SYNTHESIS AND PROPERTIES OF sym-TRIAZINE DERIVATIVES.

7.* SYNTHESIS OF PYRIDYL-SUBSTITUTED 2-AMINO- AND 2,4-DIAMINO-sym-TRIAZINES

V. I. Kelarev, R. A. Karakhanov, M. Bellul',
R. L. Ushakova, and A. I. Mikaya

UDC 547.826'495.9'491.8:
542.953:543.51

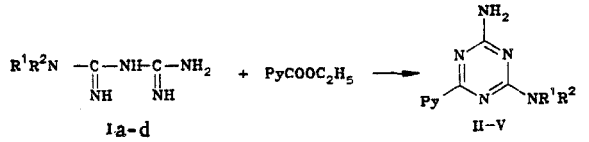
N-Substituted 2,4-diamino-6-pyridyl-sym-triazines were synthesized by cyclocondensation of pyridinecarboxylic acid esters with biguanides. 4,6-Disubstituted 2-amino-sym-triazines containing pyridyl residues were obtained by the reaction of pyridinecarboxylic acid nitriles with guanidine or of pyridinecarboxylic acid esters with N-imidoylguanidines. Aminotriazines of this type are also formed in the condensation of N-acylguanidines with nitriles or imino esters. The general principles of the fragmentation of 2-amino-4-dialkylamino-6-pyridyl-sym-triazines under the influence of electron impact were established.

Some pyridyl-substituted sym-triazines display high biological activity [2-4] and also have pesticide activity [5, 6]. In order to expand research in this direction we synthesized mono- and diamino derivatives of sym-triazine that contain one or two pyridyl fragments in their molecules. Heterocyclic systems of this type may be of definite interest as intermediates for obtaining chemical agents for the protection of plants, cationic and nonionogenic surfactants, and stabilizers and additives for polymeric materials.

To obtain the indicated compounds, in the present research we used the condensation of derivatives of pyridinecarboxylic acids (esters, nitriles, and imino esters) with guanidine, N-acyl- and N-imidoylguanidines, and biguanides.

It is known [7] that 2,4-diamino-6-alkyl(aryl, hetaryl)-sym-triazines (guanamines) are formed as a result of the condensation of esters with biguanides. However, despite the widespread use of various esters in this reaction, only individual instances [3, 8] of the participation of esters of acids of the pyridine series in condensations with biguanides have thus far been described. It was noted that 2,4-diamino-6-pyridyl-sym-triazines are formed in yields no higher than 35-40%.

In the present research we made a more detailed study of the condensation of ethyl esters of pyridinecarboxylic acids with N-substituted biguanides Ia-d in the presence of bases.



Ia-d II-V

Ia IIa-d $R^1=R^2=CH_3$; Ib, IIIa-c $R^1=R^2=C_2H_5$; Ic, IVa,c $R^1=R^2=C_4H_9$; Id, Va-c $R^1=H$, $R^2=C_6H_5$; II—V a R = 2-pyridyl b R = 3-pyridyl c R = 4-pyridyl

N-Substituted 2,4-diamino-6-pyridyl-sym-triazines II-V are formed in high yields (Table 1) by heating equimolar amounts of the reagents in ethanol for 18-20 h in the presence of sodium ethoxide. Replacement of ethanol by higher-boiling solvents — dioxane, butanol, and DMF — did not lead to an appreciable increase in the yields of aminotriazines.

To synthesize the 4,6-disubstituted 2-amino-sym-triazines containing pyridyl fragments we used the three methods that are usually employed to obtain monoamino-sym-triazines of this type: condensation of nitriles of pyridinecarboxylic acids with guanidine [9] (method A), condensation of N-imidoylguanidines VIIa-c with ethyl nicotinate [10] (method B), and con-

*See [1] for Communication 6.

I. M. Gubkin Moscow Institute of Petroleum and Gas, Moscow 117917. Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 5, pp. 674-680, May, 1988. Original article submitted July 31, 1986; revision submitted December 25, 1987.

 0009-3122/88/2405-0550$12.50

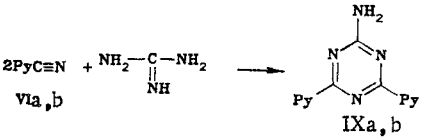
TABLE 1. N-Substituted 2,4-Diamino-6-pyridyl-sym-triazines II-IV

Compound	mp,* °C		Found, %			Empirical formula	Calc., %			Yield, %
	our data	literature data	C	H	N		C	H	N	
IIa	237—238	238—240 [8]				$C_{10}H_{12}N_6$				76
IIb	205—206	200—202 [3]				$C_{10}H_{12}N_6$				81
IIc	233—233,5	229—231 [3]				$C_{10}H_{12}N_6$				75
IIIa	204—205	—	59,1	6,7	34,3	$C_{12}H_{16}N_6$	59,0	6,6	34,4	70
IIIb	138—139	—	58,9	6,5	34,5	$C_{12}H_{16}N_6$	59,0	6,6	34,4	68
IIIc	169,5—171	—	59,2	6,7	34,2	$C_{12}H_{16}N_6$	59,0	6,6	34,4	77
IVa	154—155	—	63,9	8,1	28,2	$C_{16}H_{24}N_6$	64,0	8,0	28,0	67
IVc	145—146	—	64,1	8,0	28,3	$C_{16}H_{24}N_6$	64,0	8,0	28,0	71
Va	232—233	—	63,5	4,5	32,0	$C_{14}H_{12}N_6$	63,6	4,5	31,8	69
Vb	216—217	217—218 [8]			31,6	$C_{14}H_{12}N_6$			31,8	78
Vc	214—215	211—213 [8]				$C_{14}H_{12}N_6$				70

*Compounds IIa and Vb, c were crystallized from acetonitrile; the remaining compounds were crystallized from alcohol.

densation of N-acylguanidines VIIIa-d with nitriles VIa-f [11] (method C). It should be noted that methods B and C make it possible to synthesize amino-sym-triazines that contain two different substituents in the ring.

2-Amino-4,6-dipyridyl-sym-triazines IXa, b are formed as a result of the condensation of nitriles VIa, b with guanidine.



VIa,b IXa,b

VI, IX a Py = 3-pyridyl, b Py = 4-pyridyl

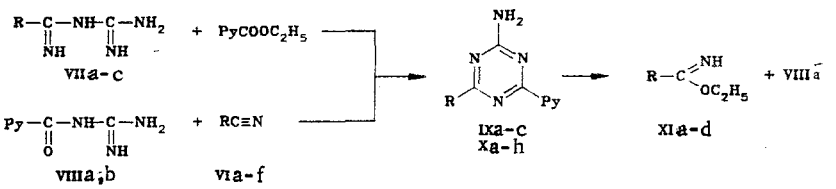
The best yields of sym-triazines IXa, b (60-65%) are obtained in the case of prolonged heating of the reagents taken in a molar ratio of 2:1 in butanol in the presence of sodium ethoxide; in addition to aminotriazines IXa, b, unidentified high-melting products of side reactions (mp > 350°C) that are only slightly soluble in most organic solvents were isolated.

The condensation of N-imidoylguanides VIIa, c with ethyl nicotinate proceeds under mild conditions (refluxing of equimolar amounts of the reagents for several hours in alcohol) and is not accompanied by the formation of side compounds.

The condensation of N-(pyridylcarbonyl)guanidines VIIIa, b with nitriles VIa-f was carried out by heating the reagents for several hours. It was found that the reaction of guanidines VIIIa, b with nitriles VIa-c, f should be carried out by heating to 165-180°C for 8-10 h. When acetonitrile VId or trichloroacetonitrile VIe are used, the corresponding 6-substituted 2-amino-4-pyridyl-sym-triazines Xc, d, f are formed in good yields (Table 2) by prolonged refluxing (18-20 h) of guanidines VIIIa, b in excess nitrile.

In addition, for the synthesis of some of the compounds of this type (Xa, d, e) we also used an alternative method — the reaction of N-benzoyl- (VIIIc) and N-trichloroacetylguanidine (VIIId) with pyridinecarboxylic acid nitriles.

A logical development of the latter method for obtaining aminotriazines is the introduction of imino esters into the reaction with N-acylguanidines (method D). In this case amino-triazines are formed in high yields (Table 2) after brief refluxing of equimolar amounts of the reagents in absolute alcohol.



VIIa, IXc, XIa R = 3-pyridyl; VIc, VIIb, Xa,e XIb R = C_6H_5; VIIc, Xb, XIc R = 3-indolyl; VIIIa, Xa,b Xa-d,h Py = 3-pyridyl; VIIIb, IXc, Xe-g Py = 4-pyridyl; VId Xc,f R = CH_3; VIe Xd R = CCl_3; VIf, Xg R = $C_{13}H_{27}$; Xh, XId R = 3-indolylmethyl

TABLE 2. 6-Substituted 2-Amino-4-pyridyl-sym-triazines IX and X

Compound	mp,* °C	R_f (solvent system)	Found				Empirical formula	Calculated				Yield, % (method of synthesis)
			C, %	H, %	N, %	M		C, %	H, %	N, %	M	
IXa	324—325 (320—323 [28])	0,46 (A)	62,3	3,9	33,7	250	$C_{13}H_{10}N_6$	62,4	4,0	33,6	250	64 (A), 88 (B), 71 (C), 91 (D)
IXb	349—350	0,32 (A)	62,5	4,0	33,5	250	$C_{13}H_{10}N_6$	62,4	4,0	33,6	250	60 (A), 74 (C)
IXc	318—320	0,41 (A)	62,2	4,1	33,7	250	$C_{13}H_{10}N_6$	62,4	4,0	33,6	250	70 (C)
Xa	202—204	0,32 (B)	67,4	4,5	28,0	249	$C_{14}H_{11}N_5$	67,5	4,4	28,1	249	78 (B), 77 (C), 93 (D)
Xb	269—270	0,24 (A)	66,0	4,9	28,8	290	$C_{16}H_{14}N_6$	66,2	4,8	29,0	290	84 (B), 92 (D)
Xc	170—171	0,50 (A)	57,5	5,0	37,3	187	$C_9H_9N_5$	57,7	4,8	37,4	187	72 (C)
Xd	178—180	0,24 (B)	37,0	2,2	24,3	289**	$C_9H_6Cl_3N_5$	37,2	2,1	24,1	289**	77 (C)
Xe	155—157	0,54 (A)	67,3	4,5	28,0	249	$C_{14}H_{11}N_5$	67,5	4,4	28,1	249	76 (C)
Xf	191—193	0,62 (A)	57,5	4,8	37,5	187	$C_9H_9N_5$	57,7	4,8	37,4	187	60 (C)
Xg	138—139	0,68 (A)	70,8	9,4	19,6	355	$C_{21}H_{33}N_5$	71,0	9,3	19,7	355	76 (C)
Xh	188—189,5	0,35 (B)	67,4	4,5	28,0	302	$C_{17}H_{14}N_6$	67,5	4,6	27,8	302	95 (D)

*The compounds were recrystallized: IXa from ethylene glycol, IXb from methylcellulose, IXc and Xb, d, g from aqueous DMF, Xa from aqueous ethylene glycol, Xc, e, f from alcohol, and Xh from butanol.
**The ion containing the ^{35}Cl isotope.

TABLE 3. Mass Spectra of 2-Amino-4-dialkylamino-6-sym-triazines*

Compound	m/z (relative intensities of the peaks, %)
IIa	216 (M^+, 100), 215 (2), 201 (45), 187 (14), 173 (22), 172 (6), 159 (8), 105 (70), 78 (22), 69 (15)
IIb	216 (M^+, 100), 215 (6), 201 (50), 187 (19), 173 (25), 159 (37), 126 (27), 105 (45), 87 (16), 69 (37)
IIc	216 (M^+, 100), 215 (8), 201 (40), 187 (20), 173 (25), 172 (11), 131 (10), 108 (18), 105 (30), 69 (25)
IIIa	244 (M^+, 92), 229 (6), 216 (17), 215 (100), 201 (26), 173 (10), 159 (17), 105 (62), 79 (23), 69 (13)
IIIb	244 (M^+, 100), 229 (29), 215 (90), 201 (30), 173 (9), 126 (42), 105 (40), 86 (9), 78 (5), 72 (66)
IIIc	244 (M^+, 100), 229 (45), 216 (17), 215 (90), 201 (40), 173 (8), 105 (22), 101 (9), 78 (10), 69 (24)
IVa	300 (M^+, 100), 271 (60), 258 (30), 257 (35), 243 (27), 216 (70), 215 (52), 201 (75), 173 (10), 105 (80)
IVb	300 (M^+, 71), 271 (31), 258 (36), 257 (80), 243 (16), 216 (45), 215 (100), 201 (75), 173 (6), 105 (21)

*The 10 most intense peaks are presented.

Absorption bands with different intensities that are characteristic for the stretching and deformation vibrations of the triazine ring [12-14] are observed in the IR spectra of all of the 2,4-diamino- (II-IV) and 2-aminotriazines (IX, X) at 1530-1575, 1405-1420, 1090-1120, 980-1010, 805-810, and 695-710 cm^{-1}. The spectra of the synthesized compounds also contain absorption bands that are characteristic for pyridyl fragments [14]. The intense absorption bands at 1635-1670 cm^{-1} are related to the scissors vibrations of the N–H bonds in the primary amino groups; this is characteristic for amino-sym-triazines [15].

The mass spectra of the synthesized amino- and diaminotriazines also confirm the proposed structures. The experimentally found molecular masses of the substances correspond to the calculated values, and the character of the subsequent fragmentations, which was confirmed by metastable transitions, is in agreement with the structures presented. All of the investigated sym-triazine derivatives form stable molecular ions (M^+), the peaks of which, as a rule, have the maximum intensity. It should be noted that the spectra of the isomeric diaminotriazines that differ with respect to the type of pyridyl radical differ slightly only with respect to the relative intensities of the peaks of the common fragment ions, which cannot be linked with the type of isomer.

The most characteristic fragmentation of 2-amino-4-dialkyl-amino-6-pyridyl-sym-triazines II-IV involves fragmentation of the dialkylamino group, while fragmentation due to cleavage of the triazine ring is uncharacteristic [16]. However, attention should be directed to the presence in the mass spectra of these compounds of peaks of medium intensity with m/z 105, which may be due to $[PyCNH]^+$ ions formed by ring cleavage and migration of a hydrogen atom to the charged fragment.

A fundamental peculiarity of the fragmentation of sym-triazines II-IV is the fact that splitting out of an alkyl group from M^+ occurs more readily than fragmentation due to cleavage of the C–C bond adjacent to the nitrogen atom of the amino group. This is seen quite clearly when one compares the intensities of the peaks of the $[M - R]^+$ and $[M - CH_2R]^+$ ions (Table 3). The former ions are formed somewhat more readily than the latter only in the case of dibutylamino derivatives IVa, b. This character of the fragmentation sharply distinguishes dialkylamino-sym-triazines from the dialkylamino derivatives of aromatic and heteroaromatic compounds [7].

Low-intensity peaks of ions with m/z 173 due to the ejection of a dialkylamino group from M^+ are observed in the spectra of all of the sym-triazines II-IV. Of definite interest is the presence in the mass spectra of dimethylamino-sym-triazines IIa-f of peaks of $[M - 29]^+$ ions, the formation of which is probably associated with splitting out of a CH_2NH particle, which is characteristic for dimethylamino derivatives of other heteroaromatic systems [18].

A more complex fragmentation pattern is observed in the case of dibutylamino-sym-triazines IVa, c. In addition to cleavage at the α-C–C bond, the butyl group is capable of undergoing cleavage at the β- and γ-C–C bonds, which leads to the appearance of $[M - C_3H_7]^+$, $[M - C_2H_5]^+$, and $[M - CH_3]^+$ ions. Specific cleavage of the N-butyl group is also associated with the elimination of C_3H_6 and C_4H_8 molecules from M^+. The second N-butyl group, which is retained in all of the resulting ions, is fragmented only via ejection of C_3H_6 and C_4H_8 molecules, which leads to extremely intense peaks of ions with m/z 216, 215, and 201. It should be noted that such processes are usually not observed for dialkylamino derivatives of the aromatic series.

EXPERIMENTAL

The IR spectra of KBr pellets of the compounds were recorded with a UR-20 spectrometer. The PMR spectra of solutions in deuteromethanol or d_6-DMSO were obtained with a Tesla BS-487C spectrometer with hexamethyldisiloxane (HMDS) as the internal standard. The mass spectra were recorded with an LKB-2091 spectrometer using a system for direct introduction of the samples into the ion source; the ionizing-electron energy was 70 eV, the emission current was 25 μA, and the temperature of the ion source was 200°C vis-à-vis a sample-vaporization temperature of 130-140°C. The compositions of the reaction mixtures and the purity of the compounds obtained were monitored by TLC on activity II (Brockmann) Al_2O_3 in benzene–methanol systems [10:1 (A) or 20:1 (B)] with development by iodine vapors.

N-Substituted 2,4-Diamino-6-pyridyl-sym-triazines II-IV. A 0.30 mole sample of the hydrochloride of biguanide Ia-d was added in portions at 0°C to a solution of sodium ethoxide obtained from 7.0 g (0.30 mole) of sodium in 200 ml of absolute alcohol, and the reaction

mixture was stirred for 1 h at 20°C. It was then cooled to 0°C, and the precipitated NaCl was removed by filtration and washed on the filter with 25 ml of cold absolute alcohol. A solution of 0.30 mole of ethyl pyridinecarboxylate in 70 ml of absolute alcohol was added dropwise with stirring to the resulting solution of the biguanide base, after which the reaction mixture was refluxed with stirring for 14-18 h (with monitoring by means of TLC until the starting ester had vanished). The mixture was then evaporated to dryness at reduced pressure, and the residue was washed with hot hexane (three 50-ml portions) and crystallized from a suitable solvent (Table 1).

PMR spectrum of IIc (d_6-DMSO): 3.08 (6H, d, J = 7.0 Hz, NCH_3), 6.74 (2H, broad s, NH_2), 7.05-7.27 ppm (4H, m, aromatic protons). PMR spectrum of IIIa (d_6-DMSO): 1.43 (6H, t, CH_3), 3.34 (4H, q, CH_2), 6.88 (2H, broad s, NH_2), 7.12-7.44 ppm (4H, aromatic protons).

N-(3-Pyridylimidoyl)guanidine (VIIa). A solution of 3.75 g (25 mmoles) of nicotinic acid ethyl imino ester in 25 ml of absolute alcohol was added dropwise at 20-25°C to a stirred solution of the guanidine base in absolute alcohol, which was obtained from 2.02 g (30 mmole) of guanidine hydrochloride and 0.74 g (32 mmole) of sodium in 50 ml of absolute alcohol, and the reaction mixture was maintained for 3 days at 20°C. The solvent was then removed at reduced pressure, and the residue was crystallized from aqueous alcohol to give 3.3 g (80%) of guanidine VIIa with mp 149-150°C and R_f 0.46 (B). Found: C 51.3; H 5.6; N 43.0%; M^+ 163. $C_7H_9N_5$. Calculated: C 51.5; H 5.5; N 42.9%; M 163.

N-Benzimidoylguanidine (VIIb). This compound was similarly obtained from imino ester XIb. The product was obtained in 82% yield and had mp 106-108°C (from aqueous alcohol) and had mp 106-108°C (from aqueous alcohol) and R_f 0.74 (B) (mp 107-109°C [19]).

N-(3-Indolylimidoyl)guanidine (VIIc). This compound was similarly obtained from imino ester XIc [20]. The product was obtained in 91% yield and had mp 232-233°C (from aqueous DMF) and R_f 0.12 (B). Found: C 62.9; H 5.7; N 36.5%. $C_{10}H_{11}N_5$. Calculated: C 62.8; H 5.8; N 36.6%.

N-(3-Pyridylcarbonyl)guanidine (VIIIa). A solution of 18.1 g (0.120 mole) of ethyl nicotinate in 30 ml of absolute alcohol was added dropwise at 20°C to a stirred solution of the guanidine base prepared from 11.5 g (0.120 mole) of guanidine hydrochloride and 2.87 g (0.125 mole) of sodium in 100 ml of absolute alcohol, and the reaction mixture was refluxed with stirring for 12 h. It was then evaporated to dryness at reduced pressure, and the residue was washed with ether (three 50-ml portions) and crystallized from absolute alcohol to give 18.7 g (95%) of guanidine VIIIa with mp 195-196°C and R_f 0.38 (B) (mp 196-197°C [21]).

N-(4-Pyridylcarbonyl)guanidine (VIIIb). This compound was similarly obtained from ethyl isonicotinate. The product was obtained in 91% yield and had mp 230-231.5°C (from absolute alcohol) and R_f 0.34 (B) (mp 231°C [21]).

6-Substituted 2-Amino-4-pyridyl-sym-triazines IX and X. A. A solution of the guanidine base obtained from 2.02 g (30 mmole) of guanidine hydrochloride and 0.74 g (32 mmole) of sodium in 50 ml of absolute alcohol was evaporated at reduced pressure (the bath temperature did not exceed 40°C), the residue was dissolved in 100 ml of butanol, 6.24 g (60 mmole) of nitrile VIa, b was added, and the solution was refluxed with stirring for 20-25 h until ammonia evolution ceased. The reaction mixture was evaporated to dryness at reduced pressure, the residue was extracted with hot alcohol (four 35-ml portions), and the extract was evaporated to give sym-triazines IXa, b.

B. A solution of 1.7 g (12 mmole) of ethyl nicotinate in 10 ml of absolute alcohol was added dropwise at 20°C to a stirred solution of 10 mmole of VIIa-c in 25 ml of absolute alcohol, and the reaction mixture was refluxed with stirring for 2 h. It was then cooled to 20°C and poured into 200 ml of cold water. The precipitate was removed by filtration, washed on the filter with water, dried in vacuo over P_2O_5, and crystallized from a suitable solvent (Table 2). This procedure was used to obtain sym-triazines IXa, and Xa, b.

C. A mixture of 12 mmoles of acylguanidine VIIIa, b and 50 mmoles of nitrile VIa-c, f was stirred for 8-10 h at 165-180°C (when nitriles VId, e were used, the mixture was refluxed with stirring for 18-20 h), after which it was evaporated at reduced pressure, and the residue was washed with ether (three 15-ml portions) and crystallized from a suitable solvent to give sym-triazines IXa-c and Xa, c-g.

D. A mixture of 1.64 g (10 mmoles) of nicotinoylguanidine VIIIa and 10 mmoles of imino ester XIa-d in 35 ml of absolute alcohol was refluxed with stirring for 2 h, after which it

was cooled to 20°C and poured into 150 ml of cold water. The precipitate was removed by filtration, washed on the filter with water, dried in vacuo over P_2O_5, and crystallized from a suitable solvent. This procedure was used to obtain sym-triazines IXa and Xa, b, h.

2-Amino-4-(3-pyridyl)-6-phenyl-sym-triazine (Xa). A mixture of 1.63 g (10 mmoles) of N-benzoylguanidine VIIIc [22] and 5.2 g (50 mmoles) of nitrile VIa was heated for 10 h at 165°C, after which the excess nitrile was removed at reduced pressure, and the residue was crystallized from aqueous ethylene glycol to give 1.8 g (74%) of triazine Xa.

sym-Triazine Xd (68% yield) was similarly obtained from N-trichloroacetylguanidine VIIId [22] and nitrile VIa, and sym-triazine Xe (72% yield) was similarly synthesized from N-benzoylguanidine VIIIc and nitrile VIb.

No melting-point depressions were observed for mixtures of samples of sym-triazines IX and X obtained by the different methods, and their IR spectra were identical.

PMR spectrum of sym-triazine Xa (d_6-DMSO): 6.52 (2H, s, NH_2), 6.97-7.18 ppm (9H, m, aromatic protons). PMR spectrum of sym-triazine Xc (d_6-DMSO): 2.54 (3H, s, CH_3), 6.63 (2H, broad s, NH_2), 7.32-7.54 ppm (4H, m, aromatic protons). PMR spectrum of sym-triazine Xg (d_6-DMSO): 1.12 (3H, t, CH_3), 1.84-2.26 (22H, m, CH_2), 3.12 (2H, t, α-CH_2), 6.57 (2H, broad s, NH_2), 7.27-7.40 ppm (4H, m, aromatic protons). PMR spectrum of sym-triazine Xh (deuteromethanol): 4.10 (2H, s, CH_2), 6.75 (2H, broad s, NH_2), 7.12-7.84 (9H, m, aromatic protons), 8.14 ppm (1H, s, indole NH).

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