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TWO DIRECTIONS OF CYCLIZATION OF α-DIAZO-β-DITHIOAMIDES. NEW REARRANGEMENTS OF I,2,3-TRIAZOLE-4-CARBOTHIAMIDES.

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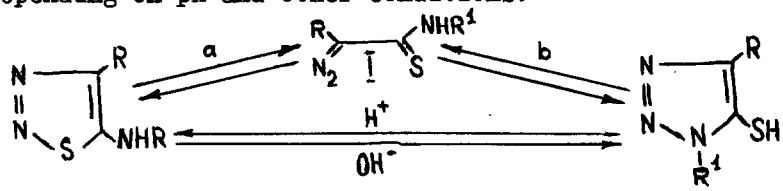
The cyclization processes of 2-diazomalondithioamides, generated by five different methods, have been studied. The ambient character of the derivatives of 2-diazomalondithioamide is the cause of previously unknown rearrangements of 5-mercapto-I,2,3-triazole- and 5-amino-I,2,3-thiadiazole-4-N-R-carbothioamides to 5-N-R-amino-I,2,3-thiadiazole-4-carbothioamides. NMR ^{1}H and ^{13}C spectra and mass-spectra are presented for the series of 5-amino-I,2,3-thiadiazole derivatives.

It is known that 5-amino-I,2,3-thiadiazoles in the presence of bases rearrange into 5-mercapto-I,2,3-triazoles [I]. The reverse reaction is described only for a few compounds and takes place in the hot acid solutions [2]. Both processes proceed via intermediate α-diazothioamides I which, unlike diazoamides, are very reactive compounds and cyclise to either 5-amino-I,2,3-thiadiazoles or the isomeric 5-mercapto-I,2,3-triazoles, depending on pH and other conditions.

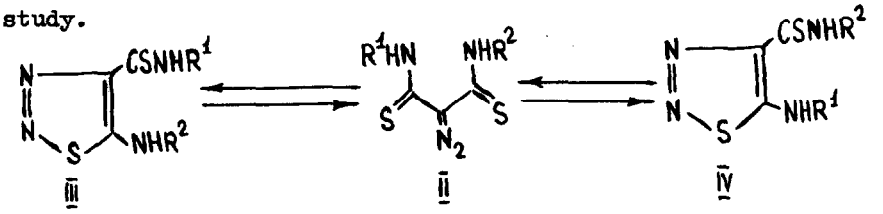


Several methods for the generation of diazo-compounds I are known:

I. Reaction of arylsulphonylazides with thioacetamide derivatives in the presence of bases (diazotransfer reaction [3]).
2. Reaction of α-diazonitriles with H_2S [4-7].
3. Reaction of chloral hydrasones with Na_2S [8].
4. Diazotization of 2-aminothioacetamide derivatives [9].

Nevertheless, we could not find in the literature any report on the synthesis or reactivity of diazo-compounds with two thioamide groups. It may be supposed that compounds of that type II with various substituents on nitrogen atoms of thioamide groups should cyclise in close to neutral media by one of the sulphur atoms. The influence of the nature of the substituents on the direction of such a cyclization is the object of

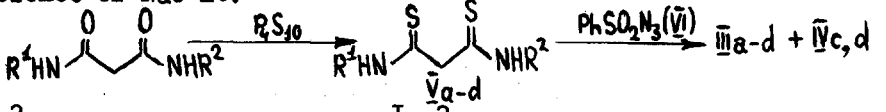
this study.



III II IV

Results and discussion

For generation of 2-diazomalondithioamides II we studied the reaction of malondithioamide derivatives Va-d with benzenesulphonyl azide VI in the presence of NaO-Et.

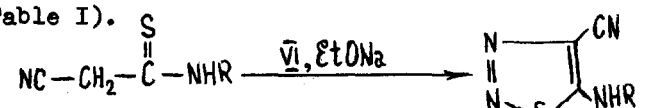


Va-d

R^I=H, R^2=H(a); CH_3 (b); Ph (c); R^I=R^2=Ph (d).

It was shown that this reaction takes place under mild conditions and represents a good method for obtaining 5-amino-I,2,3-thiadiazole-4-carbothioamides III and IV. The individual compounds IIIa,b,d appear in the case of thioamides Va,b,d. However, 2-thiocarbamoyl-N-phenylthioacetamide Vc transforms to the mixture of isomeric thiadiazoles IIIc and IVc. Both isomers have equal Rf values for all the TLC systems studied and in the case of HPLC. Mass spectra of these compounds provide abundant peaks of molecular ions. The dominant process of fragmentation is connected with the elimination of N_2 molecule (see Table I) which is characteristic for 5-amino-I,2,3-thiadiazoles [I0,II]. $[M-N_2]^{+\cdot}$ ions have thiiren structure, including one of the sulphur atoms depending on the nature of the substituents.

^{I}H and ^{I3}C NMR spectra of IIIc and IVc mixture provide double peaks for the NH-groups and for carbon atoms of the cycle and of the carbothioamide group respectively. To assign the signals we studied ^{I3}C NMR spectra of 5-amino-I,2,3-thiadiazole-4-carbonitriles VIIa-c obtained via diazotransfer reaction with cyanothioacetamide derivatives [I2] and of ethyl-5-amino-I,2,3-thiadiazole-4-carboxylate VIII, obtained by known methods [4,6,7] (see Table I).



VIIa-c

R=H (a), Me (b), Ph (c)

The signals in the weak field (I80-I87 ppm) were referred to the carbon atoms of thioamide groups on the basis of literature data [I3]. Two signals (I30.04 and I65.78 ppm) are characteristic for 5-amino-I,2,3-

thiadiazole VIIIa. They are referred to the carbon atoms C_4 and C_5 of the cycle, respectively, since in the high resolution spectrum of this compound the signal at 130.04 ppm is split into doublet of triplets ($^1J_{C_4-H}$= 189.0Hz, $^3J_{C_4-C_5-NH_2}$ = 4.27 Hz).

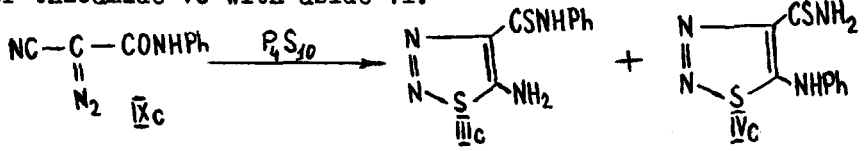
These data permitted to refer the signals in the spectra of compounds III and IV (160....165 ppm to C_5 and 130....135 ppm to C_4 of the cycle). ^{13}C NMR spectra support such a conclusion (see Table I, comp.VII,VIIIb).

Comparison of spectra of the compounds IIIa,b,d permits to make a conclusion, that substitution of hydrogen atom in aminogroup with methyl or phenyl groups shifts the signal of the corresponding carbon atom to the strong field for 3-4 ppm [12]. Therefore the signals at 168.4 and 184.1, 162.8 and 186.8 ppm are due to compounds IIIc and IVc respectively.

There are 4 signals of NH-protons of amino- and carbothioamide-groups in the PMR spectra of the mixture of compounds IIIc and IVc. Comparison of spectra of the compounds VIIa, VIII (free aminogroups), IIIa (unsubstituted amino- and carbothioamide groups), IIIb (free aminogroup and monosubstituted carbothioamide group), VIIb,c (monosubstituted aminogroup), IIId (monosubstituted amino- and thioamide groups) demonstrates a shift of NH signals into weak field with substitution of hydrogen atoms for alkyl or aryl groups. These facts permit us to refer the signals in the PMR spectra of compounds IIIc and IVc (see Table I). The ratio of isomers IIIc and IVc, defined from the integral curves in PMR spectra, is 1:3.

All the spectral parameters for compounds IIIa,b are equal to the analogous values for samples obtained previously by interaction of 2-diazo-2-cyanoacetamides IXa,b [14] and 5-amino-1,2,3-thiadiazol-4-carboxamides Xa,b [15] with P_4S_{10} [16] and Lawesson's reagent [17].

The reaction of diazo-compound IXc with P_4S_{10} leads to the mixture of isomeric thiadiazoles IIIc and IVc in the same ratio as in the case of reaction of thioamide Vc with azide VI.



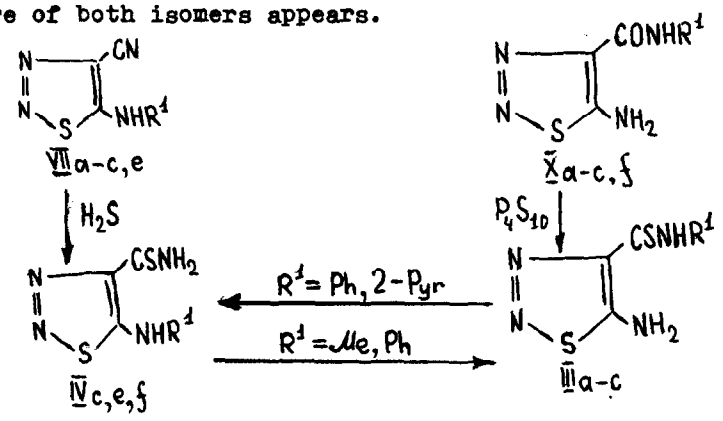
In order to obtain IVa-c compounds with evident structures, reactions of 5-amino-1,2,3-thiadiazole-4-carbonitriles VIIa-c with H_2S were carried out. These reactions take place in the presence of triethylamine at 0-5°C and finish in 30 minutes. Nitrile VIIa transforms into thioamide IIIa with high yields. However, 5-amino-1,2,3-thiadiazole-4-N-methylthiocarboxamide IIIb was obtained in the reaction of nitrile VIIb with H_2S

in the place of expected 5-methylamino-I,2,3-thiadiazole-4-carbothioamide IVb.

In the case of 5-phenylamino-I,2,3-thiadiazole-4-carbonitrile VIIc the rearrangement takes place only for the definite part of the molecules. As a result the mixture of thiadiazoles IIIc and IVc appear. 5-acetamide-I,2,3-thiadiazole-4-carbothioamide IVe, obtained via reaction of nitrile VIIe with H_2S, does not rearrange.

Since thiadiazole IVc rearranges to isomer IIIc only partly, one can suppose, that the reverse rearrangement should be probable in some conditions. In fact, reaction of 4-N-phenylcarboxamide Xc with P_4S_{10} in the boiling dioxane leads to the mixture of thiadiazoles IIIc and IVc in the same ratio, as in reactions of malondithioamide Vc with azide VI and nitrile VIIc with H_2S. 5-amino-I,2,3-thiadiazole-4-N-2'-pyridylcarbothioamide IIIf, obtained from amide Xf, rearranges totally to 5-2'-pyridylamino-I,2,3-thiadiazole-4-carbothioamide IVf.

Hence, these experiments permit us to find out the influence of substituents on the rearrangements III ⟶ IV and IV ⟶ III. If R^I is electrondonating group, compound III is obtained, if R^I is electronaccepting group, compound IV is obtained, if R^I is amphoteric (e.g.phenyl) group, the mixture of both isomers appears.



R^I=H (a), CH_3 (b), Ph (c), $COCH_3$ (e), 2-Pyr (f).

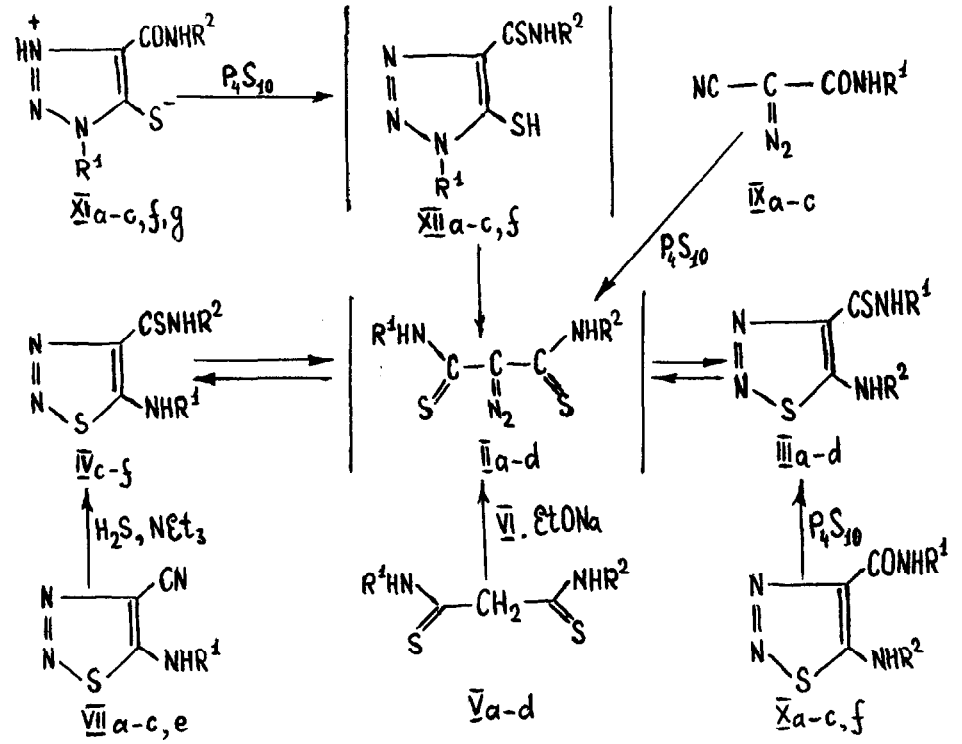
It is worthwhile mentioning that reaction of 5-mercapto-I,2,3-triazole-4-carboxamides XIa,c,f with P_4S_{10} leads not to triazoles XIIa,c,f, but to the products of their rearrangement, thiadiazoles IIIa,c and IVc, f, i.e. in this reaction the influence of substituents R on the composition of the products is absolutely analogous to the one mentioned above.

Since in all the transformations (diazo-transfer with malondithioamides V, thionisation of α-diazonitriles, I,2,3-thiadiazole-4-carbo-

nitriles, I,2,3-thiadiazole- and I,2,3-triazole-4-carboxamides) the ratio of products is the same, one can suppose, that all the processes mentioned proceed via 2-diazomalondithioamides II. The ratio of isomers depends not on the method of their formation, but on the nature of the substituents at nitrogen atoms of carbothioamide groups. The substitution for the methyl group, posessing electrondonating properties, gives the individual thiadiazole III. The electronaccepting groups (acetyl or pyridyl) provide exclusively thiadiazoles IV, whereas the phenyl group (an intermediate case) produces the mixture of thiadiazoles III and IV.

All the new rearrangements of 5-mercapto-I,2,3-triazole- and 5-amino-I,2,3-thiadiazole-4-N-R-carbothioamides to 5-N-Ramino-I,2,3-thiadiazole-4-carbothioamides described above are similar to the Dimroth-type rearrangements [I]. On the other hand they are different from the view point of reaction conditions, structures of products and quantities of atoms in the side chains participating in the isomerisations.

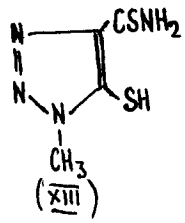
Scheme Methods of generation and directions of cyclisation of 2-diazomalondithioamides.

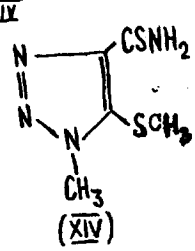


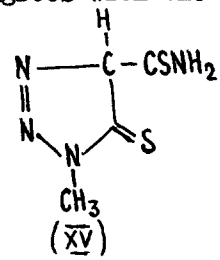
$R^1 = R^2 = H$ (a), Ph (d); $R^1 = H$, R^2 = Me (b), Ph (c), CO_2Me (e)

 (f); R^1 = Me; R^2 = H (g)

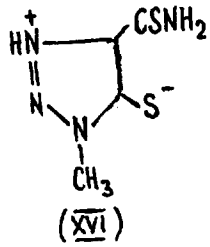
The traditional mercapto-form (XIII) has been excluded on the base of UV-spectral data: λ_{max}(lgε)234(2.95), 296(2.88) for XIg and 215 (4.18), 267(3.92), 335(4.16) for XIV. Structure XV disagrees with the PMR data (see Experimental).

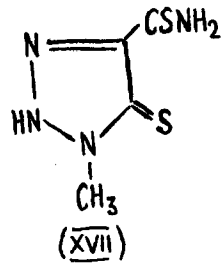

(XIII)


(XIV)


(XV)

The distinction between most probable zwitterionic structure XVI and alternative structure XVII has been done on the basis of earlier data for mercaptoimidazoles [18], which have been shown existing only in the zwitterionic forms.


(XVI)


(XVII)

Experimental

The melting points of all the substances I-XII were measured in open capillaries without correction. The IR spectra were obtained with Karl Zeiss UR-20 spectrometer in KBr tablets. PMR spectra were obtained with Perkin-Elmer SR-12B(60MHz) and Bruker WP-80(80.13MHz) instruments in CD_3OD and DMSO-d_6. ^{13}C NMR spectra were obtained with Bruker WP-80 (20.13MHz) spectrometer in DMSO-d_6 (internal standard - TMS). Mass spectra were obtained with Finnigan MAT-212 instrument, using the direct inlet system. Electron energy was 70eV, source temperature -180°C.

All the mixtures and substances obtained were analysed using TLC on Silufol-UW-254 and HPLC (Millichrom) on Siluforb-600.

The properties of the obtained compounds are listed in Tables I and 2.

I. Malondithioamides Va-d. 1M of P_4S_{10} was added to 1M of corresponding malondiamide in 1l of dry dioxane at 50°C with stirring. The mixture was boiled for 1,5 hours and then filtered. The liquid phase was vaporised in vacuo and the substance was recrystallised from water with activated carbon.

II. Reaction of malondithioamides Va-d with benzenesulphonylazide VI. 1mM of compound V was added to $NaOC_2H_5$ solution made from 0.1mM of Na

and Iml of ethanol. Then I.ImM of azide VI was added at 0-5°C. The precipitate formed was filtered off and recrystallised from water or ethanol.

III. The general procedure of thionisation of 2-diazo-2-cyanoacetamides IXa-c (A) [I4], 5-mercapto-I,2,3-triazole-4-carboxamides (B) [I5], 5-amino-I,2,3-thiadiazole-4-carboxamides (C) [5,I5].

0.5mM of P_4S_{10} was added to ImM of compounds IX, X or XI in I5ml of dry dioxane at 50°C with stirring. The mixture was boiled for I hour and then filtered off. The liquid phase was concentrated in vacuo. Then 0.5l of water was added. The precipitate formed was filtered off and recrystallised from water.

IV. 5-acetylamino-I,2,3-thiadiazole-4-carbonitrile VIIe.

I.72ml (I7.5mM) of acetylchloride and I.Iml (I7.5mM) of triethylamine was added to the suspension of Ig (6.0mM) of 5-amino-I,2,3-thiadiazole-4-carbonitrile [9] in 20ml of chloroform. The reaction has been carried out at 50°C during 5 hours. The liquid phase was concentrated in vacuo and the percipitate formed was recrystallised from water. The crystals are colourless.

V. 5-N-R-amino-I,2,3-thiadiazole-4-carbonitriles VIIb,c were obtained by method II from 2-cyanothioacetamides [I2].

VI. Sulphohydration of 5-amino-I,2,3-thiadiazole-4-carbonitriles VIIa-c,e

0.ImM of triethylamine (VIIa,b,e) or $NaOC_2H_5$ (VIIc) was added to a solution of ImM of nitrile VIIa-c,e in Iml of chloroform (VIIa-c) or ethanol (VIIe). The mixture was treated by H_2S at -5-0°C until saturation. The reaction of compounds VIIa,b takes place at 0°C and of compounds VIIc,e at heating up to 60-80°C for 4 hours. The precipitate formed was filtered off and recrystallised from water or ethanol.

VII. 5-mercapto-I,2,3-triazole-4-N-R-carboxamides XIc,f.

ImM of corresponding 5-amino-I,2,3-triazole-4-carboxamide Xc or Xf was suspended in 5ml of 25% solution of NH_3 in water and refluxed for dissolving of the precipitate. The reaction mixture was concentrated to I/3 of the previous volume. Acid was added to pH I. The precipitate formed was filtered off and recrystallised from water.

VIII. 5-amino-I,2,3-thiadiazole-4-N-(2'-pyridyl)-carboxamide Xf.

This compound was obtained using method II and 2-thiocarbamoyl-N-(2-pyridyl)acetamide XIIIf.

IX. 2-thiocarbamoyl-N-(2-pyridyl)acetamide XIIIf.

I5g (90mM) of 2-cyano-N-2'-pyridylacetamide was added to a solution of 2g (90mM) of Na in 300ml of ethanol. The mixture was saturated by H_2S at 0°C. Then the mixture was heated at 80°C for 2 hours. The precipitate formed after cooling was filtered off and recrystallised from ethanol.

Table I. The spectral characteristics of the obtained compounds.

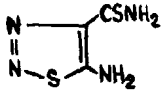
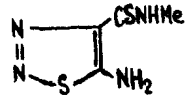
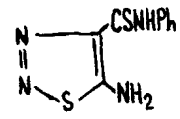
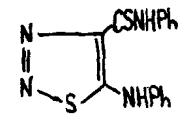
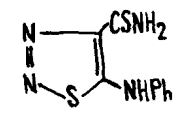
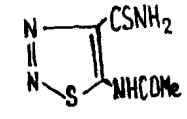
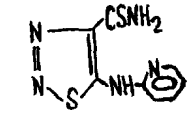
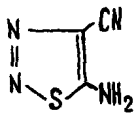
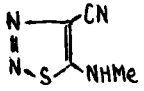
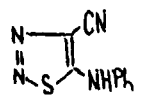
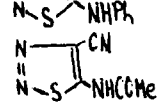
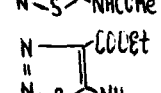
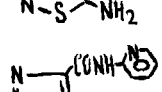
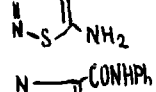
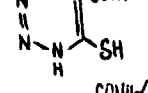
N comp.	Formulae	Mass-spectrum m/z(relative abundance)	PMR spectrum, δ, ppm	^{13}C NMR spectrum, δ, ppm[d]
IIIa		$M^{+\cdot}$ 160(100), 132(33), 105(33) 67(27), 60(34), 46(43)	9.1(2H, s., NH_2), 9.4(2H, s., $CSNH_2$)	136.7(C_4), 168.4(C_5), 187.3(C=S)
IIIb		$M^{+\cdot}$ 174(100), 146(15), 117(20), 105(24), 74(17), 67(18)	3.1(3H, doubl., J=5.5Hz, CH_3), 8.9(2H, s., NH_2), 10.4 (1H, quad., J=5.5Hz, NHMe)	31.1(1H, 3H, quad. of doubl., ^{2}J=11.23Hz, CH_3NH), 167.2 (C_5), 184.7(C=S), 136.9(C_4)
IIIc[a]		$M^{+\cdot}$ 236(100), 208(3), 175(31) 148(27), 105(33), 77(53)	7.1-7.9(5H, mult., Ph), 9.1(2H, s., NH_2), 11.8 (1H, s., CSNH)	137.5(C_4), 168.4(C_5), 184.1(C=S)
IIId		$M^{+\cdot}$ 312(100), 284(13), 219(31) 191(42), 148(52), 77(59)	7.2-7.8(10H, mult., 2Ph), 12.1(1H, s., NH), 12.6 (1H, s., NHCS)	139.6(C_4), 162.9(C_5), 183(C=S)
IVc[b]		$M^{+\cdot}$ 236(100), 208(3), 175(31), 148(27), 105(32), 77(53)	7.1-7.9(5H, mult., Ph), 9.9(2H, s., $CSNH_2$), 12.7 (1H, s., NH)	137.9(C_4), 162.8(C_5), 186.8(C=S)
IVe		$M^{+\cdot}$ 202(84), 174(63), 141(74), 132(100), 105(79), 60(48)	2.4(3H, s., Me), 10.2(2H, s., $CSNH_2$), 13.1(1H, s., NH)	22.7(3H, quad., Me), 141.0 (C_4), 153.9(C_5), 168.9 (C=O), 186.9(C=S)
IVf		$M^{+\cdot}$ 237(6), 209(20), 176(100), 105(26), 78(60), 60(4)	7.0-8.6(4H, mult., C_5H_4N), 10.0(2H, doubl., $CSNH_2$), 13.2(1H, s., NH)	172.7(C_4), 118.6(C_2), 138.9 (C_4), 139.2(C_3), 146.4(C_5), 148.1(C_1), 155.5(C_5), 187(CS)
VIIa			8.5(2H, s., NH_2)	112.9(CN), 115.8(C_4) 170.6(C_5)

Table I. The spectral characteristics of the obtained compounds.[e]

N comp.	Formulae	Mass-spectrum m/z(relative abundance)	PMR spectrum, δ, ppm	^{13}C NMR spectrum, δ, ppm
VIIb		$M^{+\cdot}$ 216(57), 188(100), 187(46) 173(44), 155(59), 91(41)	2.2(3H,s.,Me), 10.9 (1H,s.,NH)	23.8(Me), 112.5(CN), 118.8(C_4), 167.6(C_5)
VIIc		$M^{+\cdot}$ 202(40), 174(100), 146(26), 142(46), 77(51), 51(22)	7.3-7.5(5H,mult.,Ph), 11.1(1H,s.,NH)	
VIIe		$M^{+\cdot}$ 168(100), 140(84), 98(59), 71(60), 70(66)	2.4(3H,s.,Me), 8.4 (1H,s.,NH)	
VIII			1.3(3H,tripl.,J=5.5Hz, Me), 4.3(2H,quad.,J=5.5Hz, CH_2), 8.4(2H,s.,NH_2)	14.2(Me), 60.3(CH_2), 132.5(C_4), 162.5(C_5), 169.7(C=O)
Xf		$M^{+\cdot}$ 221(31), 193(28), 160(62), 121(50), 78(100), 51(29)	7.1-8.4(5H,mult.,Pyr, NHCO), 9.7(2H,s.,NH_2)	
XIc		$M^{+\cdot}$ 220(40), 128(16), 93(100), 92(12), 77(18), 45(8)	7.0-7.9(5H,mult.,Ph), 10.5(1H,s.,NH), 11.8 (1H,s.,NH)	
XIf		M 221(100), 128(26), 94(37), 78(63), 72(89), 43(74)	7.0-8.5(4H,mult.,C_5H_4N), 7.4(1H,s.,NH), 9.9(1H,s.,NH)	

a) In the mixture with IVc b) In the mixture with IIIc c) 6 most abundant ions are listed

d) The signals of phenyl carbon atoms are omitted e) All the substances are colorless needle-shaped crystals, darkening on the open air.

Table 2. Melting points, yields, IR spectra and elemental analysis data of the obtained compounds.

N comp.	$T_{melt.}$ °C	Obtained,%				Brutto-formulae	Calculated,%				IR spectrum (KBr), sm^{-1}	Yields,% method N
		C	H	N	S		C	H	N	S		
IIIa	171-172	22.7	2.6	34.9	39.8	$C_3H_4N_4S_2$	22.5	2.5	35.0	40.0	3330, 3190(NH)	30(II) 34(IIIC) 98(VI)
IIIb	138-140	27.9	3.3	32.0	36.3	$C_4H_6N_4S_2$	27.6	3.5	32.2	36.7	3300, 3200(NH)	65(II) 25(IIIC) 98(VI)
IIIc + IVc	120-121	46.1	3.5	23.5	27.2	$C_9H_8N_4S_2$	45.8	3.4	23.7	27.1	3318, 3170(NH) 2940(CH)	82(II) 35(IIIA) 80(IIIB) 82(IIIC) 96(VI)
IIId	189	57.8	3.5	17.9	20.7	$C_{15}H_{12}N_4S_2$	57.7	3.9	17.9	20.5	3295(NH) 2890(CH)	80(II)
IVe	156	30.1	3.1	27.9	32.1	$C_5H_6N_4OS_2$	29.7	3.0	27.7	31.7	3340, 3305, 3212(NH)	77(VI)
IVf	235	40.5	2.9	29.9	27.3	$C_8H_7N_5S_2$	40.5	3.0	29.5	27.0	3280, 3170(NH) 3000(CH)	35(IIIB) 34(IIIC)
Vb	128-130	33.0	5.6	19.1	43.4	$C_4H_8N_2S_2$	32.4	5.4	18.9	43.2	3335, 3330, 3230, 3165(NH) 2940(CH)	17
Ve	141-142	51.1	4.8	12.8	30.9	$C_9H_{10}N_2S_2$	51.4	4.8	13.3	30.5	3335, 3300(NH)	13
VIIb	138	34.6	2.9	40.5	23.0	$C_4H_4N_4S$	34.3	2.9	40.0	22.9	3240, 3130(NH) 3045(CH), 2248(CN)	64
VIIc	165-168	53.6	3.0	28.0	15.4	$C_9H_6N_4S$	53.5	3.0	27.7	15.8	3244, 3151(NH), 3035(CH), 2248(CN)	52
VIIe	125	36.1	2.4	32.8	19.3	$C_5H_4N_4OS$	35.7	2.4	33.3	19.1	3283(NH), 2252(CN), 1720(C=O)	77

Table 2. Melting points, yields, IR spectra and elemental analysis data of the obtained compounds.

N comp.	$T_{melt.}$ °C	Obtained,%				Brutto-formulae	Calculated,%				IR spectrum (KBr), sm^{-I}	Yields,%
		C	H	N	S		C	H	N	S		
Xf	68	42.7	3.2	3I.I	I3.9	$C_8H_7N_5OS$	43.4	3.2	3I.7	I4.5	3255,3I90(NH), I675(C=O)	90
XIc	I95-I96	49.4	3.8	25.3	I4.6	$C_9H_8N_4OS$	49.I	3.7	25.4	I4.6	3255,3I90(NH), I630(C=O)	89
XIf	285	43.I	3.5	3I.9	I4.7	$C_8H_7N_5OS$	43.4	3.2	3I.7	I4.5	3I30,3065(NH), 2985(CH), I650(C=O)	8I
XIIIf	I50-I5I	49.9	4.7	2I.3	I7.0	$C_8H_9N_5OS$	49.2	4.6	2I.5	I6.4	3300,3I00(NH), I680(C=O)	37

References

I. G.L'Abbē. J.Heterocycl.Chem. I984, 2I, 627.
2. Bakulev V.A., Mokrushin V.S. Khim.Geterotsikl.Soed. I986, I42I.
3. Regitz M., Sherer H. Chem.Ber. I969, I02, 4I7.
4. Bakulev V.A., Shafran Yu.M., Mokrushin V.S., Pushkareva Z.V. Khim. Geterotsikl.Soed. I982, I696.
5. Kolobov M.Yu., Bakulev V.A., Mokrushin V.S. Z.Org.Khim. I987, 23, II20.
6. Konstantinova N.V. Byulletem'Izobreteniy. I982, 33, I33, Pat.2990I62 (USSR).
7. Nakai M., Harada K., Mori Y. Pat.3I548 (Eur); C.A. I984, I00, 72730.
8. Kumkazu S. Pat.I03840 (Eur); C.A. I984, I00, 72730.
9. Shafran Yu.M., Bakulev V.A., Mokrushin V.S., Kondratiev P.N. Pat. I05574I (USSR). Byulleten'Izobreteniy. I983, 43, 79.
I0. Lebedev A.T., Shevchenko V.E., Kazaryan A.G., Bakulev V.A., Shafran Yu.M., Kolobov M.Yu., Petrosyan V.S. Khim.Geterotsikl.Soed. I987, 68I.
II. Lebedev A.T., Samguina T.Yu., Sharbatyan P.A., Bakulev V.A., Dankova E.F., Petrosyan V.S. Khim.Geterotsikl.Soed. I988, II8.
I2. Grabenko A.D., Pelkis P.S., Kulaeva L.N. Z.Ob.Khim. I962, 32, 2240.
I3. Levi G., Nelson G. Carbon-I3 Nuclear Magnetic Resonance for Organic Chemists. Wiley-Interscience I972.
I4. Shafran Yu.M., Bakulev V.A., Mokrushin V.S., Alekseev S.G. Khim.Geterotsikl.Soed. I984, I266.
I5. Shafran Yu.M., Bakulev V.A., Mokrushin V.S., Validuda G.I. Khim.Geterotsikl.Soed. I986, 69I.
I6. Dankova E.F., Bakulev V.A., Mokrushin V.S., Sidorov E.O., Lebedev A.T. Khim.Geterotsikl.Soed. I987, 845.
I7. Cava M.P., Levinson M.I. Tetrah.Lett. I985, 4I, 506I.
I8. Bakulev V.A., Mokrushin V.S., Grishakov A.N., Pushkareva Z.V. Khim. Geterotsikl.Soed. I982, 957.