Three-component condensation of 2-aminothiophene-3-carboxylic acid derivatives with aldehydes and Meldrum's acid

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New approach to the synthesis of 4,7-dihydro-5*H*-thieno[2,3-*b*]pyridin-6-ones using 2-aminothiophenes unsubstituted at position 3 of the thiophene ring has been developed. Labile 2-aminothiophenes have been obtained by the *in situ* decarboxylation of unstable 2-amino-3-thiophenecarboxylic acids. The three-component condensation of 2-aminothiophenes with aromatic aldehydes and the Meldrum's acid is the key step of the process.

Key words: 2-aminothiophenes, the Meldrum's acid, three-component condensation, decarboxylation, 4,7-dihydro-5*H*-thieno[2,3-*b*]pyridin-6-ones.

In the literature, there are data on the reaction of arylmethylidene derivatives of the Meldrum's acid **1** with stable heterocyclic analogs of enamines, *viz.*, aminopyrazoles,[1,2] aminopyridazinones,[3] and aminopyrimidinones,[4] resulting in the annulation of the dihydropyridinone ring with the Meldrum's acid (**2**) which plays the role of C_2-synthon. However, the number of such stable aminoheterocycles is limited. In particular, 2-aminothiophenes unsubstituted at position 3 have not been used in the condensation described due to their instability,[5] though it is absolutely obvious that transformations, in which aminothiophenes **3** are used as the 1,3-dinucleophiles, open access to development of fused heterocyclic system of the type **4**, which contain thiophene fragment and possess various biological activity.[6,7] In the present communication, we suggest to *in situ* generate these unstable enamines[3] with free position 3 from 2-aminothiophene-3-carboxylic acids **5**, which are known to readily undergo decarboxylation.[8—10] Acids **5** were generated from esters **6a—d** through sodium salts **7a—d** and used without purification and, as a rule, without isolation (Scheme 1).

The purpose of this research consisted in development of methods for the synthesis of substituted 4,7-dihydro-5*H*-thieno[2,3-*b*]pyridin-6-ones **4** on the basis of the *in situ* obtained 2-aminothiophenes **3**, aromatic aldehydes **8**, and the Meldrum's acid (**2**). Such thienopyridinones have been obtained earlier[11] by the Beckmann rearrangement of 4-aryl-4,5-dihydrocyclopenta[*b*]thiophen-6-one oximes. However, in this case the target products were formed as a mixture of two possible isomers in low yield. In the present work, we suggest a general efficient approach to the synthesis of structures **4** using readily decarboxylating acids **5** as the starting compounds. The latter were obtained by the alkaline hydrolysis of the corresponding 2-amino-3-thiophenecarboxylic acid esters **6**, the products of the Gewald reaction. The hydrolysis was carried out in aqueous ethanol since in the alkali alcohol solution, a rearrangement with the formation of sodium salts of 3-cyano-2-hydroxythiophenes is possible[12,13] and the presence of electron-withdrawing substituents at position 5 of the thiophene ring makes the alkaline hydrolysis considerably more difficult.

The procedures for the synthesis of target thieno[2,3-*b*]-pyridinones developed by us differed in the methods of generation of labile 2-aminothiophenes **3**. When esters **6a—c** were used, the alkaline hydrolysis resulted in the corresponding sodium salts **7a—c**, which in this case were not converted to acid **5**, rather they were directly introduced into the reaction; subsequent addition of acetic acid to the reaction mixture led to neutralization, decarboxylation, and *in situ* generation of 2-aminothiophenes **3a—c** (method *A*). It should be noted that in the case of ester **6a**, acetic acid can be also used as the solvent, however, this leads to the insignificant decrease in the yields of target compounds **4**, therefore, excess of acetic acid in ethanol was used. In the case of esters **6b,c**, the use of acetic acid as the solvent is the optimum for the synthesis of compounds **4j—s**. To obtain products **4k—x** on the basis of ester **6d**, the reaction conditions have been modified since the use of salt **7d** led to final compounds in low yields, apparently, due to the side reactions. In this case, we isolated acid **5d**, the condensation of which

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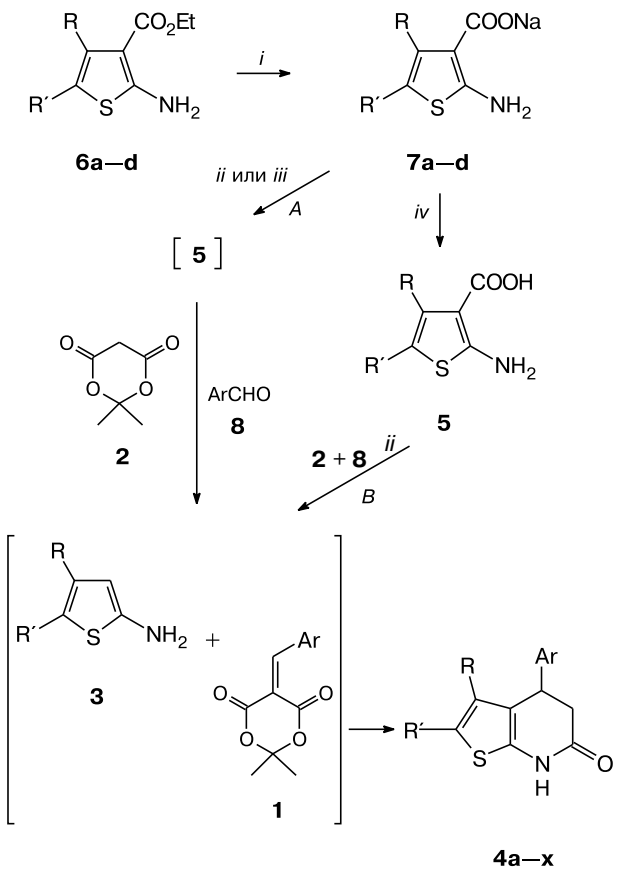
Scheme 1



Conditions and reagents: *i.* EtOH, H_2O, NaOH, Δ; *ii*. AcOH, Δ; *iii*. AcOH, EtOH, Δ; *iv*. HCl.

5—7: RR′ = $(CH_2)_4$ (**a**); R = H, R′ = Et (**b**); R = Me, R′ = PhNHCO (**c**); R = Me, R′ = Ac (**d**).

4	R	R′	Ar′
a	$(CH_2)_4$	$(CH_2)_4$	Ph
b	$(CH_2)_4$	$(CH_2)_4$	4-$MeOC_6H_4$
c	$(CH_2)_4$	$(CH_2)_4$	3,5-$(MeO)_2$-4-HOC_6H_2
d	$(CH_2)_4$	$(CH_2)_4$	Thiophen-3-yl
e	$(CH_2)_4$	$(CH_2)_4$	3,4-$(MeO)_2C_6H_3$
f	$(CH_2)_4$	$(CH_2)_4$	4-ClC_6H_4
g	$(CH_2)_4$	$(CH_2)_4$	2,3-$(MeO)_2C_6H_3$
h	$(CH_2)_4$	$(CH_2)_4$	Pyridin-3-yl
i	$(CH_2)_4$	$(CH_2)_4$	4-$CF_3C_6H_4$
j	H	Et	3,4-$(MeO)_2C_6H_3$
k	Me	PhNHCO	3-ClC_6H_4
l	Me	PhNHCO	3-$MeOC_6H_4$
m	Me	PhNHCO	3,4,5-$(MeO)_3C_6H_2$
n	Me	PhNHCO	Pyridin-3-yl
o	Me	PhNHCO	Thiophen-3-yl
p	Me	PhNHCO	3-$CF_3C_6H_4$
q	Me	PhNHCO	3-MeO-4-HOC_6H_3
r	Me	PhNHCO	3,4-$OCH_2OC_6H_3$
s	Me	PhNHCO	3-FC_6H_4
t	Me	Ac	Thiophen-3-yl
u	Me	Ac	Ph
v	Me	Ac	4-$MeOC_6H_4$
w	Me	Ac	4-ClC_6H_4
x	Me	Ac	3,4-$(MeO)_2C_6H_3$

with aldehyde **8** and the Meldrum's acid (**2**) in acetic acid allowed us to obtain thieno[2,3-*b*]pyridinones **4** (method *B*). It should be noted that these conditions can be also applied for ester **6c**.

The compounds obtained by us are crystalline substances with high melting points, the structures of which have been confirmed by the elemental analysis and ^{1}H NMR spectroscopic data. In the ^{1}H NMR spectra of products obtained, characteristic signals for the protons of the methyne fragment in the region δ 4.11—4.49 and nonequivalent protons of the methylene unit in the region δ 2.51—3.20 are observed, that is in good agreement with the literature data for analogous subjects.[1—4]

We have shown that, in contrast to approaches described earlier,[1—4] arylmethylidene derivatives of the Meldrum's acid are reasonable to be obtained directly in the reaction mixture, that decreases the number of steps for the process and virtually does not affect the yields of target products. In conclusion, the optimum method for the synthesis of thieno[2,3-*b*]pyridinones **4** is a three-component condensation of aromatic aldehyde **8**, the Meldrum's acid (**2**), and the corresponding salts **7** or acid **5** as the synthetic equivalents of labile 2-aminothiophene **3**. Apparently, the process includes the Michael addition of 2-aminothiophene to the Meldrum's acid arylmethylidene derivative with subsequent intramolecular cyclization, which is accompanied by elimination of the acetone and CO_2 molecules.

Experimental

^{1}H NMR spectra were recorded on a Bruker AM-300 (300 MHz) and Bruker WM-250 (250 MHz) spectrometers in DMSO-d_6. Mass spectra were recorded on a FINNIGAN MAT INCOS 50 instrument (direct inlet, EI, 70 eV). Melting points were measured on a Boetius heating apparatus and were not corrected. Monitoring of the reaction course and purity of compounds obtained were performed by TLC (Merck Silica gel 60 F254 plates, eluent: ethyl acetate—hexane).

Esters **6a**,[14] **6b**,[15] **6c**,[16] and **6d** [15] were obtained according to the procedures described earlier.

Synthesis of 4,7-dihydro-5*H*-thieno[2,3-*b*]pyridin-6-ones 4a—x (general procedure). ***A***. A mixture of ester **6a-c** (2 mmol) and NaOH (0.16 g, 4 mmol) in ethanol (5 mL) and water (5 mL) was refluxed for 2 h, then evaporated to dryness. The Meldrum's acid (**2**) (0.32 g, 2.2 mol), the corresponding aldehyde (2.1 mmol), and acetic acid (4 mL) in the case of esters **6b,c** or acetic acid (0.36 g, 6 mmol) and ethanol (4 mL) in the case of ester **6a** were added to the residue obtained. The mixture was refluxed for 2 h and concentrated, the residue was recrystallized from ethanol, the precipitate was filtered off and washed with ethanol and water on the filter.

B. A mixture of ester **6c,d** (2 mmol) and NaOH (0.16 g, 4 mmol) in ethanol (5 mL) and water (5 mL) was refluxed for 2 h and cooled followed by addition of concentrated hydrochloric acid (0.4 g, 4 mmol), a precipitate formed was filtered off and dried in air. Then a mixture of the acid obtained (2 mmol), the Meldrum's acid (**2**) (0.32 g, 2.2 mol), and the corresponding

Table 1. Yields, melting points, synthetic procedures, and elemental analysis data of compounds **4a—x**

Compound	Synthetic procedure	M.p./°C	Yield (%)	Found / Calculated (%)					Molecula formula
				C	H	N	S	Cl	
4a	*A*	217—219	56	71.91 72.05	6.15 6.04	4.85 4.94	11.25 11.36	—	$C_{17}H_{17}NOS$
4b	*A*	172—174	52	70.06 70.17	6.24 6.37	4.59 4.68	10.36 10.45	—	$C_{18}H_{19}NO_2S$
4c	*A*	239—240	59	63.29 63.42	6.04 6.14	3.78 3.89	9.16 9.03	—	$C_{19}H_{21}NO_4S$
4d	*A*	217—218	47	62.39 62.51	5.35 5.24	4.71 4.82	22.29 22.37	—	$C_{15}H_{15}NOS_2$
4e	*A*	236—237	57	66.61 66.73	5.99 6.11	4.19 4.05	9.50 9.62	—	$C_{19}H_{21}NO_3S$
4f	*A*	191—193	45	64.41 64.30	5.19 5.07	4.56 4.69	10.21 10.10	11.27 11.32	$C_{17}H_{16}ClNOS$
4g	*A*	210—211	52	66.31 66.43	6.35 6.45	4.20 4.29	9.51 9.60	—	$C_{19}H_{21}NO_3S$
4h	*A*	228—229	47	67.39 67.50	5.61 5.52	9.99 10.08	11.41 11.34	—	$C_{16}H_{16}N_2OS$
4i	*A*	230—231	51	61.39 61.24	4.61 4.72	3.78 3.86	—	—	$C_{18}H_{16}F_3NOS$
4j	*A*	155—156	25	64.33 64.42	6.03 6.11	4.41 4.50	10.10 10.19	—	$C_{17}H_{19}NO_3S$
4k	*A*, *B*	266—267	44/51	63.55 63.64	4.32 4.41	7.06 6.98	8.08 8.17	8.93 9.01	$C_{21}H_{17}ClN_2O_2S$
4l	*A*, *B*	248—249	53/60	67.33 67.24	5.14 5.23	7.14 7.08	8.17 8.24	—	$C_{22}H_{20}N_2O_3S$
4m	*A*, *B*	276—277	39/45	63.70 63.81	5.35 5.27	6.19 6.11	7.09 7.16	—	$C_{24}H_{24}N_2O_5S$
4n	*A*, *B*	165—166	36/44	66.10 66.18	4.71 4.80	11.56 11.48	8.82 8.91	—	$C_{20}H_{17}N_3O_2S$
4o	*A*, *B*	274—275	43/51	61.93 61.82	4.38 4.27	7.60 7.69	17.40 17.46	—	$C_{19}H_{16}N_2O_2S_2$
4p	*A*, *B*	281—283	48/56	61.39 61.52	3.98 3.89	6.51 6.60	—	—	$C_{22}H_{17}F_3N_2O_2S$
4q	*A*, *B*	252—253	33/39	64.69 64.81	4.94 4.88	6.86 6.80	7.85 7.79	—	$C_{22}H_{20}N_2O_4S$
4r	*A*, *B*	229—230	57/63	65.01 65.13	4.46 4.59	6.89 6.97	7.89 7.80	—	$C_{22}H_{18}N_2O_4S$
4s	*A*, *B*	270—271	49/55	66.30 66.42	4.50 4.42	7.36 7.45	—	—	$C_{21}H_{17}FN_2O_2S$
4t	*B*	231—233	50	57.71 57.59	4.50 4.59	4.81 4.90	22.01 22.10	—	$C_{14}H_{13}NO_2S_2$
4u	*B*	229—231	42	67.34 67.48	5.30 5.40	4.91 4.83	11.24 11.14	—	$C_{16}H_{15}NO_2S$
4v	*B*	187—188	47	64.74 64.88	5.43 5.52	4.44 4.53	10.17 10.08	—	$C_{17}H_{17}NO_3S$
4w	*B*	219—220	51	60.09 60.23	4.41 4.53	4.38 4.29	10.03 10.12	11.09 11.17	$C_{16}H_{14}ClNO_2S$
4x	*B*	236—237	48	62.59 62.45	5.54 5.63	4.05 3.97	9.28 9.34	—	$C_{18}H_{19}NO_4S$

Table 2. ^{1}H NMR spectra (DMSO-d_6, δ, *J*/Hz) of compounds **4a—x**

Compound	R	R′	H—C—H (d, 1 H)	H—C—H (dd, 1 H)	CH (d, 1 H)	Ar	NH (br.s 1 H)
4a		1.55—1.98 (m, 5 H, CH_2); 2.32—2.46 (m, 3 H, CH_2)	2.54 (*J* = 16)	3.02 (*J* = 8, 16)	4.11 (*J* = 8)	7.04—7.42 (m, 5 H)	10.37
4b		1.51—1.98 (m, 5 H, CH_2); 2.32—2.48 (m, 3 H, CH_2)	2.54 (*J* = 16)	2.99 (*J* = 8, 16)	4.11 (*J* = 8)	6.83 (m, 2 H, *J* = 8); 6.97 (d, 2 H, *J* = 8); 3.71 (s, 3 H, OMe)	10.35
4c		1.55—1.75 (m, 4 H, CH_2); 1.09—2.07 (m, 1 H, CH_2); 2.31—2.46 (m, 3 H, CH_2)	2.54 (*J* = 16)	2.99 (*J* = 8, 16)	4.11 (*J* = 8)	6.34 (s, 2 H); 3.67 (s, 6 H, 2OMe); 8.25 (br.s, 1 H, OH)	10.35
4d		1.59—1.78 (m, 4 H, CH_2); 2.11—2.46 (m, 4 H, CH_2)	2.54 (*J* = 16)	3.03 (*J* = 8, 16)	4.39 (*J* = 8)	6.78 (d, 1 H, *J* = 3); 6.92 (dd, 1 H, *J* = 3, 5); 7.32 (d, 1 H, *J* = 5)	10.39
4e		1.53—2.11 (m, 5 H, CH_2); 2.28—2.45 (m, 3 H, CH_2)	2.54 (*J* = 16)	2.95 (*J* = 8, 16)	4.03 (*J* = 8)	6.45 (d, 1 H, *J* = 8); 6.81 (m, 2 H); 3.67 (s, 6H, 2OMe)	10.33
4f		1.53—1.96 (m, 5 H, CH_2); 2.28—2.43 (m, 3 H, CH_2)	2.54 (*J* = 16)	3.01 (*J* = 8, 16)	4.13 (*J* = 8)	7.07 (d, 2 H, *J* = 8); 7.35 (d, 2 H, *J* = 8)	10.38
4g		1.51—1.89 (m, 5 H, CH_2); 2.22—2.39 (m, 3 H, CH_2)	2.51 (*J* = 16)	2.99 (*J* = 8, 16)	4.35 (*J* = 8)	6.33 (m, 1 H); 6.89(m, 2 H);	10.33
4h		1.53—1.91 (m, 5 H, CH_2); 2.33—2.45 (m, 3 H, CH_2)	2.55 (*J* = 16)	3.07 (*J* = 8, 16)	4.22 (*J* = 8)	7.38 (m, 2 H, Py); 8.40 (m, 2 H, Py)	10.48
4i		1.53—1.98 (m, 5 H, CH_2); 2.32—2.45 (m, 3 H, CH_2)	2.54 (*J* = 16)	3.07 (*J* = 8, 16)	4.26 (*J* = 8)	7.31 (d, 2 H, *J* = 8); 7.68 (d, 2 H, *J* = 8)	10.46
4j	6.25 (s, 1 H)	1.14 (t, 3 H, CH_3, *J* = 8); 2.64 (m, 2 H, CH_2)	2.63[a]	2.78 (*J* = 7, 16)	4.41[b] (*J* = 7)	6.62 (m, 1 H); 6.86 (m, 2 H); 3.71 (s, 6 H, 2 OMe)	10.36
4k	2.19 (s, 3 H, Me)	7.05—7.67 (m, 5 H, C_6H_5); 9.76 (br.s, 1 H, NH)	2.54 (*J* = 16)	3.15 (*J* = 8, 16)	4.37 (*J* = 8)	7.05—7.67 (m, 4 H, C_6H_4)	10.93
4l	2.19 (s, 3 H, Me)	6.64—7.63 (m, 5 H, C_6H_5); 9.74 (br.s, 1 H, NH)	2.56 (*J* = 16)	3.14 (*J* = 8, 16)	4.28 (*J* = 8)	6.64—7.63 (m, 4 H, C_6H_4); 3.72 (s, 3 H, OMe)	10.87
4m	2.24 (s, 3 H, Me)	7.06 (t, 1 H, C_6H_5, *J* = 7); 7.31 (t, 2 H, C_6H_5, *J* = 8); 7.63 (t, 2 H, C_6H_5, *J* = 8); 9.74 (br.s, 1 H, NH)	2.56 (*J* = 16)	3.14 (*J* = 8, 16)	4.28 (*J* = 8)	6.42 (s, 2 H)	10.89
4n	2.20 (s, 3 H, Me)	7.05—7.71 (m, 5 H, C_6H_5); 9.76 (br.s, 1 H, NH)	2.60 (*J* = 16)	3.20 (*J* = 8, 16)	4.42 (*J* = 8)	7.05—7.71 (m, 2 H, Py); 8.43 (m, 2 H, Py)	10.95
4o	2.30 (s, 3 H, Me)	6.91—7.69 (m, 5 H, C_6H_5); 9.72 (br.s, 1 H, NH)	2.63 (*J* = 16)	3.05 (*J* = 8, 16)	4.35 (*J* = 8)	6.91—7.69 (m, 3 H, Thi)	10.83
4p	2.20 (s, 3 H, Me)	7.02—7.68 (m, 9.75 (br.s, 1 H, NH)	2.61 (*J* = 16)	3.20 (*J* = 8, 16)	4.49 (*J* = 8)	7.02—7.68 (m, 4 H, C_6H_4)	10.95
4q	2.21 (s, 3 H, Me)	6.31—6.69 (m, 5 H, C_6H_5); 9.72 (br.s, 1 H, NH)	2.57 (*J* = 16)	3.08 (*J* = 8, 16)	4.18 (*J* = 8)	6.31—6.69 (m, 3 H, C_6H_3); 8.88 (s, 1 H, OH); 3.35 (s, 1 H, OMe)	10.82
4r	2.19 (s, 3 H, Me)	7.06 (t, 1 H, C_6H_5, *J* = 7); 7.32 (t, 2 H, C_6H_5, *J* = 7); 7.63 (d, 2 H, C_6H_5, *J* = 8); 9.73 (br.s, 1 H, NH)	2.54 (*J* = 16)	3.10 (*J* = 8, 16)	4.24 (*J* = 8)	5.98 (s, 2 H, CH_2); 6.52 (d, 1 H, *J* = 7); 6.67 (s, 1 H); 6.85 (d, 1 H, *J* = 8)	10.82
4s	2.20 (s, 3 H, Me)	6.88—7.68 (m, 5 H, C_6H_5); 9.73 (br.s, 1 H, NH)	2.58 (*J* = 16)	3.16 (*J* = 8, 16)	4.37 (*J* = 8)	6.88—7.68 (m, 5 H, C_6H_4)	10.90
4t	2.20 (s, 3 H, Me)	2.40 (s, 3 H, Me)	2.56 (*J* = 16)	3.15 (*J* = 8, 16)	4.33 (*J* = 8)	7.06 (m, 2 H, Thi); 7.31 (m, 1 H, Thi)	11.02
4u	2.22 (s, 3 H, Me)	2.41 (s, 3 H, Me)	2.56 (*J* = 16)	3.15 (*J* = 8, 16)	4.33 (*J* = 8)	7.03—7.36 (m, 5 H, C_6H_5)	11.02
4v	2.22 (s, 3 H, Me)	2.41 (s, 3 H, Me)	2.58 (*J* = 16)	3.11 (*J* = 8, 16)	4.27 (*J* = 8)	6.85 (d, 2 H, *J* = 8); 6.96 (d, 2 H, *J* = 8); 3.7 (s, 3 H, OMe)	10.99
4w	2.22 (s, 3 H, Me)	2.41 (s, 3 H, Me)	2.55 (*J* = 16)	3.16 (*J* = 8, 16)	4.36 (*J* = 8)	7.07 (d, 2 H, *J* = 8); 7.37 (d, 2 H, *J* = 8)	11.02
4x	2.24 (s, 3 H, Me)	2.42 (s, 3 H, Me)	2.58 (*J* = 16)	3.11 (*J* = 8, 16)	4.26 (*J* = 8)	6.38 (d, 1 H, *J* = 8); 6.82 (m, 2 H); 3.71 (s, 6 H, 2 OMe)	11.02

[a] Multiplet.
[b] Triplet.

aldehyde (2.1 mmol) in acetic acid (4 mL) was refluxed for 2 h and concentrated, the residue was recrystallized from ethanol, the precipitate was filtered off and washed with ethanol and water on the filter.

Characteristics of compounds obtained are given in Tables 1 and 2.

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