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Reactions of 4-Oxo-1,3-benzoxazinium Perchlorates with Guanidines

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Abstract—Condensation of 2-methyl-4-oxo-1,3-benzoxazinium perchlorate with various aromatic and heterocyclic aldehydes provided previously unknown arylvinyl- and hetarylvinyl-substituted salts whose recyclization under treatment with guanidine resulted in formerly undescribed 1,3,5-triazines. The reaction of perchlorates obtained with guanidinebenzoxazole led to the formation of bishetarylamines, with guanidinebenzimidazole formed triazinebenzimidazoles, the cyanoguanidine reacted with 4-oxo-1,3-benzoxazinium perchlorates to give cyanamides.

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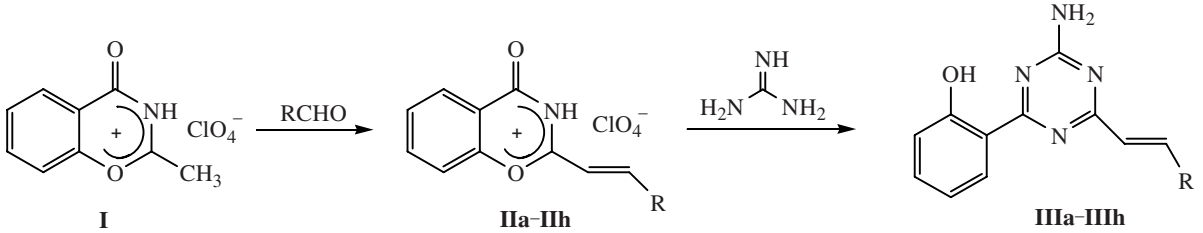
Substituted 1,3,5-triazines are extensively used in various fields of agrochemistry and pharmaceutics [1, 2]. We performed recyclization under treatment with guanidine of hetarylvinyl- and arylvinyl-substituted 4-oxo-1,3-benzoxazinium salts **II** synthesized by condensation of 2-methyl-4-oxo-1,3-benzoxazinium (**I**) with heterocyclic and aromatic aldehydes as described in [3] and thus obtained a series of previously unknown hetarylvinyl- and arylvinyl-substituted 1,3,5-triazines **III** (Scheme 1).

In order to prepare new biologically active substances we carried out recyclization of salts **I** and **II** under treatment of hetarylguanidines, of benzoxazoleguanidine with salts **I**, **IIc**, and **IIe**, and of benzimidazoleguanidine with salts **I, IIb, IIc, IIf**, and **IIg**. Therewith in the reaction with guanidinebenzoxazole triazinebenzoxazoleamines **IV** were obtained, but reaction of perchlorates **I** and **II** with guanidinebenzimidazole led instead of expected triazinebenzimidazoleamines **V** to the formation of compounds whose structure based on elemental analysis, ^{1}H NMR and mass spectra was first described as triazolebenzoxazepines [4] (Scheme 2).

However the X-ray diffraction analysis carried out on compound **VIa** showed that in this reaction formed 1,3,5-triazino[1,2-*a*]benzimidazoles **VI**.

Scheme 1.

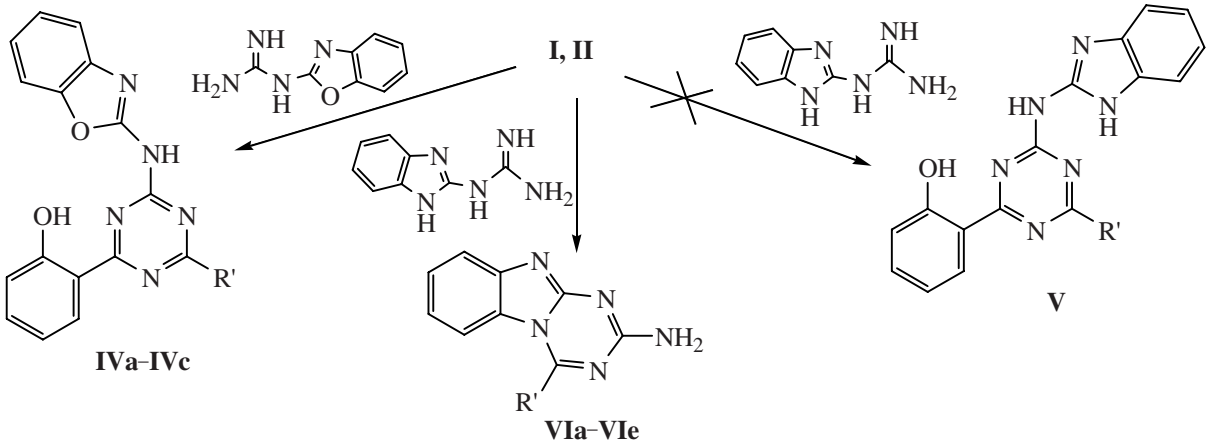


I **IIa–IIh** **IIIa–IIIh**

R = 2-pyridinyl (**a**), 4-ClC_6H_4 (**b**), 4-$Me_2NC_6H_4$ (**c**), 4-$MeSC_6H_4$ (**d**), 1-methylindol-3-yl (**e**), 2-thienyl (**f**), 4-$HOOCCH_2OC_6H_4$ (**g**), Ph (**h**).

† Deceased.

Scheme 2.



IV, R' = 4-$Me_2NC_6H_4CH$=CH (**a**), 1-methylindol-3-yl-CH=CH (**b**), Me (**c**); **VI**, R' = 4-ClC_6H_4CH=CH (**a**), 4-$Me_2NC_6H_4CH$=CH (**b**), 2-thienyl-CH=CH (**c**), 4-$HOOCCH_2C_6H_4CH$=CH (**d**), Me (**e**).

In the unit cell of compound **VIa** are located 4 molecules of unique structure (a single molecule is presented on Fig. 1). The tricyclic 1,3,5-triazino[1,2-*a*]-benzimidazole system is planar, the phenyl ring is tilted to it on the average by 19°. Geometrical parameters of the molecules are similar. The main bond lengths are compiled in the table. The accuracy attained does not provide a possibility to perform a comparative analysis of the geometry of the independent molecules. The main difference between the molecules is the variation of the phenyl ring position with respect to the double bond C^{10}=C^{11}, torsion angle $N^1C^1C^{10}C^{11}$ being ±32°. The structure is layer-like, the layers are fixed by intermolecular hydrogen bonds between the hydrogens of the amino group and nitrogen atoms of the heterocycle. Within the layer stronger hydrogen bonds N^{31}H···N^{41A} (*d*H···N 2.04 Å) result in dimers formation, and these are combined into layers by weaker hydrogen bonds N^{31}–H···N^{14A}, N^{34A}–H···N^1 (*d*H···N 2.40 Å) (Fig. 2). The outer surface of layers consists of chlorine atoms which are not involved into any short contacts.

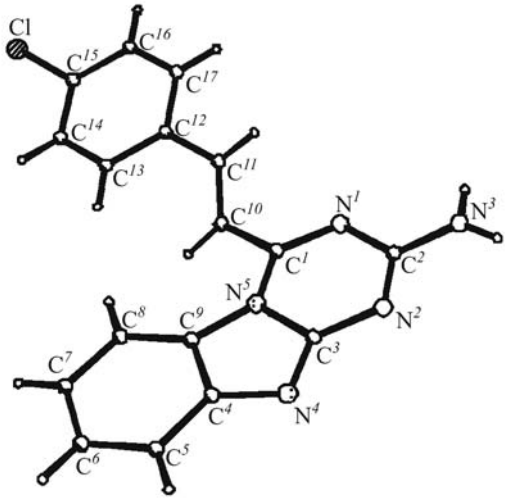


Fig. 1. Geometry of a molecule of compound **VIa**.

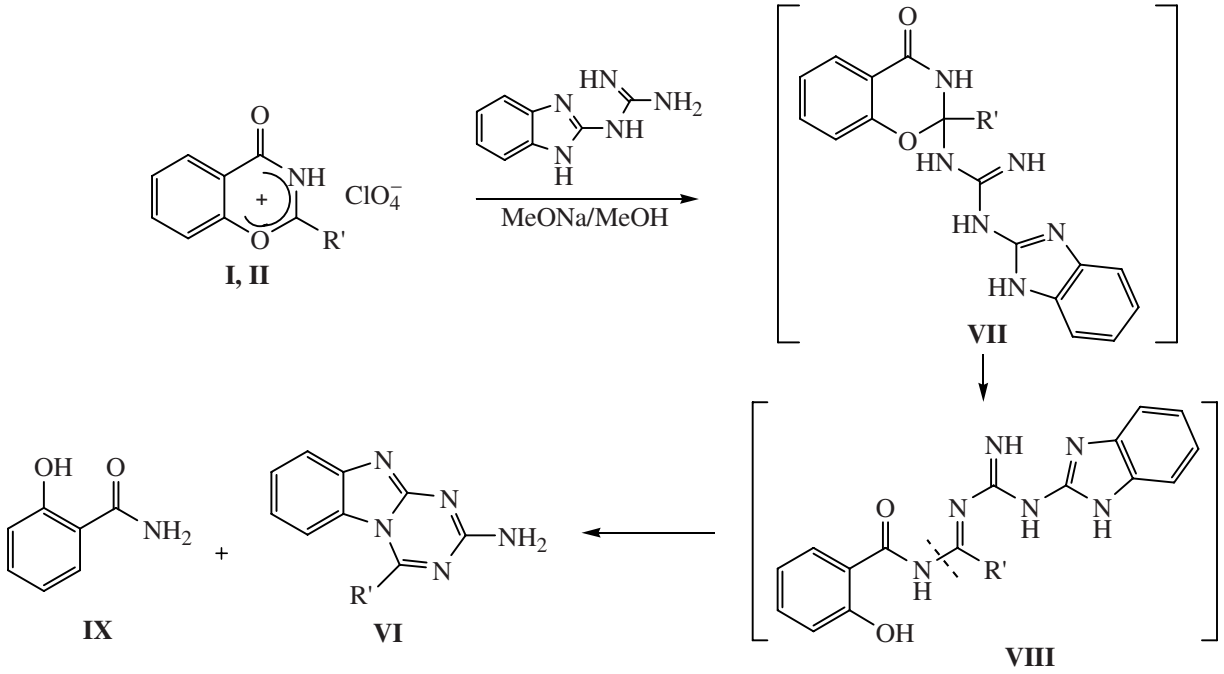
Previously 1,3,5-triazino[1,2-*a*]benzimidazoles were obtained by reaction of guanidine(amidine) benzimidazoles with orthoesters [5, 6].

The reaction presumably proceeds by the following mechanism: the guanidinebenzimidazole adds into the position *2* of the benzoxazine ring of perchlorates **I** and **II** providing benzoxazine **VII** that further opens into

Selected bond lengths (*d*, Å) in four molecules in the unit cell of compound **VIa**

Bond	No. of the molecule in the cell			
	1	2	3	4
N^1–C^1	1.30(1)	1.292(9)	1.288(9)	1.303(9)
N^1–C^2	1.41(1)	1.387(9)	1.40(1)	1.391(9)
C^2–N^2	1.30(1)	1.309(9)	1.31(1)	1.324(9)
N^2–C^3	1.35(1)	1.344(9)	1.340(9)	1.335(9)
C^3–N^5	1.440(9)	1.414(9)	1.441(9)	1.415(9)
N^5–C^1	1.355(9)	1.370(9)	1.354(9)	1.364(9)
N^4–C^3	1.291(9)	1.341(9)	1.285(9)	1.320(9)
N^4–C^4	1.384(7)	1.384(7)	1.382(7)	1.392(7)
N^5–C^9	1.384(7)	1.431(7)	1.375(7)	1.411(7)
C^2–N^3	1.32(1)	1.307(9)	1.340(9)	1.315(9)
C^1–C^{10}	1.49(1)	1.471(9)	1.48(1)	1.469(9)
C^{10}–C^{11}	1.29(1)	1.31(1)	1.30(1)	1.31(10
C^{11}–C^{12}	1.490(7)	1.490(8)	1.479(7)	1.486(9)

Scheme 3.



imide **VIII**. The latter undergoes cyclization into triazinobenzimidazole **VI** with elimination of salicylamide (**IX**) (Scheme 3).

A similar cleavage of 4-oxo-1,3-benzoxazinium perchlorates was formerly observed in reactions with indole and *o*-phenylenediamine [7].

Reaction of salts **I** and **II** with cyanoguanidine led to the formation of previously unknown cyanaminotriazines **X** containing several functional groups and capable of further chemical transformations. The reaction of azines **Xb** and **Xc** with *o*-phenylenediamine gave rise to triazinebenzimidazoleamines **V** (Scheme 4). In the IR spectra of compounds **X** the absorption bands of cyano group appeared at 2170–2155 cm^{-1}, and they disappeared after the reaction with *o*-phenylenediamine. In the ^{1}H NMR spectra of cyanaminotriazines **X** the signals are observed from the protons of R' group, of aromatic protons in the region 6.62–8.45 ppm, of groups OH and NH in the region 11–14 ppm as singlets and broadened singlets.

The structure of bisheterylamines **V** is consistent with their IR and ^{1}H NMR spectra.

EXPERIMENTAL

IR spectra of compounds obtained were registered on a spectrophotometer Specord 75IR from mulls in mineral oil, ^{1}H NMR spectra were taken on a spec-

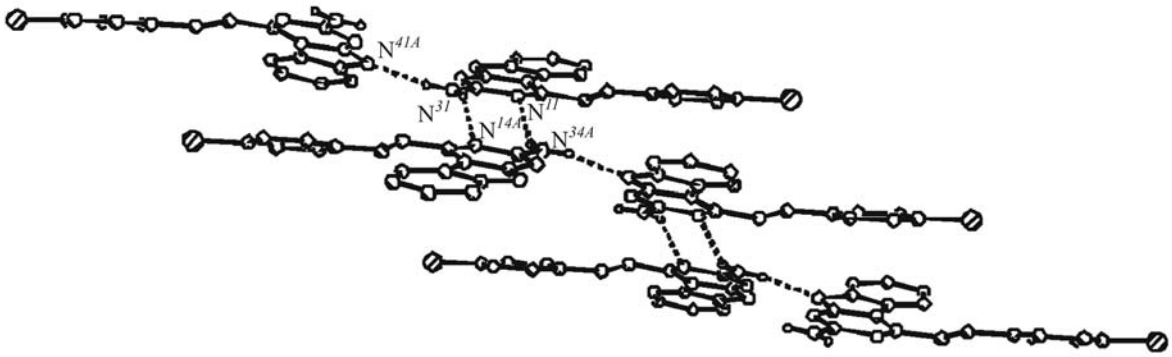
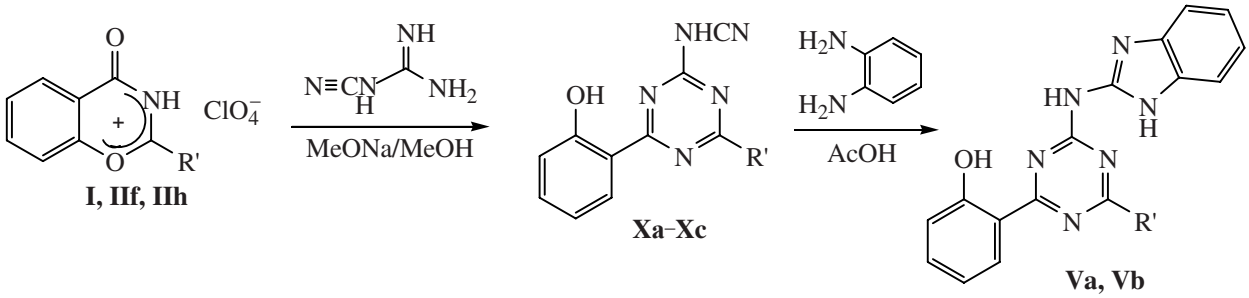


Fig. 2. Hydrogen bonds forming dimers and layers in the crystal of compound **VIa**.

Scheme 4.



I, IIf, IIh **Xa–Xc** **Va, Vb**

V, X, R' = Me (**a**), PhCH=CH (**b**), 2-thienyl-CH=CH (**c**).

trometer Varian Unity-300 (300 MHz) at 20°C in acetone-d_6 and DMSO-d_6, mass spectra were measured on a VG 7070E instrument (electron impact, 70 eV).

Perchlorates **I** and **IIh** were described in [3], compounds **IIa–IIg** were prepared by procedure described in the same paper.

2-[2-(2-Pyridyl)vinyl]-4-oxo-1,3-benzoxazinium perchlorate (IIa). Yield 99%, brown crystals, mp > 250°C. IR spectrum, ν, cm^{-1}: 1750 (C=O), 1680, 1640 (C=N), 1620 (C=C). Found, %: C 49.50; H 2.70; Cl 10.00; N 9.10. $C_{15}H_{11}ClNO_6$. Calculated, %: C 49.11; H 3.00; Cl 9.69; N 9.69.

2-[2-(4-Chlorophenyl)vinyl]-4-oxo-1,3-benzoxazinium perchlorate (IIb). Yield 79%, red crystals, mp 255–258°C. IR spectrum, ν, cm^{-1}: 1760 (C=O), 1680, 1640 (C=N), 1620 (C=C). Found, %: C 49.80; H 2.70; Cl 18.60; N 3.10. $C_{16}H_{11}Cl_2NO_6$. Calculated, %: C 50.00; H 2.86; Cl 18.49; N 3.65.

2-[2-(4-Dimethylaminophenyl)vinyl]-4-oxo-1,3-benzoxazinium perchlorate (IIc). Yield 79%, violet crystals, mp > 250°C. IR spectrum, ν, cm^{-1}: 1755 (C=O), 1680, 1650 (C=N), 1610 (C=C). Found, %: C 55.40; H 4.00; Cl 9.30; N 6.90. $C_{18}H_{17}ClN_2O_6$. Calculated, %: C 55.03; H 4.33; Cl 9.04; N 7.13.

2-[2-(4-Methylthiophenyl)vinyl]-4-oxo-1,3-benzoxazinium perchlorate (IId). Yield 99%, red crystals, mp > 260°C. IR spectrum, ν, cm^{-1}: 1680, 1640 (C=N), 1620 (C=C). Found, %: C 51.20; H 3.60; Cl 9.00; N 3.10; S 8.50. $C_{17}H_{14}ClNO_6S$. Calculated, %: C 51.58; H 3.54; Cl 8.98; N 3.54; S 8.09.

2-[2-(1-Methylindol-3-yl)vinyl]-4-oxo-1,3-benzoxazinium perchlorate (IIe). Yield 96%, red crystals, mp 235°C. IR spectrum, ν, cm^{-1}: 1640 (C=N), 1610, 1590 (C=C). Found, %: C 56.70; H 3.70; Cl 8.70; N 6.20. $C_{19}H_{15}ClN_2O_6$. Calculated, %: C 56.65; H 3.73; Cl 8.82; N 6.96.

2-[2-(2-thienyl)vinyl]-4-oxo-1,3-benzoxazinium perchlorate (IIf). Yield 98%, dark-green crystals, mp > 260°C. IR spectrum, ν, cm^{-1}: 1650 (C=N), 1620, 1600 (C=C). Found, %: C 47.30; H 2.60; Cl 9.20; N 3.70; S 8.80. $C_{14}H_{10}ClNO_6S$. Calculated, %: C 47.26; H 2.81; Cl 9.99; N 3.94; S 9.00.

2-{2-[4-(Carboxymethoxy)phenyl]vinyl}-4-oxo-1,3-benzoxazinium perchlorate (IIg). Yield 91%, light-brown crystals, mp > 270°C. IR spectrum, ν, cm^{-1}: 1670 (C=N), 1610, 1600 (C=C). Found, %: C 51.40; H 2.90; Cl 8.50; N 2.90. $C_{18}H_{13}ClNO_9$. Calculated, %: C 51.12; H 3.08; Cl 8.40; N 3.31.

2-Amino-4-(2-hydroxyphenyl)-6-[2-(2-pyridyl)-vinyl]-1,3,5-triazine (IIIa). To a solution of 10 mmol of sodium methylate (0.23 g of sodium in 10 ml of methanol) was added 1.08 g (5 mmol) of guanidine sulfate, 10 min later 1.8 g (5 mmol) of perchlorate **IIa** was added. After 16 h the reaction mixture was diluted with ice water, the separated precipitate was filtered off and washed with water. Yield 0.36 g (24%), brown crystals, mp 285°C (2-propanol). IR spectrum, ν, cm^{-1}: 3470 (NH_2), 3050 (OH), 1650, 1615 (C=N), 1600, 1525 (C=C). ^{1}H NMR spectrum, δ, ppm: 6.97–8.68 m (10H, H_{Ar}, 2H, NH_2), 13.16 s (1H, OH). Found, %: C 64.80; H 4.70; N 25.10. $C_{16}H_{13}N_5O$. Calculated, %: C 64.51; H 4.66; N 25.08.

Compounds **IIIb–IIIf** were obtained similarly.

2-Amino-4-(2-hydroxyphenyl)-6-[2-(4-chlorophenyl)vinyl]-1,3,5-triazine (IIIb). Yield 43%, yellow crystals, mp 222–224°C (2-butanol). IR spectrum, ν, cm^{-1}: 3400, 3300 (NH_2), 3180 (OH), 1625, 1610 (C=N), 1525 (C=C). ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 6.82–8.42 m (10H, H_{Ar}, 2H, NH_2), 13.29 s (1H, OH). Found, %: C 64.00; H 4.30; Cl 11.20; N 17.00. $C_{17}H_{13}ClN_5O$. Calculated, %: C 62.87; H 4.01; Cl 10.94; N 17.26.

2-Amino-4-(2-hydroxyphenyl)-6-[2-(4-dimethylaminophenyl)vinyl]-1,3,5-triazine (IIIc). Yield 38%, orange crystals, mp 255–256°C. IR spectrum, ν, cm^{-1}: 3390, 3300 (NH_2), 3150 (OH), 1630 (C=N), 1600, 1520 (C=C). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 3.09 s (6H, NMe_2) 6.65–8.39 m (10H, H_{Ar}, 2H, NH_2), 13.11 s (1H, OH). Found, %: C 68.40; H 5.70; N 21.00. $C_{20}H_{17}N_5O$. Calculated, %: C 68.47; H 5.71; N 21.02.

2-Amino-4-(2-hydroxyphenyl)-6-[2-(4-methylthiophenyl)vinyl]-1,3,5-triazine (IIId). Yield 43%, yellow crystals, mp 217–219°C. IR spectrum, ν, cm^{-1}: 3380, 3320 (NH_2), 3200 (OH), 1635 (C=N), 1595, 1520 (C=C). ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 2.45 s (3H, SMe), 6.82–8.41 m (10H, H_{Ar}, 2H, NH_2), 13.29 s (1H, OH). Found, %: C 64.30; H 4.70; N 16.60; S 9.40. $C_{18}H_{16}N_4OS$. Calculated, %: C 64.29; H 4.76; N 16.67; S 9.52.

2-Amino-4-(2-hydroxyphenyl)-6-[2-(1-methylindol-3-yl)vinyl]-1,3,5-triazine (IIIe). Yield 40%, yellow crystals, mp 210–212°C. IR spectrum, ν, cm^{-1}: 3470 (NH_2), 3050 (OH), 1680, 1630 (C=N), 1600, 1525 (C=C). ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 3.95 s (3H, NMe), 6.94–8.47 m (11H, H_{Ar}, 2H, NH_2), 13.54 s (1H, OH). Found, %: C 70.20; H 4.60; N 20.50. $C_{20}H_{17}N_5O$. Calculated, %: C 70.17; H 4.67; N 20.46.

2-Amino-4-(2-hydroxyphenyl)-6-[2-(thienyl)-vinyl]-1,3,5-triazine (IIIe). Yield 42%, light-brown crystals, mp 158–160°C (1-butanol). IR spectrum, ν, cm^{-1}: 3375, 3320 (NH_2), 3180 (OH), 1640, 1610 (C=N), 1590, 1570 (C=C). ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 6.77–8.50 m (9H, H_{Ar}, 2H, NH_2), 13.11 s (1H, OH). Found, %: C 60.70; H 4.20; N 18.70; S 10.90. $C_{15}H_{12}N_4OS$. Calculated, %: C 60.81; H 4.05; N 19.92; S 10.81.

1,3-Benzoxazol-2-yl{4-(2-hydroxyphenyl)-6-[2-(4-dimethylaminophenyl)vinyl]-1,3,5-triazin-2-yl}amine (IVa). To a solution of 10 mmol of sodium methylate (0.23 g of sodium in 10 ml of methanol) was added 1.74 g (9.9 mmol) of benzoxazolylguanidine and 3.92 g (9.9 mmol) of compound **IIc**. Then the reaction mixture was twice heated to boiling with an interval of 20 min, and then it was left standing at room temperature.After 16 h the separated reaction product was diluted with ice water, filtered off, recrystallized, and dried. Yield 40%, red crystals, mp 192–194°C (DMF). IR spectrum, ν, cm^{-1}: 3390 (NH), 3100 (OH), 1670, 1630 (C=N), 1600 (C=C). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 3.05 s (6H, NMe_2), 6.62–8.12 m (14H, H_{Ar},), 10.77 C (1H, OH), 11.57 C (1H, NH). Found, %: C 69.00; H 4.50; N 18.90. $C_{26}H_{22}N_6O_2$. Calculated, %: C 69.33; H 4.89; N 18.67.

Compounds **IVb** and **IVc** were prepared in the same way.

1,3-Benzoxazol-2-yl{4-(2-hydroxyphenyl)-6-[2-(1-methyl-1*H*-indol-3-yl)vinyl]-1,3,5-triazin-2-yl}amine (IVb). Yield 30%, yellow crystals, mp > 310°C (*i*-BuOH–DMF). IR spectrum, ν, cm^{-1}: 3370 (NH), 3050 (OH), 1680, 1630 (C=N), 1600, 1525 (C=C). ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 3.95 s (3H, NMe), 6.94–8.47 m (15H, H_{Ar}), 13.54 s (1H, OH), 13.60 s (1H, NH). Found, %: C 67.90; H 4.80; N 20.10. $C_{27}H_{20}N_6O_2$. Calculated, %: C 67.92; H 4.71; N 19.81.

1,3-Benzoxazol-2-yl[4-(2-hydroxyphenyl)-6-methyl-1,3,5-triazin-2-yl]amine (IVc). Yield 45%, pink crystals, mp 290°C (*i*-BuOH). IR spectrum, ν, cm^{-1}: 3350 (NH), 3090 (OH), 1660, 1660 (C=N), 1600 (C=C). ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 2.58 s (3H, Me) 6.91–8.43 m (8H, H_{Ar}), 12.56 s (1H, OH) 12.98 s (1H, NH). Found, %: C 63.60; H 4.20; N 21.80. $C_{17}H_{13}N_5O_2$. Calculated, %: C 63.95; H 4.08; N 21.94.

2-[4-(1*H*-Benzimidazol-2-ylamino)-6-methyl-1,3,5-triazin-2-yl]phenol (Va). A mixture of 2.27 g (10 mmol) of compound **Xa** and 1.08 g (10 mmol) of *o*-phenylenediamine was boiled for 2 h in 10 ml of acetic acid. Then the reaction mixture was diluted with cold water, the separated precipitate was filtered off and washed with water. Yield 29%, light-yellow crystals, mp > 310°C (DMTA). IR spectrum, ν, cm^{-1}: 3380 (NH), 1640, 1610 (C=N), 1525, 1500 (C=C). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.8 s (3H, Me), 6.85–8.40 m (8H, H_{Ar}) 12.1 s (1H, OH), 12.2 s (1H, NH) 13.2 s (1H, NH). Found, %: C 64.10; H 4.50; N 26.20. $C_{17}H_{14}N_6O$. Calculated, %: C 64.15; H 4.40; N 26.42.

In the same way from compound **Xb** compound **Vb** was prepared.

2-{4-(1*H*-Benzimidazol-2-ylamino)-6-[(*E*)-2-phenylethenyl]-1,3,5-triazin-2-yl}phenol (Vb). Yield 37%, yellow crystals, mp > 310°C (DMF). IR spectrum, ν, cm^{-1}: 3280 (NH), 1720, 1690, 1635 (C=N), 1600, 1540 (C=C). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 6.95–8.71 m (15H, H_{Ar}), 12.11 s (1H, OH), 12.29 s (1H, NH), 13.10 s (1H, NH). Found, %: C 70.90; H 4.50; N 20.40. $C_{24}H_{18}N_6O$. Calculated, %: C 70.94; H 4.43; N 20.69.

4-[(*E*)-2-(4-Chlorophenyl)ethenyl][1,3,5]triazino-[1.2-*a*]benzimidazol-2-amine (VIa). To a solution of 0.23 g of sodium in 10 ml of methanol was added 1.41 g (8.2 mmol) of benzimidazoleguanidine and 3.15 g (8.2 mmol) of compound **IIb**. The mixture was boiled for 60 min, then it was diluted with water, the separated

precipitate was filtered off and washed with water. Yield 1.62 g (28%), red crystals, mp 280–282°C (DMF). IR spectrum, ν, cm^{-1}: 3320 (NH_2), 1660, 1650 (C=N), 1600, 1520 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 7.22–8.49 m (12H, H_{Ar}). Found, %: C 63.40; H 3.60; Cl 11.10; N 21.70. $C_{17}H_{12}ClN_5$. Calculated, %: C 63.45; H 3.73; Cl 11.01; N 21.77.

Compounds **VIb–VIe** were prepared in the same way.

4-[(*E*)-2-(4-Dimethylaminophenyl)ethenyl][1,3,5]-triazino[1,2-*a*]benzimidazol-2-amine (VIb). Yield 32%, red crystals, mp 304–306°C (DMF). IR spectrum, ν, cm^{-1}: 1670, 1610 (C=N), 1590, 1500 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 3.11 s (6H, NMe_2), 6.72–8.19 m (12H, H_{Ar}). Found, %: C 69.10; H 5.30; N 25.40. $C_{19}H_{18}N_6$. Calculated, %: C 69.09; H 5.45; N 25.45.

4-[(*E*)-2-(2-thienyl)ethenyl][1,3,5]triazino[1,2-*a*]benzimidazol-2-amine (VIc). Yield 28%, red crystals, mp 277–279°C (1-butanol). IR spectrum, ν, cm^{-1}: 3340 (NH_2), 1650 (C=N), 1525 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 7.05–8.42 m (11H, H_{Ar}). Found, %: C 61.70; H 3.50; N 23.70; S 10.70. $C_{15}H_{11}N_5S$. Calculated, %: C 61.43; H 3.75; N 23.89; S 10.92.

{4-[(*E*)-2-(2-Amino[1,3,5]triazino[1,2-*a*]benzimidazol-4-yl)ethenyl]phenoxy}acetic acid (VId). Yield 26%, red crystals, mp 220–222°C (DMF). IR spectrum, ν, cm^{-1}: 3380 (NH_2), 1640, 1610 (C=N), 1530, 1500 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 4.69 s (2H, OCH_2), 6.82–8.15 m (12H, H_{Ar}). Found, %: C 63.10; H 4.10; N 19.30. $C_{19}H_{15}N_5O_3$. Calculated, %: C 63.16; H 4.16; N 19.39.

4-Methyl[1,3,5]triazino[1,2-*a*]benzimidazol-2-amine (VIe). Yield 30%, light-brown crystals, mp 316–318°C (DMF) (314–315°C [6]). IR spectrum, ν, cm^{-1}: 3430 (NH_2), 1640, 1610 (C=N), 1540 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 2.89 s (3H, Mε) 7.18–7.85 m (6H, H_{Ar}). Mass spectrum, *m/z* (I_{rel}, %): 199 (96), 158 (100), 131 (10), 104 (9), 90 (20), 63 (9), 42 (34). Found, %: C 60.20; H 4.50; N 35.10. $C_{10}H_9N_5$. Calculated, %: C 60.30; H 4.52; N 35.18.

***N*-[4-(2-Hydroxyphenyl)-6-methyl-1,3,5-triazin-2-yl]cyanamide (Xa).** To a solution of 10 mmol of sodium methylate (0.23 g of sodium in 10 ml of methanol) was added 0.84 g (10 mmol) of cyanoguanidine and 2.36 g (10 mmol) of perchlorate **I**. The reaction mixture was maintained at room temperature for 12 h, then ice water was added thereto, the separated precipitate was filtered off, washed with water, and recrystallized. Yield 55%, light-yellow crystals, mp 255–256°C (butanol). IR spectrum, ν, cm^{-1}: 3340 (NH), 2155 (C≡N), 1600 (C=N), 1540 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 2.37 s (3H, Me), 6.88–8.29 m (4H, H_{Ar}), 13.1 s (1H, OH), 13.78 s (1H, NH). Found, %: C 58.20; H 3.90; N 30.80. $C_{11}H_9N_5O$. Calculated, %: C 58.15; H 3.96; N 30.84.

Compounds **Xb** and **Xc** were similarly prepared.

***N*-{4-(2-Hydroxyphenyl)-6-[(*E*)-2-phenylethenyl]-1,3,5-triazin-2-yl}cyanamide (Xb).** Yield 49%, yellow crystals, mp 210°C (butanol). IR spectrum, ν, cm^{-1}: 3470 (NH), 2160 (C≡N), 1680, 1630 (C=N), 1600, 1525 (C=C). ^1H NMR spectrum (acetone-d_6), δ, ppm: 6.72–8.89 m (11H, H_{Ar}), 13.89 s (1H, OH), 14.01 s (1H, NH). Found, %: C 68.60; H 4.00; N 22.30. $C_{18}H_{13}N_5O$. Calculated, %: C 68.57; H 4.13; N 22.22.

***N*-{4-(2-Hydroxyphenyl)-6-[(*E*)-2-(2-thienyl)-ethenyl]-1,3,5-triazin-2-yl}cyanamide (Xc).** Yield 22%, brown crystals, mp > 310°C (butanol). IR spectrum, ν, cm^{-1}: 3400 (NH), 2170 (C≡N), 1650, 1605 (C=N), 1510, 1500 (C=C). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 6.65–8.39 m (9H, H_{Ar}), 13.91 s (1H, OH), 14.09 s (1H, NH). Found, %: C 59.70; H 3.30; N 20.50; S 9.80. $C_{16}H_{11}N_5OS$. Calculated, %: C 59.81; H 3.43; N 21.81; S 9.97.

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