Chemistry of Heterocyclic Compounds, Vol. 45, No. 4, 2009

N-TRIFLUOROMETHYLAZOLES

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A method for synthesizing N-trifluoromethylazole derivatives has been developed via the reaction of their sodium salts with dibromodifluoromethane and substitution of bromine by fluorine. The first ionic liquids with a trifluoromethyl group on an imidazole nitrogen atom have been synthesized.

Keywords: dibromodifluoromethane, imidazole, ionic liquids, pyrazole, 1,2,4-triazole, N-trifluoromethylazoles, fluorination.

N-Perfluoroalkyl derivatives of nitrogen heterocycles have been little studied. There is only limited information in the literature about such compounds. The introduction of a perfluoroalkyl group on the nitrogen atom *via* radical or nucleophilic reactions has not been successfully. In both cases only C-perfluoroalkyl derivatives are formed [1-3]. An electrophilic perfluoroalkylation is difficult to carry out [4]. Aliphatic precursors of heterocyclic compounds with N-perfluoroalkyl groups are also difficult to obtain or of low stability. Thus dialkylperfluoroalkylamines readily eliminated fluorine atoms from the α-position. This property has been used for exchange of an alcoholic grouping for fluorine in the Yarovenko reaction [5]. A few examples have been reported in the literature for multistage syntheses of N-trifluoromethyl derivatives, mainly of condensed heterocyclic compounds. The study [6] reported N-trifluoromethyl-4-ethoxycarbonyl pyrazole prepared via highly toxic mercury compounds. In our laboratories we have prepared N-trifluoromethylbenzotriazole through sequential dithiocarboxylation at the nitrogen atom, chlorination of the introduced function, and subsequent fluorination of the trichloromethyl group by anhydrous HF [7]. N-Trifluoromethylbenzimidazoles are formed by reaction of the corresponding sodium salts with dibromodifluoromethane and subsequent exchange of the bromine atom for fluorine using tetramethylammonium fluoride in glyme [7]. N-Trifluoromethylcarbazole was obtained by fluorination of the corresponding dithiocarboxyl derivative using AgF [8]. Using a similar sequence of reactions 1-trifluoromethyl derivatives of 4-quinolone have been synthesized and these show very high antibacterial activity [9].

In continuation of this work we have undertaken the preparation of the N-trifluoromethyl derivatives of the simplest azole representatives, *viz.* imidazole **1**, pyrazole **2**, and 1,2,4-triazole **3**. We have devised a convenient method for the introduction of a bromodifluoromethyl group on the nitrogen atom of the heterocycle through the reaction of their sodium salts with the available dibromodifluoromethane in DMF in the presence of a catalytic amount of tetrabutylammonium bromide. We have previously used this method successfully for the introduction of a bromotetrafluoroethyl group into azole molecules [10].

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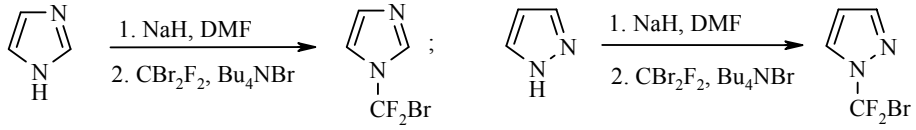
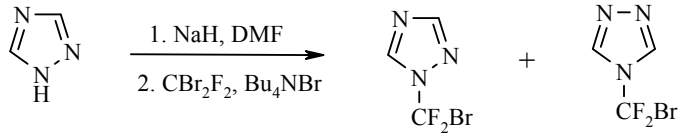
Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 4, pp. 550-556, April, 2009. Original article submitted June 23, 2008.



1 **4** **2** **5**

Bromodifluoromethyl derivatives of imidazole **4** and pyrazole **5** were obtained in high yield using this method but the difluoromethyl derivative impurities generally formed in the bromodifluoromethylation were not seen, even in the ^{19}F NMR spectra of reaction mixtures.

An unexpected result in the reaction of 1,2,4-triazole **3** with dibromodifluoromethane was the formation of a mixture of the 1-bromodifluoromethyl-1,2,4-triazole (**6**) with 4-bromodifluoromethyl-1,2,4-triazole (**7**) in equal amounts. The products are readily separated thanks to the large difference in boiling points.

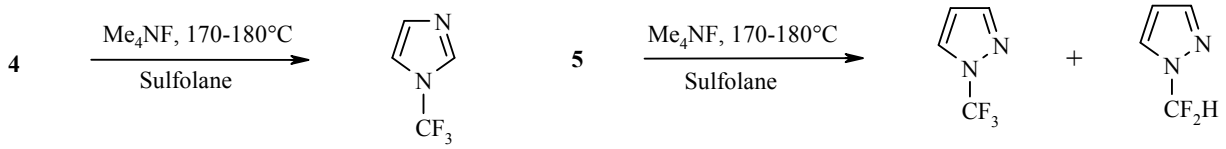


3 **6** **7**

It should be noted that the reaction of 1,2,4-triazole with 1,2-dibromotetrafluoroethane under similar conditions gives only 1-tetrafluorobromoethyl-1,2,4-triazole [10]. It is likely that the bromodifluoromethylation occurs by a halophilic mechanism similar to the reaction of CBr_2F_2 with phenols and thiophenols [11]. In such a mechanism there are intermediately formed tetrafluoroethylene in the case of the reaction with $CBrF_2CBrF_2$ and, evidently, the more reactive difluorocarbene with CBr_2F_2 hence forming a significant amount of the kinetically controlled product **7**.

It is known that the Swarts reaction is widely used for substitution of bromine atoms by fluorine [12] but the preparation of trifluoromethyl derivatives of azoles using SbF_3 was unsuccessful due to the destruction of the heterocyclic ring (even though there is an indication in the literature of the possible preparation of N-trifluoromethylimidazole with this method [13]). If the fluorination is carried out using tetramethylammonium fluoride in glyme as reported in [7] the trifluoromethyl derivatives are only formed in trace amounts and they could not be separated due to the closeness of the boiling points of products and solvent.

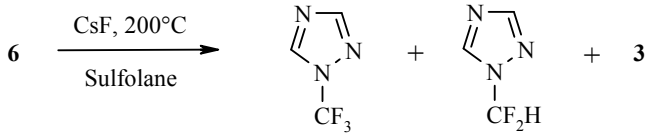
1-Trifluoromethylimidazole **8** and 1-trifluoromethylpyrazole **9** were successfully prepared by fluorination in the high boiling, polar solvent sulfolane with simultaneous distillation of the product. Pure product could be obtained in the fluorination of imidazole **4** by distillation of the reaction mixture. In the second example the difluoromethylpyrazole **10** was additionally formed. The products could be separated by fractional distillation.



8 **9** **10**

The 1-trifluoromethyl-1,2,4-triazole **11** could not be prepared by this method. At temperatures up to 180ºC the substrate is not fluorinated and at higher temperature the degradation of the fluorinating agent occurs. Substitution of the bromine atom in the triazole molecule **6** can be achieved using CsF in the presence of dibenzo-18-crown-6. Although the yield of the target product is low (14%) and the N-difluoromethyl-

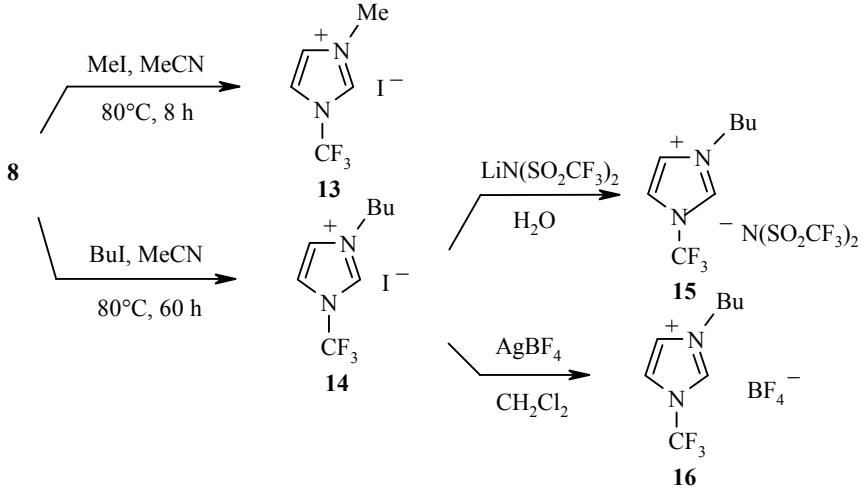
1,2,4-triazole **12** is also formed the products can be separated in the pure state by fractional distillation. In the fluorination of triazole **6** there is a significant defluoroalkylation of the heterocycle and the unsubstituted 1,2,4-triazole **3** is separated in 33% yield from the reaction mixture.



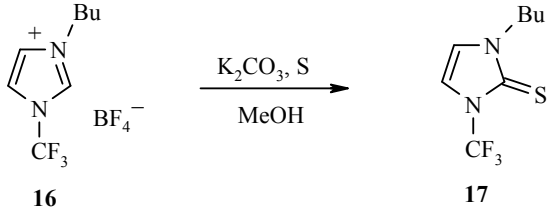
The 4-bromodifluoromethyl triazole **7** proved thermally unstable. Attempts to fluorinate it resulted in a complex mixture of products amongst which was 1-bromodifluoromethyltriazole **6** according to ^{19}F NMR spectroscopy. A similar rearrangement has also been reported in the literature for 1-alkyl-substituted 1,2,4-triazoles [14, 15].

In recent times ionic liquids have attracted the attention of chemists in organic chemistry, electrochemistry, and technology as ecologically safe alternatives to classical solvents. The search for novel materials showing ionic liquid properties is current. We have previously reported the synthesis of imidazolium salts containing an α,α-difluoromethylene fragment on the nitrogen atom [16]. Ionic liquids with trifluoromethyl groups on the nitrogen atom have not been reported to this time.

Alkylation of N-trifluoromethylimidazole **8** using alkyl iodides gave the corresponding quaternary salts **13** and **14**. Exchange of the iodide anion with ditriflylamide anion or tetrafluoroborate anion gave the corresponding low melting salts **15** and **16**. This is the first example of ionic liquids with a trifluoromethyl group on the nitrogen atom.



Salts **15** and **16** show high thermal stability (not decomposing at 250ºC) and are stable to the action of mineral acids (salt **16** can be separated unchanged after refluxing in 15% hydrochloric acid). Treatment of salt **16** with base gave an imidazolium carbene which could be fixed by reaction with sulfur. N-Tetrafluoroethyl-imidazolium salts also possess similar properties [17].



Hence we have developed a method for the synthesis of N-trifluoromethylazole derivatives and, for the first time, obtained ionic liquids with a perfluoroalkyl substituent on the nitrogen atom.

EXPERIMENTAL

^{1}H NMR spectra were recorded on a Varian VXR-300 instrument (300 MHz) with TMS as internal standard and ^{19}F NMR spectra on a Varian Gemini-200 instrument (188 MHz) with trichlorofluoromethane as internal standard using $CDCl_3$ (compounds **5, 6, 8, 9, 11, 12**) or DMSO-d_6 (compounds **13-16**). ^{13}C NMR spectra were taken on a Bruker Avance DRX 500 instrument (125 MHz) with TMS as internal standard. Mass spectra were obtained on an Agilent 1100 LS/MSD SL instrument. Melting points were determined on a Stuart Scientific SMP3 apparatus. MN-Kieslegel-60 grade silica gel was used in the work.

Reaction of Azoles 1-3 with Dibromodifluoromethane. CBr_2F_2 (15.75 g, 75 mmol) was added with vigorous stirring to a solution cooled to 0ºC of the sodium salt prepared from the corresponding heterocycle (0.05 mol), NaH (60%, 2 g, 50 mmol), and Bu_4NBr (0.1 g, 0.3 mmol) in anhydrous DMF (25 ml). The reaction mixture was gradually heated (over 2 h) to 25ºC and held at this temperature for 2 h. Water (100 ml) was added dropwise to the reaction mixture and the excess CBr_2F_2 was condensed in an entrainment trap cooled to -78ºC. The product was extracted with ether (5×30 ml) and the ether solution was washed with water (5×25 ml) and dried over $MgSO_4$. Solvent was evaporated and the product was distilled.

1-Bromodifluoromethylimidazole (4). Yield 7.68 g (78%). Bp 57-58ºC (20 mm Hg) [7].

1-Bromodifluoromethylpyrazole (5). Colorless liquid. Yield 7.38 g (75%). Bp 135-137ºC. ^{1}H NMR spectrum, δ, ppm: 6.44-6.46 (1H, m, CH); 6.85-6.93 (1H, m, CH); 7.76-7.78 (1H, m, CH). ^{19}F NMR spectrum, δ, ppm: -32.60 (s, $CBrF_2$). Found, %: C 24.46; H 1. 64; Br 40.83. $C_4H_3BrF_2N_2$. Calculated, %: C 24.39; H 1.54; Br 40.56.

1-Bromodifluoromethyl-1,2,4-triazole (6). Colorless liquid. Yield 3.26 g (33%). Bp 42-43ºC (10 mm Hg). ^{1}H NMR spectrum, δ, ppm: 8.08 (1H, s, CH); 8.49 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -34.20 (s, $CBrF_2$). Found, %: C 18.44; H 1.15; N 21.53. $C_3H_2BrF_2N_3$. Calculated, %: C 18.20; H 1.02; N 21.23.

4-Bromodifluoromethyl-1,2,4-triazole (7). Colorless liquid. Yield 3.0 g (30%). Bp 105-107ºC (10 mm Hg), mp 10-12ºC (pentane). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 8.46 (2H, s, 2CH). ^{19}F NMR spectrum ($CDCl_3$), δ, ppm: -27.92 (s, $CBrF_2$). ^{13}C {^{1}H} NMR spectrum (DMSO-d_6), δ, ppm (*J*, Hz): 139.9 (2CH); 106.0 (t, $^1J_{CF}$ = 305, $CBrF_2$). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm (*J*, Hz): 139.9 (dd, $^1J_{C,H}$ = 220, $^3J_{C,H}$ = 3, 2CH); 106.0 (t, $^1J_{C,F}$ = 305, $CBrF_2$). Found: *m/z* 198.0 $[M]^+$. $C_3H_2BrF_2N_3$. Calculated: M 198.0. Found, %: C 18.48; H 1.20; Br 40.20; N 21.37. $C_3H_2BrF_2N_3$. Calculated, %: C 18.20; H 1.02; Br 40.36; N 21.23.

Fluorination of Imidazole 4 and Pyrazole 5. Me_4NF (2.07 g, 22 mmol) was added to a solution of the corresponding heterocycle (4 g, 20 mmol) in anhydrous sulfolane (30 ml). The reaction mixture was gradually heated to 170-180ºC and the distilled product collected. The product was additionally purified by fractional distillation.

1-Trifluoromethylimidazole (8). Colorless very volatile liquid. Yield 1.0 g (36%). Bp 73-74ºC. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 7.12 (1H, s, CH); 7.16 (1H, s, CH); 7.81 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -56.40 (s, CF_3). Found, %: C 34.95; H 2.14; N 20.18. $C_4H_3F_3N_2$. Calculated, %: C 35.31; H 2.22; N 20.59.

1-Trifluoromethylpyrazole (9). Colorless very volatile liquid. Yield 1.08 g (39%). Bp 54-55ºC. ^{1}H NMR spectrum, δ, ppm: 6.93 (1H, s, CH); 7.78 (1H, s, CH); 7.86 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -60.70 (s, CF_3). Found, %: C 35.04; H 2.02; N 20.28. $C_4H_3F_3N_2$. Calculated, %: C 35.31; H 2.22; N 20.59. Fractionation gave **1-difluoromethylpyrazole (10)** (0.48 g, 20%) with bp 88-90ºC [18].

Fluorination of Triazole 6. CsF (2.43 g, 16 mmol) was added to a solution of the triazole **6** (3 g, 15 mmol) and dibenzo-18-crown-6 (0.3 g, 0.8 mmol) in anhydrous sulfolane (20 ml) at 25ºC. The reaction mixture was gradually heated to 200-205ºC and the distilled products (0.61 g) were collected. After distillation of

the products had ceased the temperature of the reaction mixture was raised to 250ºC when 1,2,4-triazole **3** (0.34 g, 33%) distilled off (mp 121ºC, heptane). The mixture of products **11** and **12** (1:0.8 according to ^{19}F NMR) contained in the first fraction was separated by fractional distillation.

1-Trifluoromethyl-1,2,4-triazole (11). Colorless very volatile liquid. Yield 0.28 g (14%). Bp 67-70ºC. ^1H NMR spectrum, δ, ppm: 8.09 (1H, s, CH); 8.49 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -60.37 (s, CF_3). Found: *m/z* 137.1 $[M]^+$. $C_3H_2F_3N_3$. Calculated: M 137.1. Found, %: N 30.28. $C_3H_2F_3N_3$. Calculated, %: N 30.65.

1-Difluoromethyl-1,2,4-triazole (12). Colorless very volatile liquid. Yield 0.11 g (6%). Bp 98-101ºC. ^1H NMR spectrum, δ, ppm (*J*, Hz): 7.22 (1H, t, $^2J_{H,F}$ = 60.3, CHF_2); 8.00 (1H, s, CH); 8.48 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -95.41 (d, $^2J_{F,H}$ = 60.3, CF_2H). Found: *m/z* 119.1 $[M]^+$. $C_3H_3F_2N_3$. Calculated: 119.1. Found, %: N 35.00. $C_3H_3F_2N_3$. Calculated, %: N 35.30.

1-Trifluoromethyl-3-methylimidazolium Iodide (13). A solution of imidazole **8** (0.045 g, 0.36 mmol) and MeI (0.1 g, 0.7 mmol) in acetonitrile (5 ml) was heated for 16 h at 70ºC. Solvent was distilled off *in vacuo* and the residue was crystallized from ethyl acetate. Yield 75 mg (75%). Colorless crystalline material. Mp 206-208ºC. ^1H NMR spectrum, δ, ppm: 4.10 (3H, s, CH_3); 7.77 (1H, s, CH); 8.22 (1H, s, CH); 10.00 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -57.20 (s, CF_3). Found, %: I 45.65; N 10.10. $C_5H_6F_3IN_2$. Calculated, %: I 45.64; N 10.08.

3-Butyl-1-trifluoromethylimidazolium Iodide (14). A solution of imidazole **8** (0.85 g, 6.25 mmol) and BuI (2.3 g, 12.5 mmol) in acetonitrile (25 ml) was heated for 96 h at 70ºC, monitoring the reaction from the ^{19}F NMR spectra. Solvent was distilled *in vacuo* and the residue was crystallized from ethyl acetate. Yield 1.8 g (90%). Colorless crystalline material. Mp 148-150ºC. ^1H NMR spectrum, δ, ppm: 0.95-1.00 (3H, m, $CH_2CH_2CH_2C\underline{H}_3$); 1.40-1.52 (2H, m, $CH_2CH_2C\underline{H}_2CH_3$); 2.02-2.11 (2H, m, $CH_2C\underline{H}_2CH_2CH_3$); 4.61-4.70 (2H, m, $C\underline{H}_2CH_2CH_2CH_3$), 8.40 (1H, s, CH); 8.47 (1H, s, CH); 10.78 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -57.90 (s, CF_3). Found, %: I 39.47; N 8.73. $C_8H_{12}F_3JN_2$. Calculated, %: I 39.65; N 8.75.

3-Butyl-1-trifluoromethylimidazolium Ditriflylamide (15). Lithium ditriflylamide (0.7 g, 2.5 mmol) was added to a solution of iodide **14** (0.65 g, 2 mmol) in water (15 ml) and stirred for 3 h at 40ºC. The product was extracted with dichloromethane (3×20 ml) and the extract was washed with water (2×10 ml) and dried over $MgSO_4$. Solvent was distilled off and the residual solvent was evaporated *in vacuo* (0.05 mm Hg, 80ºC, 10 h). Yield 0.86 g (91%). Colorless liquid. Vitrification temperature -77ºC*. ^1H NMR spectrum, δ, ppm: 0.95-1.01 (3H, m, $CH_2CH_2CH_2C\underline{H}_3$); 1.35-1.44 (2H, m, $CH_2CH_2C\underline{H}_2CH_3$); 1.82-2.00 (2H, m, $CH_2C\underline{H}_2CH_2CH_3$); 4.20-4.33 (2H, m, $C\underline{H}_2CH_2CH_2CH_3$); 7.60 (1H, s, CH); 7.67 (1H, s, CH); 9.78 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -58.89 (3F, s, NCF_3); -80.30 (6F, s, $2SO_2CF_3$). Found, %: C 25.24; H 2.82; N 8.48; I < 0.5. $C_{10}H_{12}F_9N_3O_4S_2$. Calculated, %: C 25.38; H 2.56; N 8.88.

3-Butyl-1-trifluoromethylimidazolium Tetrafluoroborate (16). A solution of salt **14** (1.46 g, 4.56 mmol) in dichloromethane (10 ml) was added to a solution of $AgBF_4$ (0.89 g, 4.56 mmol) in dichloromethane (20 ml). The reaction mixture was stirred for 15 min at 25ºC. The residue was separated by centrifugation and washed with dichloromethane (10 ml). Solvent was distilled off *in vacuo* (15 mm Hg) and the residual solvent was removed *in vacuo* (0.05 mm Hg, 80ºC, 8 h). Yield 1.26 g (99%). Colorless crystalline material. Mp 82-83ºC. ^1H NMR spectrum, δ, ppm (*J*, Hz): 0.14 (3H, t, $^2J_{H,H}$ = 7.5, $CH_2CH_2CH_2C\underline{H}_3$); 0.63 (2H, sext, $^2J_{H,H}$ = 7.5, $CH_2CH_2C\underline{H}_2CH_3$); 1.23 (2H, q, $^2J_{H,H}$ = 7.5, $CH_2C\underline{H}_2CH_2CH_3$); 3.71 (2H, $^2J_{H,H}$ = 7.5, $C\underline{H}_2CH_2CH_2CH_3$); 7.34 (1H, s, CH); 7.59 (1H, s, CH); 9.10 (1H, s, CH). ^{19}F NMR spectrum, δ, ppm: -58.68 (3F, s, CF_3); -151.17 (4F, s, BF_4). Found. %: C 34.00; H 4.52; N 9.96; I < 0.5. $C_8H_{12}BF_7N_2$. Calculated, %: C 34.32; H 4.32; N 10.00.

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*The vitrification temperature was determined using a DSK-D differential scanning calorimeter. Heating rate 2ºC/min. We express our sincere thanks to G. V. Titov (Institute of Macromolecular Chemistry of the National Academy of Sciences of Ukraine) for determining the vitrification temperature

3-Butyl-1-trifluoromethylimidazole-2-thione (17). Finely divided sulfur (0.3 g, 9 mmol) and powdered, calcined K_2CO_3 (0.83 g, 6 mmol) were added to a solution of salt **16** (0.84 g, 3 mmol) in anhydrous methanol (10 ml). The suspension was stirred for 24 h at room temperature. The precipitate was filtered, the methanol distilled *in vacuo* (15 mm Hg), and the residue was column chromatographed (eluent $CHCl_3$–CCl_4, 1:1, R_f 0.5). Yield 0.6 g (90%). Colorless liquid. Bp 97-98ºC (1 mm Hg). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 0.90 (3H, t, $^2J_{H,H}$ = 7.5, $CH_2CH_2CH_2C\underline{H}_3$), 1.32 (2H, sext, $^2J_{H,H}$ = 7.5, $CH_2CH_2C\underline{H}_2CH_3$); 1.70 (2H, q, $^2J_{H,H}$ = 7.5, $CH_2C\underline{H}_2CH_2CH_3$); 3.95 (2H, t, $^2J_{H,H}$ = 7.5, $C\underline{H}_2CH_2CH_2CH_3$); 6.65 (1H, d, $^2J_{H,H}$ = 2.7, CH); 7.59 (1H, d, $^2J_{H,H}$ = 2.7, CH). ^{19}F NMR spectrum, δ, ppm: -59.30 (s, CF_3). Found, %: C 42.74; H 5.05; N 12.55. S 14.33. $C_8H_{11}F_3N_2S$. Calculated, %: C 42.84; H 4.95; N 12.50; S 14.29.

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