1,3-Dipolar cycloaddition of diazocyclopropane to strained cycloalkenes and conjugated dienes to give spiro(1-pyrazoline-3,1′-cyclopropanes)

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Polycyclic spiro(1-pyrazoline-3,1′-cyclopropanes) were obtained in 32–70 % yields by the reaction of diazocyclopropane generated *in situ* with 2-methyltricyclo[3.2.1.$0^{2,4}$]oct-6-ene, spiro[2,4]hepta-4,6-diene dimer, benzvalene, spiro[2,3]hex-1-ene, methyl 1-methylcyclopropene-3-carboxylate, buta-1,3-diene, and 2-methylbuta-1,3-diene.

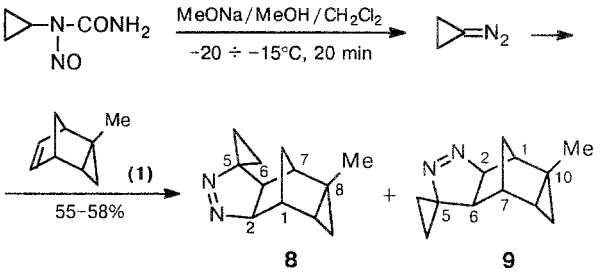
Key words: spiro(1-pyrazoline-3,1′-cyclopropanes), 1,3-dipolar cycloaddition, NMR spectra.

It is well known that generation of diazacyclopropane (DCP) by alkaline decomposition of *N*-nitroso-*N*-cyclopropylurea in the presence of unsaturated acceptors results in products of its 1,3-dipolar cycloaddition[1–3] or formal adducts of cyclopropylidene[4–8] formed by elimination of a nitrogen molecule from DCP. We showed previously[3] that strained unsaturated hydrocarbons, such as norbornene and 3,3-dimethylcyclopropene, are effective traps of DCP, which react with the latter exclusively by 1,3-dipolar cycloaddition to give the corresponding 1-pyrazolines.

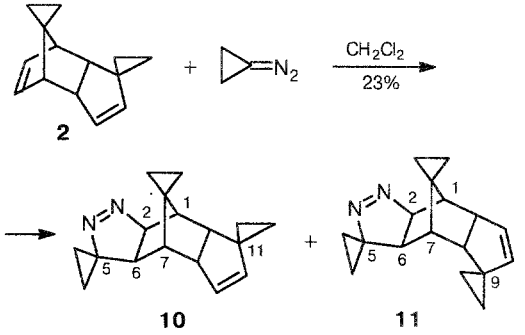
In a continuation of these investigations, we studied reactions of DCP generated *in situ* with other strained unsaturated hydrocarbons, *i.e.*, 2-methyl-*endo*-tricyclo[3.2.1.$0^{2,4}$]oct-6-ene (**1**), spiro[2,4]hepta-4,6-diene dimer (**2**), benzvalene (**3**), spiro[2,3]hex-1-ene (**4**), and methyl 1-methylcyclopropene-3-carboxylate (**5**), as well as conjugated dienes, buta-1,3-diene (**6**) and 2-methylbuta-1,3-diene (**7**). The reactions were carried out according to the usual procedure[3] by adding small portions of *N*-nitroso-*N*-cyclopropylurea to a vigorously stirred mixture of a 1.5-fold molar excess of the corresponding unsaturated compound with MeONa in a MeOH–CH_2Cl_2 (~1:1) mixture at −25 to −15 °C.

As expected, the reaction of DCP with tricyclooctene **1** occurs exclusively as *exo*-addition of DCP to the double bond in compound **1** to give polycyclic spiro(pyrazolinecyclopropanes) **8** and **9**, which differ in orientation of the methyl group relative to the pyrazoline moiety (the isomer ratio is ~1 : 1 according to ^{1}H NMR data).

Similarly, generation of DCP in the presence of asymmetric polycyclic diene **2** affords two isomeric spiro(pyrazolinecyclopropanes) **10** and **11** formed due to different access of DCP to the norbornene bond in diene **2** (according to ^{1}H NMR data, the ratio **10** : **11** is ≈1.4 : 1). Despite the steric hindrance due to spiro-



cyclopropane moiety located at the head of the bridge, pyrazolines **10** and **11** are exclusively formed as *exo*-isomers, although the overall yield of these compounds (23 %) is markedly lower than that in the case of norbornenes unsubstituted at position 7. It should be noted that adducts of DCP (or cyclopropylidene) to the cyclopentene double bond were not found among the reaction products.



Separation of pyrazolines **10** and **11** by preparative TLC (silica gel, hexane–ether, 1 : 1) resulted in two zones with R_f 0.80–0.84 and 0.75–0.80, characterized by different regioisomer ratios. In both cases, ^{1}H and ^{13}C NMR spectra displayed a double set of signals with

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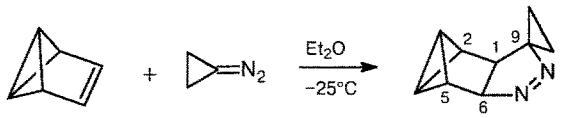
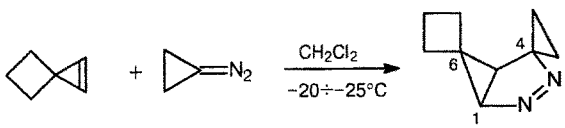
different intensity corresponding to a mixture of isomers **10** and **11** in ~8 : 1 and 1 : 1.8 ratios. The *exo*-configuration of the pyrazoline moiety in compounds **10** and **11** as well as pyrazolines **8** and **9** follows from the multiplicity of the signals of protons at C(2) in the ^{1}H NMR spectra, which in all cases are broadened doublets with $J_{2,6}$ = 7 Hz and $J_{1,2} \leq 1$ Hz. The assignment of the pyrazolines obtained to structures **10** and **11** was performed taking into account differences in the chemical shifts of the H(1) and H(7) protons (δ_{anti} = 1.90 and 1.37, δ_{syn} = 2.30 and 0.99, respectively) differently screened by the spiro-bonded cyclopropane moiety, depending on its position in the cyclopentene ring; the highest upfield shift of the H(7) signal for isomer **11** is caused by the overall anisotropy of the cyclopropane moieties.

Benzvalene (**3**) used as an ethereal solution* was also found to be a good DCP trap. The reaction occurs as 1,3-dipolar cycloaddition to give polycyclic pyrazoline **12** in 65 % yield; C_9H_{10} hydrocarbons, which could result from addition of cyclopropylidene to benzvalene, were not found. Unlike benzvalene (**3**), the resulting compound **12** is quite stable and can be distilled *in vacuo* without decomposition.



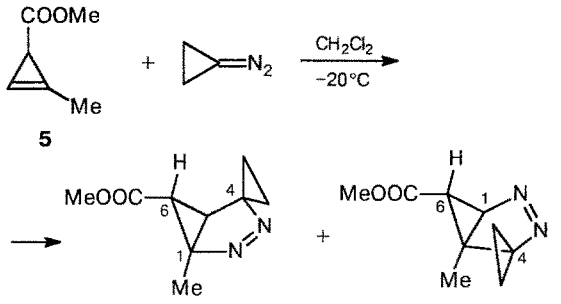
3 **12**, 60%

As we showed previously,[3] DCP readily forms products of 1,3-dipolar cycloaddition to 3,3-dimethylcyclopropene. It was found that other cyclopropenes can also serve as DCP traps. For example, generation of DCP in the presence of spiro[2,3]hex-1-ene (**4**) results in stable dispirane pyrazoline **13**, whose yield reaches 65 % with respect to the distilled product.



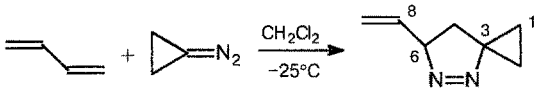
4 **13**, 62–65%

Products of 1,3-dipolar cycloaddition are also formed by the reaction of DCP with cyclopropenecarboxylate **5**; the addition of DCP to the asymmetric double bond results in two isomeric pyrazolines **14** and **15** in ~2 : 1 ratio. According to ^{1}H and ^{13}C NMR spectral data, the signals of linked angular H and C atoms in one of the isomers have chemical shifts δ_H 4.84 and δ_C 72.7, while in the other, δ_H 1.97 and δ_C 30.1, which suggests that this moiety is bonded to N or C atoms in the pyrazoline ring and that isomers **15** and **14** are formed, respectively. The ester group in both isomers has the most favorable *anti*-orientation, which follows from the coupling constant of ^3J = 2.3–4.0 Hz typical of *trans*-protons of the cyclopropane ring. In addition, the ^{1}H NMR spectrum of the reaction mixture does not contain signals with coupling constants J >5 Hz typical of cisoid protons of the cyclopropane ring, which suggests the absence of isomers with *syn*-orientation of the methoxycarbonyl group.



5 **14**, ~30% **15**, ~16%

Apart from strained cyclic unsaturated compounds, conjugated aliphatic dienes can also serve as DCP traps. For example, generation of DCP in the presence of butadiene (**6**) at ~25 °C forms 6-ethenyl-4,5-diazaspiro[2,4]hept-4-ene (**16**), yield ~42 %, as the main addition product, which rather readily undergoes polymerization to give a rubbery bulk. The structure of pyrazoline **16** unambiguously follows from an analysis of spectral data. A calculation of the multiplicity of signals of all protons in pyrazoline **16** by the CALM program[9] showed that they coincide completely with the experimental ^{1}H NMR spectrum and made it possible to estimate the coupling constants (see Experimental), which was found to be quite similar to those of 3-ethenylpyrazoline synthesized previously by addition of diazomethane to butadiene.[10]



6 **16**, 42%

The reaction of DCP with 2-methylbutadiene (**7**) also results in 1,3-dipolar addition products, but the main product is 6-methylethylidene-4,5-diazaspiro[2,4]-hept-4-ene (**18**) isolated in 32 % yield by vacuum distillation of the reaction mixture, rather than the corresponding 5-ethenylpyrazoline **17**. The structure of pyrazoline **18** unambiguously follows from the ^{1}H and ^{13}C NMR spectra, primarily from an analysis of signals in the olefin-related region of the spectrum. The addition of DCP to compound **7** probably occurs at the

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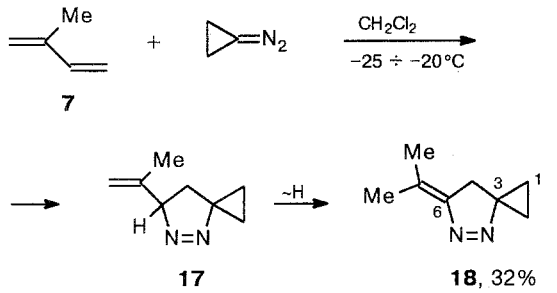
unsaturated double bond, and the resulting pyrazoline **17** isomerizes into pyrazoline **18** under the reaction conditions. It is interesting that compound **16** does not undergo such isomerization under similar conditions, and, moreover, 1-pyrazolines **16** and **17** do not undergo the apparently more favorable isomerization into 2-pyrazolines.



Thus, DCP generated from *N*-nitroso-*N*-cyclopropylurea is readily trapped by strained unsaturated compounds or conjugated dienes to give, in all cases, 1-pyrazolines incorporating a spiro-bonded cyclopropane moiety. All of the pyrazolines obtained are thermally stable and can be isolated by routine distillation *in vacuo*.

Experimental

^{1}H and ^{13}C NMR spectra were recorded on Bruker WM-250 (250 and 62.9 MHz) and Bruker AC-200 (200 and 50.3 MHz) spectrometers from 7–15 % solutions in $CDCl_3$ containing 0.1 % $SiMe_4$ as the internal standard. GLC-MS spectra were obtained on a Finnigan MAT INCOS-50 instrument (70 eV, RSL-200 capillary column, 30 m length). Spiro[2,4]hepta-4,6-diene dimer (**2**),[11] spiro[2,3]hex-1-ene (**4**),[12] and methyl 1-methylcyclopropene-3-carboxylate (**5**)[13] were obtained by procedures reported in the literature. 2-Methyl-*endo*-tricyclo[3.2.1.0^{2,4}]oct-6-ene (**1**) was synthesized by Diels–Alder reaction of 1-methylcyclopropene with cyclopentadiene, yield 90 %, b.p. 139–140 °C. ^{13}C NMR, δ: 132.8 and 130.7 (HC=CH), 61.6 (C(8)), 47.8 and 44.0 (C(1) and C(5)), 25.9 (C(2)), 24.1 (C(3)), 20.7 (CH_3), 19.5 (C(4)). The procedure used freshly distilled unsaturated compounds of at least 97 % purity according to GLC and ^{1}H NMR data. *N*-Nitroso-*N*-cyclopropylurea was obtained by the method reported previously.[3,14]

Synthesis of spiro(1-pyrazoline-3,1′-cyclopropanes). General procedure. *N*-Nitroso-*N*-cyclopropylurea (2.6 g, 20 mmol) was added at −25 to −15 °C in small portions over a period of 20 min to a vigorously stirred mixture of sodium methoxide (1.8 g, 33 mmol), methanol (5 mL), CH_2Cl_2 (5 mL), and 30 mmol of the corresponding unsaturated compound (**1**–**7**). The mixture was stirred for 20 min at the same temperature and then slowly heated to 20 °C. CH_2Cl_2 (15 mL) was then added to the reaction mixture, and the organic layer was separated and dried with anhydrous $MgSO_4$. The solvent was removed, and the residue was fractionated *in vacuo*.

8-Methyl- and 10-methylspiro{3,4-diazatetracyclo-[5.3.1.0^{2,6}.0^{8,10}]undec-3-ene-5,1′-cyclopropanes} (8 and 9) were obtained as a mixture of isomers in ~1 : 1 ratio, yield 55–58 %, b.p. 112–115 °C (0.4 Torr). ^{1}H NMR, δ: 4.61 and 4.57 (two br.d, $J_{2,6}$ = 7.2 Hz, H(2) in **8** and **9**), 2.89 (dq, $J_{1,10}$ = 4.9 Hz, J = 1.4 Hz, H(1) in **8**), 2.59 (pseudoquint., J = 1.4 Hz, H(1) in **9**), 2.00 (m, $J_{7,8}$ = 4.8 Hz, H(7) in **9**), 1.76 (m, J_{gem} = 9.6 Hz, *anti*-H(11) in **8** and **9**), 1.54–1.72 (m, H(7) in **8**, *syn*-H(11) and HCCH in cyclopropane at C(5) in **8** and **9**), 1.25 (s, CH_3 in **9**), 1.15 (s, CH_3 in **8**), 0.92–1.14 (m, H(8), *syn*-H(9), H(10) and HCCH in cyclopropane at C(5) in **8** and **9**), 0.68 (m, *anti*-H(9) in **8** and **9**). ^{13}C NMR, δ: 93.8, 93.1 (C-2), 72.6 (C-5), 45.0, 44.9 (C-1), 44.4 (C-11), 41.1 (C-7), 40.8, 40.3 (C-6), 27.1 s, 26.8 d, 26.1 s, 25.5 d (C-8, C-10), 24.0 (C-9), 21.6 (CH_3), 15.3, 10.8 (CH_2CH_2). MS, *m/z* (*I*(%)): 173 [M−CH_3]$^+$ (0.4), 145 (25), 131 (26), 117 (50), 91 (100).

Trispiro{3,4-diazatetracyclo[5.5.1.0^{2,6}.0^{8,10}]trideca-3,9-diene-5,1′:11,1″:13,1‴-tricyclopropane} (10) and trispiro{3,4-diazatetracyclo[5.5.1.0^{2,6}.0^{8,10}]trideca-3,10-diene-5,1′:9,1″:13,1‴-tricyclopropane} (11). The solvents and most of the starting diene **2** were distilled off *in vacuo*, and the resudue was separated by preparative TLC (silica gel L, ether–hexane, 1 : 1) to give two zones containing a mixture of regioisomers **10** and **11** in ~8 : 1 ratio (R_f 0.80–0.84) and 1:1.8 (R_f 0.75–0.80), overall yield 23 %. ^{1}H NMR, δ: **10**, 5.39 (dd, $J_{9,10}$ = 5.3 Hz, $J_{8,9}$ = 2.1 Hz, H(9)), 5.22 (d, $J_{2,6}$ = 7.2 Hz, H(2)), 5.15 (dd, $J_{9,10}$ = 5.3 Hz, $J_{8,10}$ = 1.9 Hz, H(10)), 3.40 (ddt, $J_{8,12}$ = 9.5 Hz, $J_{7,8}$ = 4.7 Hz, J ~2.0 Hz, H(8)), 2.54 (dd, $J_{8,12}$ = 9.5 Hz, $J_{1,12}$ = 4.7 Hz, H(12)), 1.97 (br.d, $J_{2,6}$ = 7.2 Hz, H(6)), 1.90 (br.d, $J_{1,12}$ =4.7 Hz, H(1)), 1.37 (br.d, $J_{7,8}$ = 4.7 Hz, H(7)), 1.60 and 0.91 (two m, 2 HCCH in cyclopropane at C(5)), 0.80–1.00, 0.67, 0.45, 0.31, 0.14 (five m in cyclopropanes at C(11) and C(13)); **11**, 5.58 (dd, $J_{10,11}$ = 5.3 Hz, $J_{11,12}$ = 2.1 Hz, H(11)), 5.15 (dd, $J_{10,11}$ = 5.3 Hz, $J_{10,12}$ = 1.9 Hz, H(10)), 4.81 (br.d, $J_{2,6}$ = 7.2 Hz, H(2)), 3.51 (m, $J_{8,12}$ = 9.5 Hz, $J_{1,12}$ = 4.7 Hz, J ~ 2.0 Hz, H(12)), 2.40 (dd, $J_{8,12}$ = 9.5 Hz, $J_{7,8}$ = 4.7 Hz, H(8)), 2.20 (br.d, $J_{2,6}$ = 7.2 Hz, H(6)), 2.30 (br.d, $J_{1,12}$ = 4.7 Hz, H(1)), 0.99 (br.d, $J_{7,8}$ = 4.7 Hz, H(7)), 1.60 and 0.90 (two m, 2 HCCH in cyclopropane at C(5)), 0.80–1.00, 0.67, 0.45, 0.31, 0.14 (five m in cyclopropanes at C(11) and C(13)). ^{13}C NMR, δ: **10**, 138.9 (C-9), 128.8 (C-10), 92.1 (C-2), 72.1 (C-5), 52.4, 48.2, 47.9, 47.6, 40.1 (C-1, C-6, C-7, C-8, C-12), 32.0 (C-13), 27.7 (C-11), 15.6, 15.2, 12.1, 11.1 (C-2′, C-3′, C-2″, C-3″ in cyclopropanes at C(5) and C(11)), 6.3, 0.6 (C-2‴, C-3‴ in cyclopropane at C(13)); **11**, 139.2 (C-11), 128.9 (C-10), 93.7 (C-2), 72.5 (C-5), 51.5, 48.7, 48.5, 47.5, 38.5 (C-1, C-6, C-7, C-8, C-12), 31.8 (C-13), 27.7 (C-11), 15.5, 14.9, 11.6, 10.5 (C-2′, C-3′, C-2″, C-3″ in cyclopropanes at C(5) and C(11)), 6.2, 0.6 (C-2‴, C-3‴ in cyclopropane at C(13)). MS, *m/z* (*I*(%)): 252 M$^+$ (2), 209 (2), 195 (7), 181 (10), 132 (31), 117 (100).

Spiro{7,8-diazatetracyclo[4.3.0.0^{2,4}.0^{3,5}]non-7-ene-9,1′-cyclopropane} (12). A weakly colored liquid, yield ~60 %, b.p. 87–89 °C (1 Torr). ^{1}H NMR, δ: 5.19 (dt, 1 H, $J_{1,6}$ = 7.0 Hz, J ~1.3 Hz, H(6)), 2.75 (d.quint, 1 H, J = 3.2 Hz, J ~1.4 Hz, H(5)), 2.07 (m, 1 H, H(4)), 2.02 (dt, 1 H, $J_{1,6}$ = 7.0 Hz, J ~ 1.0 Hz, H(1)), 1.89 (m, 1 H, H(2)), 1.86 (m, 1 H, H(3)), 1.67, 1.52 and 0.9–1.1 (three m, 1 H, 1 H and 2 H, CH_2CH_2). ^{13}C NMR, δ: 96.1 (J_{C-H} = 146 Hz, C-6), 62.3 (C-9), 42.3 (J_{C-H} = 138 Hz, C-1), 38.4 (J_{C-H} = 165 Hz, C-5), 36.2 (J_{C-H} = 163 Hz, C-2), 14.4, 10.4 (J_{C-H} = 160 Hz, CH_2CH_2), 6.5 (J_{C-H} = 208 Hz, C-4), 0.1 (J_{C-H} = 210 Hz, C-3). MS, *m/z* (*I*(%)): 146 M$^+$ (2), 117 (42), 115 (49), 103 (14), 91 (70), 78 (100).

Dispiro{2,3-diazabicyclo[3.1.0]hex-2-ene-4,1′-cyclopropane-6,1″-cyclobutane} (13). A weakly colored liquid, yield 62–65 %, b.p. 76–78 °C (0.6 Torr). ^{1}H NMR, δ: 4.67

(d, 1 H, $J_{1,5}$ =4.8 Hz, H(1)), 1.55–2.25 (m, 8 H, 3 CH_2 in cyclobutane and CHCH in cyclopropane), 1.52 (d, 1 H, $J_{1,5}$ =4.8 Hz, H(5)), 1.42, 0.92 (two m, 1+1 H, CHCH in cyclopropane). ^{13}C NMR, δ: 72.3 (C-1), 69.2 (C-4), 31.0 (C-6), 28.9 (C-5), 28.5, 21.8 and 15.5 (3 CH_2 in cyclobutane), 13.8, 10.2 (CH_2CH_2 in cyclopropane). MS, *m/z* (*I*(%)): 148 M^+ (0.6), 147 (0.9), 120 (38), 105 (21), 92 (100).

6-Methoxycarbonyl-1-methyl- and 6-methoxycarbonyl-5-methylspiro{2,3-diazabicyclo[3.1.0]hex-2-ene-4,1′-cyclopropanes} (14 and 15). Yellow oil, a mixture of regioisomers (~2 : 1), yield 45–48 %, b.p. 108–112 °C (0.5 Torr). ^1H NMR, δ: **14**, 3.50 (s, OCH_3), 1.97 (d, $J_{5,6}$ = 4.0 Hz, H(6)), 1.75 (s, CH_3), 0.92 (d, $J_{5,6}$ = 4.0 Hz, H(5)), 1.55–1.7, 1.0–1.3 (two m, CH_2CH_2); **15**, 4.84 (d, $J_{1,6}$ = 2.3 Hz, H(1)), 3.50 (s, OCH_3), 1.03 (d, $J_{1,6}$ = 2.3 Hz, H(6)), 1.01 (s, CH_3), 1.55–1.7, 1.0–1.2 (two m, CH_2CH_2). ^{13}C NMR, δ: **14**, 169.6 (CO), 72.9 (C-1), 69.0 (C-4), 51.3 (OCH_3), 32.1 (C-6), 30.1 (C-5), 16.9, 11.3 (CH_2CH_2), 11.1 (CH_3); **15**, 169.5 (CO), 72.7 (C-1), 69.8 (C-4), 51.3 (OCH_3), 32.5 (C-6), 23.8 (C-5), 14.1, 11.8 (CH_2CH_2), 8.0 (CH_3).

6-Ethenylspiro-4,5-diazaspiro[2,4]hept-4-ene (16). A slightly yellow, readily polymerizable liquid, yield 42 %, b.p. 80–82 °C (13 Torr). ^1H NMR, δ: 5.94 (ddd, 1 H, J_{trans} = 17.2 Hz, J_{cis} = 10.4 Hz, $J_{6,8}$ = 7.0 Hz, =CH), 5.30 (dt, 1 H, J_{trans} = 17.2 Hz, J = 1.2 Hz) and 5.23 (dt, 1 H, J_{cis} = 10.4 Hz, J = 1.2 Hz, $=CH_2$), 5.05 (m, 1 H, $J_{6,7a}$ = 9.6 Hz, $J_{6,7b}$ = 7.3 Hz, $J_{6,8}$ = 7.0 Hz, $J_{6,9}$ = 1.2 Hz, H(6)), 1.91 (dd, 1 H, J_{gem} = 12.7 Hz, $J_{6,7a}$ = 9.6 Hz, H(7a)), 1.70 (m, 1 H, J_{cis} = 9.4 Hz, J_{trans} = 4.2 Hz, J_{gem} = 6.6 Hz, H(1a)), 1.56 (m, 2 H, H(2a) and H(7b)), 1.03 (m, 2 H, H(1b) and H(2b)); the H(1), H(2), H(6), and H(7) protons have additional far coupling constants J 0.4–0.5 Hz.

6-(1-Methylethylidene)-4,5-diazaspiro[2,4]hept-4-ene (18). Yellow liquid, yield 32 %, b.p. 70–72 °C (0.6 Torr). ^1H NMR, δ: 2.41 (sept, 2 H, J ~2.1, H(7)), 2.33 (br.t, 3 H, J = 2.1 Hz, CH_3), 1.82 (br.t, 3 H, J = 2.0 Hz, CH_3), 1.68 and 1.09 (two m, 2×2 H, CH_2CH_2). ^{13}C NMR, δ: 157.6 (C-6), 133.2 ($=CMe_2$), 66.2 (C-3), 29.1 (J_{C-H} = 136 Hz, C-7), 21.9 and 19.5 (J_{C-H} = 126 Hz, 2 CH_3), 15.5 (J_{C-H} = 163 Hz, CH_2CH_2).

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