

**HUGERSHOFF REACTION OF *N*-1- OR *N*-2-NAPHTHOYL-*N'*-MONO-SUBSTITUTED AND *N',N'*-DISUBSTITUTED THIOUREA DERIVATIVES**

Milan DZURILLA\*, Peter KUTSCHY, Jan IMRICH and Stanislav BRTOS

*Department of Organic Chemistry,**P. J. Safarik University, 041 67 Kosice, The Slovak Republic*

Received September 1, 1994

Accepted November 16, 1994

*Dedicated to Professor Milan Kratochvil on the occasion of his 70th birthday.*

Hugershoff reaction of *N*-1- or *N*-2-naphthoyl-*N'*-monosubstituted and *N',N'*-disubstituted thiourea derivatives, prepared by the reaction of 1-naphthoyl or 2-naphthoyl isothiocyanate with primary or secondary amines, was studied. Depending on substituents the corresponding urea or benzothiazole derivatives were obtained as reaction products. Their structure was determined on the basis of infrared, <sup>1</sup>H and <sup>13</sup>C NMR and mass spectra.

In our previous papers we have studied the Hugershoff reaction<sup>1</sup> i. e. oxidation of thiourea derivatives with bromine in chloroform. Thioureas prepared from  $\alpha,\beta$ -unsaturated acyl isothiocyanates afforded in the dependence on the substituents of thiourea grouping thiazolines or benzothiazoles<sup>2-4</sup>. In the case of *O*-alkyl-*N*-(3-phenylpropenyl)monothiocarbamates the sole reaction product was 5-benzylidene-1,3-thiazolidine-2,4-dione<sup>5</sup>.

We were interested in the reaction products of Hugershoff reaction of thioureas prepared from 1-naphthoyl or 2-naphthoyl isothiocyanate and primary and secondary amines (Table I). It was found that by the action of bromine as an oxidation agent in chloroform the different products are formed in the dependence on the nature of *N'*-substituents (Scheme 1).

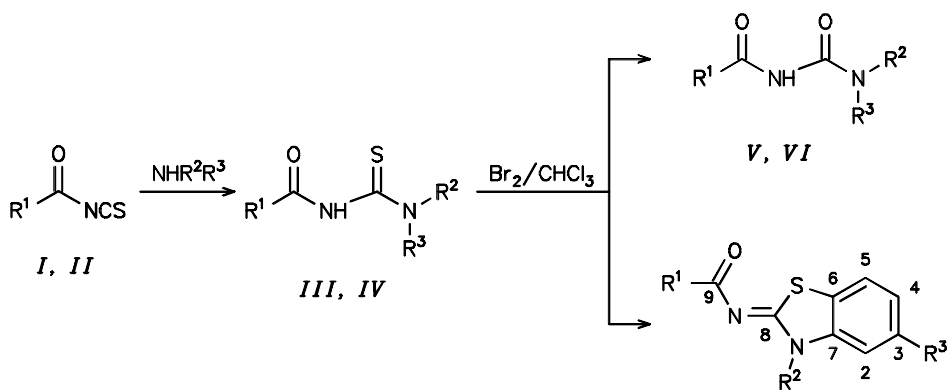
The Hugershoff reaction of thioureas with *N'*-alkyl substituents (*IIIa*, *IIIb*, *IVa* – *IVf*) and *N'*-aryl substituents which have the *ortho*-position not activated for an electrophilic substitution (*IIIc* – *IIIg*, *IIIi* – *IIIo*, *IVg* – *IVk*, and *IVm* – *IVs*) resulted in the oxidation to corresponding urea derivatives *Va* – *Vn* and *Vla* – *Vlr* (Table II). The exchange of sulfur to oxygen can be explained by hydrolysis of transiently formed sulfenyl bro-

\* The author to whom correspondence should be addressed.

amide<sup>2</sup> during the work-up of the reaction mixture because the same urea derivatives were obtained even when the reaction was carried out in anhydrous media. Similar oxidation of thiourea to urea derivatives was previously observed in the study of the Hegershoff reaction of *N*-phenyl-*N'*-(2-phenyl-4-thiazolylmethyl)thiourea<sup>6</sup> and *N*-phenyl-*N'*-(4-pentinoyl)thiourea<sup>7</sup>. In the case of *N*-(3-methoxyphenyl) derivatives *IIIh* and *IVl* as well as *N'*-methyl-*N'*-phenyl derivatives *IIIp* and *IVt* which have the *ortho*-position of the phenyl ring activated for an electrophilic reaction<sup>2,3</sup> the intermediate sulfenyl bromide undergoes intramolecular electrophilic substitution with the formation of benzothiazole derivatives *VIIa*, *VIIb*, *VIIIa*, and *VIIIb*.

All of investigated thiourea derivatives *III* and *IV* reacted with the formation of either urea or benzothiazole derivatives. Simultaneous formation of both types of products was not observed. The structure of prepared thioureas *III* and *IV* was confirmed by elemental analysis and spectral data (Table I).

The infrared spectra of obtained ureas exhibit two absorption bands of carbonyl vibrations corresponding to the transformation of thiocarbonyl to carbonyl group. In <sup>13</sup>C NMR spectra of ureas *Va*, *Vb*, *VIa*, and *VIb* there are present the signals of carbon



In formulae *I*, *III*, *V*, *VII* : R<sup>1</sup> = 1-Naphthyl

*II*, *IV*, *VI*, *VIII* : R<sup>1</sup> = 1-Naphthyl

*VIIa*, R<sup>2</sup> = H; R<sup>3</sup> = CH<sub>3</sub>O

*VIIb*, R<sup>2</sup> = CH<sub>3</sub>; R<sup>3</sup> = H

*VIIIa*, R<sup>2</sup> = H; R<sup>3</sup> = CH<sub>3</sub>O

*VIIIb*, R<sup>2</sup> = CH<sub>3</sub>; R<sup>3</sup> = H

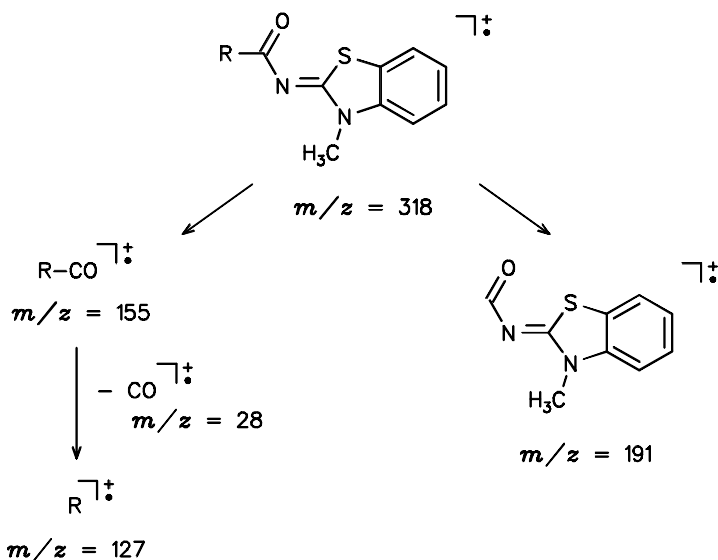
For R<sup>2</sup> and R<sup>3</sup> in compounds *III* – *VI* see Tables I and II

SCHEME 1

atoms of C=O groups instead of C=S carbon atoms present in starting compounds. The structure of urea derivatives was also confirmed by mass spectra of compounds *Via* and *Vib*.

Benzothiazole derivatives *VII* and *VIII* were identified on the basis of their IR,  $^1\text{H}$  and  $^{13}\text{C}$  NMR and mass spectroscopy. Compared to ureas products *VII* and *VIII* exhibit in the infrared spectra only one absorption band of carbonyl group. The mass spectra of compounds *VIIb* and *VIIIb* are in agreement with the structure of benzothiazolines (Scheme 2).

In the case of derivatives *VIIa* and *VIIIa* there are two possibilities of ring-closure with the formation of product having methoxy group in position 5 or 7. The structure of obtained product was determined by  $^1\text{H}$  NMR spectroscopy. In the spectra of compounds *VIIa* and *VIIIa* the signal of H-4 was split into doublet-doublet due to interaction with H-2 and H-5. H-5 and H-2 exhibited doublet signals owing to interaction with proton H-4. These data indicate the presence of two vicinal and one isolated proton, which corresponds to the structure with methoxy group in position 5 of benzothiazole ring.



SCHEME 2

TABLE I  
*N*-Naphthoyl-*N'*-monosubstituted and *N,N'*-disubstituted thiourea derivatives *IIIa* – *IIIp* and *IVa* – *IVt*

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup>	
				% C	% H	% N	v(C=O)	v(N-H)
<i>IIIa</i> <sup>d</sup>	H	C <sub>15</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (272.4)	159 – 161	66.14	5.92	10.28	1 675	3 403
	CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub>		95	66.31	5.98	10.40		3 262
<i>IIIb</i> <sup>d</sup>	H	C <sub>15</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (272.4)	165 – 167	66.14	5.92	10.28	1 670	3 408
	(CH <sub>3</sub> ) <sub>2</sub> CH		93	66.39	6.01	10.43		3 255
<i>IIIc</i>	H	C <sub>18</sub> H <sub>14</sub> N <sub>2</sub> O <sub>2</sub> S (306.4)	151 – 153	70.56	4.61	9.14	1 680	3 407
	C <sub>6</sub> H <sub>5</sub>		85	70.82	4.75	9.28		3 235
<i>IIIId</i>	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (320.4)	147	71.23	5.03	8.74	1 685	3 420
	2-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>		89	71.36	5.13	8.83		3 205
<i>IIIe</i>	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (320.4)	118 – 120	71.23	5.03	8.74	1 675	3 405
	3-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>		93	71.29	5.09	8.89		3 230
<i>IIIff</i>	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (320.4)	176 – 178	71.23	5.03	8.74	1 676	3 405
	4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>		80	71.39	5.17	8.92		3 235
<i>IIIg</i>	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (336.4)	148 – 150	67.84	4.79	8.33	1 680	3 400
	2-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>		76	67.96	4.87	8.52		3 195
<i>IIIh</i> <sup>d</sup>	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (336.4)	123 – 124	67.84	4.79	8.33	1 675	3 400
	3-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>		87	68.02	4.90	8.47		3 228

TABLE I  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup>	
				% C	% H	% N	v(C=O)	v(N-H)
<i>IIIi</i>	H 4-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> S (336.4)	172 – 174 84	67.84 67.91	4.79 4.93	8.33 8.39	1 680 3 240	3 405 3 240
<i>IIIj</i>	H 2-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> OS (340.8)	184 – 185 83	63.44 63.58	3.85 3.97	8.22 8.41	1 680 3 170	3 400 3 170
<i>IIIk</i>	H 3-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> OS (340.8)	147 – 149 88	63.44 63.51	3.85 4.01	8.22 8.30	1 678 3 210	3 400 3 210
<i>IIIl</i>	H 4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> OS (340.8)	207 – 209 94	63.44 63.60	3.85 4.03	8.22 8.33	1 680 3 228	3 410 3 228
<i>IIIm</i>	H 2-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> OS (385.3)	174 – 176 76	56.11 56.38	3.40 3.53	7.27 7.38	1 680 3 170	3 405 3 170
<i>IIIn</i>	H 3-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> OS (385.3)	154 – 156 91	56.11 56.42	3.40 3.47	7.27 7.35	1 675 3 210	3 480 3 210
<i>IIIo</i>	H 4-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> OS (385.3)	213 – 215 93	56.11 56.29	3.40 3.43	7.27 7.29	1 680 3 235	3 415 3 235
<i>IIIp</i>	CH <sub>3</sub> C <sub>6</sub> H <sub>5</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> OS (320.4)	134 – 136 74	71.23 71.41	5.03 5.09	8.74 8.87	1 700 3 395	3 395 3 395
<i>IV<sup>d</sup></i>	H CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub>	C <sub>15</sub> H <sub>16</sub> N <sub>2</sub> OS (272.4)	85 – 87 80	66.14 66.29	5.92 5.99	10.28 10.37	1 670 3 265	3 430 3 265

TABLE I  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup>	
				% C	% H	% N	v(C=O)	v(N-H)
<i>IV</i> <sup>b</sup>	H (CH <sub>3</sub> ) <sub>2</sub> CH	C <sub>15</sub> H <sub>16</sub> N <sub>2</sub> O (272.4)	117 – 118 79	66.14 66.34	5.92 6.01	10.28 10.41	1 675	3 430 3 255
<i>IV</i> <sup>c</sup>	H CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub>	C <sub>16</sub> H <sub>18</sub> N <sub>2</sub> O (286.4)	75 – 77 65	67.10 67.26	6.34 6.48	9.78 9.91	1 675	3 430 3 265
<i>IV</i> <sup>d</sup>	H (CH <sub>3</sub> ) <sub>2</sub> CHCH <sub>2</sub>	C <sub>16</sub> H <sub>18</sub> N <sub>2</sub> O (286.4)	68 – 70 58	67.10 67.32	6.34 6.39	9.78 9.86	1 675	3 425 3 260
<i>IV</i> <sup>e</sup>	H (CH <sub>3</sub> ) <sub>3</sub> C	C <sub>16</sub> H <sub>18</sub> N <sub>2</sub> O (286.4)	120 – 122 84	67.10 67.28	6.34 6.41	9.78 9.92	1 665	3 420 3 255
<i>IV</i> <sup>f</sup>	CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub>	C <sub>18</sub> H <sub>22</sub> N <sub>2</sub> O (314.5)	94 – 96 80	68.74 68.83	7.05 7.08	8.91 8.97	1 695	3 390
<i>IV</i> <sup>g</sup>	H C <sub>6</sub> H <sub>5</sub>	C <sub>18</sub> H <sub>14</sub> N <sub>2</sub> O (306.4)	147 – 149 91	70.56 70.77	4.61 4.78	9.14 9.31	1 667	3 415 3 260
<i>IV</i> <sup>h</sup>	H 2-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O (320.4)	136 – 138 86	71.23 71.37	5.03 5.16	8.74 8.92	1 670	3 425 3 240
<i>IV</i> <sup>i</sup>	H 3-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O (320.4)	153 – 155 88	71.23 71.41	5.03 5.20	8.74 8.84	1 673	3 420 3 230

TABLE I  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup>	
				% C	% H	% N	v(C=O)	v(N-H)
IVj	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>5</sub>	180 – 182	71.23	5.03	8.74	1 675	3 434
	4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	(320.4)	87	71.39	5.22	8.97		3 240
IVk	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>5</sub> S	153 – 155	67.84	4.79	8.33	1 675	3 425
	2-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	(336.4)	88	68.01	4.92	8.51		3 210
IVl <sup>r</sup>	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>5</sub> S	139 – 141	67.84	4.79	8.33	1 675	3 424
	3-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	(336.4)	87	67.93	4.90	8.47		3 240
IVm	H	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>5</sub> S	183 – 185	67.84	4.79	8.33	1 673	3 420
	4-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	(336.4)	95	67.97	4.95	8.48		3 230
IVn	H	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>5</sub>	184 – 186	63.44	3.85	8.22	1 675	3 420
	2-ClC <sub>6</sub> H <sub>4</sub>	(340.8)	90	63.61	3.94	8.37		3 210
IVo	H	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>5</sub>	157 – 159	63.44	3.85	8.22	1 675	3 420
	3-ClC <sub>6</sub> H <sub>4</sub>	(340.8)	89	63.62	4.01	8.43		3 223
IVp	H	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>5</sub>	194 – 196	63.44	3.85	8.22	1 673	3 430
	4-ClC <sub>6</sub> H <sub>4</sub>	(340.8)	91	63.58	3.99	8.29		3 235
IVq	H	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>5</sub>	186 – 188	56.11	3.40	7.27	1 670	3 425
	2-BrC <sub>6</sub> H <sub>4</sub>	(385.3)	86	56.38	3.51	7.41		3 200

TABLE I  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup>	
				% C	% H	% N	v(C=O)	v(N-H)
<i>IVr</i>	H 3-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O (385.3)	158 – 160 89	56.11 56.29	3.40 3.49	7.27 7.39	1 678	3 428 3 230
<i>IVs</i>	H 4-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O (385.3)	201 – 203 89	56.11 56.41	3.40 3.52	7.27 7.43	1 680	3 425 3 230
<i>IVt</i>	CH <sub>3</sub> C <sub>6</sub> H <sub>5</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O (320.4)	123 – 125 60	71.23 71.46	5.03 5.12	8.74 8.90	1 697	3 395

<sup>1</sup>H NMR spectra of compounds: *IIIa*: 1.11 t, 3 H (CH<sub>3</sub>); 1.84 m, 2 H and 3.80 m, 2 H (2 × CH<sub>2</sub>); 7.98 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.2 t, 1 H and 10.85 s, 1 H (2 × NH). *IIIb*: 1.41 d, 6 H (2 × CH<sub>3</sub>); 3.30 m, 1 H (CH); 7.96 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.08 d, 1 H and 10.68 s, 1 H (2 × NH). *IIIc*: 3.85 s, 3 H (CH<sub>3</sub>O); 6.84 dt, 1 H (1 H of C<sub>6</sub>H<sub>4</sub>); 7.68 m, 9 H (6 H of C<sub>10</sub>H<sub>7</sub> and 3 H of C<sub>6</sub>H<sub>4</sub>); 8.40 s, 1 H (1 H of C<sub>10</sub>H<sub>7</sub>); 9.10 s, 1 H and 12.65 s, 1 H (2 × NH). *IVa*: 1.00 t, 3 H (CH<sub>3</sub>); 1.78 m, 2 H and 3.67 m, 2 H (2 × CH<sub>2</sub>); 7.86 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.10 t, 1 H and 10.73 s, 1 H (2 × NH). *IVb*: 1.31 d, 6 H (2 × CH<sub>3</sub>); 4.56 m, 1 H (CH); 7.90 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.09 d, 1 H and 10.59 s, 1 H (2 × NH). *IVc*: 0.95 t, 3 H (CH<sub>3</sub>); 1.55 m and 3.70 m, 6 H (3 × CH<sub>2</sub>); 7.94 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.19 t, 1 H and 10.73 s, 1 H (2 × NH). *IVd*: 0.98 t, 6 H (2 × CH<sub>3</sub>); 2.04 m, 2 H (CH<sub>2</sub>); 3.55 m, 1 H (CH); 7.90 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.23 t, 1 H and 10.80 s, 1 H (2 × NH). *IVe*: 1.65 s, 9 H (3 × CH<sub>3</sub>); 7.79 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.05 s, 1 H and 10.98 s, 1 H (2 × NH). *IVf*: 0.99 t, 6 H (2 × CH<sub>3</sub>); 1.83 m, 4 H and 3.82 m, 4 H (2 × CH<sub>2</sub>); 8.05 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.76 s, 1 H (NH). *IVi*: 3.85 s, 3 H (CH<sub>3</sub>O); 6.84 dt, 1 H (1 H of C<sub>6</sub>H<sub>4</sub>); 7.63 m, 9 H (6 H of C<sub>10</sub>H<sub>7</sub> and 3 H of C<sub>6</sub>H<sub>4</sub>); 8.43 s, 1 H (1 H of C<sub>7</sub>H<sub>10</sub>); 9.26 s, 1 H and 12.69 s, 1 H (2 × NH). <sup>13</sup>C NMR spectra of compounds: *IIIa*: 11.50 q (CH<sub>3</sub>); 21.61 t (CH<sub>2</sub>); 47.59 t (CH<sub>2</sub>); 124.49, 124.64, 126.20, 126.88, 127.96, 128.67, 129.82, 130.98, 132.74 and 133.78 (C<sub>10</sub>H<sub>7</sub>); 169.02 s (CO); 179.54 s (CS). *IIIb*: 21.77 q (2 × CH<sub>3</sub>); 47.85 d (CH); 124.49, 124.67, 126.20, 126.88, 127.96, 128.67, 129.86, 131.02, 132.77 and 133.82 (C<sub>10</sub>H<sub>7</sub>); 169.02 s (CO); 178.68 s (CS). *IVa*: 11.46 q (CH<sub>3</sub>); 21.65 t (CH<sub>2</sub>); 47.63 t (CH<sub>2</sub>); 123.07, 127.32, 127.85, 128.82, 128.98, 129.00, 129.12, 129.30, 132.44 and 135.54 (C<sub>10</sub>H<sub>7</sub>); 167.00 s (CO); 179.95 s (CS). *IVb*: 21.76 q (2 × CH<sub>3</sub>); 47.97 d (CH); 122.99, 127.32, 127.88, 128.67, 128.89, 129.08, 129.23, 129.30, 132.44 and 135.57 (C<sub>10</sub>H<sub>7</sub>); 166.89 s (CO); 178.46 s (CS). Mass spectra, *m/z* (%), of compounds: *IVa*: 272 (M<sup>+</sup>, 27), 155 (91), 127 (10), 58 (45). *IVb*: 272 (M<sup>+</sup>, 39), 155 (65), 127 (79), 58 (100).



TABLE II  
*N*-Naphthoyl-*N'*-monosubstituted and *N,N'*-disubstituted urea derivatives *Va* – *Vn* and *VIa* – *VIr*

Compound	$R^2/R^3$	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, $\text{cm}^{-1}$ $\nu(\text{CO-NH-CO})$
				% C	% H	% N	
<i>Va</i> <sup>d</sup>	H $\text{CH}_3\text{CH}_2\text{CH}_2$	$\text{C}_{15}\text{H}_{16}\text{N}_2\text{O}_2$ (256.3)	144 – 146 56	70.29 70.47	6.29 6.24	10.93 11.08	1 690 1 660
<i>Vb</i> <sup>d</sup>	H $(\text{CH}_3)_2\text{CH}$	$\text{C}_{15}\text{H}_{16}\text{N}_2\text{O}_2$ (256.3)	147 – 149 43	70.29 70.38	6.29 6.38	10.93 11.04	1 700 1 660
<i>Vc</i>	H $\text{C}_6\text{H}_5$	$\text{C}_{18}\text{H}_{14}\text{N}_2\text{O}_2$ (256.3)	212 – 214 85	74.47 74.71	4.86 4.98	9.65 9.83	1 705 1 680
<i>Vd</i>	H $2\text{-CH}_3\text{C}_6\text{H}_4$	$\text{C}_{19}\text{H}_{16}\text{N}_2\text{O}_2$ (304.4)	214 – 216 45	74.97 75.10	5.30 5.42	9.20 9.38	1 695 1 665
<i>Ve</i>	H $3\text{-CH}_3\text{C}_6\text{H}_4$	$\text{C}_{19}\text{H}_{16}\text{N}_2\text{O}_2$ (304.4)	176 – 178 47	74.97 75.21	5.30 5.48	9.20 9.41	1 697 1 672
<i>Vf</i>	H $4\text{-CH}_3\text{C}_6\text{H}_4$	$\text{C}_{19}\text{H}_{16}\text{N}_2\text{O}_2$ (304.4)	224 – 226 61	74.97 75.15	5.30 5.43	9.20 9.40	1 700 1 673
<i>Vg</i>	H $2\text{-CH}_3\text{OC}_6\text{H}_4$	$\text{C}_{19}\text{H}_{16}\text{N}_2\text{O}_3$ (320.4)	228 – 230 60	71.23 71.47	5.03 5.14	8.74 8.89	1 695 1 680
<i>VI</i> <sup>d</sup>	H $4\text{-CH}_3\text{OC}_6\text{H}_4$	$\text{C}_{19}\text{H}_{16}\text{N}_2\text{O}_3$ (320.4)	220 – 222 41	71.23 71.43	5.03 5.12	8.74 8.91	1 705 1 673
<i>Vi</i>	H $2\text{-ClC}_6\text{H}_4$	$\text{C}_{18}\text{H}_{13}\text{ClN}_2\text{O}_2$ (324.8)	210 – 212 66	66.56 66.74	4.03 4.17	8.63 8.79	1 700 1 680
<i>Vj</i>	H $3\text{-ClC}_6\text{H}_4$	$\text{C}_{18}\text{H}_{13}\text{ClN}_2\text{O}_2$ (324.8)	217 – 219 82	66.56 66.68	4.03 4.14	8.63 8.74	1 703 1 673

TABLE II  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup> ν(CO-NH-CO)
				% C	% H	% N	
Vk	H 4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>2</sub> (324.8)	226 – 228 74	66.56 66.71	4.03 4.16	8.63 8.73	1 708 1 675
VI	H 2-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>2</sub> (369.2)	251 – 253 69	58.56 58.72	3.55 3.71	7.59 7.74	1 705 1 680
Vm	H 3-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>2</sub> (369.2)	223 – 225 71	58.56 58.79	3.55 3.76	7.59 7.79	1 700 1 690
Vn	H 4-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>2</sub> (369.2)	228 – 230 84	58.56 58.67	3.55 3.69	7.59 7.68	1 705 1 670
VId <sup>a</sup>	H CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub>	C <sub>15</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (256.3)	142 – 144 36	70.29 71.43	6.29 6.39	10.93 11.07	1 705 1 685
VIf <sup>a</sup>	H (CH <sub>3</sub> ) <sub>2</sub> CH	C <sub>15</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (256.3)	139 – 141 46	70.29 71.41	6.29 6.35	10.93 10.99	1 700 1 680
VIl <sup>a</sup>	H CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub>	C <sub>16</sub> H <sub>18</sub> N <sub>2</sub> O <sub>2</sub> (270.3)	146 – 148 58	71.10 71.27	6.71 6.88	10.36 10.46	1 710 1 690
VId <sup>f</sup>	H (CH <sub>3</sub> ) <sub>2</sub> CHCH <sub>2</sub>	C <sub>16</sub> H <sub>18</sub> N <sub>2</sub> O <sub>2</sub> (270.3)	141 – 143 37	71.10 71.31	6.71 6.90	10.36 10.52	1 710 1 690
VIl <sup>e</sup>	H (CH <sub>3</sub> ) <sub>3</sub> C	C <sub>16</sub> H <sub>18</sub> N <sub>2</sub> O <sub>2</sub> (270.3)	149 – 151 36	71.10 71.34	6.71 6.91	10.36 10.49	1 725 1 680
VIf <sup>e</sup>	CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub>	C <sub>18</sub> H <sub>22</sub> N <sub>2</sub> O <sub>2</sub> (298.4)	101 – 103 14	72.45 72.61	7.43 7.57	9.39 9.53	1 710 1 670

TABLE II  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup> ν(CO-NH-CO)
				% C	% H	% N	
<i>Vlg</i>	H C <sub>6</sub> H <sub>5</sub>	C <sub>18</sub> H <sub>14</sub> N <sub>2</sub> O <sub>2</sub> (290.3)	204 – 206 46	74.47 74.62	4.86 4.93	9.65 9.79	1 693 1 665
<i>Vlh</i>	H 2-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (304.4)	206 – 208 48	74.97 75.08	5.30 5.42	9.20 9.31	1 665 1 660
<i>Vli</i>	H 3-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (304.4)	175 – 177 66	74.97 75.11	5.30 5.45	9.20 9.38	1 697 1 668
<i>Vlj</i>	H 4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (304.4)	210 – 212 22	74.97 75.09	5.30 5.38	9.20 9.29	1 682 1 670
<i>Vlk</i>	H 2-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>3</sub> (320.4)	207 – 210 61	71.23 71.43	5.03 5.12	8.74 8.91	1 695 1 670
<i>Vlf</i>	H 4-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>3</sub> (320.4)	152 – 154 67	71.23 71.46	5.03 5.16	8.74 8.95	1 690 1 663
<i>Vlm</i>	H 2-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>2</sub> (324.8)	216 – 218 85	66.56 66.72	4.03 4.21	8.63 8.81	1 705 1 675
<i>Vln</i>	H 3-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>2</sub> (324.8)	217 – 219 82	66.56 66.71	4.03 4.09	8.63 8.82	1 694 1 666
<i>Vlo</i>	H 4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>2</sub> (324.8)	232 – 234 78	66.56 66.64	4.03 4.10	8.63 8.78	1 690 1 665
<i>Vlp</i>	H 2-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>2</sub> (369.2)	219 – 221 85	58.56 58.81	3.55 3.71	7.59 7.74	1 700 1 670

TABLE II  
(Continued)

Compound	R <sup>2</sup> /R <sup>3</sup>	Formula (M.w.)	M.p., °C Yield, %	Calculated/Found			IR, cm <sup>-1</sup> ν(CO-NH-CO)
				% C	% H	% N	
<i>Vlq</i>	H 3-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>2</sub> (369.2)	204 – 206 82	58.56 58.72	3.55 3.69	7.59 7.68	1 694 1 665
<i>Vlr</i>	H 4-BrC <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>13</sub> BrN <sub>2</sub> O <sub>2</sub> (369.2)	242 – 244 89	58.56 58.68	3.55 3.62	7.59 7.70	1 695 1 670

<sup>a</sup> <sup>1</sup>H NMR spectra of compounds: *Va*: 1.01 t, 3 H (CH<sub>3</sub>); 1.66 m, 2 H and 3.25 m, 2 H (2 × CH<sub>2</sub>); 7.98 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.85 t, 1 H and 9.45 s, 1 H (2 × NH). *Vb*: 1.25 d, 6 H (2 × CH<sub>3</sub>); 4.06 m, 1 H (CH); 7.80 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.73 d, 1 H and 9.55 s, 1 H (2 × NH). *Vla*: 0.99 t, 3 H (CH<sub>3</sub>); 1.69 m, 2 H and 3.43 m, 2 H (2 × CH<sub>2</sub>); 7.98 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.96 t, 1 H and 10.26 s, 1 H (2 × NH). *Vlb*: 1.31 d, 6 H (2 × CH<sub>3</sub>); 4.19 m, 1 H (CH); 8.13 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.85 d, 1 H and 10.39 s, 1 H (2 × NH). *Vlc*: 1.14 t, 3 H (CH<sub>3</sub>); 1.68 m and 3.50 m, 6 H (3 × CH<sub>2</sub>); 8.88 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.93 t, 1 H and 11.00 s, 1 H (2 × NH). *Vld*: 1.00 t, 6 H (2 × CH<sub>3</sub>); 1.91 m, 2 H (CH<sub>2</sub>); 3.30 m, 1 H (CH); 8.15 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.03 d, 1 H and 10.29 s, 1 H (2 × NH). *Vle*: 1.68 s, 9 H (3 × CH<sub>3</sub>); 8.26 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.35 s, 1 H and 9.06 s, 1 H (2 × NH). *Vlf*: 1.35 t, 6 H (2 × CH<sub>3</sub>); 2.10 m, 4 H and 2.89 m, 4 H (2 × CH<sub>2</sub>CH<sub>2</sub>); 8.28 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.79 s, 1 H (NH). <sup>13</sup>C NMR spectra of compounds: *Va*: 11.42 q (CH<sub>3</sub>); 21.81 t (CH<sub>2</sub>); 41.68 t (CH<sub>2</sub>); 124.45, 125.08, 126.28, 126.58, 127.58, 128.52, 130.12, 131.50, 132.29 and 133.78 (C<sub>10</sub>H<sub>7</sub>); 154.31 s and 170.32 s (2 × CO). *Vb*: 22.70 q (2 × CH<sub>3</sub>); 42.25 d (CH); 124.44, 125.08, 126.34, 126.43, 127.58, 128.52, 130.09, 131.62, 132.25 and 132.82 (C<sub>10</sub>H<sub>7</sub>); 153.27 s and 170.21 s (2 × CO). *Vla*: 11.46 q (CH<sub>3</sub>); 22.92 t (CH<sub>2</sub>); 41.73 t (CH<sub>2</sub>); 124.11, 126.42, 126.76, 127.73, 128.41, 129.38, 129.38, 129.75, 132.55 and 135.42 (C<sub>10</sub>H<sub>7</sub>); 155.13 s and 168.66 s (2 × CO). *Vlb*: 22.84 q (2 × CH<sub>3</sub>); 42.29 d (CH); 124.08, 126.58, 126.76, 127.77, 128.48, 129.30, 129.38, 129.86, 132.62 and 135.46 (C<sub>10</sub>H<sub>7</sub>); 154.20 s and 168.61 s (2 × CO). Mass spectra, *m/z* (%), of compounds: *Vla*: 256 (M<sup>+</sup>, 22), 155 (100), 127 (91), 58 (46). *Vlb*: 256 (M<sup>+</sup>, 54), 155 (100), 127 (91), 58 (46).

## EXPERIMENTAL

The infrared absorption spectra were recorded on an IR-75 spectrometer (Zeiss, Jena) in chloroform (compounds *II*, *IIIa* – *IIIp*, *IVa* – *IVt*) or in KBr pellets (compounds *I*, *Va* – *Vn*, *VIa* – *VIr*, *VIIa*, *VIIb*, *VIIIa*, *VIIIb*); the wavenumbers are given in  $\text{cm}^{-1}$ .  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were measured on Tesla BS 487 (80 MHz for  $^1\text{H}$ ) and Tesla BS 567 (25.15 MHz for  $^{13}\text{C}$ ) spectrometers in deuteriochloroform (compounds *I*, *II*, *IIIa*, *IIIb*, *IIIh*, *IVa* – *IVf*, *IVh*, *Va*, *Vb*, *VIa*, *VIb*, *VIc*) or in hexadeuteriodimethyl sulfoxide (compounds *VIc*, *VIe*, *VIg*, *VIIa*, *VIIb*, *VIIIa*, *VIIIb*) with tetramethylsilane as internal standard. Chemical shifts are given in ppm ( $\delta$ -scale), coupling constants (*J*) in Hz. The mass spectra of compounds *IVa*, *IVb*, *VIa*, *VIb*, *VIIa*, and *VIIb* were recorded on a JMS 100D spectrometer (Jeol) at ionization energy 70 eV. The reaction course was monitored by thin-layer chromatography (TLC) on Silufol plates (Kavalier, The Czech Republic).

1-Naphthoyl Isothiocyanate (*I*)

A solution of potassium thiocyanate (2.43 g, 25 mmol) in acetone (40 ml) was added to a solution of 1-naphthoyl chloride (4.77 g, 25 mmol) in acetone (20 ml). After stirring for 10 min, benzene (80 ml) was added and the precipitate of potassium chloride was filtered off. The solvent was evaporated and the residue was crystallized from hexane. Yield 68%, m.p. 30 – 32 °C. For  $\text{C}_{12}\text{H}_7\text{NOS}$  (213.3) calculated: 67.57% C, 3.31% H, 6.57% N; found: 67.74% C, 3.42% H, 6.71% N. IR spectrum: 1 697 (C=O); 1 969 (N=C=S).  $^{13}\text{C}$  NMR spectrum: 124.37, 125.57, 126.13, 126.91, 128.82, 129.19, 131.54, 133.71, 132.89, and 136.28 ( $\text{C}_{10}\text{H}_7$ ); 147.89 s (NCS); 161.52 s (CO).

2-Naphthoyl Isothiocyanate (*II*)

Title compound was prepared from potassium thiocyanate (2.43 g, 25 mmol) and 2-naphthoyl chloride (4.77 g, 25 mmol) according to the above-described procedure. Yield 85%, m.p. 68 – 69 °C. For  $\text{C}_{12}\text{H}_7\text{NOS}$  (213.3) calculated: 67.57% C, 3.31% H, 6.57% N; found: 67.68% C, 3.48% H, 6.69% N. IR spectrum: 1 680 (C=O); 1 950 (N=C=S).  $^{13}\text{C}$  NMR spectrum: 124.75, 127.21, 127.85, 127.99, 128.89, 129.49, 129.71, 132.25, 133.03, and 136.39 ( $\text{C}_{10}\text{H}_7$ ); 147.97 s (NCS); 161.63 s (CO).

General Procedure for Preparation of *N*-Naphthoyl-*N'*-monosubstituted *IIIa* – *IIIp* and *N,N'*-Disubstituted Thioureas *IVa* – *IVt*

The corresponding amine (5 mmol) was added dropwise with stirring to a solution of 1- or 2-naphthoyl isothiocyanate (1.07 g, 5 mmol) in cyclohexane (20 ml; preparation of compounds *IIIa*, *IIIb*, *IVa* – *IVf*) or benzene (15 ml; preparation of compounds *IIIc* – *IIIp*, *IVg* – *IVt*). The precipitate separated within 1 h was filtered with suction, dried and crystallized from ethanol. Yields, melting points, analytical data and IR spectra are given in Table I.

General Procedure for Preparation of *N*-Naphthoyl-*N'*-monosubstituted *Va* – *Vn* and *N,N'*-Disubstituted Ureas *VIa* – *VIr*

To a stirred solution of substituted thiourea (3 mmol) in chloroform (15 ml) bromine (155  $\mu\text{l}$ , 3 mmol) was added dropwise. The stirring was continued 1 h until starting compound completely reacted (monitored by TLC). The solvent was evaporated and the residue crystallized from ethanol. Yields, melting points, analytical data and IR spectra are given in Table II.

General Procedure for Preparation of Naphthalene-1- or 2-Carboxylic Acid  
(Substituted-3*H*-benzothiazol-2-ylidene)amides VIIa, VIIb, VIIIa, and VIIIb

To a stirred solution of thiourea IIIh, IIIp, IVl, or IVt (2.5 mmol) in chloroform (12.5 ml) bromine (130  $\mu$ l, 2.5 mmol) was added dropwise and stirring was continued for 1 h. The precipitate was filtered with suction, dried and crystallized from an appropriate solvent.

*Naphthalene-1-carboxylic acid (5-methoxy-3H-benzothiazol-2-ylidene)amide* (VIIa); yield 92%, m.p. 228 – 230 °C (acetone). For C<sub>19</sub>H<sub>14</sub>N<sub>2</sub>O<sub>2</sub>S (334.4) calculated: 68.24% C, 4.22% H, 8.37% N; found: 68.32% C, 4.38% H, 8.56% N. IR spectrum: 3 490 (N–H); 1 672 (C=O); 1 535 (C=N). <sup>1</sup>H NMR spectrum: 3.87 s, 3 H (CH<sub>3</sub>O); 7.06 dd, 1 H, *J*(2,4) = 2.4, *J*(4,5) = 8.8 (H-4); 7.42 d, 1 H, *J*(2,4) = 2.4 (H-2); 7.82 d, 1 H, *J*(4,5) = 8.8 (H-5); 8.07 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 9.29 s, 1 H (NH). <sup>13</sup>C NMR spectrum: 55.41 (C-1); 103.97 (C-2); 112.89 (C-4); 122.08 (C-5); 123.12 (C-6); 149.47 (C-7); 158.66 (C-3); 159.48 (C-8); 167.58 (C-9); 124.80, 126.44, 127.11, 127.37, 127.91, 128.42, 129.69, 131.03, 131.52 and 133.09 (C<sub>10</sub>H<sub>7</sub>).

*Naphthalene-1-carboxylic acid (3-methyl-3H-benzothiazol-2-ylidene)amide* (VIIb); yield 90%, m.p. 199 – 201 °C (acetone). For C<sub>19</sub>H<sub>14</sub>N<sub>2</sub>OS (318.4) calculated: 71.67% C, 4.43% H, 8.80% N; found: 71.79% C, 4.58% H, 8.96% N. IR spectrum: 1 670 (C=O); 1 525 (C=N). <sup>1</sup>H NMR spectrum: 4.05 s, 3 H (CH<sub>3</sub>); 7.68 m, 9 H (3 H of C<sub>6</sub>H<sub>4</sub> and 6 H of C<sub>10</sub>H<sub>7</sub>); 8.51 dd, 1 H (1 H of C<sub>6</sub>H<sub>4</sub>); 9.13 t, 1 H (1 H of C<sub>10</sub>H<sub>7</sub>). <sup>13</sup>C NMR spectrum: 32.03 q (CH<sub>3</sub>); 111.70, 122.26, 123.46, 124.28, 125.21, 125.73, 126.63, 127.79, 128.95, 130.62, 131.26, 133.20, 133.31, 136.71 (C<sub>10</sub>H<sub>7</sub> and C<sub>6</sub>H<sub>4</sub>); 165.97 s (CO); 175.27 s (NCS). Mass spectrum, *m/z* (%): 318 (M<sup>+</sup>, 88), 197 (67), 191 (37), 155 (100), 127 (96), 80 (59).

*Naphthalene-2-carboxylic acid (5-methoxy-3H-benzothiazol-2-ylidene)amide* (VIIIa); yield 85%, m.p. 226 – 227 °C (ethanol). For C<sub>19</sub>H<sub>14</sub>N<sub>2</sub>O<sub>2</sub>S (334.4) calculated: 68.24% C, 4.22% H, 8.37% N; found: 68.47% C, 4.39% H, 8.51% N. IR spectrum: 3 460 (N–H); 1 670 (C=O); 1 545 (C=N). <sup>1</sup>H NMR spectrum: 3.86 s, 3 H (CH<sub>3</sub>O); 7.00 dd, 1 H, *J*(2,4) = 2.4; *J*(4,5) = 8.7 (H-4); 7.35 d, 1 H, *J*(2,4) = 2.4 (H-2); 7.93 d, 1 H, *J*(4,5) = 8.7 (H-5); 7.68 and 8.08 m, 7 H (C<sub>10</sub>H<sub>7</sub>); 8.85 s, 1 H (NH). <sup>13</sup>C NMR spectrum: 55.45 (C-1); 103.71 (C-2); 112.71 (C-4); 122.07 (C-5); 124.24 (C-6); 149.06 (C-7); 158.66 (C-3); 160.26 (C-8); 165.97 (C-9); 123.04, 126.89, 127.59, 128.13, 128.35, 128.56, 129.20, 129.39, 131.89 and 134.69 (C<sub>10</sub>H<sub>7</sub>).

*Naphthalene-2-carboxylic acid (3-methyl-3H-benzothiazol-2-ylidene)amide* (VIIIb); yield 83%, m.p. 165 – 167 °C (acetone). For C<sub>19</sub>H<sub>14</sub>N<sub>2</sub>OS (318.4) calculated: 71.67% C, 4.43% H, 8.80% N; found: 71.82% C, 4.54% H, 8.89% N. IR spectrum: 1 680 (C=O); 1 545 (C=N). <sup>1</sup>H NMR spectrum: 4.05 s, 3 H (CH<sub>3</sub>); 7.65 m, 9 H (C<sub>6</sub>H<sub>4</sub> and 5 H of C<sub>10</sub>H<sub>7</sub>); 8.38 dd, 1 H and 8.88 s, 1 H (2 × 1 H of C<sub>10</sub>H<sub>7</sub>). Mass spectrum, *m/z* (%): 318 (M<sup>+</sup>, 26), 191 (26), 155 (16), 127 (96), 82 (100), 80 (96).

## REFERENCES

1. Hugershoff A.: Ber. Dtsch. Chem. Ges. 36, 3153 (1903).
2. Kutschy P., Dzurilla M., Kristian P., Kutschyova K.: Collect. Czech. Chem. Commun. 46, 436 (1981).
3. Dzurilla M., Kristian P., Imrich J., Stec J.: Collect. Czech. Chem. Commun. 48, 3134 (1983).
4. Dzurilla M., Kutschy P., Imrich J., Koscik D., Kraus R.: Collect. Czech. Chem. Commun. 56, 1287 (1991).
5. Dzurilla M., Imrich J., Kutschy P., Koscik D.: Chem. Papers 46, 102 (1992).
6. Silberg I. A., Silberg S.: Rev. Roum. Chim. 28, 139 (1983).
7. Kutschy P., Kristian P., Dzurilla M., Koscik D., Nadaskay R.: Chem. Papers 41, 519 (1987).