Chemistry of Heterocyclic Compounds, Vol. 49, No. 9, December, 2013 (Russian Original Vol. 49, No. 9, September, 2013)

SUBSTITUTED 2-FORMYLBENZOIC ACIDS IN THE SYNTHESIS OF 11*H*-ISOINDOLO[2,1-*a*]BENZIMIDAZOL-11-ONES, 5*H*-ISOINDOLO[2,1-*a*][3,1]BENZOXAZINE-5,11(6a*H*)-DIONES, AND 6,6a-DIHYDROISOINDOLO-[2,1-*a*]QUINAZOLINE-5,11-DIONES

E. V. Gromachevskaya[1]*, A. V. Fin'ko[1], A. V. Butin[1,2], K. S. Pushkareva[1], V. D. Strelkov[1], L. I. Isakova[1], and G. D. Krapivin[1]

Optimal conditions were developed for the synthesis of 11H-isoindolo[2,1-a]benzimidazol-11-one, 5H-isoindolo[2,1-a][3,1]benzoxazine-5,11(6aH)-dione, and 6,6a-dihydroisoindolo[2,1-a]quinazoline-5,11-dione derivatives in the reaction of substituted 2-formylbenzoic acids with o-phenylenediamine, anthranilic acid, and anthranilamide, respectively. The bifolded structure of 6,6a-dihydroisoindolo-[1,2-a]quinazoline-5,11-dione was verified and investigated.

Keywords: anthranilamide, anthranilic acid, 2-formylbenzoic acid, isoindolone, *o*-phenylenediamine, heterocyclization domino reaction, N,N-bis-nucleophiles, N,O-bis-nucleophiles.

In the previous communication we described the domino reactions of substituted 2-formylbenzoic acids (FBAs) with 2-(1-aminoalkyl)phenols and 2-aminophenyl(diethyl, diphenyl)carbinols, which lead to the formation of 10*H*-isoindolo[2,1-*b*][1,3]benzoxazin-12(4b*H*)-ones and 5*H*-isoindolo[2,1-*a*][3,1]benzoxazin-11(6a*H*)-ones, respectively [1]. The derivatives of the latter compound were found to have a pesticidal activity [2].

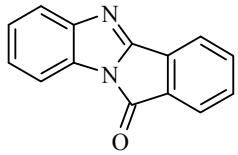
These results served as prerequisite for the reaction of FBAs with *o*-phenylenediamine (PDA), anthranilic acid, and anthranilamide aimed for the synthesis of new derivatives of tetracyclic heterosystems containing an isoindolone fragment: 11*H*-isoindolo[2,1-*a*]benzimidazol-11-one, 5*H*-isoindolo[2,1-*a*][3,1]benz-oxazine-5,1(6a*H*)-dione, and 6,6a-dihydroisoindolo[2,1-*a*]quinazoline-5,11-dione. The isoindolone fragment is present in the composition of natural compounds (alkaloids [3, 4]) and apparently makes a definite contribution to the display of one or other type of biological activity [5-8].

Various products from the reaction of FBA with PDA have been described in the literature. Thus, 2-(1*H*-benzimidazol-2-yl)benzoic acid was obtained by refluxing equimolar amounts of the reagents in methanol under acid catalysis [9], 4b,5-dihydro-11*H*-isoindolo[2,1-*a*]benzimidazol-11-one was obtained by

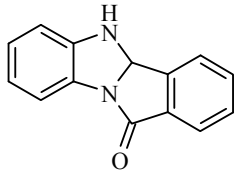
*To whom correspondence should be addressed, e-mail: organics@kubstu.ru.

[1]Kuban State Technological University, 2 Moskovskaya St., Krasnodar 350072, Russia.
[2]Perm State University, 15 Bukireva St., 15, Perm 614990, Russia; e-mail: alexander_butin@mail.ru.

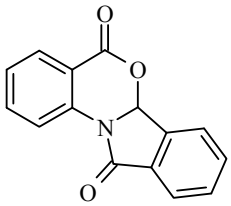
Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 9, pp. 1428-1442, September, 2013. Original article submitted June 26, 2013.



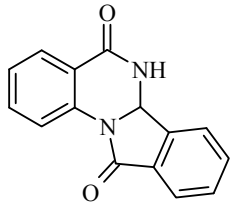
11*H*-Isoindolo-
[2,1-*a*]benzimidazol-11-one



4b,5-Dihydro-11*H*-isoindolo-
[2,1-*a*]benzimidazol-11-one



5H-Isoindolo-
[2,1-*a*][3,1]benzoxazine-5,11(6a*H*)-dione

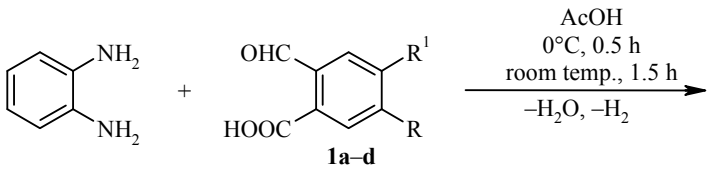


6,6a-Dihydroisoindolo-
[2,1-*a*]quinazoline-5,11-dione

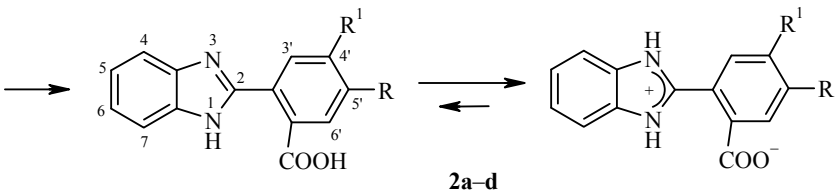
refluxing in the presence of TsOH with azeotropic distillation of the water [6]. A. Cul et al. [10] obtained 11*H*-isoindolo[2,1-*a*]benzimidazol-11-one by refluxing the starting materials (12 h) in toluene in the presence of TsOH.

Various versions of the reaction of PDA with phthalic anhydride have been also used for the preparation of 11*H*-isoindolo[2,1-*a*]benzimidazol-11-ones: refluxing equimolar amounts of the reagents in *n*-amyl alcohol [5]; fusion without a solvent [11]; with microwave irradiation in solution [12].

In the present work, we used 2-(1*H*-benzimidazol-2-yl)benzoic acids **2a-d** to produce 11*H*-isoindolo[2,1-*a*]benzimidazol-11-one derivatives. We found that compounds **2a-d** are formed readily in the reaction of equimolar amounts of the corresponding FBAs **1a-d** and PDA at room temperature in acetic acid, which apparently serves as both solvent and a catalyst. Although the process occurs as a sequence of cyclization and dehydrogenation (oxidation) reactions [13] it is impossible to isolate the intermediates under the given conditions. The reaction is complete in 1.5-2.0 h, following the filtration of colorless crystals of the products **2a-d** from the reaction mixture.



1a–d



2a–d

a R = R^1 = H; **b** R = Cl, R^1 = H; **c** R = NO_2, R^1 = H; **d** R = R^1 = OMe

The composition and structure of the synthesized compounds were established by elemental analysis, ^{1}H and ^{13}C NMR spectroscopy, and mass spectrometry (Tables 1-3). The IR spectra of compounds **2a-d** do not contain bands for the stretching vibrations of the C=O bond of the carboxyl group in the region of 1720-1680 cm^{-1} characteristic of the starting acids **1a-d**, but there are strong absorption bands at 1605-1580 and 1390-1360 cm^{-1}

TABLE 1. Physicochemical Characteristics of the Synthesized Compounds

Compound	Empirical formula	Found, % / Calculated, %				Mp, °C	R_f	Yield, %
		C	H	N	Hal			
2a	$C_{14}H_{10}N_2O_2$	70.21 / 70.58	4.42 / 4.23	11.58 / 11.76	—	243-245 (245 [33])	0.11	72
2b	$C_{14}H_9ClN_2O_2$	61.73 / 61.66	3.26 / 3.33	10.18 / 10.27	13.07 / 13.00	228-290	0.05	74
2c	$C_{14}H_9N_3O_4$	59.45 / 59.37	3.09 / 3.20	14.71 / 14.84	—	290-293	0.07	65
2d	$C_{16}H_{14}N_2O_4$	64.22 / 64.42	4.85 / 4.73	9.21 / 9.39	—	230-233	0.10	77
3a	$C_{14}H_8N_2O$	76.52 / 76.35	3.45 / 3.66	12.52 / 12.72	—	>290 (decomp.) (>290 [5])	0.60	70
3b	$C_{14}H_7ClN_2O$	66.36 / 66.03	2.50 / 2.77	11.35 / 11.00	14.20 / 13.92	190-192	0.71	72
3c	$C_{14}H_7N_3O_3$	63.75 / 63.40	2.31 / 2.66	15.48 / 15.84	—	242-245	0.80	80
3d	$C_{16}H_{12}N_2O_3$	68.71 / 68.56	4.05 / 4.32	10.21 / 9.99	—	230-233	0.37	75
4b	$C_{15}H_{10}ClNO_4$	59.28 / 59.32	3.51 / 3.32	4.36 / 4.61	11.82 / 11.67	211-213	0.53	70
4c	$C_{15}H_{10}N_2O_6$	57.10 / 57.33	3.35 / 3.21	8.64 / 8.91	—	207-209	0.71	75
5a	$C_{15}H_9NO_3$	71.52 / 71.71	3.83 / 3.61	5.37 / 5.58	—	210-212 (219 [19])	0.45	77
5b	$C_{15}H_8ClNO_3$	63.26 / 63.06	2.65 / 2.82	4.78 / 4.90	12.32 / 12.41	>250 (decomp.)	0.52	60
5c	$C_{15}H_8N_2O_5$	60.65 / 60.82	2.92 / 2.72	9.58 / 9.46	—	261-263	0.80	65
5d	$C_{17}H_{13}NO_5$	65.32 / 65.59	4.05 / 4.21	4.71 / 4.50	—	232-235	0.68	52
6a	$C_{15}H_{10}N_2O_2$	72.39 / 71.99	3.90 / 4.03	11.56 / 11.19	—	255-258 (250-253 [25])	0.54	80
6b	$C_{15}H_9ClN_2O_2$	63.10 / 63.28	3.25 / 3.19	9.75 / 9.84	12.31 / 12.45	292-295	0.53	78
6c	$C_{15}H_9N_3O_4$	61.41 / 61.02	3.27 / 3.07	14.15 / 14.23	—	212-215	0.10	72
6d	$C_{17}H_{14}N_2O_4$	65.45 / 65.80	4.71 / 4.55	8.85 / 9.03	—	298-300	0.15	52
6e	$C_{15}H_9BrN_2O_2$	54.75 / 54.74	2.35 / 2.76	8.63 / 8.51	24.07 / 24.28	265-267	0.56	80
6f	$C_{15}H_9IN_2O_2$	47.55 / 47.90	2.23 / 2.41	7.30 / 7.45	33.52 / 33.74	282-284	0.60	73
6g	$C_{16}H_{12}N_2O_3$	68.57 / 68.56	4.28 / 4.32	10.00 / 9.99	—	286-289	0.20	80

belonging to the asymmetric and symmetric stretching vibrations of the carboxylate anion. In the region of 2250-1800 cm^{-1} there are broad absorption bands belonging to the protonated amino groups [14, 15]. In the crystalline state the compounds **2a-d** are present in the zwitterionic form. In the ^{1}H NMR spectra recorded in DMSO-d_6 the signals of the protons of the carboxyl and amine groups are not recorded due to fairly fast exchange on the NMR time scale. The spectra contain signals for the aromatic protons in the region of 7.22-8.54 ppm, while the spectrum of compound **2d** also contains signals for the two methoxy groups (Table 2).

TABLE 2. Spectral Characteristics of the Synthesized Compounds

Com-pound	IR spectrum, ν, cm^{-1}	^1H NMR spectrum, δ, ppm (*J*, Hz)
1	2	3
2a	2000-1900 (N^+H); 1590, 1370 (COO^-)	7.26 (1H, dd, *J* = 6.0, *J* = 3.0, H-7); 7.28 (1H, dd, *J* = 6.0, *J* = 3.0, H-4); 7.61 (1H, dt, *J* = 6.0, *J* = 3.0, H-6); 7.63 (1H, dt, *J* = 6.0, *J* = 3.0, H-5); 7.65 (1H, dt, *J* = 7.0, *J* = 1.5, H-4'); 7.72 (1H, dt, *J* = 7.0, *J* = 1.5, H-5'); 7.82 (1H, dd, *J* = 7.0, *J* = 1.5, H-3'); 7.88 (1H, dd, *J* = 7.0, *J* = 1.5, H-6')
2b	2250 (N^+H); 1580, 1370 (COO^-)	7.23 (1H, dd, *J* = 6.0, *J* = 3.0, H-7); 7.25 (1H, dd, *J* = 6.0, *J* = 3.0, H-4); 7.59 (1H, dt, *J* = 6.0, *J* = 3.0, H-6); 7.61 (1H, dt, *J* = 6.0, *J* = 3.0, H-5); 7.77 (1H, dd, *J* = 8.7, *J* = 2.0, H-4'); 7.83-7.87 (2H, m, H-3',6')
2c	2000-1800 (N^+H); 1580, 1390 (COO^-); 1510, 1328 (NO_2)	7.27 (1H, dd, *J* = 6.0, *J* = 3.0, H-7); 7.29 (1H, dd, *J* = 6.0, *J* = 3.0, H-4); 7.64 (1H, dt, *J* = 6.0, *J* = 3.0, H-6); 7.66 (1H, dt, *J* = 6.0, *J* = 3.0, H-5); 8.12 (1H, dd, *J* = 7.0, *J* = 2.4, H-4'); 8.50-8.54 (2H, m, H-3',6')
2d	1900-1800 (N^+H); 1590, 1360 (COO^-)	3.89 (3H, s, OCH_3), 3.91 (3H, s, OCH_3); 7.24 (1H, dd, *J* = 6.0, *J* = 3.0, H-7); 7.26 (1H, dd, *J* = 6.0, *J* = 3.0, H-4); 7.38 (1H, s, H-3'); 7.53 (1H, s, H-6'); 7.60 (1H, dt, *J* = 6.0, *J* = 3.0, H-6); 7.62 (1H, dt, *J* = 6.0, *J* = 3.0, H-5)
3a	1735 (C=O); 1648 (C=N)	7.10-7.30 (2H, m, H-7,8); 7.35 (1H, d, *J* = 8.1, H-6); 7.52 (1H, d, *J* = 8.1, H-9); 7.71-7.75 (4H, m, H-1,2,3,4)
3b	1750 (C=O); 1610 (C=N)	7.28 (1H, t, *J* = 6.0, H-8); 7.37 (1H, t, *J* = 6.0, H-7); 7.60-7.75 (2H, m, H-6,9); 7.77-7.83 (2H, m, H-3,4); 7.90 (1H, s, H-1)
3c	1750 (C=O); 1600 (C=N); 1450, 1320 (NO_2)	7.39 (1H, dt, *J* = 7.5, *J* = 1.2, H-8); 7.47 (1H, dt, *J* = 7.5, *J* = 1.2, H-7); 7.78 (1H, dd, *J* = 7.5, *J* = 1.2, H-6); 7.81 (1H, dd, *J* = 7.5, *J* = 1.2, H-9); 8.14 (1H, d, *J* = 8.1, H-4); 8.35 (1H, dd, *J* = 8.1, *J* = 2.1, H-3); 8.48 (1H, d, *J* = 2.1, H-1)
3d	1730 (C=O); 1590 (C=N)	3.90 (3H, s, OCH_3); 4.00 (3H, s, OCH_3); 7.27 (1H, t, *J* = 7.0, H-8); 7.32 (1H, t, *J* = 7.0, H-7); 7.40 (1H, s, H-4); 7.45 (1H, s, H-1); 7.58-7.72 (2H, m, H-6,9)
4b	3200-2500, 1780 (COOH); 3350 (NH); 1680 (C=O)	6.93 (1H, dd, *J* = 7.9, *J* = 7.5, H-7); 7.27 (1H, d, *J* = 7.9, H-8); 7.31 (1H, d, *J* = 8.0, H-5); 7.58 (1H, dd, *J* = 7.5, *J* = 8.0, H-6); 7.83 (1H, d, *J* = 8.7, 2-CH); 7.91-7.96 (2H, m, H-3',4'); 7.98 (1H, d, *J* = 1.5, H-6'); 8.70 (1H, d, *J* = 8.7, NH); 13.13 (1H, br. s, COOH)
4c	3200-2500, 1770 (COOH); 3340 (NH); 1650 (C=O); 1500, 1330 (NO_2)	6.94 (1H, dd, *J* = 8.0, *J* = 7.5, H-7); 7.32 (1H, d, *J* = 8.0, H-8); 7.43 (1H, d, *J* = 8.0, H-5); 7.59 (1H, dd, *J* = 8.0, *J* = 7.5, H-6); 7.93 (1H, d, *J* = 9.8, H-3'); 8.07 (1H, d, *J* = 10.3, 2-CH); 8.57 (1H, d, *J* = 1.6, H-6'); 8.68 (1H, dd, *J* = 9.8, *J* = 1.6, H-4'); 8.79 (1H, d, *J* = 10.3, NH); 13.20 (1H, br. s, COOH)

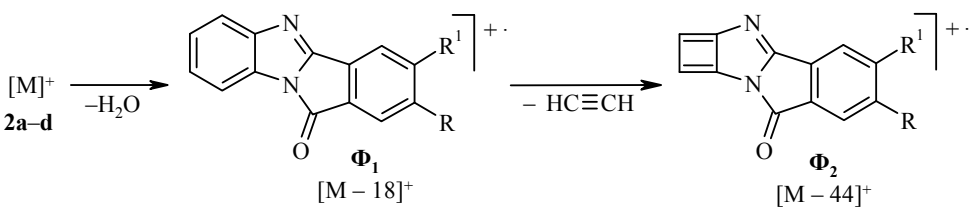
TABLE 2 (continued)

1	2	3
5a	1750 (COO); 1720 (CON)	6.66 (1H, s, 6a-CH); 7.35 (1H, ddd, *J* = 7.8, *J* = 7.5, *J* = 1.0, H-2); 7.66 (1H, ddd, *J* = 7.5, *J* = 7.4, *J* = 1.2, H-3); 7.72-7.79 (3H, m, H-7,8,9); 7.97 (1H, dd, *J* = 7.4, *J* = 1.0, H-4); 8.15 (1H, dd, *J* = 7.8, *J* = 1.2, H-1); 8.18 (1H, dd, *J* = 7.4, *J* = 0.6, H-10)
5b	1735 (COO); 1700 (CON)	7.13 (1H, s, 6a-CH); 7.45 (1H, dd, *J* = 8.5, *J* = 8.0, H-3); 7.85 (1H, dd, *J* = 8.5, *J* = 8.0, H-2); 7.90 (1H, d, *J* = 7.7, H-8); 7.97 (1H, d, *J* = 7.7, H-7); 8.00 (1H, s, H-10); 8.06 (1H, d, *J* = 8.5, H-4); 8.08 (1H, d, *J* = 8.5, H-1)
5c	1750 (COO); 1720 (CON); 1530, 1310 (NO_2)	7.25 (1H, s, 6a-CH); 7.47 (1H, dd, *J* = 8.4, *J* = 7.7, H-2); 7.89 (1H, dd, *J* = 8.4, *J* = 7.7, H-3); 8.08 (1H, d, *J* = 6.7, H-7); 8.12 (1H, d, *J* = 6.7, H-8); 8.19 (1H, d, *J* = 8.4, H-4); 8.55 (1H, s, H-10); 8.65 (1H, d, *J* = 8.4, H-1)
5d	1740 (COO); 1720 (CON)	3.86 (3H, s, OCH_3); 3.89 (3H, s, OCH_3); 6.96 (1H, s, 6a-CH); 7.32-7.38 (2H, m, H-3,7); 7.40 (1H, s, H-10); 7.79 (1H, dd, *J* = 8.0, *J* = 7.5, H-2); 7.98 (1H, d, *J* = 8.0, H-4); 8.00 (1H, d, *J* = 8.0, H-1)
6a	3170 (NH); 1720, 1680 (C=O)	6.41 (1H, s, 6a-CH); 7.30 (1H, dd, *J* = 8.1, *J* = 8.0, H-9); 7.61-7.65 (2H, m, H-2,8); 7.72 (1H, dd, *J* = 7.9, *J* = 7.5, H-3); 7.84 (1H, d, *J* = 7.9, H-4); 7.90 (1H, d, *J* = 7.6, H-7); 7.98 (1H, d, *J* = 8.1, H-10); 8.08 (1H, d, *J* = 8.3, H-1); 8.92 (1H, br. s, NH)
6b	3210 (NH); 1710, 1676 (C=O)	6.47 (1H, s, 6a-CH); 7.33 (1H, dd, *J* = 8.0, *J* = 7.6, H-3); 7.68 (1H, dd, *J* = 8.4, *J* = 7.6, H-2); 7.83 (1H, d, *J* = 8.0, H-8); 7.86 (1H, s, H-10); 7.89 (1H, d, *J* = 8.0, H-7); 7.97 (1H, d, *J* = 8.4, H-4); 8.05 (1H, d, *J* = 8.0, H-1); 9.39 (1H, s, NH)
6c	3210 (NH); 1750, 1670 (C=O); 1510, 1330 (NO_2)	6.92 (1H, d, *J* = 9.4, 6a-CH); 6.96 (1H, dd, *J* = 7.7, *J* = 7.6, H-2); 7.29 (1H, d, *J* = 8.5, H-4); 7.49-7.53 (2H, m, H-1,3); 7.87 (1H, d, *J* = 8.2, H-7); 8.61 (1H, dd, *J* = 8.2, *J* = 2.1, H-8); 8.76 (1H, d, *J* = 2.1, H-10); 9.01 (1H, d, *J* = 9.4, NH)
6d	3200 (NH); 1730, 1680 (C=O)	3.85 (3H, s, OCH_3); 3.90 (3H, s, OCH_3); 6.25 (1H, s, 6a-CH); 7.21 (1H, s, H-7); 7.25 (1H, dd, *J* = 7.7, *J* = 7.3, H-2); 7.40 (1H, s, H-10); 7.60 (1H, dd, *J* = 7.3, *J* = 7.5, H-3); 7.88 (1H, d, *J* = 7.5, H-4); 7.98 (1H, d, *J* = 7.7, H-1); 9.27 (1H, br. s, NH)
6e	3180 (NH); 1710, 1680 (C=O)	6.45 (1H, s, 6a-CH); 7.33 (1H, ddd, *J* = 7.8, *J* = 7.5, *J* = 1.0, H-3); 7.67 (1H, ddd, *J* = 8.0, *J* = 7.5, *J* = 1.5, H-2); 7.78 (1H, d, *J* = 8.1, H-10); 7.84 (1H, dd, *J* = 8.1, *J* = 1.6, H-9); 7.92 (1H, dd, *J* = 7.8, *J* = 1.5, H-4); 8.00 (1H, dd, *J* = 8.0, *J* = 1.0, H-1); 8.07 (1H, d, *J* = 1.6, H-7); 9.31 (1H, s, NH)
6f	3172 (NH); 1729, 1674 (C=O)	6.39 (1H, s, 6a-CH); 7.32 (1H, t, *J* = 8.3, H-2); 7.65-7.71 (2H, m, H-3,7); 7.98 (1H, d, *J* = 8.3, H-4); 8.06 (1H, d, *J* = 8.5, H-8); 8.09 (1H, d, *J* = 8.3, H-1); 8.14 (1H, s, H-10); 8.96 (1H, br. s, NH)
6g	3190 (NH); 1725, 1670 (C=O)	3.89 (3H, s, OCH_3); 6.41 (1H, s, 6a-CH); 7.30 (1H, dd, *J* = 8.2, *J* = 1.5, H-8); 7.32 (1H, d, *J* = 1.5, H-10); 7.33 (1H, dd, *J* = 7.7, *J* = 7.6, H-2); 7.66 (1H, ddd, *J* = 8.0, *J* = 7.6, *J* = 1.5, H-3); 7.77 (1H, d, *J* = 8.2, H-7); 7.93 (1H, dd, *J* = 7.7, *J* = 1.5, H-1); 8.01 (1H, d, *J* = 8.0, H-4); 9.33 (1H, s, NH)

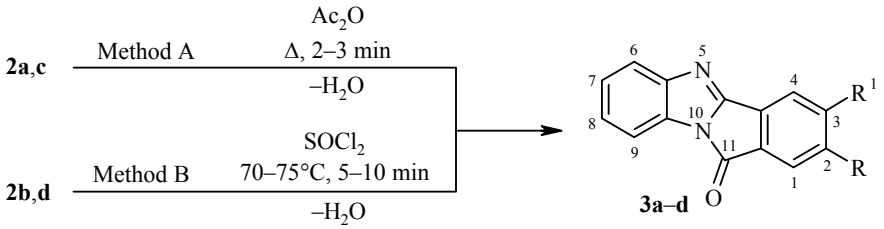
TABLE 3. Mass spectra of the Synthesized Compounds

Compound	m/z (I_{rel}, %)
2a	238 $[M]^+$ (10), 220 (15), 194 (100), 193 (20), 165 (8), 130 (12), 104 (20), 102 (25), 97 (12), 90 (40), 76 (42), 63 (67)
2b	274 $[M\ (^{37}Cl)]^+$ (4), 272 $[M\ (^{35}Cl)]^+$ (12), 256 (16), 254 (50), 230 (35), 228 (100), 211 (10), 193 (8), 191 (15), 167 (10), 149 (11), 137 (10), 124 (15), 104 (17), 101 (18), 92 (18), 77 (20)
2c	283 $[M]^+$ (26), 265 (7), 239 (96), 219 (17), 193 (100), 192 (35), 164 (16), 90 (20), 77 (16), 63 (48)
2d	298 $[M]^+$ (40), 280 (82), 265 (20), 254 (100), 252 (55), 239 (29), 237 (30), 211 (32), 195 (22), 168 (45), 140 (36), 92 (67), 77 (25)
3a	220 $[M]^+$ (4), 207 (10), 206 (100), 194 (12), 179 (20), 177 (18), 152 (11), 151 (20), 104 (7), 103 (22), 89 (20), 76 (33)
3b	256 $[M\ (^{37}Cl)]^+$ (37), 254 $[M\ (^{35}Cl)]^+$ (100), 239 (3), 191 (12), 165 (5), 136 (7), 127 (6), 90 (8), 76 (5), 63 (9)
3c	265 $[M]^+$ (100), 235 (53), 219 (62), 191 (22), 165 (12), 164 (19), 140 (7), 90 (7), 76 (8), 75 (10)
3d	280 $[M]^+$ (100), 265 (27), 237 (40), 235 (15), 207 (12), 194 (38), 179 (10), 166 (12), 152 (8), 140 (15), 104 (5), 91 (8), 77 (8)
4b	305 $[M\ (^{37}Cl)]^+$ (7), 303 $[M\ (^{35}Cl)]^+$ (22), 285 (5), 259 (7), 257 (8), 230 (10), 214 (13), 180 (10), 169 (30), 167 (100), 139 (35), 119 (22), 75 (35)
4c	314 $[M]^+$ (100), 296 (60), 268 (18), 251 (15), 241 (19), 225 (24), 205 (10), 178 (80), 132 (39), 119 (23), 104 (12), 76 (22)
5a	251 $[M]^+$ (47), 250 (17), 223 (22), 207 (100), 179 (30), 178 (32), 151 (13), 140 (6), 105 (8), 95 (8), 77 (15)
5b	287 $[M\ (^{37}Cl)]^+$ (11), 285 $[M\ (^{35}Cl)]^+$ (30), 259 (3), 257 (10), 243 (36), 241 (100), 213 (27), 206 (7), 178 (27), 164 (12), 151 (18), 139 (10), 123 (15), 110 (20), 90 (45), 77 (35), 75 (76)
5c	296 $[M]^+$ (35), 268 (5), 252 (55), 206 (100), 194 (17), 177 (18), 164 (10), 151 (23), 103 (12), 90 (50), 77 (25), 75 (73)
5d	311 $[M]^+$ (40), 283 (20), 267 (46), 252 (41), 251 (72), 250 (38), 224 (30), 223 (60), 208 (20), 207 (100), 179 (20), 178 (40), 165 (20), 151 (20), 101 (11), 77 (17)
6a	250 $[M]^+$ (41), 249 (100), 222 (28), 220 (15), 192 (6), 179 (5), 146 (6), 132 (11), 130 (45), 119 (23), 102 (13), 90 (19), 77 (37)
6b	286 $[M\ (^{37}Cl)]^+$ (35), 285 (27), 284 $[M\ (^{35}Cl)]^+$ (100), 283 (85), 258 (7), 256 (20), 249 (9), 248 (7), 239 (8), 192 (27), 187 (20), 119 (45), 105 (28), 75 (12)
6c	295 $[M]^+$ (25), 294 (70), 293 (51), 265 (44), 263 (50), 249 (40), 247 (57), 236 (43), 219 (55), 191 (33), 164 (55), 146 (70), 119 (35), 101 (98), 90 (100), 77 (45)
6d	310 $[M]^+$ (10), 309 (100), 308 (25), 294 (10), 293 (13), 265 (32), 249 (29), 222 (10), 173 (25), 119 (12), 101 (18), 95 (13), 77 (12)
6e	331 $[M\ (^{81}Br)]^+$ (12), 330 (98), 329 $[M\ (^{79}Br)]^+$ (13), 328 (100), 303 (10), 301 (12), 249 (25), 248 (31), 210 (58), 208 (53), 192 (13), 182 (18), 155 (21), 119 (37), 103 (20), 90 (18), 77 (9)
6f	376 $[M]^+$ (100), 375 (45), 374 (20), 295 (25), 287 (19), 249 (25), 248 (40), 222 (10), 206 (12), 191 (12), 175 (10), 119 (13), 102 (20), 90 (29), 77 (50)
6g	280 $[M]^+$ (100), 279 (72), 278 (7), 252 (15), 248 (5), 236 (17), 195 (6), 191 (5), 181 (9), 119 (10), 107 (8), 76 (31)

A characteristic feature of the mass spectra (EI ionization) of the acids **2a-d** is the successive removal from their molecular ion $[M]^+$ of the water and acetylene molecules, which leads to the radical-cations $\mathbf{\Phi_1}$ and $\mathbf{\Phi_2}$ (Table 3).



As mentioned earlier [16], closure of the lactam ring in acids **2a-d** takes place under harsher conditions: the isoindolobenzimidazolones **3a**,**c** and **3b**,**d**, were obtained by refluxing compounds **2a**,**c** in acetic anhydride (method A) or by refluxing compounds **2b**,**d** in thionyl chloride (method B), respectively.

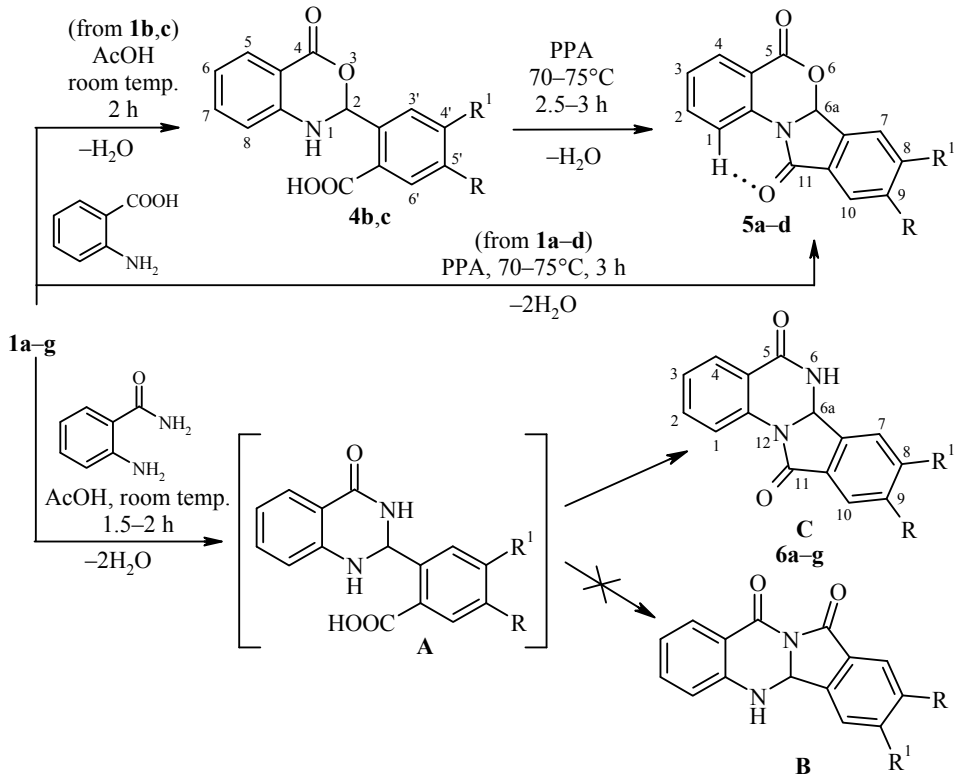


a R = R^1 = H; **b** R = Cl, R^1 = H; **c** R = NO_2, R^1 = H; **d** R = R^1 = OMe

The structure of the isoindolobenzimidazolones **3a-d** was confirmed by the presence of a strong band at 1750-1730 cm^{-1}, characteristic of the stretching vibrations of the carbonyl group in condensed cyclic γ-lactams [14], in their IR spectra. Another important indication confirming the structure of compounds **3a-d** is the fact that their molecular ions correspond to the radical-cations **Φ$_1$** formed at the first stage of fragmentation of the molecular ions of the acids **2a-d** (Table 3).

Researchers have use various methods, based on cyclization of *N*-(2-dibromomethylbenzoyl)anthranilic acid [17], oxidation of 1*H*-isoindolo[2,1-*a*]indole-11-carboxaldehyde [18], and pyrolysis of *N*-benzylbenzisoxazolone derivatives [19], for the formation of 5*H*-isoindolo[2,1-*a*][3,1]benzoxazine-5,11(6a*H*)-diones.

During the reaction of substituted FBAs **1b**,**c** with anthranilic acid in acetic acid at room temperature with the reagents in an equimolar ratio we isolated the acids **4b**,**c** (Tables 1-3). The ^{1}H NMR spectra of these compounds contain a characteristic pair of doublets (signals of the 2-CH and 1-NH protons with identical vicinal constants) (Table 2) and a broadened singlet for the proton of the carboxyl group.

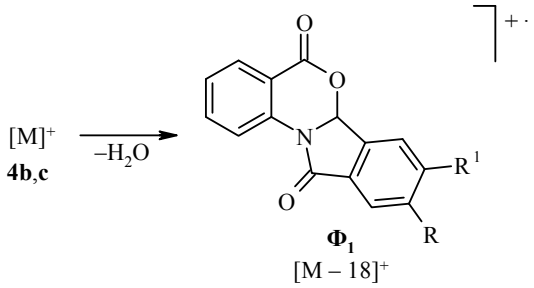


Intramolecular dehydration of the acids **4b**,**c** in polyphosphoric acid (PPA) leads to the 5*H*-isoindolo[2,1-*a*][3,1]benzoxazine-5,11(6a*H*)-diones **5b**,**c**. The tetracyclic compounds **5a-d** are also formed in a single stage when formylbenzoic acids **1a-d** are heated with anthranilic acid in PPA. Compounds **4b**,**c** and **5a-d** are colorless crystals that are readily soluble in most organic solvents.

The IR spectra of isoindolobenzoxazinediones **5a-d** contain characteristic bands for the stretching vibrations of the ester and lactam carbonyl groups in the regions of 1735-1750 and 1700-1720 cm^{-1}. In the ^1H NMR spectra there are singlet signals for the methine (6a-CH) and aromatic protons at 6.66-7.25 and 7.32-8.65 ppm, respectively (Table 2).

In the ^1H NMR spectra of compounds **5b**,**c** we observe a downfield shift (by 0.81 and 1.33 ppm, respectively) of the signal of the aromatic proton H-1 compared with the signal of the analogous proton H-8 in the spectra of compounds **4b**,**c** (Table 2). This effect is clearly the result of a change in the electronegativity of the nitrogen atom after its acylation and the appearance of an intramolecular contact between the H-1 proton and the oxygen atom of the amide group in the structures of the isoindolobenzoxazinediones **5b**,**c**. The same relationship is observed during comparison of the ^1H NMR spectra of the isoindolobenzimidazolones **3a-d** and benzimidazoles **2a-d** with respect to the signals of the aromatic protons in the benzimidazole rings H-9 and H-7 (Table 2).

The mass spectra of compounds **4b**,**c** and **5a-d** contain molecular-ion peaks with fairly high intensity (Table 3). The fragmentation of the molecular ions of the acids **4b**,**c** begins with the elimination of a water molecule; the radical-cations **Φ1** $[M\text{-}H_2O]^+$ are actually the molecular ions of the corresponding molecules **5a-d**. The first step in the fragmentation of the molecular ions of compounds **5a-d** affects the ester fragment of the molecules (concurrent ejection of CO and CO_2), indicating that the charge and spin in the molecular ion are located at the carbonyl oxygen of the ester group (Table 3).



The domino reaction of the anthranilic acid amides and *o*-aminocarboxamides with *o*-formyl(acyl)-benzoic acids, during which two rings (pyrimidine and isoindolone) are formed in sequence, is typically used for the formation of the isoindoloquinazoline structure **6**. The authors of the papers [20-29] consider *a priori* that both primary and secondary aminocarboxamides in reaction with *o*-formyl(acyl)benzoic acids form tetracyclic structures with the same angular skeleton **C**, although in our opinion none of the cited papers present convincing evidence for an angular and not linear structure of the skeleton of the products from the reaction of primary amides of anthranilic acid with the FBA.

In a recent work [8], the isoindoloquinazolinedione structures **6** were obtained by a new three-component reaction of isatoic anhydride with unsubstituted FBA and ammonium carbonate or primary amines. In the case of 6-(4-chlorophenyl)methyl-6,6a-dihydroisoindolo[2,1-*a*]quinazoline-5,11-dione an angular structure of the heterocyclic skeleton was confirmed by X-ray structural analysis for the obtained compounds, which in fact being the products from reaction of the secondary anthranilic acid amide with FBA must have a skeleton with an angular structure. The 6-unsubstituted derivative of isoindoloquinazolidinedione obtained with ammonium carbonate was assigned as an angular structure evidently by analogy with the previously reported data, but this was not proved.

TABLE 4. Results from Homonuclear (NOESY) and Heteronuclear (HMBC) Correlation Experiments for Compound **6b**

Position of atom	^{1}H NMR spectrum, δ, ppm	HSQC (^{13}C)	HMBC	NOESY
NH	9.22	—	67.4; 120.5	7.89; 6.47
1	8.05	120.0	125.4; 120.5	7.68
4	7.97	128.7	163.9; 137.4; 133.9	7.33
7	7.89	133.8	135.6; 133.5	6.47
10	7.86	123.9	135.6; 163.5	—
8	7.83	126.5	139.8	7.89
2	7.68	133.9	137.4; 128.7; 120.0	8.05; 7.33
3	7.33	125.4	120.5	7.97; 7.68
6a	6.47	67.4	163.5; 139.8; 133.8	9.22; 7.89

Our experiments showed that the reaction of equimolar amounts of the series of FBAs **1a-g** with anthranilamide in acetic acid leads to the formation of the corresponding 6,6a-dihydroisoindolo[2,1-*a*]-quinazoline-5,11-diones **6a-g** without isolation of the intermediate acids **A**. The synthesis of 9-nitroiso-indoloquinazolinedione **6c** is proceeded at room temperature, while compounds **6a**,**b**,**d-g** are only formed if the temperature is increased to 70-75°C. We significantly simplified the procedure for the formation of compounds **6a-g** compared to the methods described in the literature [8, 25] by using acetic acid as solvent and catalyst. The obtained compounds **6a-g** are colorless crystals, poorly soluble in most organic solvents.

The ^{1}H NMR spectra of the tetracyclic compounds **6a-g** contain characteristic singlet signals for the methyne protons 6a-CH in the region of 6.25-6.92 ppm and singlet signals for the NH protons at 8.92-9.39 ppm, which is more characteristic of amide than of amine protons [30]. In the IR spectra of these products there are strong bands for the stretching vibrations of the lactam and amide carbonyl groups at 1710-1750 cm^{-1} and 1670-1680 cm^{-1}, respectively (Table 2).

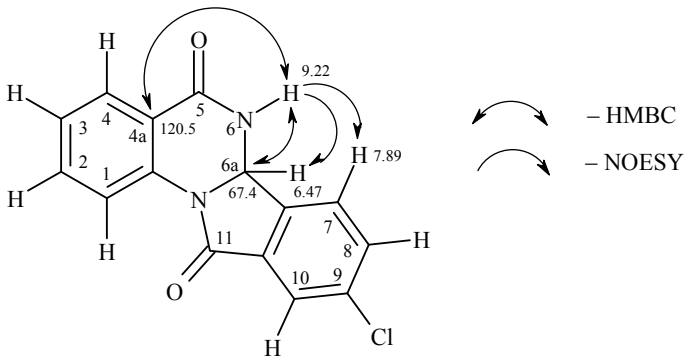


Fig. 1. The structurally significant HMBC and NOESY correlation spectra for compound **6b**.

Analysis of the mass spectra of the isoindoloquinazolinediones **6a-g** indicates that all the samples give molecular-ion peaks in which hydrogen atom abstraction is a characteristic feature of primary dissociation (Table 3).

The HSQC, HMBC, and NOESY correlation experiments for isoindoloquinazolinedione **6b** (Table 4, Fig. 1) do not provide a conclusive choice between structures **B** and **C**, although the absence of cross peaks corresponding to interaction of the NH and H-1 protons, which should be characteristic of structure **B**, thus indirectly favoring structure **C**. Similar evidence in favor of structure **C** may also be supported by the absence of long-range spin-spin couplings between the NH proton and the C-1 and C-13 carbon atoms, which must be characteristic of structure **B**.

To make an unambiguous choice between the linear **B** and angular **C** structures of the heterocyclic skeleton in isoindoloquinazolinediones **6**, we used X-ray structural analysis of one of the representatives of this series, 8-bromo-6,6a-dihydroisoindolo[2,1-*a*]quinazoline-5,11-dione (**6e**). A general view of this molecule is presented in Figure 2, from which it is seen that compound **6e** is actually the angular isomer **C**.

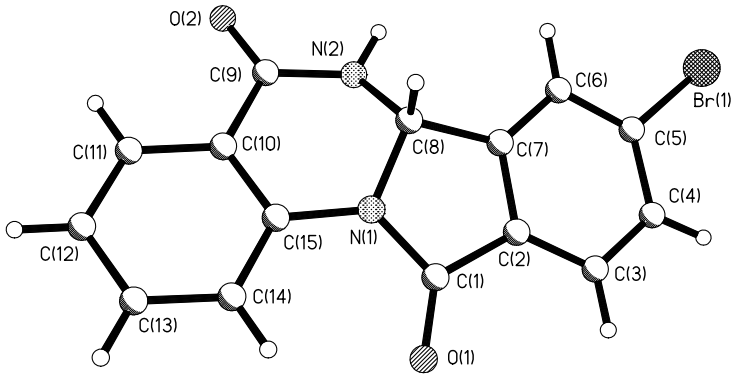


Fig. 2. Molecular structure of compound **6e** according to X-ray structural analysis.

In the heterocyclic skeleton of the molecule there is a single sp^3-hybridized atom (the C(8) carbon), and in the end it is this that determines the bifolded structure of the molecule core as a whole. The molecule of compound **6e** is not planar and can be described by a set of three intersecting planes. The benzene ring C(10)···C(15) and the C(9), N(1), and N(2) atoms adjacent to it form plane 1 (mean deviation of the atoms from the plane 0.0323 Å). The C(8) atom deviates from this plane by 0.4223 Å and together with the N(1) and N(2) atoms forms plane 2. The angle between planes 1 and 2 is 143.7°. The isoindolone fragment of the molecule (the C(1)···C(8) and N(1) atoms) is planar (plane 3, mean deviation of the atoms from the plane 0.382 Å). The angle between planes 2 and 3 is 127.4°. Such geometry of the heterocycle ensures the absence of angular strains at the tetrahedral carbon atom C(8) and a planar trigonal environment at both nitrogen atoms.

Some of the compounds that we obtained were tested under laboratory and field conditions for pesticidal activity at the All-Russian Research Institute of Biological Plant Protection (Krasnodar).

Compounds **2c**, **5b**, **6a**,**d**,**f** exhibited antidotal activity in relation to the hormonal herbicide 2,4-dichlorophenoxyacetic acid (2,4-D) on seedlings and vegetative plants of the Master sunflower variety. Laboratory trials of the isoindoloquinazolinediones **6a**,**d**,**f** showed that after treatment of sunflower seedlings with aqueous solutions of the herbicide (at a concentration of 10^{-3} %) and the antidote (at concentrations of 10^{-2}, 10^{-3}, 10^{-4}, and 10^{-5} %, Table 5) the length of the hypocotyl increases from 116 to 131% and the length of the root from 116 to 154% in relation to the standard. From the results of the field trials of compounds **2c**, **5b** it follows that the increase in the crop yield of sunflowers in relation to the standard amounts to 14% with benzimidazole **2c** as antidote and 12% with isoindolobenzoxazinedione **5b** (Table 6). The results of the trials demonstrate the prospects of using the products in the cultivation of sunflowers and in the search for new antidotes in this series of heterocycles.

Thus, we have demonstrated the possibility of producing the tetracyclic condensed heterosystems 11*H*-isoindolo[2,1-*a*]benzimidazol-11-one, 5*H*-isoindolo[2,1-*a*][3,1]benzoxazine-5,11(6a*H*)-dione, and 6,6a-dihydroisoindolo[2,1-*a*]quinazoline-5,11-dione and also 2-substituted benzimidazolones by choosing optimal conditions for the reactions of substituted FBAs with bifunctional compounds and have determined some of their useful properties.

TABLE 5. Antidotal Activity of the Synthesized Compounds in Relation to 2,4-D on Sunflower Seedlings (numerator – hypocotyl, denominator – root)*

Compound	Control	Herbicide (standard)		Herbicide + antidote at concentration, %							
				10^{-2}		10^{-3}		10^{-4}		10^{-5}	
	A	A	C	A	B	A	B	A	B	A	B
6a	103/180	49/52	52/71	51/59	104/113	57/67	116*²/129*²	57/75	116*³/144*²	52/57	106/110
6d	103/180	49/52	52/71	64/80	131*²/154*²	50/54	102/104	51/53	104/102	55/59	112/113
6f	103/180	49/52	52/71	60/65	122*²/125*²	55/59	112/116*³	64/73	131*²/140*²	58/67	118*³/129*²

*A is the average length of the hypocotyl (root), mm; B is % to the standard; C is the suppression of growth of the hypocotyl (root), %.
*²Significantly at probability level 0.95.
*³Significantly at probability level 0.90.

TABLE 6. Effect of the Antidotal Activity of Compounds **2c** and **5b** on the Crop Yield (C/ha) of the Sunflower Master Variety during Field Trials

Antidote	Control (untreated plants)	2,4-D (standard)	2,4-D + antidote	Increase in relation to the standard
2c	43.4	15.3	17.5	2.2 (14%)
5b	41.8	16.4	18.4	2.0 (12%)

EXPERIMENTAL

The IR spectra were recorded on a Specord-71 instrument as suspensions in vaseline oil (compounds **2**, **3a-d**, **4b**,**c**) and a Spectrum Two instrument with a DTIR attachment (compounds **5a-d**, **6a-g**). The ^{1}H and ^{13}C NMR spectra of compounds **6a-g** and also the HSQC, HMBC, and NOESY hetero- and homonuclear correlation spectra of compound **6b** were recorded on an Agilent 400/54 spectrometer (400 MHz for ^{1}H nuclei, 100 MHz for ^{13}C nuclei). The ^{1}H NMR spectra of the remaining compounds were recorded on a Bruker DRX-500 instrument (500 MHz, compounds **4b**,**c** and **5b**,**c**) and a Bruker AM-300 instrument (300 MHz, compounds **2a-d**, **3a-d**, and **5a**,**d**). Solvents: $CDCl_3$ (compounds **5a**, **6c**) and DMSO-d_6 (the other compounds) with TMS as internal standard. The mixing time in the NOESY experiment was 200 msec. The mass spectra were recorded on a Varian CH-6 instrument with direct injection of the sample into the ion source at 50-180°C with electron ionization energy 70 eV. Elemental analysis was performed on a Hewlett-Packard HP-185B C,H,N analyzer. The melting points were determined on a Stuart SNP 30 heating apparatus and were not corrected. The reactions were monitored by TLC on Silufol UV-254 plates in the PhH–Me_2CO, 4:1 (compounds **3a-d**, **4b**,**c**, **5a-d**, **6a-g**) and PhH–EtOH, 4:1 (compounds **2a-d**) systems with iodine vapor for visualization.

The laboratory and field trials of compounds **2c**, **5b**, **6a**,**d**,**f** were conducted by the described procedure [31].

2-(1*H*-Benzimidazol-2-yl)benzoic Acids 2a-d (General Method). A mixture of the acid **1a-d** (4 mmol) and PDA (0.43 g, 4 mmol) in glacial acetic acid (5 ml) was stirred with cooling on an ice bath for 30 min and then at room temperature for 1.5 h. When the starting compounds had disappeared from the reaction mixture the colorless precipitate was filtered off, recrystallized from a 1:1 mixture of Me_2CO and DMF, washed with water, and dried.

Synthesis of 11*H*-isoindolo[2,1-*a*]benzimidazol-11-ones 3a-d. A. A mixture of the acid **2a**,**c** (1 mmol) and Ac_2O (2 ml) was heated until dissolved, refluxed for 2-3 min, and then cooled. The precipitate was filtered off, washed with petroleum ether, recrystallized from EtOH, and dried. Compounds **3a**,**c** were obtained.

B. $SOCl_2$ (1.0-1.5 ml) was added to the acid **2b**,**d** (1 mmol). The mixture was heated, and when the release of gas had stopped and a uniform mass had formed it was poured onto ice. The yellow precipitate that separated was filtered off and recrystallized from EtOH.

5-Nitro-2-(4-oxo-1,4-dihydro-2*H*-3,1-benzoxazin-2-yl)benzoic Acid (4c). A mixture of 2-formyl-5-nitrobenzoic acid (**1c**) (0.78 g, 4 mmol) and anthranilic acid (0.55 g, 4 mmol) in glacial acetic acid (10 ml) was stirred at room temperature for 2 h. The sediment of the starting substances was seen to disappear, and a new one separated. The colorless precipitate was filtered off, washed with a 3:1 mixture of EtOH and H_2O, dried, and recrystallized from EtOH. Yield 0.94 g (75%).

Compound **4b** was obtained similarly.

9-Nitro-5*H*-isoindolo[2,1-*a*][3,1]benzoxazine-5,11(6a*H*)-dione (5c). A. The acid **4c** (1.25 g, 4 mmol) was added with stirring to FBA (10 ml). The mixture was heated on a water bath at 70-75°C for 2.5-3 h, and H_2O (15-20 ml) was then added in small portions to the mixture while it was cooled on an ice bath. The precipitate that separated was extracted with CH_2Cl_2. The organic layer was separated and dried, the solvent was distilled, and the colorless residue was recrystallized from a 4:1 mixture of EtOH and PhH. Yield 0.77 g (65%).

Compound **5b** was obtained similarly.

B. The acid **1c** (0.78 g, 4 mmol) and anthranilic acid (0.55 g, 4 mmol) were added with stirring to PPA (10 ml). The mixture was heated on a water bath at 70-75°C for 2.5-3 h. The product was isolated and purified by method A. Yield 0.91 g (77%).

Compounds **5a**,**b**,**d** (colorless substances) were obtained similarly.

6,6a-Dihydroisoindolo[2,1-*a*]quinazoline-5,11-diones 6a-g (General Method). A mixture of the acid **1a-g** (4 mmol) and anthranilamide (0.54 g, 4 mmol) in glacial acetic acid (10 ml) was stirred at 70-75°C (at room temperature in the case of the nitro derivative **1c**) for 1.5-2 h. At the end of the reaction, the precipitate was filtered off and recrystallized from EtOH, dried. Yield 0.85 g (72%).

X-ray Structural Investigation of Compound 6e. The colorless monoclinic crystals of compound **6e** ($C_{15}H_9BrN_2O_2$) were grown by crystallization from glacial acetic acid. Crystal parameters: 0.55×0.23×0.15 mm. At 20°C: *a* 13.6774(12), *b* 7.4891(6), *c* 24.544(2) Å; α 90, β 103.258(2), γ 90°; *V* 2450.0(4) Å^3; *M* 329.15; *Z* 8; $d_{calc.}$1.785 g/cm^3. Space group *P*21/*n*. X-ray structural analysis was performed on a CAD4 automatic four-circle diffractometer (graphite monochromator, MoKα radiation, ω-scan, $2\theta_{max}$ 24.98°); 16959 reflections, of which 7136 were unique, were obtained. The structure was solved by the direct method with SHELXTL program set [32] and was refined in anisotropic approximation (isotropic for the hydrogen atoms) to probability factors R^1 0.0389 and wR^2 0.0925. The complete set of atomic coordinates was deposited at the Cambridge Crystallographic Data Center (deposit CCDC 934695).

The authors thank the Ministry of Education and Science of the Russian Federation for financial support of the investigation.

REFERENCES

1. E. V. Gromachevskaya, A. S. Pilipenko, A. V. Butin, V. E. Zavodnik, and G. D. Krapivin, *Khim. Geterotsikl. Soedin.*, 125 (2010). [*Chem. Heterocycl. Compd*., **46**, 106 (2010).]
2. E. V. Gromachevskaya, A. V. Butin, A. S. Pilipenko, G. D. Krapivin, A. A. Borodavko, L. I. Isaeva, and V. D. Strelkov, RF Pat. 2345083; *Byul. Izobret.*, No. 3, 1222 (2009).
3. E. V. Boltukhina, F. I. Zubkov, and V. A. Varlamov, *Khim. Geterotsikl. Soedin.*, 963 (2006). [*Chem. Heterocycl. Compd*., **42**, 831 (2006).]
4. E. V. Boltukhina, F. I. Zubkov, and V. A. Varlamov, *Khim. Geterotsikl. Soedin.*, 1123 (2006). [*Chem. Heterocycl. Compd*., **42**, 971 (2006).]
5. S. K. Meegalla, G. J. Stevens, C. A. McQueen, A. Y. Chen, C. Yu, L. F. Liu, L. R. Barrows, and E. J. LaVoie, *J. Med. Chem.*, **37**, 3434 (1994).
6. S. N. Khattab, S. Y. Hassan, A. El-Faham, A. M. M. El Massry, and A. Amer, *J. Heterocycl. Chem.*, **44**, 617 (2007).
7. V. V. Lyaskovskii, Z. V. Voitenko, and V. A. Kovtunenko, *Khim. Geterotsikl. Soedin.*, 323 (2007). [*Chem. Heterocycl. Compd*., **43**, 253 (2007).]
8. K. S. Kumar, P. M. Kumar, K. A. Kumar, M. Sreenivasulu, A. A. Jafar, D. Rambabu, G. R. Krishna, C. M. Reddy, R. Kapavarapu, K. Shivakumar, K. K. Priya, K. V. L. Parsa, and M. Pal, *Chem. Commun*., 5010 (2011).
9. L. Yu. Ukhin, L. G. Kuz'mina, L. V. Belousova, and E. N. Shepelenko, *Second International Scientific Conference. New Directions in the Chemistry of Heterocyclic Compounds* [in Russian], Zheleznovodsk, Russia (2011), p. 241.
10. A. Cul, A. Daïch, B. Decroix, G. Sanz, and L. Van Hijfte, *Tetrahedron*, **60**, 11029 (2004).
11. A. Bistrzycki and A. Lecco, *Helv. Chim. Acta*, **4**, 425 (1921).
12. P. V. L. Boddupally, Y. Tallapally, S. R. Pamulaparthy, and N. Banda, *Heterocycles*, **65**, 2329 (2005).

13. K. N. Zelenin, I. V. Ukraintsev, and V. V. Alekseev, *Khim. Geterotsikl. Soedin.*, 363 (1998). [*Chem. Heterocycl. Compd*., **34**, 329 (1998).]
14. A. D. Cross, *Introduction to Practical Infrared Spectroscopy* [Russian translation], Izd. Inostr. Lit., Moscow (1961), p. 93.
15. L. A. Kazitsyna and N. B. Kupletskaya, *Application of UV, IR, NMR, and Mass Spectrometry in Organic Chemistry* [in Russian], Izd. MGU, Moscow (1979), p. 74.
16. J. Arient, *Usp. Khim.*, **34**, 1908 (1965).
17. H. Diesbach, A. Schürch, and G. Cavin, *Helv. Chim. Acta*, **31**, 716 (1948).
18. E. Desarbre and J.-Y. Mérour, *Heterocycles*, **41**, 1987 (1995).
19. J. Khalafy and R. H. Prager, *Aust. J. Chem*., **51**, 925 (1998).
20. E. F. M. Stephenson, *J. Chem. Soc.*, 2354 (1954).
21. J. Honzl, *Chem. Listy*, **49**, 1671 (1955).
22. J. Honzl, *Collect. Czech. Chem. Commun.*, **21**, 725 (1956).
23. M. Lamchen, *J. Chem. Soc. C*, 573, (1966).
24. I. Butula and W. Otting, *Monatsh. Chem.*, **99**, 1320 (1968).
25. M. Chelardoni and V. Pestellini, *Ann. Chim.*, **64**, 421 (1974).
26. V. Pestellini, M. Ghelardoni, C. Bianchini, and A. Liquori, *Boll. Chim. Farm*., **117**, 54 (1978).
27. V. Balasubramaniyan and N. P. Argade, *Tetrahedron*, **45**, 835 (1989).
28. P. Pigeon, J. Sikoraiova, S. Marchalin, and B. Decroix, *Heterocycles*, **56**, 129 (2002).
29. P. Aeberli and W. J. Houlihan, *J. Org. Chem*., **33**, 2402 (1968).
30. A. Gordon and R. Ford, *The Chemist's Companion* [Russian translation], Mir, Moscow (1976), p. 293.
31. V. D. Strelkov, L. V. Dyadyuchenko, L. I. Isakova, and I. G. Dmitrieva, *Agrokhimiya*, 28 (2010).
32. G. M. Sheldrick, *SHELXL-97: Program for the Solution of Crystal Structures*, University of Göttingen, Germany (1997).
33. C. J. Perry, *J. Chem. Soc*., *Perkin Trans. 2,* 977 (1997).