Reaction of 2-aza-1,3,5-trienes with superbases: an unexpected approach towards exomethylene tetrahydroazepines*

N. A. Nedolya, O. G. Volostnykh, O. A. Tarasova, A. I. Albanov, and B. A. Trofimov*

A. E. Favorsky Irkutsk Institute of Chemistry, Siberian Branch of the Russian Academy of Sciences, 1 ul. Favorskogo, 664033 Irkutsk, Russian Federation. Fax: +7 (395) 241 9346. E-mail: nina@irioch.irk.ru

Reaction of 2-methoxy-*N*-(1-methylpropylidene)-1-(methylsulfanyl)buta-1,3-dien-1-amine with Bu^{t}OK resulted in 2-ethylidene-6-methoxy-7-(methylsulfanyl)-3,4,5,6-tetrahydro-2*H*-azepine, the first example of 2-ethylidene-3,4,5,6-tetrahydro-2*H*-azepines, along with hitherto unknown 7-ethyl-3-methoxy-2-(methylsulfanyl)-4,5-dihydro-3*H*-azepine and 2-ethyl-6-methoxy-3*H*-azepine.

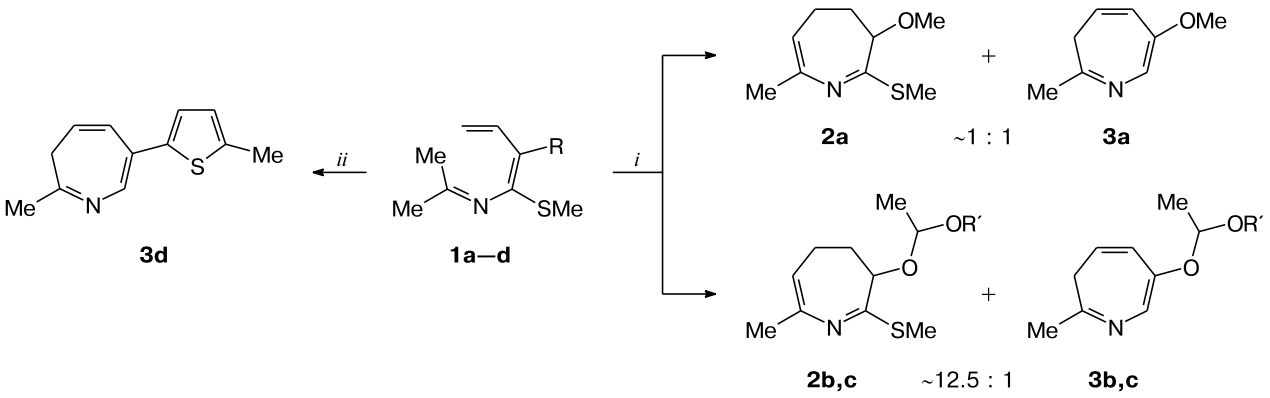
Key words: azatrienes, azepines, dihydroazepines, methoxyallene, isothiocyanate, superbases, deprotonation, electrocyclic ring closure.

It has been recently shown[1] that treatment of 2-aza-1,3,5-trienes **1**, which are readily available from lithiated allenes or alkynes, isopropyl isothiocyanate and methyl iodide,[2—4] with potassium *tert*-butoxide in THF or in the THF—DMSO mixture readily and usually in high yields afforded hitherto unknown 4,5-dihydro-3*H*-azepines **2** and/or 3*H*-azepines **3**. Under compatible reaction conditions, the nature of the substituent R in the starting azatriene **1** determined the ratio **2** : **3** (Scheme 1).

In continuation of this research, we involved 2-aza-1,3,5-triene **4** in the reaction under consideration in order to synthesize novel derivatives of 4,5-dihydro-3*H*-azepines and 3*H*-azepines promising for the search for biologically and pharmacologically active compounds and to study in detail azatriene deprotonation, which were almost not explored. Compound **4** was prepared from 1-lithio-1-methoxyallene and *sec*-butyl isothiocyanate (*via* 1-aza-1,3,4-triene **5**) (Scheme 2).[5]

Isomerization of 1-aza-1,3,4-triene **5** into 2-aza-1,3,5-triene **4** accompanied by the competing heterocyclizations of azatrienes occurring even at low temperatures,[2] the latter reactions furnished pyrrole **6** and 2,3-dihydropyridine **7** in

Scheme 1



1: R = OMe (**a**), OCH(Me)OEt (**b**), OCH(Me)OBu (**c**), 5-methyl-2-thienyl (**d**); **2:** R′ = Et (**b**), Bu (**c**)

Reagents and conditions: *i*. Bu^{t}OK (1.0—1.2 equiv.), THF—DMSO ((4.3—5.0) : 1, v/v), ~—30 °C, 30 min (Method *A*) or THF, 0 °C, 10 min (Method *B*). *ii*. Method *A*.

* Dedicated to Academician of the Russian Academy of Sciences R. Z. Sagdeev on the occasion of his 70th birthday.

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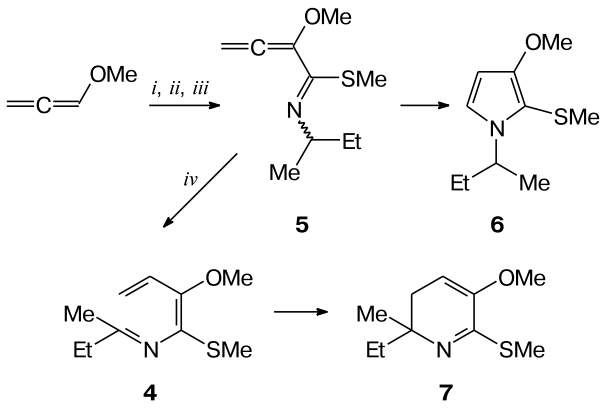
Scheme 2



Reagents and conditions: *i*. Bu^{n}Li, THF—*n*-C_6H_{14}, −100—−50 °C, 10 min. *ii*. Et(Me)CHN=C=S, THF, −90—−30 °C, 20 min. *iii*. MeI, −80—20 °C, 30 min. *iv*. ~60 °C, 15 min, [1,5]-H shift.

small amounts. At the increased temperatures, the major reaction product was 2,3-dihydropyridine **7** (up to 90% in the mixture with pyrrole **6**).[5] Whereas, in the presence of CuX (X = Cl, Br, I) pyrrole **6** is the only product.[4]

Unlike azatrienes **1** (see Scheme 1), in the case of 2-aza-1,3,5-triene **4**, superbase can attack not only the methyl group at the C atom of the azomethyne fragment but also the methylene group of the same moiety (N=C(Me)CH_2Me).[6] Eventually, it could led to 7-ethyl-4,5-dihydro-3*H*-azepine **8** and the corresponding 3*H*-azepine **9** as well as to 6,7-dimethyl-3-methoxy-2-(methylsulfanyl)-4,5-dihydro-3*H*-azepine (**10**) and 3*H*-azepine **11**, or to their mixture (Scheme 3).

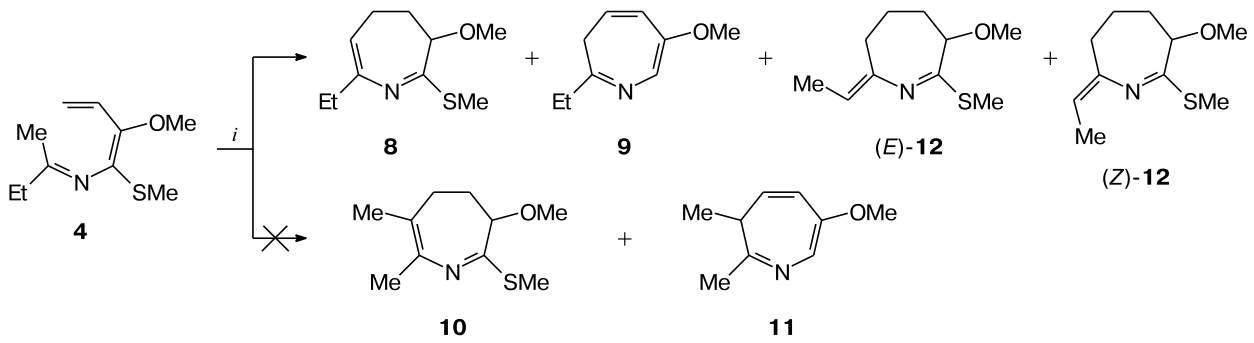
It was found that under the described conditions (Method *A*)[1] deprotonation of 2-aza-1,3,5-triene **4** involved exclusively the methyl group of the azomethyne moiety and unexpectedly yielded the exomethylene compound, 2-ethylidene-6-methoxy-7-(methylsulfanyl)-3,4,5,6-tetrahydro-2*H*-azepine (**12**) (predominantly (*Z*)-isomer), as a major product along with 3*H*-azepine **9** and 4,5-dihydro-3*H*-azepine **8** (see Scheme 3). The ratio of compounds **8**, **9**, and **12** was ~7 : 1 : 8.7, the total yield was ~93% (the ^{1}H NMR data), and the isolated yield was ~80%. The signals for the (*E*)-isomer of 3,4,5,6-tetrahydro-2*H*-azepine **12** in the ^{1}H NMR spectra of the reaction products were of low intensity.

Reaction proceeds most likely *via* intramolecular cyclization of the carbanion **A**, which was generated *in situ* by the action of Bu^{t}OK, into azacycloheptadienyl anion **B** and accompanied by the competing [1,5]-H^1 or [1,5]-H^2 shift (Scheme 4). Protonation of the isomerized intermediates resulted in endo- and exomethylene azacycloheptadienes **8** and **12**, while the competing elimination of methylsulfanyl anion afforded 3*H*-azepine **9**.

3*H*-Azepine **9** was isolated as the individual compound. The attempts to separate the mixtures of 4,5-dihydro-3*H*-azepine **8** with 3,4,5,6-tetrahydro-2*H*-azepine **12** by column chromatography or using diluted HCl (varying the concentration of the solution) failed. In the latter case, we managed to affect the ratio **8** : **12** and separate a fraction enriched with 4,5-dihydro-3*H*-azepine **8** (up to ~65% by the ^{1}H NMR data). All other fractions contained predominantly 3,4,5,6-tetrahydro-2*H*-azepine **12** (either (*Z*)-isomer or the mixture of (*E*)- and (*Z*)-isomers).

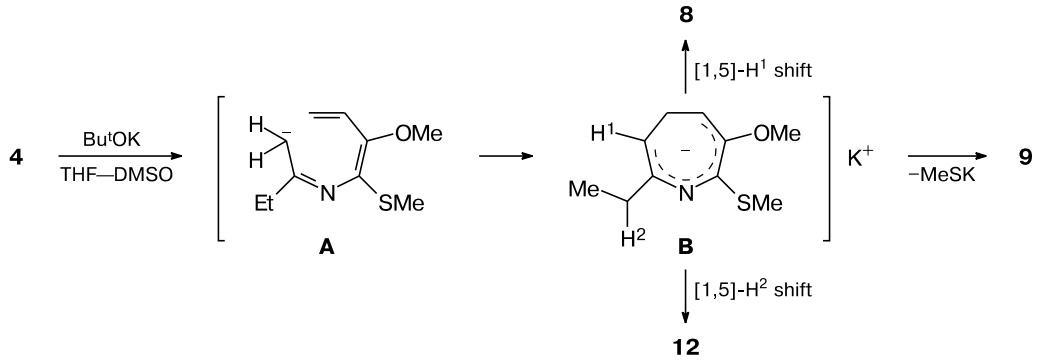
It should be noted that despite the great interest in seven-membered azaheterocycles, which is due primarily to their pronounced biological activity (the preparations based on these compounds are widely used in the medical practice as psychotropic drugs, antidepressants, and anticonvulsants for the treatment of various forms of mental disorders and as well as the antiviral, antimicrobial, and anticancer drugs[7]), the data on the synthesis of exomethylene azepine compounds of the type of **12** are apparently absent or of difficult access. Only few examples were described, *e.g.*, synthesis of several derivatives of tetrasubstituted 1-benzoyl-2-methylidene-2,3-dihydro-1*H*-azepines by the reaction of the corresponding 2-methyl(ethyl)-3*H*-azepines (obtained from 2*H*-azirines and cyclopenta-2,4-

Scheme 3



Reagents and conditions: *i*. Bu^{t}OK (1.5 equiv.), THF—DMSO (5 : 1, v/v), ~−30 °C, 30 min.

Scheme 4



dien-1-one) with benzoyl chloride in benzene in the presence of DABCO,[8] synthesis of 2-[bromo(triphenyl)-phosphoranyl]methylidene-2,3,4,7-tetrahydro-1*H*-azepines from 2-vinylaziridines and bromo(triphenyl)prop-2-ynylphosphorane,[9] and preparation of 1-[2-(phenylmethylidene)-3,4,5,6-tetrahydro-2*H*-azepin-7-yl]pyridinium tosylate from 2-[(*E*)-phenylmethylidene]cyclohexanone oxime and tosyl chloride in the anhydrous pyridine.[10]

In summary, we synthesized the first representative of 2-ethylidene-3,4,5,6-tetrahydro-2*H*-azepines and obtained the novel data on the deprotonation of 2-aza-1,3,5-trienes and properties of the carbanions formed.

Experimental

IR spectra were recorded on a Bruker Vertex 70 spectrometer in a thin layer. ^{1}H (400.13 MHz) and ^{13}C (100.62 MHz) NMR spectra were run on a Bruker DPX-400 instrument, homo- and heteronuclear correlation experiments were performed an a Bruker AV-400 spectrometer in $CDCl_3$ with hexamethyldisiloxane as internal standard. Mass spectra (EI, 70 eV) were obtained on a Shimadzu GCMS-QP5050A instrument. Liquid nitrogen was used for the cooling. THF was purified by reflux with powdered KOH (~50 g L^{-1}) and subsequent distillation over Na in the presence of benzophenone under argon. Methoxyallene[11] and *sec*-butyl isothiocyanate[2] were synthesized by the known procedure. Commercially available butyllithium (~2.5 *M* solution in hexane, Chemetall, Germany) was used.

2-Methoxy-*N*-(1-methylpropylidene)-1-(methylsulfanyl)-buta-1,3-dien-1-amine (4). To a solution of Bu^{n}Li (128 mmol) in a mixture of hexane (51.2 mL) and THF (120 mL) cooled to −100 °C, methoxyallene (10.21 g, 146 mmol) was added, the mixture was stirred at ~−50 °C for 10 min, cooled to −90 °C, and then a mixture of *sec*-butyl isothiocyanate (11.52 g, 100 mmol) and THF (5 mL) was added in one portion. After stirring at ~−30 °C for 20 min, the mixture was cooled to −80 °C, MeI (22.84 g, 161 mmol) was added, and the cooling was removed. After stirring for 30 min at ambient temperature, the mixture was cooled to −80 °C and saturated aqueous NH_4Cl (150 mL) was added. The products were extracted with diethyl ether (3×50 mL), the combined organics were washed with water (three times) and dried with $MgSO_4$. The solvent was removed *in vacuo* maintaining the bath temperature below 20 °C to give 19.66 g (~99%) of light brown mobile liquid. According to the ^{1}H NMR data, the mixture contained 2-aza-1,3,5-triene **4**[5] (~45%), methyl *N*-(*sec*-butyl)-2-methoxybuta-2,3-dienimidothioate (**5**)[5] (~38%), 1-(*sec*-butyl)-3-methoxy-2-(methylsulfanyl)-1*H*-pyrrole (**6**)[5] (~14%), and 2-ethyl-5-methoxy-2-methyl-6-(methylsulfanyl)-2,3-dihydropyridine (**7**)[5] (~3%). Additional vacuum drying of the reaction products at ~60 °C (bath temperature) for 15 min, afforded the mixture of three following products: 2-aza-1,3,5-triene **4** (~82%), pyrrole **6** (~10%), and 2,3-dihydropyridine **7** (~8%).

Deprotonation of 2-aza-1,3,5-triene 4. To a product mixture (7 g), which contained ~29 mmol of 2-aza-1,3,5-triene **4**, in THF (42 mL) cooled to −65 °C, a mixture of DMSO (10.5 mL) and THF (10.5 mL) was added followed by addition of Bu^{t}OK (4.73 g, 42 mmol). The reaction mixture was stirred at ~−30 °C for 30 min, cooled to −60 °C, and quenched with water (50 mL). After separation of the layers, the water phase was extracted with diethyl ether (3×50 mL), the combined organic layers were washed with water (three times), and dried with $MgSO_4$. Removal of the solvents under a reduced pressure at 20 °C afforded 6.60 g of light brown mobile liquid, which according to the ^{1}H NMR data contained (*Z*)-isomer of 3,4,5,6-tetrahydro-2*H*-azepine **12** (~41%), 4,5-dihydro-3*H*-azepine **8** (~33%), pyrrole **6** (~14%), 2,3-dihydropyridine **7** (~7%), 3*H*-azepine **9** (~5%), and trace amounts of (*E*)-isomer of **12**. Treatment of the mixtures with ~1—3% aqueous HCl and subsequent column chromatography (neutral Al_2O_3, subsequent elution with petroleum ether, petroleum ether—Et_2O) provided 0.50 g (11%) of 3*H*-azepine **9** as orange liquid and 3.98 g (69%) of the mixtures of 4,5-dihydro-3*H*-azepine **8** and 3,4,5,6-tetrahydro-2*H*-azepine **12** (liquids with color from yellow to orange) with different ratio of (*E*)- and (*Z*)-isomers of compound **12**.

2-Ethyl-6-methoxy-3*H*-azepine (9). n_D^{21} 1.5244. IR (neat), ν/cm^{-1}: 3149 w, 3072 w, 3037, 2970 s, 2934 s, 2875, 2830, 2753 w, 2429 w, 2239 w, 2204, 2166 w, 2034 w, 1913 w, 1849 w, 1718 w, 1674 w, 1655 w, 1619 s, 1594 sh, 1573 sh, 1535, 1490 w, 1459 s, 1443, 1427 s, 1422 s, 1398, 1376, 1361 sh, 1343, 1313 w, 1287, 1263 w, 1217 s, 1201 s, 1178, 1153 s, 1146 sh, 1128, 1094, 1069, 1057, 1045, 1018 s, 969, 945 w, 933 w, 921 w, 902 w, 863, 838, 823, 774, 750, 734 sh, 713, 698, 655 sh, 622, 590 w, 575 w, 556 w, 543 w, 503 sh, 482 w, 464 w, 443 w. ^{1}H NMR, δ: 1.10 (t, 3 H, $CH_2C\underline{H}_3$, ^3J = 7.3 Hz); 2.40 (q, 2 H, $C\underline{H}_2CH_3$, ^3J = 7.3 Hz); 2.49 (m, 2 H, C(3)H_2); 3.68 (s, 3 H, OMe); 5.32 (dt, 1 H,

C(4)H=, ^3J = 9.4 Hz, ^3J = 7.1 Hz); 6.17 (dd, 1 H, C(5)H=, ^3J = 9.4 Hz, ^4J = 2.4 Hz); 6.97 (d, 1 H, C(7)H=, ^4J = 2.4 Hz). ^{13}C NMR, δ: 11.67 ($CH_2\underline{C}H_3$); 32.87 ($\underline{C}H_2CH_3$); 35.88 ($C(3)H_2$); 55.80 (OMe); 117.90 (C(7)H=); 120.81 (C(5)H=); 124.16 (C(4)H=); 150.05 (C(6)=); 153.03 (N=C). MS (EI, 70 eV), m/z (I_{rel} (%)): 151 $[M]^{\cdot+}$ (46), 136 $[M - Me]^+$ (100), 123 $[M - C_2H_4]^{\cdot+}$ (34), 108 $[M - Me - CO]^+$ (31). Found (%): C, 71.19; H, 8.44; N, 9.52. $C_9H_{13}NO$. Calculated (%): C, 71.49; H, 8.67; N, 9.26.

Mixture of isomeric compounds 8, *Z*-12, and *E*-12. Found (%): C, 60.50; H, 8.42; N, 7.27; S, 16.00. $C_{10}H_{17}NOS$. Calculated (%): C, 60.26; H, 8.60; N, 7.03; S, 16.09.

7-Ethyl-3-methoxy-2-(methylsulfanyl)-4,5-dihydro-3*H*-azepine (8). ^{1}H NMR, δ: 1.02 (t, 3 H, $CH_2C\underline{H}_3$, ^3J = 7.5 Hz); 1.78, 1.91 (both m, 1 H each, $C(4)H_2$); 2.08, 2.47 (both m, 1 H each, $C(5)H_2$); 2.18 (q, 2 H, $C\underline{H}_2CH_3$, ^3J = 7.5 Hz); 2.32 (s, 3 H, SMe); 3.37 (s, 3 H, OMe); 3.99 (dd, 1 H, C(3)H, ^3J = 10.4 Hz, ^3J = 7.8 Hz); 5.18 (m, 1 H, C(6)H=). ^{13}C NMR (j-mod), δ: 11.72 (SMe); 12.21 ($CH_2\underline{C}H_3$); 20.73 ($C(4)H_2$); 28.96 ($\underline{C}H_2CH_3$); 42.41 ($C(5)H_2$); 58.95 (OMe); 82.03 (C(3)H); 107.92 (C(6)H=); 152.65 (C(7)=); 174.90 (N=C). Signals in the NMR spectra were unambiguously assigned using homonuclear and heteronuclear 2D NMR experiments (COSY and HSQC). MS (EI, 70 eV), m/z (I_{rel} (%)): 199 $[M]^{\cdot+}$ (88), 184 $[M - Me]^+$ (57), 152 $[M - MeS]^+$ (20), 126 $[M - MeSCN]^{\cdot+}$ (100).

2-(*Z*)-Ethylidene-6-methoxy-7-(methylsulfanyl)-3,4,5,6-tetrahydro-2*H*-azepine (*Z*-12). ^{1}H NMR, δ: 1.55 (d, 3 H, $C\underline{H}_3CH$=, ^3J = 6.7 Hz); 1.62, 1.78 (both m, 1 H each, $C(4)H_2$); 1.63, 1.90 (both m, 1 H each, $C(5)H_2$); 1.92, 2.15 (both m, 1 H each, $C(3)H_2$); 2.28 (s, 3 H, SMe); 3.39 (s, 3 H, OMe); 3.80 (dd, 1 H, C(6)H, ^3J = 9.6 Hz, ^3J = 2.3 Hz); 4.83 (q, 1 H, $CH_3C\underline{H}$=, ^3J = 6.7 Hz). ^{13}C NMR (j-mod), δ: 12.05 (SMe); 12.53 ($\underline{C}H_3CH$=); 26.95 ($C(5)H_2$); 29.75 ($C(4)H_2$); 32.51 ($C(3)H_2$); 58.51 (OMe); 82.74 (C(6)H); 108.54 ($CH_3\underline{C}H$=); 147.09 (N—C=); 171.24 (N=C). Signals in the NMR spectra were unambiguously assigned using homonuclear and heteronuclear 2D NMR experiments (COSY, NOESY, and HSQC).

2-(*E*)-Ethylidene-6-methoxy-7-(methylsulfanyl)-3,4,5,6-tetrahydro-2*H*-azepine (*E*-12). ^{1}H NMR, δ: 1.64 (d, 3 H, $C\underline{H}_3CH$=, ^3J = 7.0 Hz); 2.23 (s, 3 H, SMe); 3.37 (s, 3 H, OMe); 3.77 (m, 1 H, C(6)H); 4.99 (q, 1 H, $CH_3C\underline{H}$=, ^3J = 7.0 Hz). The signals of the protons for the $C(4)H_2$—$C(6)H_2$ groups overlap with more intense signals of (*Z*)-isomer. ^{13}C NMR (j-mod), δ: 11.99 (SMe); 12.31 ($\underline{C}H_3CH$=); 26.21 ($C(5)H_2$); 30.06 ($C(4)H_2$); 32.45 ($C(3)H_2$); 58.49 (OMe); 81.99 (C(6)H); 109.46 ($CH_3\underline{C}H$=); 146.97 (N—C=); 171.71 (N=C).

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