QUATERNIZATION OF TETRAZOLES UNDER HIGH PRESSURE AND ESTABLISHMENT OF THE STRUCTURES OF QUATERNARY TETRAZOLIUM SALTS BY ^{13}C NMR SPECTROSCOPY

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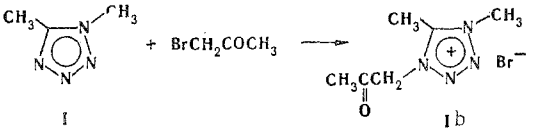
The quaternization of 1- and 2-acetonyl-5-R-tetrazoles, as well as dimethyltetrazole, was accomplished under high pressure conditions (5000-14,000 kgf/cm^2) by means of bromoacetone and methyl iodide and at atmospheric pressure and room temperature under the influence of methyl fluorosulfate. The structure of the salts were ascertained by ^{13}C NMR spectroscopy, and the principles of the change in the chemical shifts and the spin–spin coupling constants during quaternization and protonation of the tetrazoles were studied.

The quaternization of tetrazoles with alkyl halides and various alkylating agents has been quite adequately studied [1, 2]. The overwhelming majority of tetrazoles are alkylated exclusively in the 4 position; however, the formation of products of quaternization at the N_3 atom also is observed in some cases [3]. The structures of the salts were proved primarily by PMR spectroscopy, and ^{13}C NMR spectroscopy was used only in a few cases [4, 5].

Very little study has been devoted to the quaternization of tetrazoles with functionally substituted alkylating agents. Reagents in which the group undergoing replacement and the functional grouping are maximally close could be of greatest interest, since the effect of the substituent on the direction of quaternization should be manifested to a greater degree in this case. With this in mind, we studied the quaternization of tetrazoles with bromoacetone, having in view the possible use of the products for the synthesis of two-ring systems. The quaternization of weak bases such as tetrazoles ($pK_{BH^+} = -2$ to -3 [6]) is a slow process and takes place at high temperatures, and in a number of cases the reaction is reversible and is accompanied by side processes [3]. At the same time, it is known that the quaternization of nitrogen bases is markedly accelerated by pressure (the volume effect of activation $\Delta V^{\neq} \sim -30$ cm^3/mole), and we also took this into account in our experiments. The optimal conditions for reactions of this sort were studied in the case of the reaction of 1,5-dimethyltetrazole (I) in acetone. At atmospheric pressure and room temperature the yield of 1,5-dimethyl-4-acetonyltetrazolium bromide (Ib) is 37% after 18 days; however, the



I Ib

yield is 65% after 7 h at 50°C and a pressure of kgf/cm^2. Pronounced resinification is observed when the temperature is increased to 80°C (at a pressure of 5000 kgf/cm^2), and no more than 10% of salt Ib can be isolated after 6 h. The reaction proceeds rather slowly at room temperature even at 14,000 kgf/cm^2 — the yield of bromide Ib is 20% after 24 h.

Quaternary tetrazolium salts with a CH_2COR grouping can also be synthesized by direct quaternization of the corresponding tetrazolyl ketones with various alkylating agents. For this purpose, we obtained the previously undescribed ketones II and III by a known method [7], viz., by alkylation of 5-R-tetrazole anions with bromoacetone.

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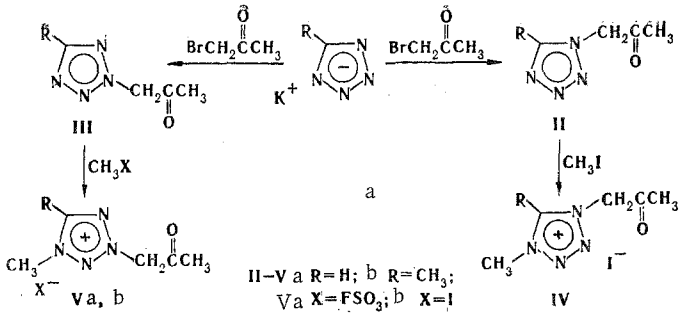
TABLE 1. Physicochemical Data and IR and PMR Spectra of Acetonyltetrazoles and Acetonyltetrazolium Salts

Compound	mp,[a] °C	IR (KBr), CO, cm^{-1}	PMR, ppm						Found, %				Empirical formula	Calculated, %				Yield, %
			solvent	CH_2	CH_3CO	$C-CH_3$	$N-CH_3$	CH	C	H	Hal	N		C	H	Hal	N	
Ib	157—158	1735	CD_3OD	6,0	2,42	2,92	4,37	—	31,0	5,0	34,9	23,8	$C_6H_{11}BrN_4O$	30,7	4,7	34,0	23,8	65
IIa	87—88	1738	$CDCl_3$	5,42	2,28	—	—	8,82	38,3	4,9	—	44,1	$C_4H_6N_4O$	38,1	4,8	—	44,4	23
IIb	98,5—99,5	1729	$CDCl_3$	5,23	2,25	2,36	—	—	43,0	5,9	—	40,0	$C_5H_8N_4O$	42,9	5,8	—	40,0	44
IIIa	55—57	1732	$CDCl_3$	5,60	2,17	—	—	8,62	38,3	4,8	—	44,4	$C_4H_6N_4O$	38,1	4,8	—	44,4	23
IIIb	69—70	1729	$CDCl_3$	5,42	2,12	2,40	—	—	43,0	5,9	—	39,8	$C_5H_8N_4O$	42,9	5,8	—	40,0	23,5
IVa	130	1740	CD_3OD	5,98	2,43	—	4,47	10,98	22,7	3,5	47,4	20,7	$C_5H_9IN_4O$	22,4	3,4	47,3	20,9	35
			CD_3CN	6,01	2,45	—	4,47	11,10										
IVb	162—163	—	—	—	—	—	—	—	—	—	—	—		—	—	—	—	43
Va	84—86 b	1745	CD_3CN	6,00	2,36	—	4,43	9,80	—	—	—	—	$C_5H_9FN_4O_4S$	—	—	—	—	92
Vb	144	1730	CD_3OD	—[c]	2,42	2,90	4,44	—	25,5	3,9	45,1	19,5	$C_6H_{11}IN_4O$	25,6	3,9	45,0	19,9	46
			CD_3CN	6,12	2,41	2,86	4,37	—										
VIb	266		D_2O	—	—	3,33	4,67	—	—	—	—	—	$C_4H_9IN_4$	—	—	—	—	50
VIIb	157—158	1735	D_2O	6,00	2,47	2,80	—	—	—	—	—	—	$C_7H_{13}BrN_4O_2$	—	—	—	—	36

[a]The compounds were crystallized: IIa, b from benzene, IIIb and VIb from water, and IVa, Vb, and VIIb from isopropyl alcohol. [b]From CH_3NO [sic], precipitated with CH_2Cl_2. [c]Not visible because of deuterium exchange.

TABLE 2. ^{13}C NMR Spectra (ppm) and $J_{^{13}C,^1H}$ Values (Hz) of Tetrazoles and Tetrazolium Salts

Compound	5-R	1-R	2-R	4-R	Solvent	C_5	5-CH_3	N—CH_3	N—CH_2	CH_3	CO
1-Methyl-tetrazole	H	CH_3	—	—	D_2O	145,1 (220)	—	35,1 (144)	—	—	—
I	CH_3	CH_3	—	—	$CDCl_3$	153,1	9,5	34,5	—	—	—
					D_2O	154,4	8,5	34,3	—	—	—
					H_2SO_4 (98%)	153,7	9,2	37,2	—	—	—
Ib	CH_3	CH_3	—	CH_2COCH_3	H_2SO_4 (98%)	154,8	10,0	38,8	59,7	28,9	200,6
IIa	H	CH_2COCH_3	—	—	$CDCl_3$	145,3	—	—	57,6	28,5	—
					CH_3NO_2	145,3 (220)	—	—	56,8 (143)	26,8 (129)	200,6 m
					H_2SO_4 (98%)	143,9 (238)	—	—	60,0 (148)	28,3 (131)	204,4 m
IIb	CH_3	CH_2COCH_3	—	—	$CDCl_3$	154,5	9,9	—	56,8	28,6	199,6
					CH_3NO_2	154,4	8,2 (132)	—	56,0 (142)	26,9 (129)	200,7
					H_2O	155,8	8,9 (132)	—	56,8 (143)	28,0 (129)	204,0
					H_2SO_4 (98%)	155,2	9,2	—	59,1	28,3	203,1
IIIa	H	—	CH_2COCH_3	—	$CDCl_3$	154,2 (216)	—	—	61,9 (144)	27,9 (130)	200,2 m
					CH_3NO_2	153,7 (215)	—	—	61,3 (144)	26,9 (130)	200,2 m
					H_2O	154,6 (218)	—	—	62,3 (144)	28,0 (129)	203,9
					H_2SO_4 (98%)	149,4 (234)	—	—	65,6 (147)	28,3 (130)	200,9
IIIb	CH_3	—	CH_2COCH_3	—	$CDCl_3$	164,5	12,1	—	62,0	28,4	200,0
					D_2O	165,0	10,8	—	62,0	27,8	204,4
					H_2SO_4 (98%)	161,2	10,5	—	65,1	28,5	203,6
IVa	H	CH_2COCH_3	—	CH_3	H_2O	145,3 (236)	—	41,1 (149)	61,8 (148 t, 3 q)	30,1 (130)	200,9 m
IVb	CH_3	CH_2COCH_3	—	CH_3	D_2O	154,7	10,2	38,7	59,7	28,8	201,1
Va	H	—	CH_2COCH_3	CH_3	H_2O	151,5 (237)	—	40,3 (148)	66,1 (149)	28,6 (130)	201,2
Vb	CH_3	—	CH_2COCH_3	CH_3	H_2O	161,7	12,1	39,9	66,6	30,0	200,8
VIb	CH_3	CH_3	—	CH_3	D_2O	153,6	10,7	38,7	—	—	—
VIIb	CH_3	CH_2COCH_3	—	CH_2COCH_3	H_2O	156,2	10,1 (136)	—	60,0 (148)	28,7 (130)	201,2

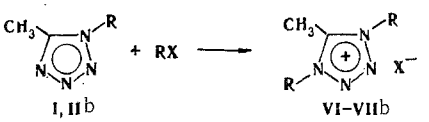


II–V a R=H; b R=CH$_3$; Va X=FSO$_3$; b X=I

Isomers II and III are easily separated owing to their different solubilities and the differences in their boiling points and were identified by means of their NMR spectra (see Tables 1 and 2). The ratios of the isomers increase in favor or isomer II on passing from tetrazole to 5-methyltetrazole, i.e., as the electron-donor properties of the substituent in the 5 position increase, as noted previously in [1, 8].

The quaternization of ketones II-III was accomplished by means of methyl iodide under a pressure of 5000-14,000 kgf/cm^2 in acetone at 40-50°C (see the experimental section). The IR spectrum of iodide IVb is similar to the spectrum of bromide Ib; this is explained by the absence of Br^- and I^- absorption at 800-4000 cm^{-1}. It is not difficult to note that both compounds can have identical positions of the substituents in the cation only in the case of alkylation of I and IIb in the 4 position. The identical nature of the cations of salts I and IVb was also established from the identical character of their ^{13}C NMR and PMR spectra. We were unable to quaternize 2-acetonyltetrazole (IIIa) with methyl iodide even at 14,000 kgf/cm^2. However, a powerful methylating agent such as FSO_2OCH_3 reacts readily at atmospheric pressure (at 20°C for 4 h) in nitromethane with tetrazole IIIa to give 1-methyl-3-acetonyltetrazolium fluorosulfonate (Va) in almost quantitative yield.

Reactions in which salts with two identical substituents attached to the nitrogen atoms are formed are convenient for establishing the site of quaternization in tetrazoles. Thus trimethyltetrazolium iodide (VIb), in which the two methyl groups are equivalent, according to the data from the PMR and ^{13}C NMR spectra (Tables 1 and 2), is obtained in the alkylation of 1,5-dimethyltetrazole (I) with methyl iodide.



I, IIb VI–VIIb

I, VI b R=CH$_3$; IIb, VII b R=CH$_2$COCH$_3$; VI b X=I; VII b X=Br

With this in mind, we carried out the quaternization of 1-acetonyl-5-methyltetrazole (IIb) with bromoacetone (at 14,000 kgf/cm^2) at 30°C. In the salt obtained (VIIb) both acetonyl groupings are also indistinguishable in the PMR and ^{13}C NMR spectra (Tables 1 and 2). In other words, quaternization took place in the 4 position in both cases.

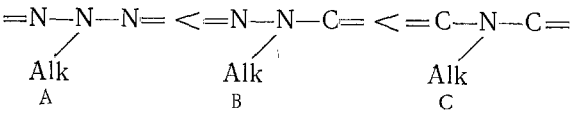
In principle, all of the synthesized ketones I-V and VIIb can exist in keto or enol forms. According to the data from PMR and ^{13}C NMR spectroscopy in various solvents (nitromethane, chloroform, methanol, water, acetonitrile, and sulfuric acid), only the keto form is present. The IR spectra of these compounds in KBr constitute evidence that even in the solid state they also exist primarily in the keto form (from the intense CO band at 1730-1745 cm^{-1} and the absence of bands of the enol form at 1600-1650 and 3500 cm^{-1}).

In the case of ketones Ib, IVa, Vb, and VIIb with a quaternized tetrazole ring the methylene protons are labile and in solutions in D_2O and CD_3OD undergo virtually complete deuterium exchange in a few hours, according to the PMR and ^{13}C NMR data. It is known that in 1,4-dialkyltetrazolium cations that are not substituted in the 5 position the ring C–H bond is capable of readily undergoing ionization, as a consequence of which the ring proton undergoes deuterium exchange in deuterium-donor media [9]. The 1-acetonyl-4-methyltetrazolium cation (IVa), in the PMR spectrum of which in CD_3OD the ring proton vanishes completely (at least after 1 h), has the same property.

All of the synthesized tetrazolium salts are stable in neutral and acidic aqueous solutions; however, they decompose rapidly even in weakly alkaline media (for example, in the

presence of Na_2CO_3). The bromides and iodides release HBr and HI in concentrated sulfuric acid; however, the structure of the cation remains unchanged (according to the ^{13}C NMR data).

The ^{13}C spectra of the tetrazoles (Table 2) that contain an acetonyl group illustrate the rule that shielding of the carbon atoms of the N–Alk groups increases for azole systems in the following order [10]:



The δCH_2 value for the 1-substituted isomer (fragment B) is substantially lower than for the 2-substituted isomer (fragment A). In addition, the $\delta^{13}C$ values for the C_5 atom are considerably different for these two isomers. The C_5 signal of the 2-substituted isomer is always found at weaker field [11-14].

The spin–spin coupling constant (SSCC) $^1J_{C_5,^1H}$, which is larger (by 4-5 Hz) in the spectra of the 1-substituted isomers and is subjected to the effect of the solvent to a smaller degree than δC, can be used to solve problems of isomerism in the tetrazole series. Since for tetrazole IIa in nitromethane $^1J_{C_5,^1H}$ = 220 Hz, whereas for tetrazole IIIa under the same conditions it decreases to 215 Hz. Let us note that the difference is also retained for protonated tetrazoles (in H_2SO_4),* although the SSCC values change in the case of protonation (238 Hz for the 1-substituted isomer and 234 Hz for the 2-substituted isomer). These differences level out in the case of quaternization of tetrazoles: 236 Hz for salt IVa, and 237 Hz for salt Va.

It has been previously shown that significant changes in the chemical shifts in both the PMR [17] and ^{13}C NMR [4] spectra are observed in the case of protonation of tetrazoles with acids (CF_3COOH, CH_3COOH). In [12], for example, it was established that protonation of 1- and 2-phenyltetrazoles always takes place at the N_4 atom; δC_5 (in H_2SO_4) decreases, while $^1J_{C_5,^1H}$ increases to virtually the same value for both isomers (234-235 Hz).

Different changes in δC_5 are observed in the case of protonation of acetonyltetrazoles II-III for different R values (Table 2). Thus when R = H (IIa and IIIa), δC_5 in H_2SO_4, as in the case of N-phenyltetrazoles, decreases for both isomers; when R = CH_3, δC_5 decreases for 2-isomer IIIb but increases for 1-isomer IIb. In addition, as we noted above, the increase in the $^1J_{C_5,^1H}$ SSCC values does not eliminate their difference for the two isomers. In the case of protonation δ5-CH_3 decreases, whereas δCH_2 in the acetonyl group increases in both isomers.

In contrast to protonation, the quaternization of aromatic heterocycles that contain two or three nitrogen atoms is, in the opinion of Fayet and co-workers [18], more reliable in ascertaining the effect of the positive charge in the ring on the ^{13}C NMR spectra. This is evidently also valid for compounds of the tetrazole series. We have found data only for two compounds of the tetrazole series, viz., 1-phenyl-4-ethyl- and 2-phenyl-4-ethyltetrazolium tetrafluoroborates, which are used as model samples in the investigation of the protonation of N-phenyltetrazoles [4]. The $\delta^{13}C$ values have also been determined for condensed aromatic systems that include a tetrazole fragment and their salts obtained by alkylation at the tetrazole nitrogen atom [5].

An increase in $^1J_{^{13}C,^1H}$ for all of the C atoms is observed in the case of quaternization: for C_5 and for the carbon atoms of the substituents that are bonded directly to the tetrazole ring atoms. The $^1J_{C_5,^1H}$ value undergoes the greatest increase: from 220 to 236 in the 1-isomer, and from 218 to 237 Hz in the 2-isomer (Table 2); from 216 to 234 Hz in

*Protonation of acetonyltetrazoles takes place at the nitrogen atom rather than at the carbonyl group. Evidence in favor of this is provided by the slight change in $\delta^{13}C$ of the carbonyl carbon atom in H_2SO_4 as compared with $CDCl_3$ ($\sim$3 ppm) and the virtually identical $\delta^{13}C$ values in H_2SO_4 and H_2O. In the case of protonation at the CO group one should expect a much greater shift of CO to weak field (by $\sim$40 ppm), as, for example, in the case of acetone [15]. In the case of protonation of acetonyltetrazoles $^1J_{^{13}C,^1H}$ for C_5 increases by approximately the same value as in tetrazoles that do not contain acetonyl groups (see below); in addition, it is known that the tetrazole ring [6] is approximately five orders more basic than the CO group of ketones [16].

1-phenyl- and from 211 to 235 Hz in 2-phenyltetrazoles [4]. The SSCC (^1J) for the N—CH_2, N—CH_3, and 5-CH_3 groups increase to a lesser extent: from 143 to 148, from 114 to 149, and from 132 to 136 Hz, respectively. The increase in all of the SSCC (^1J) in the case of quaternization indicates that the positive charge that develops in this case in tetrazoles is delocalized over the entire system, as in the case of azoles with a smaller number of nitrogen atoms [18].

One might have expected that the presence of a positive charge on passing from tetrazoles to their salts would lead to a characteristic change in δC_5. However, an analysis of the data in [4, 14] shows that this is not the case. For N-phenyltetrazoles [4] in the case of the 2-isomer the introduction of an ethyl group leads to a decrease in δC_5 (a shift of the signal to strong field), regardless of the solvent [85% H_2SO_4, $(CD_3)_2SO$]; at the same time, for the 1-isomer this decrease is observed only for a solution of the salt in 85% H_2SO_4, whereas the same δC_5 value that obtains for the starting tetrazole (in $CDCl_3$) is observed in $(CD_3)_2CO$. For condensed aromatic systems such as, for example, for thiazolo-[3,2]tetrazole [14] one observes the strong-field shift of the carbon atom this is customary for tetrazole and thiazole rings (3.8-4.9 ppm in the case of alkylation at the N_1 atom). The data in Table 2 confirm that a regular change in δC_5 also is not observed in acetonyltetrazoles in the case of quaternization and protonation.

More characteristic changes in the chemical shifts during quaternization are observed for the signals of the carbon atoms of the 5-CH_3, N—CH_3, and N—CH_2 substituents. They undergo deshielding in the spectra recorded in identical solvents (for example, in water).

Thus the observed increase in $\delta^{13}C$ for substituents R^1, R^2, R^4, and R^5 can be used for identification purposes, just as can the $^1J_{^{13}C,^1H}$ values. This is true when the same solvent is used for the tetrazole and its salt (water in the present research) and also if solutions of the tetrazole in some other solvents ($CDCl_3$, CH_3NO_2) were used.

It follows from the data presented above that there is virtually no change in δC_5 (as compared with the starting tetrazoles) in the case of quaternization of the 1- and 2-isomers in the 4 position, whereas in the case of quaternization of the 1-isomer in the 3 position one observes a weak-field shift of 5-7 ppm for δC_5 (quaternization of the 1-isomer in the 3 position is equivalent to quaternization of the 2-isomer in the 4 position). This makes it possible to determine the site of alkylation for the 1-isomer in the case of different substituents attached to the nitrogen atoms.

EXPERIMENTAL

The ^{13}C NMR spectra were recorded with a Bruker WP-60 spectrometer under pulse conditions with Fourier transformation. The ^{13}C chemical shifts (±0.1 ppm) were measured relative to external hexamethyldisiloxane (HMDS) [δ $HMDS_{ext}$ = 2.0 ppm for tetramethylsilane (external)]. The PMR spectra were recorded with a Varian DA-60-JL spectrometer (60 MHz), while the IR spectra were recorded with a UR-20 spectrometer.

1,5-Dimethyl-4-acetonyltetrazolium Bromide (Ib). A solution of 13.7 g (0.14 mole) of 1,5-dimethyltetrazole [19] and 12.6 g (0.15 mole) of bromoacetone in 30 ml of acetone was heated at 50°C under a pressure of 5000 kgf/cm² for 7 h, after which it was cooled, and the precipitated crystals of Ib were removed by filtration, washed with acetone and ether, and air dried.

Alkylation of Tetrazole and 5-Methyltetrazole with Bromoacetone. A 0.1-mole sample of the tetrazole was added to a solution of 0.1 mole of KOH in 20 ml of water, and after the solid had dissolved, the solution was mixed with a solution of 0.1 mole of bromoacetone in 20 ml of acetone. The reaction mixture was stirred for 1 h until a homogeneous solution formed, and the solution was allowed to stand overnight. It was then evaporated to dryness, and the residue was diluted with 50 ml of acetone. The potassium bromide was removed by filtration, the acetone was removed from the filtrate by distillation, and the residue was extracted with three 100-ml portions of hot benzene with vigorous stirring. The insoluble resin was extracted with hot chloroform, the solvent was evaporated, and the residue was extracted with hot benzene. The crystals of 1-isomer II that separated from the benzene solutions when they were cooled were removed by filtration and air dried. The mother liquor was evaporated, and the residue was distilled in vacuo to give 2-isomer IIIa, with bp 89°C (1 mm), and IIIb with bp 90°C (1 mm). The still residue was extracted with hot benzene to give an additional small amount of the 1-isomer.

Quaternization of Acetonyltetrazoles II and IIIb with Methyl Iodide under Pressure. A solution of 8 mmole of acetonyltetrazole and 10 mmole of methyl iodide in 3 ml of acetone was heated at 50°C under pressure (6000 kgf/cm^2 for IIb and 14,000 kgf/cm^2 for IIa and IIIb) for 5.5 h, after which it was cooled and diluted with 10 ml of ether. The precipitated crystals of IVa and Vb (or the oil in the case of IVb) were recrystallized from isopropyl alcohol, washed with ether, and air dried.

1-Methyl-3-acetonyltetrazolium Fluorosulfonate (Va). A 0.5-ml sample of methyl fluorosulfonate was added to a solution of 0.5 g (4 mmole) of 2-acetonyltetrazole (IIIa) in 2 ml of absolute nitromethane, and the reaction mixture was allowed to heat up spontaneously (to 45-50°C) at room temperature for 4 h. It was then diluted with absolute CH_2Cl_2, and the precipitated Va was removed by filtration, washed with absolute CH_2Cl_2, and dried in vacuo for 10 min to give white hygroscopic needles.

1,4,5-Trimethyltetrazolium Iodide (VIb). A solution of 0.6 g (6.1 mmole) of 1,5-dimethyltetrazole (I) and 1 ml (16 mmole) of methyl iodide in 1 ml of acetone was allowed to stand for 9 days at room temperature. The precipitated long white needles of VIb were removed by filtration, washed with ether, and air dried.

1,4-Diacetonyl-5-methyltetrazolium Bromide (VIIb). A solution of 0.7 g (5 mmole) of 1-acetonyl-5-methyltetrazole (IIb) and 0.46 ml (5.5 mmole) of bromoacetone in 2.5 ml of acetone was heated at 30°C under a pressure of 14,000 kgf/cm^2 for 5.5 h, after which it was cooled, and the precipitated crystals of VIIb were removed by filtration, washed with acetone and ether, and air dried. The mother liquor was evaporated, and the residue was recrystallized from isopropyl alcohol to give 0.14 g (20%) of starting IIb. The yield of VIIb was somewhat higher (39%) at 40°C; however, its isolation was less convenient, and more side products were formed.

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