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UTILIZATION OF THE AMIDOALKYLATION REACTION IN THE SYNTHESIS OF HYDROGENATED PYRIMIDINE-2-THIONES

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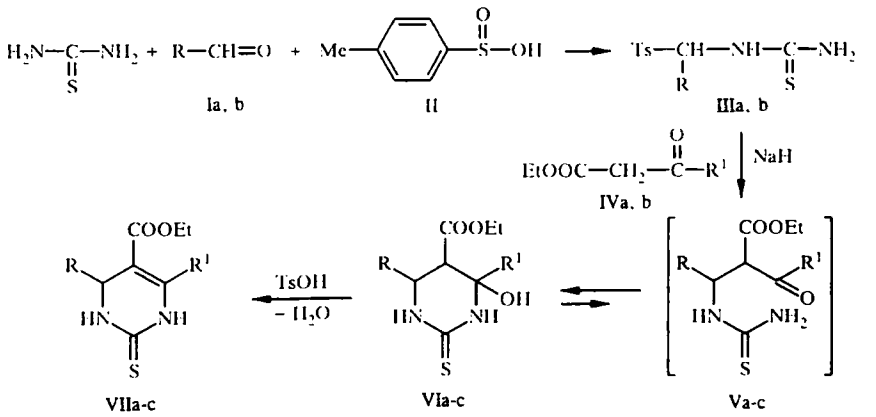
Reaction of thiourea, propionic aldehyde, or benzaldehyde and p-toluenesulfinic acid gave α-substituted N-(tosylmethyl)thioureas. Interaction of the last with sodioacetoacetic ester or sodiobenzoylacetic ester led to the synthesis of the corresponding 4-hydroxy-5-ethoxycarbonylhexahydropyrimidine-2-thiones, the dehydration of which afforded 5-ethoxycarbonyl-1,2,3,4-tetrahydropyrimidine-2-thiones.

We previously [1, 2] developed a method for the synthesis of 5-acyl-4-hydroxyhexahydropyrimidine-2-thiones based on the amidoalkylation reaction of 1,3-dicarbonyl compounds of N-(azidomethyl)- and N-(tosylmethyl)thiourea. In the development of this investigation, it seemed expedient to study the possible utilization in the synthesis of other amidoalkylating reagents, particularly N-(tosylmethyl)thioureas substituted in the α-position. The present communication describes the isolation of these compounds and their utilization in the synthesis of hydrogenated pyrimidine-2-thiones.

We showed that thiourea reacts with propionic aldehyde (Ia) or benzaldehyde (Ib) and p-toluenesulfinic acid (II) [the molar ratio thiourea:(I):(II) = 1:1:1.1] in water at 20°C to form N-(1-tosylpropyl)thiourea (IIIa) or N-(α-tosylbenzyl)thiourea (IIIb) correspondingly with yields of 84-85%. The synthesis of compound (IIIb) from thiourea, benzaldehyde, and sodium p-toluenesulfinate in the presence of excess formic acid was described previously [3]. However, since the molar ratio of the reagents indicated was 1:2:2.2 correspondingly, N,N'-bis(α-tosylbenzyl)thiourea was also formed in a significant amount besides the product (IIIb).



I, III a R = Et, b R = Ph. IV a R^1 = Me, b R^1 = Ph. V—VII a R = Et, R^1 = Me; b R = Ph, R^1 = Me; c R = R^1 = Ph

The reaction of the thioureas (IIIa,b) in acetonitrile at room temperature with sodioacetoacetic ester, obtained from acetoacetic ester (IVa) and sodium hydride, results in the formation of 6-substituted 4-hydroxy-4-methyl-5-ethoxycarbonylhexahydropyrimidine-2-thiones (VIa,b) correspondingly with yields of 70-82%. The last are products of the

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spontaneous intramolecular cyclization of the intermediate N-(oxoalkyl)thioureas (Va,b). Under analogous conditions, the thiourea (IIIb) reacts with sodiobenzoylacetic ester, being converted to 4-hydroxy-4,6-diphenyl-5-ethoxycarbonylhexahydropyrimidine-2-thione (VIc) with the yield of 86%. It should be noted that both stages in the formation of the products (VIa-c) — the reaction of the thioureas (IIIa,b) with anions of the CH-acids (IVa,b), and the heterocyclization of the thioureas (Va-c) — proceed with high diastereoselectivity: compounds (VIa,c) are obtained as the only stereoisomer, and compound (VIb) is formed as the 88:12 mixture of two stereoisomers.

The synthesized 4-hydroxyhexahydropyrimidine-2-thiones (VIa-c) undergo ready dehydration when their solutions in ethanol are boiled in the presence of p-toluenesulfonic acid; this results in the formation of the 5-ethoxycarbonyl-1,2,3,4-tetrahydropyrimidine-2-thiones (VIIa-c) correspondingly with yields of 49-84%.

The structure of the compounds (IIIa,b), (VIa-c), and (VIIa-c) was established from the data of PMR, IR, and UV spectra (see the Experimental section).

The IR spectra of the sulfonylthioureas (IIIa,b) are characterized by a series of broad absorption bands of the stretching vibrations of the NH groups at 3044-3413 cm^{-1} and two intense absorption bands at 1530-1609 cm^{-1}, associated with the vibrations of atoms of the thioureide part of the molecules [4], as well as two strong bands of the stretching vibrations of the SO_2 group at 1285-1301 and 1135-1139 cm^{-1}.

The IR spectra of the 4-hydroxyhexahydropyrimidine-2-thiones (VIa-c) have absorption bands of the stretching vibrations of the NH and OH groups at 3170-3379 cm^{-1} and a strong band of the stretching vibrations of the ester carbonyl group at 1732-1737 cm^{-1}, as well as two intense "thioamide-II" absorption bands at 1500-1586 cm^{-1}. Moreover, the spectra of the pyrimidines (VIb,c) show bands determined by the presence of phenyl substituents.

It is known that the phenomenon of ring—chain isomerism is possible for 4-hydroxyhexahydropyrimidine-2-thiones [2, 5, 6]. On the basis of the absence from the IR spectra of compounds (VIa-c) of the absorption band of the ketonic carbonyl group at 1680-1720 cm^{-1}, which should be expected in the case of the acyclic isomeric forms (Va-c), we concluded that these substances exist mainly in the cyclic form (VI), indicated in the Scheme, in the crystalline state.

A characteristic feature of the IR spectra of the 1,2,3,4-tetrahydropyrimidine-2-thiones (VIIa-c) by comparison with the spectra of the 4-hydroxyhexahydropyrimidine-2-thiones (VIa-c) is the significant shift of the band of the stretching vibrations of the ester carbonyl group to the long-wave region due to the formation of a conjugated system: it is observed at 1642-1693 cm^{-1} in the spectra of the compounds (VIIa-c).

The UV spectra of the thiones (VIa-c) have two intense absorption bands with maximums at 206-211 and 248-251 nm, caused by the unconjugated thioureide chromophore [7]. In the transition from the compounds (VIa-c) to the tetrahydropyrimidinethiones (VIIa-c), the short-wave absorption band maintains its position whereas the long-wave band, associated with the $\pi—\pi^*$-transition in the thioureide chromophore [8], undergoes a significant bathochromic shift (56-62 nm) and is observed at 304-313 nm. It should be noted that both bands of the compounds (VIIa-c) have a shoulder: for the band at 207-210 nm, it is situated on the side of the long-wave part, and for the band at 304-313 nm, it is situated on the side of the short-wave part and probably pertains to the $\pi—\pi^*$-transition in the C=C bond [2, 8].

The PMR spectra of the hydroxypyrimidines (VIa,c), recorded before their recrystallization, contain only one set of proton signals indicating that individual diastereomers are formed. On the basis of the SSCC of the 5-H and 6-H protons (11.9-12.0 Hz), we concluded that there was the equatorial orientation of substituents at the carbon atoms $C_{(5)}$ and $C_{(6)}$ of the compounds considered. The presence of the equatorially orientated substituent at the $C_{(6)}$ atom is also confirmed by the SSCC $J_{NH,6}$ = 0 Hz, which, as was shown in the work [9], is only possible with the axial position of the 6-H proton. The possibility of determining the position of the substituents at the quaternary $C_{(4)}$ carbon atom from the PMR spectra of compounds (VIa,c) does not arise. However, it can be assumed, as was done in the work [2], that the hydroxyl group in the molecules of the compounds indicated has the axial orientation.

The PMR spectrum of the thione (VIb) contains two sets of signals of the analogous protons, the ratio of the integral intensities of which is 88:12. On the basis of this, it was concluded that two diastereomers are formed. Analysis of the SSCC shows that the main stereoisomer of compound (VIb) has the equatorial position of the substituents at the $C_{(5)}$ and $C_{(6)}$ carbon atoms ($J_{5,6}$ = 12.0 Hz, $J_{NH,6}$ = 0 Hz), and the minor stereoisomer has the axial orientation of the substituent at $C_{(5)}$ and the equatorial orientation of the substituent at $C_{(6)}$ ($J_{5,6}$ = 5.2 Hz, $J_{NH,6}$ = 0 Hz).

The absence of spectral indications of the acyclic isomeric forms (Va-c) in the PMR spectra of the hydroxypyrimidines (VIa-c) favored the conclusion that these compounds exist exclusively in the cyclic form in the solution of DMSO-D_6, as well as in the crystalline state (see above).

EXPERIMENTAL

The IR spectra were recorded on the Shimadzu IR-435 instrument for suspensions in mineral oil. Electronic spectra in the region of 200-400 nm were obtained on the Beckman DU-6 spectrophotometer for solutions at the concentration of $5 \cdot 10^{-5}$ M in methanol. The PMR spectra were registered on the Bruker MSL-200 spectrometer (200 MHz) for solutions of the samples in $CDCl_3$ or DMSO-D_6; the internal standard was HMDS. The monitoring of the course of reactions and the purity of the products was accomplished by the method of TLC on plates of Kieselgel 60 F_{254} (Merck) in the 9:1 system of chloroform—methanol; spots were detected in iodine vapor.

N-(1-Tosylpropyl)thiourea (IIIa). To the solution of 1.000 g (13.14 mmole) of thiourea in 13 ml of water are added 0.764 g (13.15 mmole) of propionic aldehyde and 2.254 g (14.43 mmole) of p-toluenesulfinic acid [10]. The resulting heterogeneous mixture is stirred at room temperature for 48 h and cooled to 0°C. The residue of the product is filtered off, washed with ice-cold water and with hexane, and dried. Compound (IIIa) is obtained with the yield of 3.033 g (84.8%); it is purified by recrystallization from acetonitrile. The mp is 123.5-124.5°C (decomp.). The IR spectrum is as follows: 3413 cm^{-1}, 3315 cm^{-1}, 3185 cm^{-1}, 3044 cm^{-1} (ν NH), 1606 cm^{-1}, 1570 cm^{-1} (thioamide-II), 1492 cm^{-1} (ν C=C), 1301 cm^{-1} (ν_{as} SO_2), 1139 cm^{-1} (ν_s SO_2), and 808 cm^{-1} (δ C–H_{arom}). Found, %: C 48.61, H 5.93, and N 10.41. $C_{11}H_{16}N_2O_2S_2$. Calculated, %: C 48.51, H 5.92, and N 10.29.

N-(α-Tosylbenzyl)thiourea (IIIb). The product (IIIb) is obtained analogously to compound (IIIa) from thiourea, benzaldehyde, and p-toluenesulfinic acid. The yield is 83.8%. The mp is 139.5-140.5°Ç (acetonitrile); the literature gives the mp 153-154°C (ethanol) [3]. The IR spectrum is as follows: 3347 cm^{-1} shoulder, 3270 cm^{-1}, 3168 cm^{-1} (ν NH), 1609 cm^{-1}, 1530 cm^{-1} (thioamide-II), 1493 cm^{-1} (ν C=C), 1285 cm^{-1} (ν_{as} SO_2), 1135 cm^{-1} (ν_s SO_2), 802 cm^{-1}, 736 cm^{-1}, and 697 cm^{-1} (δ C-H_{arom}).

4-Hydroxy-4-methyl-6-ethyl-5-ethoxycarbonylhexahydropyrimidine-2-thione (VIa). To the mixture of 0.049 g (2.06 mmole) of sodium hydride in 2 ml of dry acetonitrile at 0°C is added, dropwise with stirring in the course of 5 min, the solution of 0.273 g (2.10 mmole) of acetoacetic ester in 2 ml of acetonitrile. To the resulting suspension of sodioacetoacetic ester is added 0.539 g (1.98 mmole) of compound (IIIa). The reaction mass is stirred at room temperature for 48 h prior to the distillation of the solvent *in vacuo*. To the residue are added 3 ml of water, and the mixture is cooled to 0°C. The precipitated residue of the product is filtered off, washed with ice-cold water, and dried. Compound (VIa) is obtained with the yield of 0.340 g (69.7%); it has the mp 149-150°C (decomp., ethanol). The IR spectrum is as follows: 3244 cm^{-1}, 3208 cm^{-1} shoulder (ν NH, ν OH), 1732 cm^{-1} (ν C=O), 1586 cm^{-1}, 1532 cm^{-1} shoulder, 1516 cm^{-1} (thioamide-II), 1179 cm^{-1}, and 1135 cm^{-1}. The PMR spectrum (DMSO-D_6) is as follows: 8.56 ppm (1H, broad s, $N_{(3)}$–H), 8.07 ppm (1H, broad s, $J_{NH,6\text{-}Ha}$ = 0 Hz, $N_{(1)}$–H), 5.90 ppm (1H, s, OH), 4.15 ppm (1H, d q, J_{AB} = 10.8 Hz, H_A in OCH_2), 4.08 ppm (1H, d q, H_B in OCH_2), 3.75 ppm (1H, d t, J_{5a6a} = 12.0 Hz, $J_{6aH\alpha}$ + $J_{6\text{-}Ha\text{-}H\alpha'}$ = 8.3 Hz, 6-Ha), 2.40 ppm (1H, d, 5-H_a), 1.59-1.79 ppm (2H, m, CH_2 in 6-C_2H_5), 1.41 ppm (3H, s, 4-CH_3), 1.18 ppm (3H, t, J = 7.1 Hz, $CH_2\underline{CH_3}$), and 0.79 ppm (3H, t, J = 7.3 Hz, CH_3 in 6-C_2H_5). The UV spectrum, given as the λ_{max} (log ε), is as follows: 206 nm (4.05) and 248 nm (4.26). Found, %: C 48.57, H 7.47, N 11.30, and S 12.76. $C_{10}H_{18}N_2O_3S$. Calculated, %: C 48.76, H 7.37, N 11.37, and S 13.02.

4-Hydroxy-4-methyl-6-phenyl-5-ethoxycarbonylhexahydropyrimidine-2-thione (VIb). The product (VIb) is obtained analogously to compound (VIa) from the thiourea (IIIb) and sodioacetoacetic ester. The yield of the 88:12 mixture of the diastereomers of (VIb) is 82.4%. The mp is 212-213°C (decomp., ethanol). The IR spectrum is as follows: 3305 cm^{-1}, 3232 cm^{-1}, 3186 cm^{-1} (ν NH, ν OH), 3087 cm^{-1}, 3061 cm^{-1} (νC–H_{arom}), 1737 cm^{-1} (νC=O), 1604 cm^{-1}, 1496 cm^{-1} (νC=C), 1566 cm^{-1}, 1517 cm^{-1} (thioamide-II), 1235 cm^{-1}, 1178 cm^{-1}, 1116 cm^{-1}, 1082 cm^{-1}, 754 cm^{-1}, and 693 cm^{-1}. The PMR spectrum of the main diastereomer (DMSO-D_6) is as follows: 8.76 ppm (1H, broad s, $N_{(3)}$–H), 8.36 ppm (1H, broad s, $J_{NH,6\text{-}Ha}$ = 0 Hz, $N_{(1)}$–H), 7.20-7.50 ppm (5H, m, C_6H_5), 6.10 ppm (1H, s, OH), 4.77 ppm (1H, d, J_{5a6a} = 12.0 Hz, 6-H_a), 3.92 ppm (1H, d q, J_{AB} = 10.9 Hz, H_A in OCH_2), 3.84 ppm (1H, d q, H_B in OCH_2), 2.82 ppm (1H, d, 5-H_a), 1.46 ppm (3H, s, 4-CH_3), and 0.94 ppm (3H, t, J = 7.1 Hz, CH_3 in C_2H_5). The PMR spectrum of the minor diastereomer (DMSO-D_6)† is as follows: 8.65 ppm (1H, broad s, $N_{(3)}$–H), 8.46 ppm (1H, broad s, $J_{NH,6\text{-}Ha}$ = 0 Hz, $N_{(1)}$–H), 6.27 ppm (1H, s, OH), 4.94 ppm (1H, d, $J_{5\text{-}He,6\text{-}Ha}$ = 5.2 Hz, 6-H_a), 3.71 ppm (2H, q, OCH_2), 1.32 ppm (3H, s, 4-CH_3), and 0.79 ppm (3H, t, J = 7.2 Hz, $CH_2\underline{CH_3}$). The UV spectrum, given as λ_{max} (log ε), is as follows: 210 nm (4.36) and 250 nm (4.36). Found, %: C 57.14, H 6.32, N 9.43, and S 10.65. $C_{14}H_{18}N_2O_3S$. Calculated, %: C 57.12, H 6.16, N 9.52, and S 10.89.

†The signals of the 5-H_e protons and the protons of the aromatic ring are superimposed with the signals of the analogous protons of the second isomer.

4-Hydroxy-4,6-diphenyl-5-ethoxycarbonylhexahydropyrimidine-2-thione (VIc). The product (VIc) is obtained analogously to the compound (VIa) from the thiourea (IIIb) and sodiobenzoylacetic ester. The yield is 85.7%. The mp is 165-166°C (decomp., ethanol). The IR spectrum is as follows: 3379 cm^{-1}, 3249 cm^{-1}, 3170 cm^{-1} (ν NH, ν OH), 3072 cm^{-1}, 3062 cm^{-1}, 3042 cm^{-1} ($\nu C-H_{arom}$), 1733 cm^{-1} ($\nu C=O$), 1606 cm^{-1}, 1589 cm^{-1} ($\nu C=C$), 1564 cm^{-1}, 1500 cm^{-1} (thioamide-II), 1177 cm^{-1}, 1035 cm^{-1}, 751 cm^{-1}, and 694 cm^{-1}. The PMR spectrum (DMSO-D_6) is as follows: 8.71 ppm (1H, broad s, $N_{(3)}-H$), 8.63 ppm (1H, broad s, $J_{NH,6\text{-}Ha} = 0$ Hz, $N_{(1)}-H$), 7.25-7.55 ppm (10H, m, 4-C_6H_5, 6-C_6H_5), 6.83 ppm (1H, s, OH), 4.94 ppm (1H, d, $J_{5\text{-}Ha,6\text{-}Ha'} = 11.9$ Hz, 6-H_a), 3.50 ppm (1H, d q, $J_{AB} = 10.5$ Hz, H_A in OCH_2), 3.45 ppm (1H, d q, H_B in OCH_2), 3.02 ppm (1H, d, 5-H_a), and 0.51 ppm (3H, t, J = 7.1 Hz, $CH_2\underline{CH_3}$). The UV spectrum, given as λ_{max} (log ε), is as follows: 211 nm (4.42) and 251 nm (4.25). Found, %: C 64.14, H 5.49, N 7.72, and S 9.12. $C_{19}H_{20}N_2O_3S$. Calculated, %: C 64.02, H 5.66, N 7.86, and S 9.00.

6-Methyl-4-ethyl-5-ethoxycarbonyl-1,2,3,4-tetrahydropyrimidine-2-thione (VIIa). The solution of 0.103 g (0.42 mmole) of the hydroxyhexahydropyrimidine (VIa) and 0.008 g of $TsOH \cdot H_2O$ in 2 ml of abs. ethanol is boiled with stirring for 1 h. The solution is cooled to −5°C, and the residue is filtered off, washed with cold ethanol, and dried. Compound (VIIa) is obtained with the yield of 0.047 g (49.4%); it has the mp 150.5-151.5°C (ethanol). The IR spectrum is as follows: 3316 cm^{-1}, 3182 cm^{-1}, 3109 cm^{-1} ($\nu N-H$), 1656 cm^{-1} ($\nu C=O$), 1575 cm^{-1} (thioamide-II), 1276 cm^{-1}, 1188 cm^{-1}, and 1122 cm^{-1}. The PMR spectrum ($CDCl_3$) is as follows: 8.10 ppm (1H, broad s, $N_{(1)}$-H), 7.60 ppm (1H, broad s, $N_{(3)}-H$), 4.32 ppm (1H, ddd, $J_{4\text{-}H,NH} + J_{4\text{-}H,H\alpha} + J_{4\text{-}H,H\alpha'} = 14.7$ Hz, 4-H), 4.19 ppm (1H, d q, $J_{AB} = 11.0$ Hz, H_A in OCH_2), 4.16 ppm (1H, d q, H_B in OCH_2), 2.30 ppm (3H, s, 6-CH_3), 1.45-1.72 ppm (2H, m, CH_2 in 4-C_2H_5), 1.27 ppm (3H, t, J = 7.2 Hz, $CH_2\underline{CH_3}$), and 0.91 ppm (3H, t, J = 7.5 Hz, CH_3 in 4-C_2H_5). The UV spectrum, given as λ_{max} (log ε), is as follows: 207 nm (4.04) and 304 nm (4.22). Found, %: C 52.52, H 7.15, N 12.34, and S 13.90. $C_{10}H_{16}N_2O_2S$. Calculated, %: C 52.61, H 7.06, N 12.27, and S 14.04.

6-Methyl-4-phenyl-5-ethoxycarbonyl-1,2,3,4-tetrahydropyrimidine-2-thione (VIIb). The product (VIIb) is obtained analogously to compound (VIIa) from the hydroxyhexahydropyrimidine (VIb). The yield is 83.5%. The mp is 212-213°C (ethanol); the literature value for the mp is 207°C (ethanol) [11]. The IR spectrum is as follows: 3324 cm^{-1}, 3159 cm^{-1}, 3088 cm^{-1} ($\nu N-H$), 1667 cm^{-1} ($\nu C=O$), 1573 cm^{-1} (thioamide-II), 1281 cm^{-1}, 1198 cm^{-1}, 1178 cm^{-1}, 1117 cm^{-1}, 758 cm^{-1}, and 688 cm^{-1}. The PMR spectrum ($CDCl_3$) is as follows: 7.86 ppm (1H, broad s, $N_{(1)}-H$), 7.86 ppm (1H, broad s, $N_{(3)}-H$), 7.22-7.37 ppm (5H, m, C_6H_5), 5.38 ppm (1H, d, $J_{4\text{-}H,NH} = 3.2$ Hz, 4-H), 4.08 ppm (1H, d q, $J_{AB} = 10.8$ Hz, H_A in OCH_2), 4.05 ppm (1H, d q, H_B in OCH_2), 2.34 ppm (3H, s, 6-CH_3), and 1.14 ppm (3H, t, J = 7.2 Hz, $CH_2\underline{CH_3}$). The UV spectrum, given as λ_{max} (log ε), is as follows: 209 nm (4.31) and 308 nm (4.33). Found, %: C 60.94, H 6.12, N 10.01, and S 11.75. $C_{14}H_{16}N_2O_2S$. Calculated, %: C 60.85, H 5.84, N 10.14, and S 11.60.

4,6-Diphenyl-5-ethoxycarbonyl-1,2,3,4-tetrahydropyrimidine-2-thione (VIIc). The product (VIIc) is obtained analogously to compound (VIIa) from the hydroxyhexahydropyrimidine (VIc). The yield is 80.2%. The mp is 198.5-199.5°C (ethanol); the literature mp is 192°C (ethanol) [11]. The IR spectrum is as follows: 3148 cm^{-1}, 3102 cm^{-1} shoulder ($\nu N-H$), 1693 cm^{-1}, 1642 cm^{-1} (ν C=O), 1572 cm^{-1} (thioamide-II), 1492 cm^{-1} (ν C=C in C_6H_5), 1260 cm^{-1}, 1208 cm^{-1}, 1138 cm^{-1}, 1097 cm^{-1}, 769 cm^{-1}, and 690 cm^{-1}. The PMR spectrum ($CDCl_3$) is as follows: 7.78 ppm (1H, broad s, $N_{(1)}-H$), 7.26-7.47 ppm (11H, m, 4-C_6H_5, 6-C_6H_5, and $N_{(3)}-H$), 5.52 ppm (1H, d, $J_{4\text{-}H,NH} = 3.0$ Hz, 4-H), 3.85 ppm (1H, d q, $J_{AB} = 11.0$ Hz, H_A in OCH_2), 3.82 ppm (1H, d q, H_B in OCH_2), and 0.80 ppm (3H, t, J = 7.1 Hz, $CH_2\underline{CH_3}$). The UV spectrum, given as λ_{max} (log ε), is as follows: 210 nm (4.39), ~248 nm shoulder, and 313 nm (4.22). Found, %: C 67.57, H 5.31, N 8.09, and S 9.61. $C_{19}H_{18}N_2O_2S$. Calculated, %: C 67.43, H 5.36, N 8.28, and S 9.47.

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