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CONDENSED IMIDAZO-1,2,4-AZINES.

12.* SYNTHESIS AND STRUCTURE OF SUBSTITUTED 5-H-IMIDAZO[1,2-b]-1,2,4-TRIAZEPIN-4-ONES (-THIONES)

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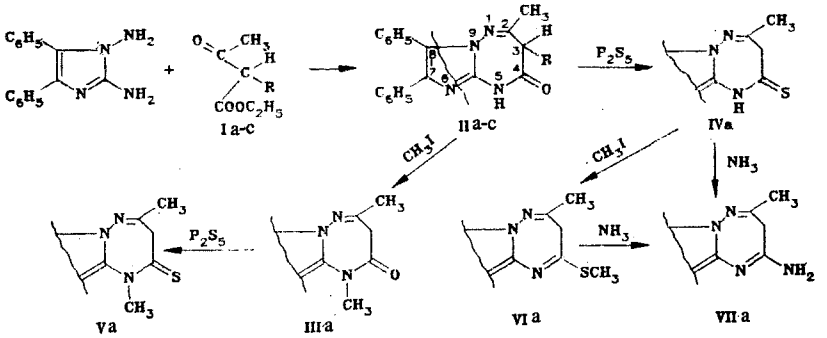
The synthesis has been effected of new representatives of the imidazo[1,2-b]-1,2,4-triazepin-4-ones (-thiones). It has been shown with the aid of IR and UV spectroscopic and mass spectrometric methods that the lactam (thione) form is the predominant one for the compounds synthesized.

In 1976, we performed the synthesis of the first representatives of a new heterocyclic system — 2,4-dimethyl-substituted imidazo[1,2-b]-1,2,4-triazepines[2]. Later, following the principle that we had proposed for forming an annelated triazepine ring with the use of a 1,2-diaminoimidazole, we studied the reaction of the latter with acetoacetic ester (AAE) [3], with fluorinated β-diketones [4], and with chalcones [5].

Continuing work in this direction, we have synthesized new 5H-imidazo[1,2-b]-1,2,4-triazepin-4-ones (IIa-c) and have studied their reactivities and we have also investigated the structure of the compounds obtained.

The imidazotriazepin-4-ones (IIa-c) were obtained by the method of Bruker et al. [3] by boiling 1,2-diamino-4,5-diphenylimidazole [6] with AAE or its ethyl or bromo derivative (Ia-c) in acetic acid in the presence of the sodium acetate.



I-VIIa R=H; b R = C_2H_5; c R = Br

When compound (IIa) was treated with methyliodide in methanol containing sodium methanolate, alkylation took place at the N(5) atom with the formation of the 2,5-dimethylimidazotriazepine (IIIa). This conclusion was based on the identity of the IR, UV, and mass spectra of compound (IIIa) and a sample with the authentic structure obtained by condensing 1-amino-2-methylamino-4,5-diphenylimidazole with AAE [3].

*For Communication 11, see [1].

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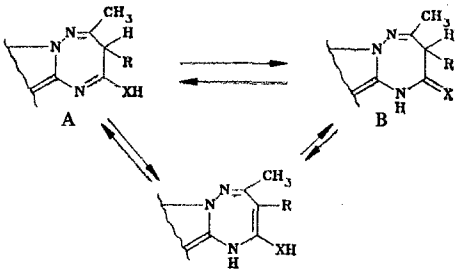
The imidazotriazepin-4-one (IIa), like its 5-methyl derivative (IIIa), readily took part in the thionylation reaction. Thus, the action of phosphorus pentasulfide in anhydrous pyridine on compounds (IIa) and (IIIa) gave their thio analogs (IVa) and (Va), respectively.

When the thione (IVa) was treated with methyl iodide under the conditions described above, alkylation did not take place in a manner analogous to that of imidazotriazepin-4-one (IIa), instead it took place at the sulfur atom, leading to the methylthio derivative (VIa).

Definite interest was presented by the investigation of the mobility of the mercapto and methylthio groups of compounds (IVa) and (VIa) in nucleophilic substitution reactions, since this opens up a route to the creation of imidazo-1,2,4-triazepines containing various substituents at the $C(_4)$ atom. In view of this, we studied the reaction of the mercapto derivatives (IVa) and (VIa) with ammonia. It was found that when compounds (IVa) and (VIa) were heated with aqueous ammonia in isopropanol 4-amino-2-methyl-7,8-diphenylimidazo[1,2-b]-1,2,4-triazepine (VIIa) was formed.

In view of the fact that compounds (IIa), (IVa), and (VIIa) are theoretically capable of prototropic tautomerism (forms A, B, and C), using the method of model compounds [7] we studied the tautomeric equilibrium for these substances present in various states of aggregation.



IIa R=H, X=O; IVa R=H, X=S; VIIa R=H, X=NH

A comparison of the electronic absorption spectra of compound (IIa) with (IIIa) and of (IVa) with (VIa) showed (Table 1) that the spectra of substances (IIIa) and (VIa) with a fixed tautomeric form were close to those of their analogs (IIa) and (IVa). Obviously, the possible lactam–lactim or thiol–thione tautomerism for forms A and B does not lead to a change in the length of the chromophoric chain while in the realization of forms C a bathochromic shift of the long-wave absorption band should appear in the UV spectrum because of the increase in the number of conjugated multiple bonds. The results obtained permit the conclusion that in solutions with different polarities the tautomeric form C is not realized for compounds (IIa-VIIa) (Table 1).

TABLE 1. Electronic Absorption Spectra of Substituted Triazepin-4-ones (thiones)

Compound	UV spectrum, λ_{max}, nm (log ϵ)		Compound	UV spectrum, λ_{max}, nm (log ϵ)	
	in propanol	in dioxane		in propanol	in dioxane
IIa	225 (sh) (4,30) 243 (4,34) 250 (sh.) (4,32) 287 (4,17)	230 (sh.) (4,29) 245 (4,39) 252 (sh.) (4,38) 294 (4,11)	IVa	242 (4,26) 330 (4,32)	246 (4,34) 255 (sh.) (4,33) 332
IIb	224 (sh.) (4,33) 246 (4,32) 253 (4,32) 286 (4,18)	230 (4,37) 246 (4,45) 253 (4,44) 296 (4,20)	Va	248 325 (4,27)	255 326 (4,24)
IIIa	224 (sh.) (4,18) 245 (4,24) 250 (sh.) (4,23) 286 (4,06)	230 (sh.) (4,30) 245 (4,40) 251 (4,39) 293 (4,13)	VIa	228 (sh.) (4,34) 248 (4,44) 312 (4,28)	233 (sh.) (4,29) 251 (4,40) 317 (4,23)
			VIIa	232 (sh) (4,30) 252 (4,36) 297 (4,21)	239 (sh.) (4,29) 258 (4,40) 306 (4,23)

TABLE 2. Lactim-Lactam (Thiol-Thione) Tautomerism for Compounds (IIa-Va)

Compound	IR spectrum							
	in $CHCl_3$			in CH_3CN			in KBr tablets	
	frequency*, cm^{-1}	ϵ, liter/mole·cm	proportion of the keto (thione) form, %	frequency,* cm^{-1}	ϵ, liter/mole·cm	proportion of the keto (thione) form, %	$D_{C=O(C=S)}/D_{710}$	proportion of the keto (thione) form, %
IIa	1720	648	100	1720	655±49	100	1,08†	100†
IIIa	1710	650	100	1710	648	100	1,06	100
IVa	1165	168±12	64	1165	191±13	70	0,77	87†
Va	1145	264±18	100	1145	271±19	100	0,93	100

*For compounds (IIa) and (IIIa) – $\nu_{C=O}$, for (IVa) and (Va) – $\nu_{C=S}$.
†Determined by the internal standard method in relation to the δ_{C-H} band of benzene at 710 cm^{-1}.

In order to estimate the position of the tautomerism for forms A and B we used the IR spectroscopic method. The assignment of the strong absorption bands at a frequency of 1715 ± 5 cm^{-1} to $\nu_{C=O}$ in compounds (IIa) and (IIIa) caused no doubts. Knowing the frequency of the $\nu_{C=O}$ vibrations it is possible to make an assignment of the $\nu_{C=S}$ band by using the ratio of the frequencies for lactams and thiones, $\nu_{C=O}/\nu_{C=S}$ = 1.49 [8]. In the IR spectra of compounds (IVa) and (Va), this ratio corresponds to $\nu_{C=S}$ absorption at 1145 cm^{-1}.

Table 2 gives figures to establish the prototropic equilibrium between forms A and B using compounds (IIa-Va) as examples. We have shown that only for the initial compound (IVa) is the tautomeric equilibrium shifted somewhat in the direction of the thiol form (from 13 to 30-36%) while for the other compounds the keto (thione) form is the dominating one, depending on the state of aggregation of the substances concerned (from crystal to solution).

The results of the study of tautomeric equilibrium well explain the behavior of substances (IIa) and (IVa) in reaction with methyl iodide.

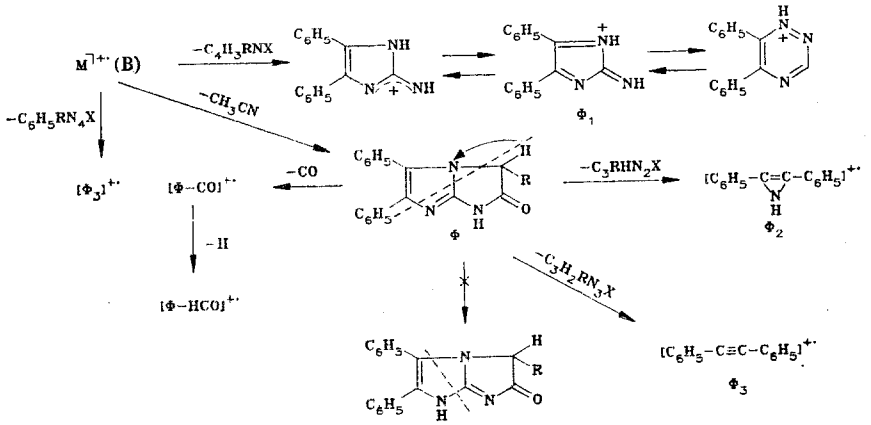
In addition to the study of the prototropic phenomena in the crystalline state and in solution, there is considerable interest in the behavior of the bicyclic compounds synthesized (IIa-c)-(VIIa) in the gas phase, free from the influence of external factors on the displacement of the tautomeric equilibrium. By using mass spectrometry (details of the mass spectra of compounds (IIa-c)-(VIIa) are given in Table 3) with ionization of the molecules by electron impact, we attempted to determine the laws of the fragmentation of the triazepine ring in compounds (IIa-c)-(VIIa) by comparing them with the known fragmentation of the bicyclic 1,4- and 1,5-benzodiazepin-2-ones [9, 10] and imidazo[1,2-b]-1,2,4-triazepin-5-ones [11].

The nature of the fragmentation of the triazepine ring differs substantially from the fragmentation processes established previously which were connected with the elimination of the methyl group in position 2, the ejection of an X particle, and the splitting out of a molecule of ketene (thioketene) or of a CH(R)CX particle from the molecular ion (M^+) [9-12] (Table 3). In this case, the splitting out of ketene (thioketene) or the particle CH(R)CX in the first stage of fragmentation was absent, which is evidence in favor of the existence of M^+ in form B, since the cleavage of a π-bond ($C_{(4)}$–$N_{(5)}$, in form A) and of a conjugated bond ($C_{(4)}$–$C_{(3)}$, in form C) is an energetically unfavorable process. The fact that the fragmentation of M^+ is due predominantly to one form was confirmed by the single fragmentation pattern for all the compounds (IIa-c)-(VIIa) (Table 3). Small differences were observed in the fragmentation pattern of compounds (IVa) and (VIa).

TABLE 3. Mass Spectra of Compounds (IIa-c)-(VIIa) (intensities in % of the maximum ion; peaks with intensities ⩾ 10% are given)

Compound	Mass spectrum, m/z (I/I_{max})														
	$[M+1]^+$	M^+	Φ	$[Φ-CX]^+$	$[Φ-HCX]^+$	$Φ_1$	$Φ_2$	$Φ_3$	$[C_{13}H_9]^+$	$[C_6H_5CNH]^+$	$[C_6H_5CN]^+$	$[C_6H_5]^+$	$[C_6H_4]^+$	$[CH_2CX]^+$	$[CH_3CN]^+$
IIa	317 (22)	316 (100)	275 (87)	247 (27)	246 (45)	234 (17)	193 (31)	178 (13)	165 (14)	104 (81)	102 (98)	77 (28)	76 (24)	42 (15)	41 (17)
IIb	345 (24)	344 (100)	303 (63)	275 (17)	274 (18)	234 (38)	193 (52)	178 (16)	165 (16)	104 (35)	103 (70)	77 (22)	76 (12)	42 (25)	41 (37)
IIc*	395 : 397 (8 : 8)	394 : 396 (34 : 34)	353 : 353 (16 : 17)	275 (10)	274 (51)	234 (18)	193 (19)	178 (10)	165 (20)	104 (62)	103 (100)	77 (50)	76 (53)	42 (23)	41 (18)
IIIa	331 (20)	330 (88)	289 (85)	261 (14)	260 (13)	248 (10)	193 (24)	178 (10)	165 (16)	104 (28)	103 (100)	77 (16)	76 (14)	42 (14)	41 (11)
IVa	333 (22)	333 (100)	291 (41)	247 —	246 —	234 (10)	193 (30)	178 (10)	165 (10)	104 (28)	103 (33)	77 (18)	76 (10)	58 (11)	41 (10)
Va	347 (18)	346 (92)	305 (88)	261 (10)	266 —	248 (18)	193 (55)	178 (14)	165 (18)	104 (28)	103 (100)	77 (32)	76 (21)	58 (23)	41 (13)
VIa	347 (23)	346 (100)	305 (42)	261 —	260 —	234 —	193 (18)	178 (14)	165 (16)	104 (34)	103 (51)	77 (35)	76 (11)	—	41 (20)
VIIa	316 (23)	315 (100)	274 (89)	247 (16)	246 (23)	234 (20)	193 (58)	178 (15)	165 (19)	104 (43)	103 (29)	77 (29)	76 (10)	41 (12)	41 (12)

*In the mass spectrum of compound (IIc) the m/z values for the following additional ions were recorded: $[M - Br]^+$ — m/z 315 (11) and $[(M - Br) - CH_3CN]^+$ — m/z 274 (28). The latter ion is equivalent to $[Φ - H]^+$.



The main fragmentary ion determining the whole course of fragmentation is the Φ ion arising from the elimination of a CH_3CN molecule from M^+. The process of cleaving the "weakest" $N(_1)$—$N(_9)$ and $C(_4)$—$N(_5)$ bonds in the triazepine ring is accompanied by the ejection of a C_4H_3RNX particle and the formation of the ion Φ_1, which is specific for form B. The subsequent fragmentation of ion Φ is connected with the loss of CX and CHX particles, and is also characteristic for cyclic ketones, thiones, and imines.

The recording of ions with m/z values of 76, 77, 103, 104, 165, 178, and 193 confirmed the structure of the imidazole moiety of the molecules of compounds (IIa-c)-(VIIa) (Table 3), and the general pattern of the fragmentation of the M^+ ions of these compounds is illustrated by the scheme (the sequence of elimination of the particles was determined by the DADI technique [13], while the empirical composition of M^+ and of the fragmentary ions Φ-Φ_3 was shown by high-resolution mass spectra for compounds (IVa)-(VIa), as examples).

The scheme given requires some refinements connected with the observed deviations in the directions of fragmentation for compounds (IVa) and (VIa) and in the determination of the mobilities of the hydrogen atoms in the $C(_3)$ and $N(_5)$ positions of the triazepine ring. The answer to the latter question will not only provide proof for the migration of hydrogen atoms but will also determine the possibility of tautomeric transformations in the gas phase.

A consideration of the mass spectra of the deuterated compounds (IIa), (Va), and (VIa) permitted the following facts to be established [14]:

1) the total percentage enrichment with deuterium at the $N(_5)H$ and $C(_3)$—HR (R = H) centers amounted to ~50%, and in the case of compound (IIa) the distribution was uniform (~25%);

2) on the formation of the ion Φ_1, the percentage retention of the label in compound (IIa) amounted to ~50%, while in compound (Va) it was only ~25%, which shows the participation of the hydrogen at the $C(_3)$ atom in the rearrangement; and

3) for the Φ_2 ion the percentage retention of the label in the case of compounds (IIa), (Va), and (VIa), likewise amounted to ~25%, which shows the migration of the hydrogen atom at $C(_3)$ into the $N(_9)$ position and excludes the possibility of the rearrangement of the ion Φ itself (see the fragmentation scheme).

In the mass spectrum of compound (VIa) with the fixed thiol form A, a process of elimination of an SCH_3 particle from the ion was observed (found — 258.1028; calculated — 258.1031 for the composition $C_{17}H_{12}N_3$). The intensity of the $[\Phi - SCH_3]^+$ ion amounted to 30% of the maximum peak of the ion in the mass spectrum of compound (VIa). The $[\Phi - SH]^+$ ion from compound (IVa) had an intensity of 23% of I_{max} and the same empirical composition (found, 258.1035). The mass spectra of both compounds lacked $[\Phi - CX]^+$ ions. According to calculation, the amount of form A in compound (IVa) amounted to ~70% [15].

For compound (Va), with the fixed thione form B, no $[\Phi - SH]^+$ ion was recorded, which shows the unreality of the existence of form C in the gas phase.

The investigation performed showed that compounds (IIa-c)-(VIIa) exist predominantly in form B, regardless of the state of aggregation. An exception is substance (IVa) in which the percentage of the thiol form (A) increased on passage from the crystal (~13%) to solution (~30-36%) to gas (~70%). The possibility of the existence of form C for compounds (IIa-c)-(VIIa) is excluded.

TABLE 4. Characteristics of the Imidazo[1,2-b]-1,2,4-triazepines ((IIa-c)-(VIIa))

Compound	mp, °C	R_f	Found, %			Empirical formula	Calculated, %			Yield, %
			C	H	N		C	H	N	
IIa	226—227	0,77	72,4	4,9	17,4	$C_{19}H_{16}N_4O$	72,1	5,0	17,7	57
IIb	212—214	0,80	74,2	6,3	15,3	$C_{23}H_{24}N_4O$	74,2	6,5	15,0	46
IIc	196—198	0,87	57,3	4,0	14,5	$C_{19}H_{15}BrN_4O$	57,7	3,8	14,2	40
IVa	208—209	0,92	68,5	4,7	16,6	$C_{19}H_{16}N_4S$	68,7	4,9	16,9	87
Va	173—175	0,90	69,6	5,4	16,0	$C_{20}H_{18}N_4S$	69,3	5,2	16,2	88
VIa	220—222	0,70	69,4	5,3	16,1	$C_{20}H_{18}N_4S$	69,3	5,2	16,2	41
VIIa	270—272	0,15	72,2	5,4	22,2	$C_{19}H_{17}N_5$	72,4	5,6	22,5	86

*The compounds were purified by crystallization: (IIa, c) and (VIa) from acetone; (IIb) from aqueous acetone; (IIIa) and (Va) from methanol; and (VIa) and (VIIa) from isopropanol.

EXPERIMENTAL

The IR spectral investigations were performed on a Hitachi ESP-3T spectrophotometer and on an UR-20 instrument (in cells where D = 10 cm and in KBr tablets). Mass spectra were obtained on a Varian MAT-311A instrument with direct introduction of the sample. The recording conditions were the standard ones [13, 16]. The temperature used to vaporize the samples in the high-energy electron source was 120-150°C. High-resolution mass spectra were obtained under the same conditions at M/ΔM = 15,000, the standard being PFK. The spectra of metastable ions were recorded in the second field-free space (DADI technique [13]). Deuteration was performed by adding an excess of CD_3OD to the sample under recording conditions for mass spectrometry.

The individuality of the compounds synthesized was checked by TLC on Silufol UV-254 plates in the butanol—toluene (1:2) system. The characteristics of the compounds synthesized are given in Table 4.

3-Substituted 5H-Imidazo[1,2-b]-1,2,4-triazepin-4-ones (IIa-c). A solution of 2.5 g (10 mmole) of 1,2-diamino-4,5-diphenylimidazole and 1.64 g (20 mmole) of sodium acetate in 20 ml of acetic acid was treated with 20 mmole of the appropriate ester (Ia-c). The mixtures were boiled for 10-12 h and cooled, and the precipitate that formed was filtered off and washed with ether, to give the corresponding compound (IIa-c).

2,5-Dimethyl-7,8-diphenylimidazo[1,2-b]-1,2,4-triazepin-4-one (IIIa). A solution of 3.2 g (10 mmole) of the imidazolotriazepin-4-one (IIa) in 30 ml of sodium methanolate, prepared from 30 ml of methanol and 0.69 g (30 mmole) of sodium, was treated with 4 g (30 mmole) of methyl iodide. The reaction mixture was boiled for 3 h and cooled, and the precipitate was filtered off and washed with cooled methanol.

Thionylation of the Imidazo[1,2-b]-1,2,4-triazepin-4-ones (IIa, IIIa). A solution of 2 mmole of one of the imidazotriazepin-4-ones (IIa) and (IIIa) in 10 ml of anhydrous pyridine was treated with 0.88 g (4 mmole) of P_2S_5. The mixture was boiled for 8 h and cooled, and the precipitate that had formed was filtered off, washed with cold water, and dried. Compounds (IVa) and (Va) were obtained.

2-Methyl-4-methylthio-7,8-diphenylimidazo[1,2-b]-1,2,4-triazepine (VIa) was synthesized from compound (IVa) in the same way as compound (IIIa).

4-Amino-2-methyl-7,8-diphenylimidazo[1,2-b]-1,2,4-triazepine (VIIa). A suspension of 3.3 g (10 mmole) of the imidazotriazepine-4-thione (IVa) or 3.5 g (10 mmole) of the methylthio derivative (VIa) in 100 ml of ethanol was treated with 15 ml of 25% ammonia. The mixture was boiled for 3 h and the resulting precipitate of compound (VIIa) was filtered off, washed with hot water, and dried.

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sym-TRIAZINE DERIVATIVES.

6.* CONVERSION OF 2,4,6-TRIETHOXYCARBONYL-1,3,5-TRIAZINE WITH ACYLHYDRAZINES INTO 3,5-DIETHOXYCARBONYL-1,2,4-TRIAZOLE

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UDC 491'792.6.07

The reaction of 2,4,6-triethoxycarbonyl-1,3,5-triazine with acylhydrazines leads to fragmentation with the formation of 3,5-diethoxycarbonyl-1,2,4-triazole, 1-acylethoxycarbonylformamidrazones N,N'-diacylhydrazines, and the amines of the corresponding carboxylic acids.

The interaction of 2,4,6-triethoxycarbonyl-1,3,5-triazine (I) with arylhydrazines (IIa-f), in contrast to the analogous reaction with ammonia, does not take place at the ester groups but at the C=N bonds of the heterocyclic system and is accompanied by a rearrangement with the formation of 2-methoxycarbonyl-4-arylhydrazino-5-oxoimidazoles (IIIa-f) [1, 2].

In order to evaluate the influence of the nature of the hydrazine component on the course of the reaction, we have used, in addition to the arylhydrazines, the acylhydrazines (IVa-c) in interaction with the ester (I). As in the case of the arylhydrazines [1], the process was performed at a ratio of the reactants (I) and (IV) of 1:4 by boiling for 2 h in ethanol, the course of the reaction being monitored by TLC.

In this case, together with the corresponding 5-oxoimidazole derivatives, from the products of the reaction of the ester (I) with all the acylhydrazines (IVa-c) we unexpectedly isolated, with yields of 60-75%, one and the same compound — the previously undescribed 3,5-diethoxycarbonyl-1,2,4-triazole (V). We also obtained the 1-acylethoxycarbonyl-formamidrazones (VIa-c) (yields 75-100%), the N,N'-diacylhydrazines (VIIa-c), and the amides of the

*For Communication 5, see [1].

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