 (amide II). ^{1}H NMR ($(CD_3)_2SO$), δ: 1.03 (t, 3 H, Me, J = 7.7 Hz); 2.13 (q, 2 H, C<u>H</u>$_2$Me, J = 7.6 Hz); 3.54 (t, 2 H, C<u>H</u>$_2$NH, J = 5.7 Hz); 4.24 (t, 2 H, CH_2N, J = 5.9 Hz); 7.18 (m, 4 H, CH of benzimidazole); 7.63 (s, 1 H, CH of benzimidazole); 7.91 (br.s, 1 H, NH).

***N*-[2-(Imidazol-1-yl)ethyl]butanamide (5)**, an oil, b.p. 162—168 °C (0.1 Torr), n_D^{20} 1.4850. Found (%): C, 59.59; H, 8.28; N, 23.15. $C_9H_{15}N_3O$. Calculated (%): C, 59.64; H, 8.34; N, 23.19. IR, ν/cm^{-1}: 3295 (NH), 1630 (amide I), 1555 (amide II). ^{1}H NMR ($(CD_3)_2SO$), δ: 0.93 (t, 3 H, Me, J = 7.3 Hz); 1.57—1.72 (m, 2 H, C<u>H</u>$_2$Me); 2.16 (t, 2 H, CH_2CO, J = 7.2 Hz); 3.55 (q, 2 H, C<u>H</u>$_2$NH, J = 6.0 Hz); 4.11 (t, 2 H, CH_2N, J = 6.0 Hz); 6.51 (br.s, 1 H, NH); 6.91, 7.02, 7.37 (all s, 1 H each, CH of imidazole).

***N*-[2-(Imidazol-1-yl)ethyl]benzamide (6)**, an oil, b.p. 172—175 °C (0.1 Torr), n_D^{20} 1.4926. Found (%): C, 66.89; H, 6.01; N, 19.45. $C_{12}H_{13}N_3O$. Calculated (%): C, 66.96; H, 6.09; N, 19.52. IR, ν/cm^{-1}: 3300 (NH), 1639 (amide I), 1550 (amide II). ^{1}H NMR ($(CD_3)_2SO$), δ: 3.60 (q, 2 H, C<u>H</u>$_2$NH, J = 7.0 Hz); 3.91 (t, 2 H, CH_2N, J = 6.0 Hz); 7.01 (s, 1 H, CH of imidazole); 7.25—7.29 (m, 3 H, CH of imidazole, CH arom.); 7.32—7.34 (m, 2 H, NH, CH arom.); 7.45—7.65 (m, 2 H, CH arom.); 7.82 (s, 1 H, CH of imidazole).

2-(Imidazol-1-yl)ethylamine hydrochloride (7a). ***A.*** Potassium iodide (1.25 g, 7.5 mmol) was added to melted azole **2a** (81.6 g, 1.2 mol). Then *N*-(2-bromoethyl)phthalimide (76.2 g, 0.3 mol) was added in portions. The melted reaction mixture was stirred at 100 °C for 3 h, transferred to a percolator, and refluxed with toluene for 20 h. The solvent was removed in water aspirator vacuum and the residue was recrystallized from Pr^{i}OH. The yield of *N*-[2-(imidazol-1-yl)ethyl]phthalimide was 54.2 g (75%), m.p. 155—157 °C (*cf.* Ref. 15: m.p. 156—157 °C). Compound **7a** was obtained from *N*-[2-(imidazol-1-yl)ethyl]phthalimide according to a known procedure.[15] The yield was 82%, m.p. 205—207 °C (*cf.* Ref. 15: m.p. 216—218 °C for $C_5H_9N_3 \cdot 2HCl$). According to the titration data, the hydrochloride obtained was formulated as $C_5H_9N_3 \cdot 1.8HCl$.

B. Concentrated HCl (100 mL, 1.2 mol) was added to a solution of compound **4a** (25 g, 0.15 mol) in EtOH (200 mL). The reaction mixture was refluxed for 3 h and concentrated in water aspirator vacuum. Ethanol (100 mL) was added to the residue and the resulting crystals were filtered off, washed with EtOH (50 mL), and dried in a desiccator over P_2O_5. The yield of compound **7a** was 21.5 g (80%), m.p. 214—216 °C. According to the titration data, the hydrochloride obtained was formulated as $C_5H_9N_3 \cdot 1.83HCl$. IR, ν/cm^{-1}: 3410 (NH_2). ^{1}H NMR ($(CD_3)_2SO$), δ: 3.31 (t, 2 H, CH_2C<u>H</u>$_2NH_2$, J = 5.7 Hz); 4.63 (t, 2 H, NC<u>H</u>$_2CH_2$, J = 5.9 Hz); 7.71, 7.90 (both d, 1 H each, CH of imidazole, J = 1.2 Hz); 8.30 (br.s, 3 H, NH_3^+); 9.31 (s, 1 H, CH of imidazole).

C. A mixture of azole **2a** (3.4 g, 0.05 mol), dihydrooxazole **1b** (4.95 g, 5.04 mL, 0.05 mol), and $ZnCl_2$ (0.1 g, 0.05 mmol) was heated in a steel vessel at 180 °C for 26 h. The reaction mixture was dissolved in EtOH (100 mL), refluxed with conc. HCl (35 mL, 0.4 mol) for 4 h, and partially concentrated *in vacuo*. Ethanol (50 mL) was added to the residue and the resulting crystals were filtered off, washed with EtOH (10 mL),

and dried in a desiccator over P_2O_5. The yield of compound **7a** was 5.3 g (87%), m.p. 200—204 °C. According to the titration data, the hydrochloride obtained was formulated as $C_5H_9N_3 \cdot 1.8HCl$.

2-(1,2,4-Triazol-1-yl)ethylamine hydrochloride (7b) was obtained as described for compound **7a** (procedure ***A***). The yield of *N*-[2-(triazol-1-yl)ethyl]phthalimide was 52%, m.p. 166—169 °C (*cf.* Ref. 16: m.p. 169—170 °C). The yield of compound **7b** was 65%, m.p. 175—180 °C (*cf.* Ref. 16: m.p. 182—183 °C for $C_4H_8N_4 \cdot 2HCl$). According to the titration data, the hydrochloride obtained was formulated as $C_5H_9N_3 \cdot 1.8HCl$.

Compound **7b** was also obtained according to procedures ***B*** and ***C***, m.p. 170—175 °C (*cf.* Ref. 16: m.p. 182—183 °C for $C_4H_8N_4 \cdot 2HCl$). IR, ν/cm^{-1}: 3448 (NH_2). ^1H NMR ($(CD_3)_2SO$), δ: 3.30 (t, 2 H, $CH_2C\underline{H}_2NH_2$, J = 5.9 Hz); 4.60 (t, 2 H, $NC\underline{H}_2CH_2$, J = 4.6 Hz); 8.50 (s, 1 H, CH of triazole); 8.52 (br.s, 3 H, NH_3^+); 9.24 (s, 1 H, CH of triazole). According to the titration data, the hydrochloride obtained was formulated as $C_4H_8N_4 \cdot 1.86HCl$.

2-(Benzimidazol-1-yl)ethylamine hydrochloride (7c) was obtained according to procedures ***B*** and ***C***, m.p. 215—217 °C (*cf.* Ref. 10: m.p. 271—277 °C for $C_9H_{11}N_3 \cdot 2HCl$). IR, ν/cm^{-1}: 3424 (NH_2). ^1H NMR ($(CD_3)_2SO$), δ: 3.39 (t, 2 H, $CH_2C\underline{H}_2NH_2$, J = 5.8 Hz); 4.60 (t, 2 H, $NC\underline{H}_2CH_2$, J = 4.8 Hz); 7.60 (m, 2 H, CH of benzimidazole); 7.90 (d, 1 H, CH of benzimidazole, J = 7.2 Hz); 8.19 (d, 1 H, CH of benzimidazole, J = 6.9 Hz); 8.65 (br.s, 3 H, NH_3^+); 9.85 (s, 1 H, CH of benzimidazole). According to the titration data, the hydrochloride obtained was formulated as $C_9H_{11}N_3 \cdot 1.81HCl$.

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References

1. DE Pat. 3 406 414; *Chem. Abstrs*, 1985, **102**, 1320369.
2. J. B. Press, W. B. Wright, Jr., P. S. Chan, J. W. Marsico, M. F. Haug, J. Tauber, and A. S. Tomcufcik, *J. Med. Chem.*, 1986, **29**, 817.
3. T. K. Venkatachalam, E. A. Sudbeck, C. Mao, and F. M. Uckun, *Bioorg. Med. Chem. Lett.*, 2001, **11**, 523.
4. DE Pat. 2 248 632; *Chem. Abstrs*, 1973, **97**, 5338g.
5. Z. Crnic and B. Gluncic, *Croat. Chem. Acta*, 1981, **54**, 225.
6. J. B. Press, W. B. Wright, P. S. Chan, J. W. Marsico, M. F. Haug, J. Tauber, and A. S. Tomcufcik, *J. Med. Chem.*, 1986, **29**, 523.
7. A. S. Pagosyan, O. S. Attaryan, and G. A. Eliazyan, *Arm. Khim. Zh.* [*Armenian Chemical Journal*], 1985, **38**, 516 (in Russian).
8. A. M. Guardo, M. P. Matia, and J. L. Garsia, *Synth. Commun.*, 1991, **21**, 535.
9. W. B. Wheatley and G. F. Stiner, *J. Chem. Soc.*, 1957, **8**, 923.
10. G. Kennedy, M. Viziano, J. A. Winders, P. Cavallini, M. Gevi, R. Fabrizio, P. Rodenger, P. Seneci, and A. Zumerle, *Bioorg. Med. Chem. Lett.*, 2000, **10**, 1751.
11. A. N. Terpigorev, M. B. Shcherbinskii, A. G. Bazanov, and I. V. Tselinskii, *Zh. Org. Khim.*, 1982, **18**, 463 [*J. Org. Chem. USSR*, 1982, **18** (Engl. Transl.)].
12. M. Fazio, *J. Org. Chem.*, 1984, **49**, 4889.
13. R. A. Sheldon, *Ros. Khim. Zh.*, 2000, **44**, 9 [*Russ. Chem. J.*, 2000, **44** (Engl. Transl.)].
14. M. Šašúnová, V. Schwarz, and Č. Michalec, *Chromatografia na tenkych vrstách vo farmácii a v klinickej biochémii* [*Thin-Layer Chromatography in Pharmacy and Clinical Biochemistry*], Vydatel´stvo Osteva, 1977 (in Czech).
15. S. I. Lur´e, M. G. Kuleshova, and N. K. Kochetkov, *Zh. Obshch. Khim.* [*Journal of General Chemistry*], 1939, **9**, 1933 (in Russian).
16. C. Ainsworth and R. G. Jones, *J. Am. Chem. Soc.*, 1955, **77**, 1538.

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