acetate (9:1). As a result, we obtained successively, 0.18 g (30%) of starting XII with R_f 0.49 (23%) (identified from a mixed melting-point determination) and 0.09 g (23%) of XIII as light-yellow crystals with mp 118-119°C (from ether). The product was quite soluble in ordinary organic solvents. IR spectrum: 1725 ($COOC_2H_5$) and 3380 cm^{-1} (NH_2). PMR spectrum: 1.28 (6H, t, CH_3—CH_2), 2.43 (3H, s, CH_3), 4.27 (4H, q, CH_2—CH_3), 4.72 (1H, s, CH), and 5.82 ppm (broad NH signal). Mass spectrum: 268 ($M^{+\cdot}$), 223 [$(M-OC_2H_5)^+$], 196 [$(M-CO_2-C_2H_4)^{+\cdot}$], 179 [$(M-OC_2H_5-OC_2H_4)^+$], 151 [$(M-CO_2-C_2H_4-OC_2H_5)^+$], 150 [$(M-CO_2-C_2H_4-C_2H_5OH)^{+\cdot}$], 124 [$(M-2CO_2-2C_2H_4)^{+\cdot}$]. Found: C 49.3; H 6.3; N 21.0%. $C_{11}H_{16}N_4O_4$. Calculated: C 49.3; H 6.0; N 20.9%.

LITERATURE CITED

1. G. M. Vakhatova and L. N. Yakhontov, Khim. Geterotsikl. Soedin., No. 2, 264 (1981).
2. G. M. Vakhatova, O. S. Anisimova, and L. N. Yakhontov, Khim. Geterotsikl. Soedin., No. 11, 1557 (1979).
3. H. R. Reimschnessel and N. T. McDevitt, J. Am. Chem. Soc., 82, 3756 (1960).
4. A. Reissert and A. More, Ber., 39, 3300 (1906).
5. J. V. Backes, R. W. West, and M. A. Whitely, J. Chem. Soc., 119, 370 (1921).

SYNTHESIS OF POLYNUCLEAR UNCONDENSED TRI- AND TETRAZOLES

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A number of the corresponding 1- and 2-propargylazoles were obtained by propargylation of 5-substituted tetrazoles and 1,2,3-triazoles with various degrees of substitution. Polyazole structures with a system of two to five uncondensed azole rings were synthesized by the reaction of the 1- and 2-propargylazoles with organic azides, diazides, and azoles, as well as by oxidative dimerization.

It is known that uncondensed polynitrogenous heterocyclic compounds have pesticidal activity [1-3]. In our search for new substances in the polyazole series that regulate plant growth we accomplished the synthesis of polynuclear uncondensed azoles with two or more heterocycles connected by methylene links. The cycloaddition of organic azides to the triple bond of propargylazoles may serve as one of the variants of this synthesis.

N-Substituted propargyltetrazoles (III, IV) and 1,2,3-triazoles (V, VI) are obtained rather smoothly and in high yields as a result of the direct action of propargyl bromide on tetrazoles (I) and 1,2,3-triazoles (II) in the presence of triethylamine or potassium hydroxide; in this case one generally observes the formation of a mixture of two isomeric propargylazoles (IIIa, b and VIa, b), the IR spectra of which contain bands of vibrations of a terminal acetylenic bond with frequencies of 2100-2140 and 3300-3320 cm^{-1}. The use of sodium ethoxide as a catalyst in the case of the reaction with 1,2,3-triazole promotes a secondary reaction of nucleophilic addition of alcohol to the triple bond and the formation of 1-(2-ethoxy-2-propen-1-yl)-1,2,3-triazole (VIIa). It is possible that an acetylene-allene rearrangement of the propargyl substituent of triazole V precedes the addition reaction. The same VIIa was obtained by treatment of 1-propargyltriazole Va with sodium ethoxide in ethanol. The IR spectrum of ethoxypropenyltriazole VIIa is characterized by the presence of a band of the vibration of a terminal vinyl group (1645-1660 cm^{-1}) and overlapped bands of the vibrations of the multiple bonds of the triazole ring and the ether bond (910, 1076, 1180, 1470, and 1600 cm^{-1}).

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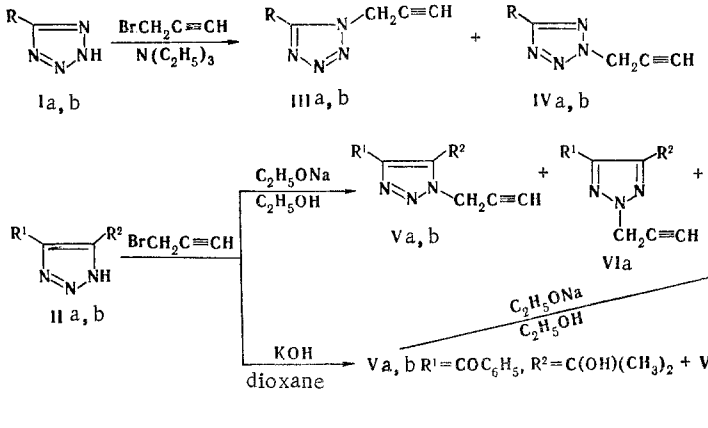
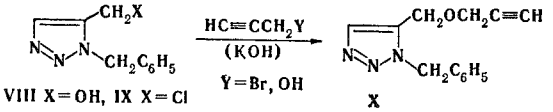


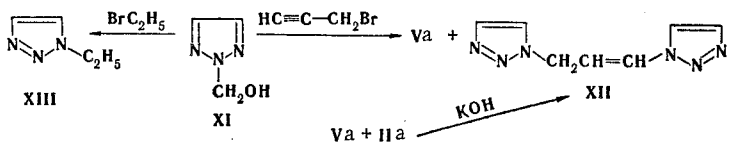
I a R=Ph; b R=$(CH_2)_2Cl$; II a $R^1=R^2=H$; b $R^1=COPh$, $R^2=C(OH)(CH_3)_2$; III a R=Ph; b R=$(CH_2)_2Cl$; IV a R=Ph; b R=$(CH_2)_2Cl$; V a $R^1=R^2=H$; b $R^1=COPh$, $R^2=C(OH)(CH_3)_2$; VI a $R^1=R^2=H$; b $R^1=COPh$, $R^2=C(OH)(CH_3)_2$; VII a $R^1=R^2=H$; b $R^1=COPh$, $R^2=C(OH)(CH_3)_2$

As expected, two isomers are formed in all cases of propargylation of 1,2,3-triazoles IIa, b (with potassium hydroxide, potassium carbonate, and sodium alcoholate as the catalyst); the principal isomer is the 1-substituted isomer (Va, b). The PMR spectrum of Va contains signals of protons of the triazole ring (8.05 and 7.71 ppm) and the propargyl grouping (5.36 and 2.10 ppm) (Table 1). Isomer VIa is interpreted unambiguously by the PMR spectrum, in which equivalent protons of the triazole ring (7.79 ppm) and methylene (5.30 ppm) and acetylenic (2.03 ppm) groupings are recorded.

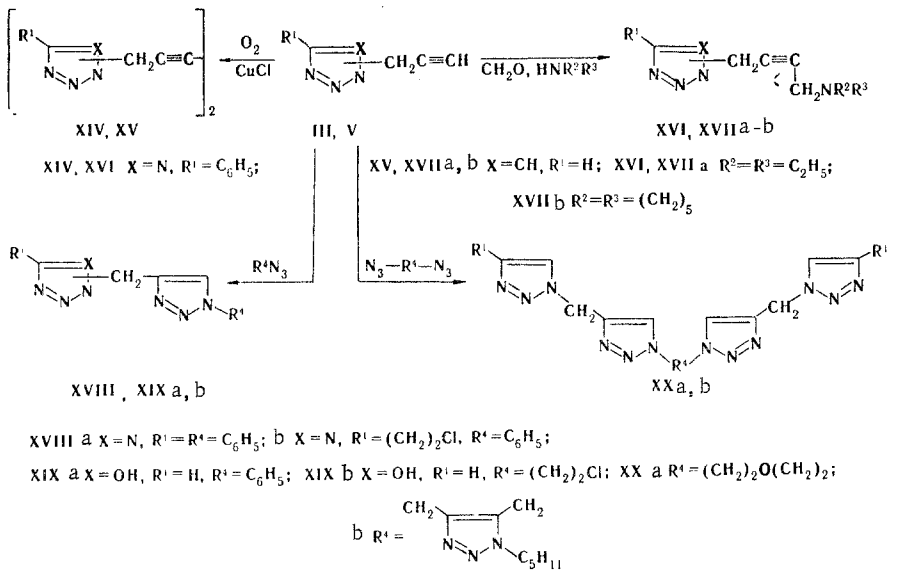
Propargyl ether X, the spectral characteristics of which are similar to those presented for propargyltriazoles V and VI, was obtained by the reaction of 1-benzyl-5(4)-hydroxymethyl-1,2,3,4-triazole (VIII) with propargyl bromide, as well as by reaction of 1-benzyl-5(4)-chloromethyl-1,2,3-triazole (IX) with propargyl alcohol in the presence of potassium hydroxide (Table 1).



In the propargylation of 2-hydroxymethyl-1,2,3-triazole (XI) in the presence of potassium hydroxide, instead of the expected ether we obtained two substances, viz., 1-propargyl-1,2,3-triazole (Va) and 1,3-bis(1,2,3-triazol-1-yl)prop-2-ene (XII). Elimination of the hydroxymethyl grouping probably occurs under the influence of potassium hydroxide under the reaction conditions; the 1,2,3-triazole liberated in the process is propargylated via the usual scheme with the formation of 1-propargyl-1,2,3-triazole. Ditriazolylpropene (XII) is the product of nucleophilic addition of 1,2,3-triazole to the multiple bond of the propargyl residue of Va. The fact of the elimination of a methylol grouping under alkaline conditions is confirmed by the reaction of 1-hydroxymethyl-1,2,3-triazole with ethyl bromide, as a result of which, 1-ethyl-1,2,3-triazole (XIII) was obtained. The PMR spectrum of ethyltriazole XIII contains signals of two nonequivalent protons of the triazole ring (7.42 and 7.53) and of methylene and methyl groups (4.35 and 1.45 ppm).



The reactions that are characteristic for a terminal acetylenic grouping are also peculiar to propargylazoles III-VI. They undergo smooth Mannich aminomethylation, undergo oxidative dehydrodimerization, and quite readily add alkyl mono- and diazides to give polyazole structures with a system of uncondensed triazole rings separated by a methylene grouping (XVIII-XX) (Table 2).



The IR spectra of XIV-XX contain absorption bands that are characteristic for azole rings (910, 980, 1090, 1180, 1470, 1600 cm^{-1}). The PMR spectra of polyazoles XVIII-XX contain signals of protons of CH_2 groups at 5.5-6.2 ppm and of protons of a triazole ring at 7.5-8.5 ppm.

EXPERIMENTAL

The IR spectra were recorded with a UR-20 spectrometer. The PMR spectra of solutions of the compounds in $CDCl_3$ and acetone were recorded with a Varian HA-100 spectrometer with hexamethyldisiloxane as the internal standard. 5-(β-Chloroethyl)-5-phenyltetrazole (Ia, b) and 4-benzoyl-5-hydroxymethyl-1,2,3-triazole (IIb) were obtained by the methods in [4-6]. The reaction products were chromatographed with a column filled with aluminum oxide (activity II) by elution with ether–petroleum ether (3:1).

1,2,3-Triazole. Oxidation of benzotriazole. A hot aqueous solution of 11.9 g (0.1 mole) of benzotriazole was added dropwise to a solution of chromic acid prepared from 50 g (0.5 mole) of chromic anhydride, 80 ml of sulfuric acid, and 100 ml of water, and the mixture was refluxed for 7 h. It was then allowed to stand overnight, and the precipitated crystals of the complex of triazolylcarboxylic acid with benzotriazole were removed by filtration to give 8.8 g (64%) of a product with mp 256-260°C (dec.). Found: C 42.7; H 2.8; N 29.8%. $C_{10}H_8N_6O_4$. Calculated: C 43.4; H 2.8; N 30.2%. Decarboxylation of the complex. A 278-g (1 mole) sample of the complex in 700 ml of benzene was placed in a 2-liter autoclave, and the mixture was heated at 195-200°C for 2 h. The pressure in the apparatus was raised to 40-50 atm. After cooling, the reaction mixture was taken out of the autoclave, the solvent was removed by distillation, and the residue was distilled *in vacuo* to give 64.8 g (94%) of 1,2,3-triazole with bp 64-67°C (2 mm). Found: C 34.7; H 4.4; N 61.0%. $C_2H_3N_3$. Calculated: C 34.4; H 4.3; N 60.8%. The residue was then distilled to give 115 g (97%) of benzotriazole with mp 100-102°C [7].

1-(2)-Propargyl-5-phenyltetrazole (IIIa, IVa). A solution of 3.6 g (0.03 mole) of propargyl bromide in 7 ml of acetone was added to a solution of 4.5 g (0.03 mole) of 5-phenyltetrazole and 3 g (0.04 mole) of triethylamine in 40 ml of acetone, and the reaction mixture was stirred at the boiling point of the solvent for 2 h. It was then cooled, and the precipitated triethylamine salt was removed by filtration. The filtrate was evaporated to a volume of 15 ml, and the concentrate was diluted with ether. The resulting mixture was washed to neutrality and dried with magnesium sulfate. The solvent was removed by distillation, and the residue was distilled *in vacuo* to give 3.5 g (63%) of a fraction with bp 129-135°C (1 mm), which, according to the PMR spectra, consisted of 53% 1-propargyl-5-phenyltetrazole (IIIa) and 47% 2-propargyl-5-phenyltetrazole (IVa), from which 1.6 g of IIIa, with mp 37-38°C, and 0.9 g of IVb, with mp 45-47°C, were isolated by chromatography.

Similarly, a mixture of isomers with bp 140-152°C (1 mm), which, according to the PMR spectra, consisted of 42% 1-propargyl-5-(β-chloroethyltetrazole) (IIIb) and 58% 2-propargyl-5-(β-chloroethyltetrazole) (IVb), was obtained from 5-(β-chloroethyl)tetrazole (Ib).

TABLE 1. Propargylazoles III-VI and X

Compound	mp, °C, or bp, (mm)	PMR spectrum, δ, ppm				Found, %			Empirical formula	Calc., %			Yields of mixt. of isomers, %	Isomer ratio in the mixt.
		CH_2	CH	1-H	2-H	C	H	N		C	H	N		
IIIa+IVa	37—38	5,36	2,67	—	—	65,0	4,5	30,5	$C_{10}H_8N_4$	65,2	4,3	30,4	63	53/47
	45—47	5,16	2,64	—	—	64,9	4,2	30,4	$C_{10}H_8N_4$	65,2	4,3	30,4		
IIIb+IVb	130—152 (1)	5,25 5,39	2,76 2,74	— —	— —	42,2	4,7	31,6	$C_6H_7ClN_4$ [a]	42,3	4,1	32,1	59	42/58
Va+VIa	90—92	2,35	2,10	8,05	7,71									
	(5)	2,31	2,03	7,79	7,09	55,9	5,1	39,1	$C_5H_5N_3$	56,0	4,6	39,2	51 [b]	83/17
Vb+VIb	67—68	5,41	3,15	—	—	66,8	5,7	15,5	$C_{15}H_{16}N_3O$	66,9	5,5	15,6	82	95/5
Xc	94—95	5,10	4,15	—	—	66,4	5,3	15,2	$C_{15}H_{16}N_3O$	66,9	5,5	15,6		
	—	4,46	2,63	7,63	—	68,6	5,9	17,4	$C_{13}H_{13}N_3O$	68,7	5,7	18,5	70	—

[a]Found: Cl 20.0%. Calculated: Cl 20.0%. [b]Compound Va was obtained by method A in 55% yield. [c]n_D^{18} 1.3987.

TABLE 2. Azoles XIV-XX

Compound	mp, °C or bp, °C (mm)	Found, %				Empirical formula	Calc., %				Yield, %
		C	H	Cl	N		C	H	Cl	N	
XIV	69	65,4	3,5	—	30,1	$C_{20}H_{14}N_8$	65,1	3,8	—	30,6	70
XV [a]	128—129	55,8	4,0	—	38,9	$C_{10}H_8N_6$	56,6	3,7	—	39,6	21
XVI	—	66,9	7,1	—	25,6	$C_{15}H_{19}N_5$	66,9	7,1	—	26,0	70
XVIIa	137—140 (3)	62,5	8,3	—	29,1	$C_{10}H_{16}N_4$	61,7	8,4	—	29,8	43
XVIIb	181 (4)	64,7	7,6	—	27,6	$C_{11}H_{16}N_4$	64,7	7,8	—	27,4	95
XVIIIa	173—174	63,5	4,0		31,9	$C_{16}H_{13}N_7$	63,3	4,3	—	32,3	80
XVIIIb	91—92	49,7	3,9	12,1	33,9	$C_{12}H_{12}ClN_7$	49,8	4,1	12,3	33,8	78
XVIIIc	104—105	49,8	4,2	12,4	33,6	$C_{12}H_{12}ClN_7$	49,7	4,1	12,3	33,8	76
XIXa	82—83	58,2	4,5	—	37,3	$C_{11}H_{10}N_6$	58,4	4,4	—	37,1	83
XIXb	93—94	39,5	4,3	16,8	39,7	$C_7H_9ClN_6$	39,6	4,2	16,5	39,6	50
XXa	148—150	45,5	4,9	—	45,6	$C_{14}H_{18}N_{12}O$	45,4	4,8	—	45,6	50
XXb	142—143	49,5	5,4	—	45,0	$C_{19}H_{25}N_{15}$	49,2	5,3	—	45,3	70

[a]n_D^{20} 1.4580.

TABLE 3. 1,2,3-Triazole Derivatives

Compound	mp, °C, or bp, °C (mm)	IR spectrum, cm^{-1}	PMR spectrum, ppm	Found, %			Empirical formula	Calc., %			Yield, %
				C	H	N		C	H	N	
VIIa	132—133 (2)	1660, 1645 (C=C), 1600, 1470, 1180, 1076 (ring)		54,6	6,8	27,9	$C_7H_{11}N_3O$	54,9	7,1	27,4	53
VIIb	120—122 (3)	1660, 1640 (C=C), 1580, 1470, 1070, 910 (ring)		68,1	7,2	14,0	$C_{17}H_{20}N_3O_2$	68,4	6,7	14,0	62
VIII	190—191 (1)	3400 (OH), 1595, 1480, 1080, 900 (ring)	7,80 (C_5—H), 7,98 (C_4—H)	63,6	5,5	22,8	$C_{10}H_{11}N_3O$	63,4	5,8	22,2	85
IX	116—117, 64—65	1595, 1475, 1070, 900 (ring)	7,74 (C_5—H), 7,96 (C_4—H)	57,9	4,6	20,2	$C_{10}H_{10}ClN_3$ [a]	57,8	4,8	20,2	76
XI	83—85 (3)	3360 (OH), 1000, 1470, 1180, 910 (ring)	7,75 (C_4, C_5—H), 5,80 (CH_2), 6,30 (OH)	36,4	5,0	42,2	$C_3H_5N_3O$	36,3	5,0	4,2	74
XII	78—81 (5)	1650 (C=C), 1600, 1460, 1110, 910 (ring)		47,3	5,1	47,1	$C_7H_8N_6$	47,7	4,5	47,7	55
XIII	120—122 (5)	1600, 1470, 1080, 900 (ring)	4,35 (CH_2), 1,45 (CH_3), 7,45 (C_5—H), 7,53 (C_4—H)	50,0	7,5	42,9	$C_7H_7N_3$	49,4	7,2	43,2	77

[a]A mixture of isomers (1-N and 2-N) was used in the reaction.

1-Propargyl-1,2,3-triazole (Va). A) A 2.5-g (0.044 mole) sample of KOH was added to a solution of 3 g (0.044 mole) of 1,2,3-triazole IIa and 5.3 g (0.044 mole) of propargyl bromide, and the mixture was stirred at the boiling point of the solvent for 4 h, after which it was cooled and filtered. The solvent was removed by distillation, and the residue was distilled *in vacuo* to give 2.6 g (55%) of Va.

Similarly, the reaction of 2.3 g (0.01 mole) of triazole IIb with 1.2 g (0.01 mole) of propargyl bromide gave 1.5 g of 1-propargyl-4-benzoyl-9-hydroxydimethyl-1,2,3-triazole (Vb) and 0.3 g of 1-propargyl-5-benzoyl-4-hydroxydimethyl-1,2,3-triazole (VIb). The characteristics are presented in Table 1.

B) A 3-g (0.044 mole) sample of triazole IIa and 5.3 g (0.044 mole) of propargyl bromide were added to a solution of sodium methoxide (from 1 g of sodium) in 20 ml of absolute methanol, and the mixture was stirred at the boiling point of the solvent for 5 h. It was then cooled, and the methoxide was removed by filtration. The residue was distilled *in vacuo*. The first fraction consisted of 2.5 g (51%) of a product with bp 90-95°C (5 mm) and was found to be a mixture of regioisomers of propargyltriazole, which consisted (on the basis of the PMR spectrum) of 83% 1-propargyl-1,2,3-triazole (Va) and 17% 2-propargyl-1,2,3-triazole (VIa). The second fraction consisted of 0.5 g (7%) of a product with bp 132°C (2 mm) and was identified as 1-(2-ethoxy-2-propen-1-yl)-1,2,3-triazole (VIIa) (Table 3).

1-(2-Ethoxy-2-propen-1-yl)-1,2,3-triazole (VIIa). A solution of 1 g (0.01 mole) of triazole Va in 10 ml of absolute ethanol was stirred at the boiling point of the solvent for 1 h in the presence of a catalytic amount of sodium. The solvent was removed by distillation *in vacuo* to give 0.8 g (52%) of a product with bp 132°C (2 mm). 1-(2-Ethoxy-2-propen-3-yl)-4-benzoyl-5-hydroxymethyl-1,2,3-triazole (VIIb) was similarly synthesized (Table 3).

1-Benzyl-4(5)-hydroxymethyl-1,2,3-triazole (VIII). This compound was obtained by the reaction of 1-propyn-3-ol with benzyl azide, as in [8] (Table 3).

1-Benzyl-4(5)-chloromethyl-1,2,3-triazole (IX). A 5.9-g (0.05 mole) sample of thionyl chloride was added to a suspension of 7.56 g (0.4 mole) of triazole VIII, cooled to 5°C, after which 3.95 g (0.05 mole) of pyridine was added slowly dropwise, and the mixture was stirred at 5°C for 1 h and allowed to stand at room temperature for 16 h. A part of the solvent was removed by distillation at reduced pressure, and the residue was dissolved in water. The aqueous solution was extracted with ether, and the ether extract was washed to neutrality and dried over magnesium sulfate. The solvent was removed, and the residue was crystallized from aqueous alcohol. The yield of the mixture of the two isomers was 6.3 g (76%). Fractional crystallization from aqueous alcohol yielded 3.1 g of 1-benzyl-4-chloromethyl-1,2,3-triazole (Table 3).

1-Benzyl-5(4)-(2-oxo-4-pentynyl)-1,2,3-triazole (X). A) A 0.3-g (5 mmole) sample of potassium hydroxide was added to a solution of 1.03 g (5 mmole) of chloromethyltriazole XI and 0.92 g (5.5 mmole) of propargyl bromide in 20 ml of acetone, and the mixture was stirred at the boiling point of the solvent for 20 h. The solvent was removed, and the residue was chromatographed to give 0.8 g (70%) of triazole X.

B) A 0.4-g (7 mmole) sample of potassium hydroxide was added to a solution of 1.5 g (7 mmole) of triazole IX and 0.4 g (7 mmole) of propargyl alcohol in 30 ml of benzene and 5 ml of acetone, and the mixture was maintained at the boiling point of the solvent for 20 h. It was then worked up as in method A to give 0.7 g (42%) of X (Table 1).

2-Hydroxymethyl-1,2,3-triazole (XI). A solution of 20.7 g (0.3 mole) of triazole IIa in 120 ml of ethanol was added to a suspension of 18 g (0.6 mole) of paraformaldehyde in 30 ml of water, and the mixture was stirred at 80°C for 5 h. The solution was evaporated partially, and the product was extracted with ether. The ether extract was washed with small portions of water to remove the paraformaldehyde and dried over magnesium sulfate. The solvent was removed, and the residue was distilled *in vacuo* (Table 3).

Propargylation of 2-Hydroxymethyl-1,2,3-triazole. A 1.12-g (0.02 mole) sample of potassium hydroxide was added to a solution of 2 g (0.02 mole) of triazole XI and 2.4 g (0.02 mole) of propargyl bromide in 30 ml of benzene, and the mixture was stirred at the boiling point of benzene for 20 h. The solvent was removed, and the residue was eluted with a mixture of petroleum ether and diethyl ether and distilled *in vacuo* to give 1,3-bis(1,2,3-triazol-1-yl)prop-2-ene (XII) and propargyltriazole Va [0.6 g (25%)] with bp 92-93°C (6 mm) (Table 1).

1,3-Bis(1,2,3-Triazolyl)propene (XII). A catalytic amount of potassium hydroxide was added to a solution of 0.01 mole of propargyltriazole Va and 0.7 g (0.01 mole) of triazole IIa in dioxane, and the mixture was stirred at 100°C for 16 h. The solvent was removed, and 0.9 g (55%) of bistriazole XII was isolated from the residue by chromatography. The principal constants were identical to those of the bistriazole obtained by the method presented above.

1-Ethyl-1,2,3-triazole (XIII). This compound was synthesized in the same way as the procedure for alkylation described above from 2-hydroxymethyl-1,2,3-triazole and ethyl bromide (Table 3).

1,6-Bis(5-phenyl-1-tetrazolyl)hexa-2,4-diyne (XIV). A 0.01-g sample of cuprous chloride in 0.5 ml of pyridine was added to a solution of 0.5 g (3 mmole) of propargyltetrazole IIIa in 10 ml of methanol, and the mixture was stirred in an oxygen atmosphere until the spot of the starting acetylene vanished on a thin layer of aluminum oxide. Water (3 ml) was added, and the precipitated crystals were removed by filtration to give 0.35 g of diacetylene XIV (Table 2). 1,6-Bis(1,2,3-triazol-1-yl)hexa-2,4-diyne (XV) was similarly obtained from propargyltriazole Va (Table 2).

5-Phenyl-1-(4-diethylaminobutyn-2-yl)tetrazole (XVI). A suspension of 0.7 g (4 mmole) of 2-propargyl-5-phenyltetrazole, 0.2 g (6.6 mmole) of paraformaldehyde, 0.4 g (5.6 mmole) of diethylamine, and 0.01 g of cuprous chloride in 5 ml of dioxane was heated on a water bath for 1 h, after which it was cooled, treated with 4 ml of 10% hydrochloric acid, and extracted with ether. The aqueous layer was neutralized with sodium bicarbonate and again extracted with ether. The ether extract was washed with water and dried over magnesium sulfate. The solvent was removed *in vacuo*, and the residue was chromatographed to give 0.7 g of tetrazole XVI (Table 2).

1-(1-Diethylaminobutyn-2-yl)-1,2,3-triazole (XVIIa) and 1-(1-piperidinobutyn-2-yl)-1,2,3-triazole (XVIIb) (Table 2) were synthesized similarly.

1-Phenyl-1,2,3-(4-triazolyl)-(5-phenyl-1-tetrazolyl)methane (XVIIIa). A suspension of 0.4 g (2.5 mmole) of 5-phenyl-1-propargyltetrazole (IIIa) and 0.5 g (4 mmole) of phenyl azide in 4 ml of toluene was maintained at the boiling point of the solvent for 3 h, after which it was cooled, and the precipitated polyazole XVIIIa was removed by filtration. Polynuclear azoles XVIIIb, XIXa, b, and XXa, b were similarly obtained. The principal characteristics are presented in Table 2.

LITERATURE CITED

1. Japanese Patent No. 21599; Ref. Zh. Khim., 9N562P (1973).
2. British Patent No. 1353699; Ref. Zh. Khim., 90458P (1975).
3. Japanese Patents Nos. 7021833 and 7021834; Chem. Abstr., 73, 77256c, 77257 (1970).
4. W. Finnegan, R. Henry, and R. Lofguist, J. Am. Chem. Soc., 80, 3908 (1958).
5. C. Arnold and D. Thatcher, J. Org. Chem., 34, 1141 (1969).
6. L. I. Vereshchagin, L. G. Tikhonova, A. V. Maksikova, L. D. Gavrilov, and G. A. Gareev, Zh. Org. Khim., 15, 612 (1979).
7. I. Heilbron and H. M. Bunbury (editors), Dictionary of Organic Compounds, Vol. 1, Eyre and Spottiswoode, London (1953).
8. S. S. Novikov, V. M. Brusnikina, and V. A. Rudenko, Izv. Akad. Nauk SSSR, Ser. Khim., 474 (1961).