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POSITIONAL ISOMERS OF THIENOPYRIMIDINONES

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Condensation of o-cyanomethylbenzoic acids with esters of 2-aminothiophene-3-carboxylic and 3-amino-thiophene-2-carboxylic acids to give isomeric 2-(4-oxo-3,4-dihydrothieno[2,3-d]pyrimidin-2-ylmethyl)-benzoic acids and their ethyl and phenacyl esters. Methylation of esters occurs at N-3. The spectroscopic characteristics of the positional isomers are compared at the level of carboxylic acids, their esters, and methylated products.

Keywords: 4-oxopyrimidinones, thienopyrimidinones, thiophenes.

We have shown previously that derivatives of 4-oxo-2-quinazolinylmethylbenzoic acids are formed by the reaction of *o*-cyanomethylbenzoic acid (**1**) with anthranilic [1] and N-methylanthranilic acids [2]. In a continuation of this work we have used in the condensation with acid **1** any derivatives of vicinal enaminoacids – esters of isomeric amino acids of the thiophene series.

It was established that boiling a chlorobenzene solution of acid **1** with 2 equivalents of the esters of the amino acids **2** or **3** for 10 min gave derivatives of thienopyrimidinones **4** and **5** in yields of not less than 70% (Table 1).

The yields of the required products were halved when equivalent amounts of the reagents were used. The structures of the products as the derivatives 2-(4-oxo-3,4-dihydrothieno[2,3-*d*]- and –[3,2-*d*]pyrimidin-2-ylmethyl)benzoic acids of the series **4** and **5** respectively were assigned on the basis of previous experiments on the condensation of acid **1** with vicinal enamine acids and spectroscopic data for the compounds synthesized. For this purpose comparisons with previously synthesized quinazolinones **10a-c** are given in Tables 2 and 3.

Compounds of the 4-oxo-3,4-dihydrothieno[2,3-*d*]series were synthesized more than 35 years ago [3]. It was established that 2-benzyl-substituted compounds of this system exhibit analgesic and anti-inflammatory properties [4]. However isomeric structures of type **5** have not been investigated until now.

2-(4-Oxo-3,4-dihydrothieno[2,3-*d*]- and –[3,2-*d*]pyrimidin-2-ylmethyl)benzoic acids of the **4** and **5** series have amphoteric properties similar to normal amino acids: they dissolve in 2 N alkaline solution and selectively in 2N HCl (derivatives of the [2,3-*d*] series dissolve considerably better). In this connection esterification of these amino acids is possible only by reaction of compounds with ethyl bromide (in DMSO) or with phenacyl bromide (in acetonitrile) in the presence of triethylamine [5].

No difference in chemical behavior of the positional isomers was noted. By alkylation at atom N-3 by standard methods [6] we obtained esters **8** and **9** of N-methylated 2-(4-oxo-3, 4-dihydrothieneo[2,3-*d*]- and –[3,2-*d*]pyrimidin-2-ylmethyl)benzoic acids.

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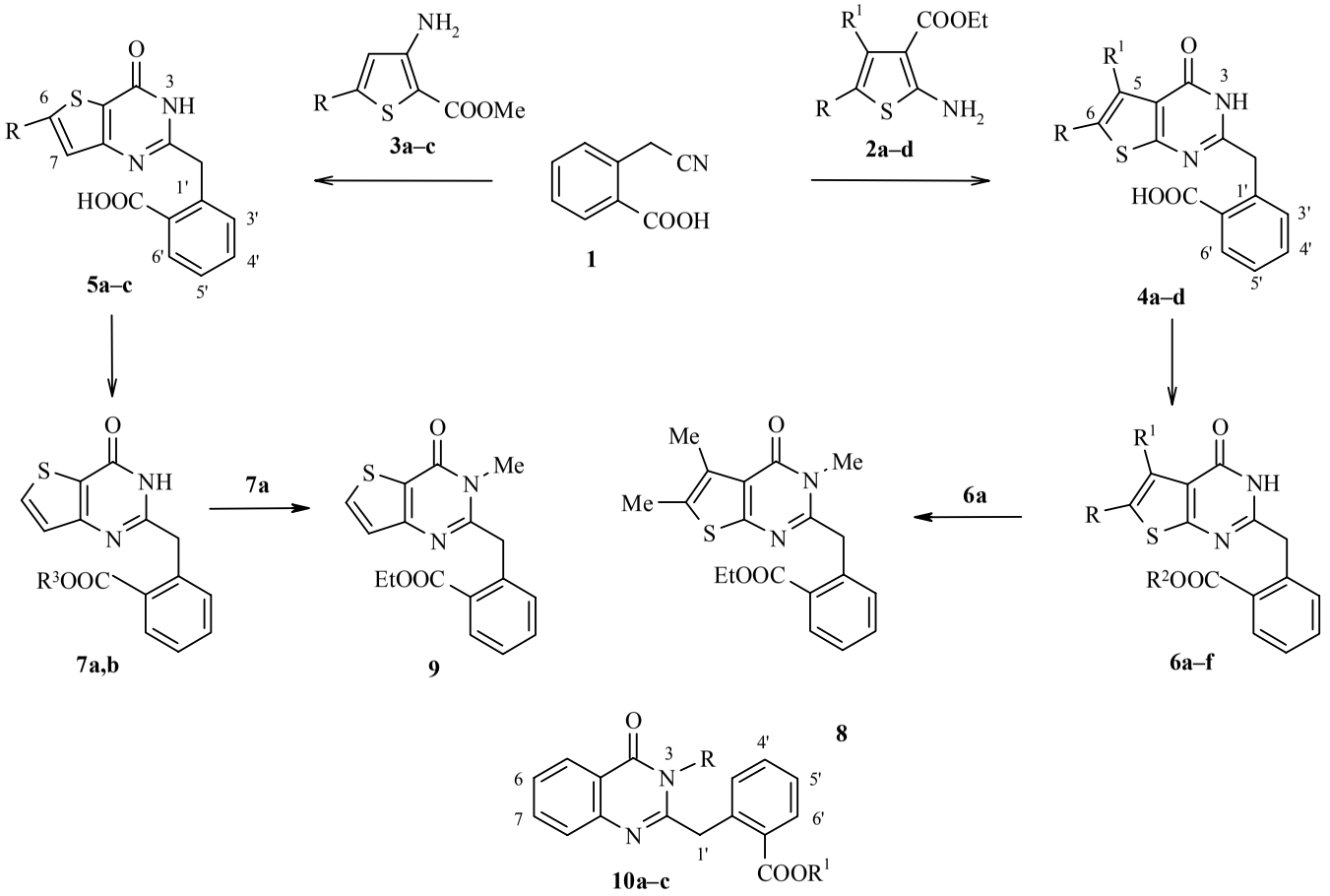

4 a R = R^1 = Me, **b** R + R^1 = $(CH_2)_3$, **c** R + R^1 = $(CH_2)_4$, **d** R = H, R^1 = Ph; **5 a** R = H, **b** R = 4'-ClC_6H_4, **c** R = 4'-FC_6H_4;
6 a R = R^1 = Me, R^2 = Et, **b** R = R^1 = Me, R^2 = CH_2COPh, **c** R + R^1 = $(CH_2)_3$, R^2 = Et, **d** R + R^2 = $(CH_2)_3$, R^2 = CH_2COPh,
e R + R^1 = $(CH_2)_4$, R^2 = Et, **f** R = R^1 = $(CH_2)_4$, R^2 = CH_2COPh; **7 a** R^3 = Et, **b** R^3 = CH_2COPh;**10 a** R = R^1 = H,
b R = H, R^1 = Et, **c** R = Me, R^1 = Et

Table 1. Characteristics of the Compounds Synthesized.

Compound	Empirical formula	Found, % / Calculated, %				mp, °C*	Yield, %
		C	H	N	S		
4a	$C_{16}H_{14}N_2O_3S$	61.21 / 61.13	4.38 / 4.49	8.76 / 8.91	10.12 / 10.20	338	73
4b	$C_{17}H_{14}N_2O_3S$	62.82 / 62.56	4.22 / 4.32	8.33 / 8.58	9.75 / 9.83	333	85
4c	$C_{18}H_{16}N_2O_3S$	63.60 / 63.51	4.61 / 4.74	8.13 / 8.23	9.33 / 9.42	350	88
4d	$C_{20}H_{14}N_2O_3S$	66.02 / 66.29	3.91 / 3.89	7.55 / 7.73	8.95 / 8.85	290	90
5a	$C_{14}H_{10}N_2O_3S$	58.93 / 58.73	3.63 / 3.52	9.52 / 9.78	11.08 / 11.20	265	70
5b	$C_{20}H_{13}ClN_2O_3S$	60.82 / 60.53	3.42 / 3.30	6.92 / 7.06	8.14 / 8.08	290*[2]	92
5c	$C_{20}H_{13}FN_2O_3S$	63.12 / 63.15	3.30 / 3.44	7.13 / 7.36	8.24 / 8.43	305*[2]	90
6a	$C_{18}H_{18}N_2O_3S$	63.41 / 63.14	5.41 / 5.30	8.35 / 8.18	9.55 / 9.36	210	80
6b	$C_{24}H_{20}N_2O_4S$	66.78 / 66.65	4.46 / 4.66	6.23 / 6.48	7.55 / 7.41	240	92
6c	$C_{19}H_{18}N_2O_3S$	64.62 / 64.39	5.01 / 5.12	7.76 / 7.90	9.20 / 9.05	240	84
6d	$C_{25}H_{20}N_2O_4S$	67.88 / 67.55	4.46 / 4.53	6.15 / 6.30	7.23 / 7.21	245	88
6e	$C_{20}H_{20}N_2O_3S$	65.62 / 65.20	5.36 / 5.47	7.58 / 7.60	8.84 / 8.70	242	82
6f	$C_{26}H_{22}N_2O_4S$	68.56 / 68.10	4.96 / 4.84	6.03 / 6.11	6.87 / 6.99	245	90
7a	$C_{16}H_{14}N_2O_3S$	61.79 / 61.13	4.63 / 4.49	9.04 / 8.91	10.45 / 10.20	130	80
7b	$C_{22}H_{16}N_2O_4S$	65.92 / 65.33	4.08 / 3.99	6.82 / 6.93	8.04 / 7.93	200*[2]	90
8	$C_{19}H_{20}N_2O_3S$	63.91 / 64.02	5.57 / 5.66	7.78 / 7.86	9.14 / 9.00	160	83
9	$C_{17}H_{16}N_2O_3S$	61.52 / 62.18	4.86 / 4.91	8.68 / 8.53	9.92 / 9.76	120	80

* Solvent: DMF (compounds **4a-d**, **6c**, **8**), HOAc (compounds **5a, 7a**, **9**), DMSO (compounds **6a,b**)
*[2] Conversion temperature.

Comparison of the ^{1}H NMR spectra of the acids with structures **4** and **5** (Table 2) showed that the hydrogen-containing groups of the *o*-carboxybenzyl substituent are practically unaffected by the change in position of the sulfur atom in the bicyclic system. The resonances of the 1′-CH_2 group, and the protons in positions 3′,4′,5′, and 6′ are observed at standard resonances with minimal shift of 0.03 ppm in either direction. The same may be observe on comparing the ^{1}H NMR spectra of the esters **6a-f** and **7a,b** and the N-methylated compounds **8** and **9**.

In the IR spectra of the acids **4** and **5** (Table 3) in the 3200-2500 cm^{-1} region there are broad absorption bands, frequently with several maxima. These bands arise, at least frequently, as a result of absorption of dimmers of the acids and are assigned to bond vibrations of the O-H bond. From the massive vibration in this region one can isolate the 2700-2500 cm^{-1}, assigned to bonding vibrations of the hydroxyl OH. Examination of the intermolecular association assists in resolving the intermolecular solution. The extremely low frequency of the valence vibration of the carbonyl group (or the COOH group) is a direct reflection of this effect. The band of the carbonyl absorption of the carboxyl group is superimposed on valence vibration of the carbonyl group of the pyrimidin-4-one unit, the latter being also an active participant in association.

TABLE 2. NMR ^{1}H Spectra of Synthesized Compounds

Compound	Chemical shifts, δ, ppm (*J*, Hz)													
	Position of resonating groups (atoms)									COOR				
	3 (s)	5	6	7	H-1' (2H, s)	H-3' (1H, d)	H-4' (1H, t)	H-5' (1H, t)	H-6' (1H, t)	H (1H, s)	CH_2CH_3	1" (2H, s)	2"+6" (2H, d)	3"+ 4"+5" (3H, m)
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
4a	12.16 (1H)	2.31 (3H, s)	2.37 (3H, s)	—	4.30	7.29 (^3J = 7.2)	7.48 (^3J = 7.2)	7.6 (^3J = 7.6)	7.91 (^3J = 7.6)	12.4	—	—	—	—
4b	12.25 (1H)	2.91 (4H, m); 2.41 (2H, m)		—	4.31	7.29 (^3J = 7.2)	7.48 (^3J = 7.2)	7.35 (^3J = 7.2)	7.93 (^3J = 8.0)	12.69	—	—	—	—
4c	12.20 (1H)	2.86 (2H, s); 2.69 (2H, s); 1.80 (4H, m)		—	4.35	7.29 (^3J = 7.2)	7.49 (^3J = 7.2)	7.36 (^3J = 7.2)	7.91 (^3J = 7.2)	12.69	—	—	—	—
4d	12.34 (1H)	7.40-7.30 (5H, m)	7.19 (1H, s)	—	4.38	7.53-7.49 (3H, m)		7.53-7.49 (m)	7.96 (^3J = 7.6)	12.79	—	—	—	—
5a	12.35 (1H)	—	7.91 (d, ^3J = 5.21)	7.16 (1H, d, ^3J = 5.61)	4.36	7.32 (^3J = 7.6)	7.48 (^3J = 7.6)	7.36 (^3J = 7.6)	7.93 (^3J = 7.2)	12.8	—	—	—	—
5b	12.47 (1H)	—	7.73 (2H, d, ^3J = 8.41, H-2',6'); 7.42 (2H, d, ^3J = 8.41 H-3',5')	7.60 (1H, s)	4.34	7.31 (^3J = 7.6)	7.47 (^3J = 7.2)	7.36 (^3J = 7.6)	7.92 (^3J = 7.6)	—*		—	—	—
5c	12.41 (1H)	—	7.76 (2H, dd, *J* = 5.2, *J* = 3.6, H-2',6',); 7.17 (2H, t, ^3J = 8.6, H-3',5')	7.54 (1H, s)	4.35	7.31 (^3J = 7.6)	7.75 (^3J = 7.0)	7.35 (^3J = 7.6)	7.92 (^3J = 7.2)	—*	—	—	—	—

TABLE 2 (continued)

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
10a	12.1 (1H)	8.05 (1H, d, ^3J = 8.0)	7.37 (1H, t, ^3J = 8.0)	7.65 (1H, t, ^3J = 7.6)*[2]	4.33	7.32 (^3J = 8.0)	7.47 (^3J = 7.8)	7.36 (^3J = 8.0)	7.91 (^3J = 7.2)	12.7	—	—	—	—
6a	12.23 (1H)	2.30 (3H, s)	2.33 (3H, s)	—	4.29	7.32 (^3J = 7.6)	7.49 (^3J = 7.6)	7.36 (^3J = 7.6)	7.88 (^3J = 7.6)	—	4.20 (2H, q, ^3J = 7.2); 1.26 (3H, t, ^3J = 6.8)	—	—	—
6b	12.17 (1H, s)	2.33 (3H, s)	2.37 (3H, s)	—	4.31	7.35 (^3J = 8.0)	7.64 (^3J = 7.6)	7.44 (^3J = 7.2)	8.09 (^3J = 8.0)	—	—	5.59	7.97 (^3J = 8.4)	—
6c	12.32 (1H, s)	2.90 (4H, m); 2.40 (2H, m)		—	4.28	7.33 (^3J = 7.6)	7.50 (^3J = 7.2)	7.37 (^3J = 7.6)	7.89 (^3J = 7.6)	—	4.20 (2H, q, ^3J = 7.2) 1.26 (3H, t, ^3J = 7.2)	—	—	—
6d	12.29 (1H)	2.87–2.93 (4H, m); 2.41 (2H, t, ^3J = 7.2)		—	4.33	7.36 (^3J = 7.6)	7.65 (^3J = 7.2)	7.45 (^3J = 7.6)	8.10 (^3J = 7.6)	—	—	5.59	7.97 (^3J = 7.2)	—
6e	12.21 (1H)	2.87 (2H, br. s); 2.69 (2H, br. s); 1.80 (4H, br. s)		—	4.27	7.32 (^3J = 7.2)	7.50 (^3J = 7.2)	7.37 (^3J = 7.6)	7.88 (^3J = 7.2)	—	4.20 (2H, q, ^3J = 6.8); 1.27 (3H, t, ^3J = 7.2)	—	—	—
6f	12.20 (1H)	2.89 (2H, br. s); 2.71 (2H, br. s); 1.80 (4H, m)		—	4.32	7.36 (^3J = 7.6)	7.45 (^3J = 7.6)	7.66 (^3J = 7.2)	8.09 (^3J = 8.0)	—	—	5.60	7.98	7.59-7.52

TABLE 2 (continued)

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
7a	12.41 (1H)	—	7.90 (d, ^3J = 5.2)	7.12 (d, ^3J = 5.2)	4.34	7.35 (^3J = 7.6)	7.51 (^3J = 7.6)	7.38 (^3J = 7.2)	7.91 (^3J = 7.2)	—	4.22 (2H, q, ^3J = 6.8); 1.27 (3H, t, ^3J = 6.8)	—	—	—
7b	12.39 (1H)	—	7.93 (d, ^3J = 5.2)	7.16 (d, ^3J = 5.2)	4.37	7.37 (^3J = 8.0)	7.64 (^3J = 7.2)	7.45 (^3J = 7.6)	8.10 (^3J = 7.6)	—	—	5.60	7.97	7.57-7.50
10b	12.22 (1H)	8.07 (1H, d, ^3J = 8.0)	7.44 (1H, t, ^3J = 6.8)	7.70 (1H, t, ^3J = 6.8)*[3]	4.31	7.39 (^3J = 8.4)	7.44 (^3J = 6.8)	7.44 (^3J = 6.8)	7.88 (^3J = 6.8)	—	4.19 (2H, q, ^3J = 7.2); 1.24 (3H, t, ^3J = 7.2)	—	—	—
8	3.56 (3H)	2.33 (3H, s)	2.40 (3H, s)	—	4.52	7.27 (^3J = 7.6)	7.52 (^3J = 7.2)	7.40 (^3J = 7.6)	7.96 (^3J = 7.6)	—	4.16 (2H, q, ^3J = 6.8); 1.27 (3H, t, ^3J = 6.8)	—	—	—
9	3.65 (3H)	—	7.92 (d, ^3J = 4.8)	7.09 (d, ^3J = 4.8)	4.58	7.30 (^3J = 7.2)	7.53 (^3J = 7.2)	7.41 (^3J = 7.2)	7.98 (^3J = 7.6)	—	4.18 (2H, q, ^3J = 6.8); 1.21 (3H, t, ^3J = 6.8)	—	—	—
10c	3.60 (3H)	8.09 (1H, d, ^3J = 7.6)	7.39 (1H, t, ^3J = 7.6)	7.64 (1H, t, ^3J = 7.6)*[4]	4.57	7.33 (^3J = 8.4)	7.52 (^3J = 7.2)	7.39 (^3J = 7.6)	7.95 (^3J = 8.0)	—	4.14 (2H, q, ^3J = 6.8); 1.16 (3H, t, ^3J = 6.8)	—	—	—

* Proton undergoing exchange.

*[2] 7.42 ppm (1H, d, ^3J = 8.0, H-8)

*[3] 7.39 ppm (1H, d, ^3J = 8.4, H-8).

*[4] 7.30 ppm (1H, d, J = 6.8, H-8).

Table 3. IR and Mass Spectra

Compound	IR spectrum, ν, cm^{-1}						Mass-spectrum, *m/z* [M$^+$ + 1] (*I* 100%)*
	C–O	C–C–O	C=N	C=O (pyrim)	C=O (carb)	C=O (phenacyl)	
4a	1274	—	1594	1665		—	315/314
4b	1274	—	1594	1664		—	327/326
4c	1271	—	1592	1671		—	
4d		—	1592	1664	1712	—	
5a	1268	—	1599	1664	1705	—	287/286
5b		—	1598	1687		—	
5c	1270	—	1597	1683		—	
10a	1250	—	1600	1670	1710		
6a	1263	1081	1595	1653	1711	—	343/342
6b		1090	1593	1667	1692*[2]	1723	
6c	1260	1081	1592	1664	1710	—	355/354
6d	1267	1092	1595	1671	1699	1723	
6e	1257	1070	1590	1671	1721	—	369/368
6f	1269	1093	1594	1670	1695	1712	
7a	1268	1079	1600	1666	1708	–	315/314
7b	1278	1095	1595	1654	1692	1724	
10b	1260	1078	1608	1670	1710	—	
8	1258	1080	1551	1676	1704	—	
9	1270	1081	1564	1668	1705	—	
10c	1265	1085	1595	1670	1700	—	

* Most intense peak in the spectrum.
*[2] Inflection

The carbonyl absorption of the pyrimidin-4-one unit as a participant in more complex structural combination was investigated in the classical work of Brown [7] and is well tabulated [8]. In the same way in the carboxylic acids **4d** and **5a** the shift of the $\nu_{C=O}$ band (from COOH, close to 1710 cm^{-1}) is not large. The carbonyl absorption of the pyrimidin-4-one unit remains close to 1670 cm^{-1}. We attribute the former difference to the influence of the phenacyl group at C-5 on the effectiveness of the hydrogen bond with participation of the carbonyl at C-4, and the latter to the electronic effect on this centre of the heteroatom in the neighboring position. On moving to the esters with structures **6-9** the picture in the carbonyl absorption region is simplified. In all cases individual bands for $\nu_{C=O}$ (from COOR) and $\nu_{C=O}$ (from the pyrimidin-4-one unit) are observed, and in the case of a phenacyl substituent, $\nu_{C=O}$ from the ketone carbonyl. Despite the remarkably low value of $\nu_{C=O}$, especially for the esters with structures **6-9**, these data agree with the recorded $\nu_{C=O}$ frequencies for quinazolone-4s [9]. Intense "alcoholic" bands of the O-C-C unit close to 1070-1080 cm^{-1} and "acidic" (the unit C-(C=O)-O close to 1260 cm^{-1} were observed for all the ethyl esters.

The electronic spectra of the carboxylic acids of the series **4** and **5** differ from that of acid **10a** not only in the number of absorption bands, but also in the distribution of their intensities in their limits (Fig. 1),

While for acid **10a** three sharp bands of different intensity and two minima are observed in the region analyzed, the contours for acids **4** and **5** are more complex and the maxima and minima described cannot be contrasted. The principal reason from the point of view of analysis of the different positional isomers is the bathochromic shift of ≈20 nm of the absorptions in the derivatives of the [2,3-*d*] series in comparison with their positions for the [3,2-*d*] positional isomers, and ≈ 15 nm for the quinazolone-4 (**10a**). The spectra of the esters with structure **6** and **7** differ little from the corresponding acids **4** and **5** Alkylation at atom N-3 (esters **8** and **9**) also does not change the electron density distribution in the esters **6** and **7** which is reflected in the similarity of their UV spectra.

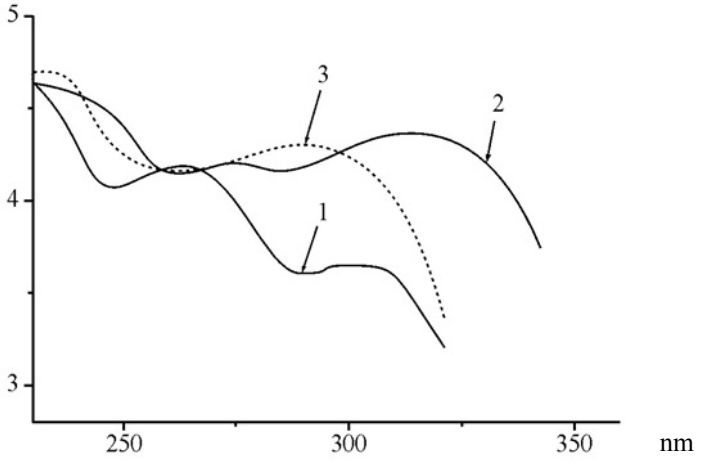

Fig. 1. UV spectra of carboxylic acids λ, nm (logε): 1 – acid **10a**: 261 (4.19), 268*, 300 (3.69), 308 (3.62). 2 – acid **4a**: 2.37 (4.59), 247*, 275 (4.20), 302 (4.31), 314 (4.36), 327*. 3 – acid **5a**: 234 (4.71), 285 (4.30), 296 (4.31), 305*.

* Inflexion.

EXPERIMENTAL

IR spectra of CsI and KBr tablets were recorded on Pye-Unicam SP3-300 and Nicolett Nexus spectrometers. ^{1}H NMR spectra of DMSO-d_6 solutions with TMS as internal standard were recorded on a Varian Mercury 400 (400 MHz). UV spectra of $5 \cdot 10^{-5}$ M methanol solutions were recorded on a Specord M-40 spectrophotometer. Mass spectra were recorded on an Agilent 1100 series machine with an Agilent LC/MSD SL selective detector, the samples were injected in a trifluoroacetic acid matrix, EI ionization, [M^+ +1] molecular ion peak. Melting points of the compounds synthesized were measured in a Pyrex capillary in a Tyl apparatus and were corrected.

2-Cyanomethylbenzoic acid (1) was prepared by a known method [10].

Compounds 3b and **3c** were obtained from Aldrich, and compounds **2a-d** were obtained by a known method [12].

Methyl 3-amino-2-thiophenecarboxylate (3a), mp 65°C, was obtained by method [11].

2-(4-Oxo-3,4-dihydrothieno[2,3-*d*]-pyrimidin-2-ylmethyl)benzoic Acids 4a-d and 2-(4-Oxo-3,4-dihydrothieno[3,2-*d*]-pyrimidin-2-ylmethyl)benzoic Acids 5a-c (General Method). A mixture of 2-cyanomethylbenzoic acid (**1**) (10 mmol) and the corresponding amino ester **2** or **3** (20 mmol) in chlorobenzene (5 ml) was boiled with stirring for 10 min, the solvent was removed in vacuum, 1,4-dioxane (40 ml) was added to the residue and the mixture was refluxed for 2h. The residue was filtered off and washed with diethyl ether.

Ethyl Esters 6a,c,e, and 7a (General Method). The corresponding thienopyrimidine **4a,c,e** or **5a** (1 mmol) was dissolved in boiling DMSO, the solution was cooled and added dropwise with stirring to triethylamine (0.1 ml, 1.5 mmol) and ethyl iodide (0.15 ml (1.5 mmol). The mixture was kept for 1 day, diluted with water, the precipitate which formed was filtered off and washed in turn with saturated sodium hydrogen carbonate solution, water, and diethyl ether.

Phenacyl Esters 6b,d,f, 7b (General Method). Triethylamine (0.2 ml, 3 mmol) was added to a suspension of the corresponding acid **4a-c** or **5a** (3 mmol) in acetonitrile (5 ml) and phenacyl bromide (3 mmol) in acetonitrile (2 ml) was added with stirring. After standing for a day, the solution was diluted with water, the precipitate which formed was filtered off and washed in turn with a saturated solution of sodium hydrogen carbonate, water, and diethyl ether.

3,5,6-Trimethyl-4-oxo-2-(2-ethoxycarbonylbenzyl)-3,4-dihydrothieno[2,3-*d*]pyrimidine (8) and 3-methyl-4-oxo-2-(2-ethoxycarbonylbenzyl)-3,4-dihydrothieno[3,2-*d*]pyrimidine (9) (General Method). The corresponding ethyl ester (1 mmol) was dissolved in boiling DMSO. To the cooled solution were added freshly prepared potash solution (0.14 g, 1 mmol) and methyl iodide (0.08 ml, 1 mmol). The mixture was left for one day, diluted with water, the precipitate was filtered off and washed with water.

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