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Modification of biologically active amides and amines with fluorine-containing heterocycles

7.* Fluorine-containing heterocyclic derivatives of the acetazolamide

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An approach is proposed to modification of a medical product acetazolamide (*N*-(5-sulfamoyl-1,3,4-thiadiazol-2-yl)acetamide) by interaction of 2-(5-acetylamino-1,3,4-thiadiazole-2-yl)sulfonyl imine methyl trifluoropyruvate, which is *in-situ* generated from acetazolamide, with 1,3-binucleophiles: 6-aminouracils, 6-aminothiouracils, and *N*-substituted ureas, to yield heterocyclic compounds with the CF_3 substituent in the nitrogen-containing mono- or bicycle.

Key words: acetazolamide, methyl trifluoropyruvate, 6-aminouracils, 6-aminothiouracils, *N*-substituted ureas, fluorine-containing hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidines, 2,5-dioxoimidazolidines, cyclocondensation.

Acetazolamide — *N*-(5-sulfamoyl-1,3,4-thiadiazol-2-yl)acetamide (**1**), or "diacarb", is one of the effective inhibitors of the carboanhydrase, which is used for conservative management of intracranial hypertension and hydrocefalus[2—4].

The aim of the present research is to expand the repertoire of actazolamide derivatives, which are furnished with trifluoromethyl-containing five-membered ring heterocycles using the multicomponent reaction of **1** with methyl trifluoropyruvate (MTFP) (**2**) and 1,3-binucleophiles, with a view to biological studies. The prerequisites for this work were the results of the research on the behavior of *N*-substituted imines of MTFP in cyclocondensation reactions with 1,3-binucleophiles. which lead to formation of trifluoromethyl-containing five-membered ring heterocycles[5—9], and the data on modification of biologically active amides with fluorine-containing heterocycles[10,11].

As the attempts to obtain the starting biselectrophilic synthon sulfonylimine of MTFP **3** in its pure state using the known methods of synthesis of *N*-substituted imines of MTFP failed, the imine **3** was generated *in-situ* by successive addition of pyridine, MTFP (**2**), and $SOCl_2$ to the solution of sulfonamide **1** in DMF with the following involvement of the compound **3** in cyclocondensation with 1,3-binucleophiles (Scheme 1). Interaction of the imine **3** with 1,3-binucleophiles is performed according to the mechanism of the cyclocondensation reaction: addition of the binucleophile to the highly reactive C=N-group with the following heterocyclization with methanol elimination in the presence of catalytic amount of Et_3N. In these intractions 6-aminouracils **4a—c**, 6-aminothiouracyls **6a—c**, and *N*-substituted ureas **8a—c** were used as 1,3-binucleophyles. Cyclocondensation of the imine **3** with **4a—c**, **6a—c**, and **8a—c** resulted in formation of the corresponding heterocyclic derivatives of the acetazolamide: 2,4,6-trioxo-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidines **5a—c**, 4,6-dioxo-2-thioxo-5-trifluoromethyl-1-aryl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidines **7a—c**, and 2,5-dioxo-4-trifluoromethylimidazolidines **9a—c**. *N*-Acetylated heterocyclic derivatives of acetazolamide **5a**,**c**, **7a**, and **9a—c** are transformed to corresponding trifluoromethyl-containing heterocyclic derivatives of 5-amino-1,3,4-thiadiazol-2-sulfonic acid **10a**,**b**, **11**, and **12a—c** under reflux in 10% HCl for 1 h.

Fluorine-containing heterocyclic derivatives of acetazolamide **5a—c**, **7a—c**, **9a—c**, **10a**,**b**, **11**, and **12a—c** are solid crystalline compounds, their composition and structure were confirmed with the elemental analysis data and ^1H and ^{19}F NMR spectroscopy data (Tables 1 and 2).

In ^{19}F NMR spectra, characteristic signals are observed at δ 4—5 ppm for **5a—c**, **7a—c**, **10a**,**b**, **11**, and at 0.1—0.6 ppm for **9a—c** and **12a—c**.

For fluorine-containing heterocyclic derivatives of 5-amino-1,3,4-thiadiazol-2-sulfonic acid **10a**,**b**, **11**, and **12a—c** bactericidal and fungicidal activity against *S. aureus, S. enteritidis, B. Anthracis, Cand.* and *E. Coli.* was

* Report 6 see lit[1].

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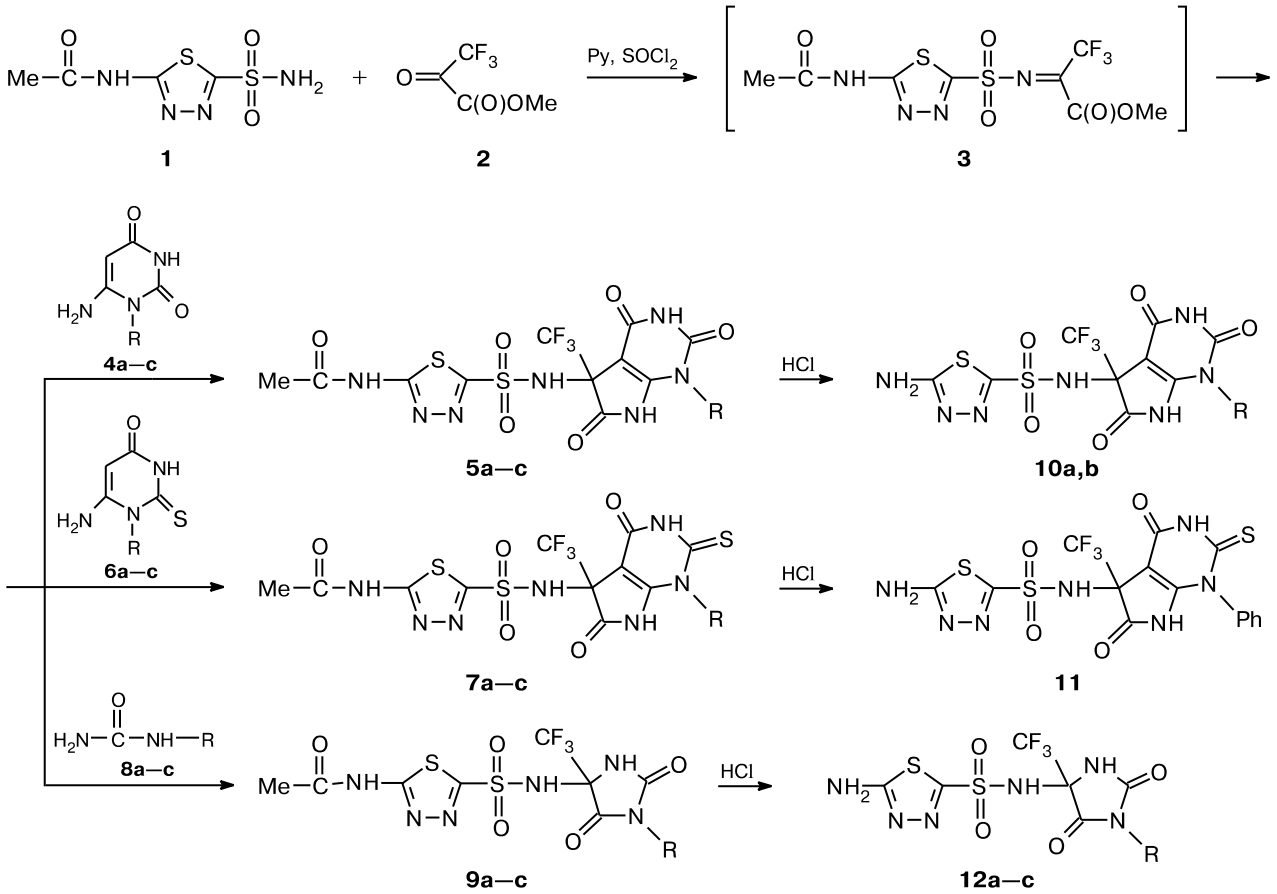
Scheme 1



5: R = Bu (**a**), 2-Me-C_6H_4 (**b**), 3,4-$(MeO)_2C_6H_3CH_2CH_2$ (**c**); **7:** R = Ph (**a**), 3-Me-C_6H_4 (**b**), 4-Me-C_6H_4 (**c**); **9:** R = Me (**a**), cyclo-C_5H_9 (**b**), $PhCH_2$ (**c**); **10:** R = Bu^n (**a**), 3,4-$(MeO)_2C_6H_3CH_2CH_2$ (**b**); **12:** R = Me (**a**), cyclo-C_5H_9 (**b**), $PhCH_2$ (**c**)

Table 1. Yields, melting points and elemental analysis data for compounds **5**, **7**, **9**, **10—12**

Compound	Yield (%)	M.p. /°C	Molecular formula	Found / Calculated (%) C	H	N
5b	73	243—245	$C_{18}H_{14}F_3N_7O_6S_2$	39.86 / 39.64	2.41 / 2.59	18.12 / 17.94
5c	77	237—239	$C_{21}H_{20}F_3N_7O_8S_2$	40.56 / 40.71	3.02 / 3.25	15.65 / 15.83
7a	69	259—261	$C_{17}H_{12}F_3N_7O_5S_3$	37.05 / 37.29	2.33 / 2.21	18.13 / 17.91
7b	81	261—263	$C_{18}H_{14}F_3N_7O_5S_3$	38.72 / 38.50	2.69 / 2.51	17.58 / 17.46
7c	75	265—267	$C_{18}H_{14}F_3N_7O_5S_3$	38.76 / 38.50	2.33 / 2.51	17.28 / 17.46
9a	73	248—250	$C_9H_9F_3N_6O_5S_2$	29.11 / 28.87	2.06 / 2.25	20.63 / 20.89
9b	76	241—243	$C_{13}H_{15}F_3N_6O_5S_2$	34.38 / 34.21	3.11 / 3.31	18.62 / 18.41
9c	79	257—259	$C_{15}H_{13}F_3N_6O_5S_2$	37.85 / 37.66	2.58 / 2.74	17.39 / 17.57
10a	86	227—229	$C_{13}H_{14}F_3N_7O_5S_2$	33.08 / 33.26	2.89 / 3.01	20.62 / 20.89
10b	85	233—235	$C_{19}H_{18}F_3N_7O_7S_2$	39.33 / 39.52	3.29 / 3.14	16.72 / 16.98
11	82	243—245	$C_{15}H_{10}F_3N_7O_4S_3$	36.44 / 36.64	2.16 / 1.99	19.18 / 19.40
12a	84	228—230	$C_7H_7F_3N_6O_4S_2$	23.19 / 23.34	1.81 / 1.96	23.18 / 23.33
12b	79	219—221	$C_{11}H_{13}F_3N_6O_4S_2$	31.99 / 31.88	2.95 / 3.16	20.11 / 20.28
12c	86	240—242	$C_{13}H_{11}F_3N_6O_4S_2$	35.59 / 35.78	2.36 / 2.54	19.42 / 19.26

Table 2. ^{1}H и ^{19}F NMR spectroscopy data for compounds **5**, **7**, **9**—**12** in DMSO-d_6

Compound	^{1}H, δ, *J*/Hz	^{19}F, δ
5b	2.12 (s, 3 H, MeC(O)); 2.25 (s, 3 H, Me); 7.24—7.62 (m, 4 H, CH_{Ar}); 10.51 (s, 1 H, NH); 10.98 (s, 1 H, NH); 11.75 (s, 1 H, NH); 13.04 (s, 1 H, NH)	5.41 (s)
5c	2.24 (s, 3 H, MeC(O)); 2.81 (t, 2 H, CH_2, *J* = 6.7); 3.73 (s, 3 H, MeO); 3.77 (s, 3 H, MeO); 4.02 (t, 2 H, CH_2, *J* = 6.7); 6.66—6.80 (m, 2 H, CH_{Ar}); 6.85 (s, 1 H, CH_{Ar}); 10.57 (s,1 H, NH); 10.83 (s, 1 H, NH); 12.32 (s, 1 H, NH); 13.02 (s, 1 H, NH)	4.78 (s)
7a	2.22 (s, 3 H, MeC(O)); 7.21—7.61 (m, 5 H, CH_{Ar}); 10.92 (s, 1 H, NH); 11.73 (s, 1 H, NH); 12.52 (s,1 H, NH); 13.15 (s, 1 H, NH)	4.54 (s)
7b	2.23 (s, 3 H, MeC(O)); 2.42 (s, 3 H, Me); 7.00—7.19 (m, 1 H, CH_{Ar}); 7.33 (t, 1 H, CH_{Ar}, *J* = 7.3); 7.38—7.51 (m, 2 H, CH_{Ar}); 10.71 (s, 1 H, NH); 11.64 (s, 1 H, NH); 12.38 (s, 1 H, NH); 13.09 (s, 1 H, NH)	5.14 (s)
7c	2.26 (s, 3 H, MeC(O)); 2.44 (s, 3 H, Me); 7.09—7.44 (m, 4 H, CH_{Ar}); 10.63 (s, 1 H, NH); 11.61 (s, 1 H, NH); 12.28 (s, 1 H, NH); 13.10 (s, 1 H, NH)	5.07 (s)
9a	2.29 (s, 3 H, MeC(O)); 3.22 (s, 3 H, Me); 9.11 (s, 1 H, NH); 11.01 (s, 1 H, NH); 13.16 (s, 1 H, NH)	0.21 (s)
9b	1.57 (m, 2 H, CH_2); 1.84 (m, 6 H, CH_2); 2.24 (s, 3 H, MeC(O)); 4.30 (quint, 1 H, CH, *J* = 7.3); 9.73 (s, 1 H, NH); 11.23 (s, 1 H, NH); 13.08 (s, 1 H, NH)	0.12 (s)
9c	2.29 (s, 3 H, MeC(O)); 4.57 (s, 2 H, CH_2); 7.27—7.48 (m, 5 H, CH_{Ar}); 9.33 (s, 1 H, NH); 11.12 (s, 1 H, NH); 13.25 (s, 1 H, NH)	0.35 (s)
10a	0.95 (t, 3 H, Me, *J* = 7.6); 1.16—1.39 (m, 2 H, CH_2); 1.53—1.69 (m, 2 H, CH_2); 3.75—3.92 (m, 2 H, CH_2); 7.82 (s, 2 H, NH_2); 10.05 (s, 1 H, NH); 10.62 (s, 1 H, NH); 11.92 (s, 1 H, NH)	5.04 (c)
10b	2.84 (t, 2 H, CH_2, *J* = 6.7); 3.71 (s, 3 H, MeO); 3.80 (s, 3 H, MeO); 4.09 (t, 2 H, CH_2, *J* = 6.7); 6.61—6.76 (m, 2 H, CH_{Ar}); 6.81 (s, 1 H, CH_{Ar}); 7.76 (s, 2 H, NH_2); 10.15 (s, 1 H, NH); 10.78 (s, 1 H, NH); 11.85 (s, 1 H, NH)	5.11 (s)
11	7.30—7.66 (m, 5 H, CH_{Ar}); 7.91 (s, 2 H, NH_2); 10.33 (s, 1 H, NH); 10.62 (s, 1 H, NH); 12.03 (s, 1 H, NH)	4.98 (s)
12a	3.34 (s, 3 H, Me); 7.72 (s, 2 H, NH_2); 9.42 (s, 1 H, NH); 10.88 (s, 1 H, NH)	0.35 (s)
12b	1.42 (m, 2 H, CH_2); 1.73 (m, 6 H, CH_2); 4.08 (quint, 1 H, CH, *J* = 7.2); 7.67 (s, 2 H, NH_2); 9.33 (s, 1 H, NH); 10.88 (s, 1 H, NH)	0.22 (s)
12c	4.48 (s, 2 H, CH_2); 7.31—7.52 (m, 5 H, CH_{Ar}); 7.58 (s, 2 H, NH_2); 9.51 (s, 1 H, NH); 11.05 (s, 1 H, NH)	0.52 (s)

investigated using the agar diffusion test. Data shown in Table 3 demonstrated high activity of compounds **12a**—**c** against *B. Anthracis* and *Candida*, in comparison with streptocide[12].

Herewith, three component reaction of *N*-(5-sulfamoyl-1,3,4-thiadiazol-2-yl)acetamide, MTFP, and 1,3-binucleophiles results in novel fluorine-containing heterocyclic derivatives of acetazolamide and presents the prospective approach to modification of this medical product. The following hydrolysis of heterocyclic derivatives of acetazolamide results in formation of fluorine-containing heterocyclic derivatives of 5-amino-1,3,4-thiadiazol-2-sulfonic acid, which are potential bactericidal sulfonamide compounds.

Experimental

^{1}H and ^{19}F NMR spectra were registered on the NMR spectrometer Bruker DPX 200 (200.13 and 188.29 MHz correspondingly) in $CDCl_3$ using Me_4Si (internal standard) and CF_3COOH (external standard). Melting point determination was performed in a sealed capillary. Starting compounds 6-aminouracils **4a**—**c** and 6-aminothiouracils **6a**—**c** were obtained according to the published earlier method[13], *N*-(5-sulfamoyl-1,3,4-thiazol-2-yl)acetamide (**1**), methyl ester of trifluoropyruvic acid (**2**) and *N*-substituted ureas **8a**—**c** were purchased from Aldrich and used without further purification.

***N*-{5-[(1-Butyl-2,4,6-trioxo-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl)sulfamoyl]-1,3,4-thiadiazol-2-yl}acetamide (5a) (general procedure).** To the stirred solution of 2.22 g (0.01 mol) of *N*-(5-aminosulfamoyl-1,3,4-thiazol-2-yl)acetamide (**1**) in 20 mL of DMF, 1.56 g (0.02 mol) of pyridine and 1.56 g (0.01 mol) of MTFP (**2**) were added consistently. The reaction mixture was stirred for 30 min, then 1.19 g (0.01 mol) of $SOCl_2$ was added, the mixture was stirred for 1 h, then 1.83 g (0.01 mol) of aminouracyl **4a** was added, the mixture was stirred for 1 h at 20 °C, then 0.1 g of Et_3N was added and the mixture was kept at 90—100 °C for 2 h. Then the reaction mixture was cooled and poured into 50 mL of 10% aqueous solution of NaCl. The residue was filtered and recrystallized from 50% EtOH. The yield of **5a** was 4.1 g (80%). M.p. 207—209 °C. Found (%)): C, 35.44; H, 3.31; N, 19.36. $C_{15}H_{16}F_3N_7O_6S_2$.

Table 3. Bactericidal and fungicidal activity of fluorine-containing heterocyclic derivatives of 5-amino-1,3,4-tiadiazol-2-sulfonic acid **10—12**

Compound	Concentration (%)	Size of the inhibitory area/mm				
		S. aureus	*S. enteritidis*	*B. anthracis*	*Candida*	*E. Coli*
Streptocide	1.00	51	58	11	11	38
	0.10	35	29	0	0	30
	0.01	22	18	0	0	20
10a	1.00	11	0	15	13	0
	0.10	0	0	11	12	0
	0.01	0	0	0	0	0
10b	1.00	0	0	0	11	0
	0.10	0	0	0	11	0
	0.01	0	0	0	0	0
11	1.00	0	0	15	11	0
	0.10	0	0	13	0	0
	0.01	0	0	0	0	0
12a	1.00	15	16	71	19	23
	0.10	13	12	23	12	22
	0.01	11	0	11	0	19
12b	1.00	15	11	12	32	11
	0.10	0	0	11	29	0
	0.01	0	0	0	30	0
12c	1.00	16	13	24	27	11
	0.10	0	16	16	25	11
	0.01	0	11	0	11	0

Calculated (%): C, 35.23; H, 3.15; N, 19.17. ^{1}H NMR (DMSO-d_6), δ: 0.98 (t, 3 H, Me, J = 7.7); 1.31—1.48 (m, 2 H, CH_2); 1.51—1.67 (m, 2 H, CH_2); 2.26 (s, 3 H, MeC(O)); 3.75—3.92 (m, 2 H, CH_2); 10.51 (s, 1 H, NH); 10.74 (s, 1 H, NH); 12.26 (s, 1 H, NH); 13.00 (s, 1 H, NH). ^{1}H NMR (DMSO-d_6), δ: 4.70 s.

***N*-[5-{[2,4,6-Trioxo-1-(*o*-tolyl)-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl]sulfamoyl}-1,3,4-thiadiazol-2-yl]acetamide (5b), *N*-[5-({1-[2-(3,4-dimethoxyphenyl)ethyl]-2,4,6-trioxo-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl}sulfamoyl)-1,3,4-thiadiazol-2-yl]acetamide (5c), *N*-{5-[(4,6-dioxo-2-thioxo-5-trifluoromethyl-1-phenyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo-[2,3-*d*]pyrimidin-5-yl)sulfamoyl]-1,3,4-thiadiazol-2-yl}acetamide (7a), *N*-{5-[(4,6-dioxo-2-thioxo-1-*meta*-tolyl-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl)sulfamoyl]-1,3,4-thiadiazol-2-yl}acetamide (7b), *N*-{5-[(4,6-dioxo-2-thioxo-1-*para*-tolyl-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl)sulfamoyl]-1,3,4-thiadiazol-2-yl}acetamide (7c), *N*-(5-{[2,5-dioxo-1-methyl-4-(trifluoromethyl)imidazolidine-4-yl]sulfamoyl}-1,3,4-thiadiazol-2-yl)acetamide (9a), *N*-(5-{1-cyclopentyl-[2,5-dioxo-4-(trifluoromethyl)imidazolidine-4-yl]sulfamoyl}-1,3,4-thiadiazol-2-yl)acetamide (9b), *N*-(5-{[1-benzyl-2,5-dioxo-4-(trifluoromethyl)imidazolidine-4-yl]sulfamoyl}-1,3,4-thiadiazol-2-yl)acetamide (9c)** were obtained similarly to the compound **5a**. Yields, melting points, elemental analysis data and NMR spectroscopy data for compounds **5b**,**c**, **7a**—**c**, and **9a**—**c** are given in Tables 1 and 2.

(1-Butyl-2,4,6-trioxo-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pirimidine-5-yl)amide of 5-amino-1,3,4-thiadiazol-2-sulfonic acid (10a), {1-[2-(3,4-dimethoxyphenyl)-ethyl]-2,4,6-trioxo-5-trifluoromethyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl}amide of 5-amino-1,3,4-thiadiazol-2-sulfonic acid (10b), (4,6-dioxo-2-thioxo-5-trifluoromethyl-1-phenyl-2,3,4,5,6,7-hexahydro-1*H*-pyrrolo[2,3-*d*]pyrimidine-5-yl)amide of 5-amino-1,3,4-thiadiazol-2-sulfonic acid (11), [2,5-dioxo-1-methyl-4-(trifluoromethyl)imidazolidine-4-yl]amide of 5-amino-1,3,4-thiadiazol-2-sulfonic acid (12a), [1-cyclopentyl-2,5-dioxo-4-(trifluoromethyl)imidazolidine-4-yl]amide of 5-amino-1,3,4-thiadiazol-2-sulfonic acid (12b), [1-benzyl-2,5-dioxo-4-(trifluoromethyl)imidazolidine-4-yl]amide of 5-amino-1,3,4-thiadiazol-2-sulfonic acid (12c) (general procedure). Acetylated heterocyclic derivative of acetazolamide (0.05 mol) **5a**,**c**, **7a** or **9a**—**c** was refluxed for 1 h in 10 mL 10% HCl. The cooled solution was diluted with 20 mL of water, and the solution was neutrolized with 10% aqueous ammonia, the residue was filtered and recrystallized from 50% EtOH. Yields, melting points, elemental analysis data and NMR spectroscopy data for compounds **10a**,**b**, **11**, and **12a**—**c** are given in Tables 1 and 2.

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