

SYNTHESIS OF 2-ARYL[1]BENZOFURO[2,3-*e*][1,2,4]TRIAZIN-3(2*H*)-ONES AND THEIR USE FOR PREPARATION OF 1-ARYL-5-(2-HYDROXYPHENYL)-6-AZAURACILS*

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Dedicated to Professor Miloslav Ferles on the occasion of his 75th birthday.

A series of arylhydrazones **1a–1i** was prepared by coupling of arene diazonium salts with ethyl *N*-(benzo[*b*]furan-2-yl)carbamate. The hydrazones **1** were thermally cyclized to the corresponding 2-aryl[1]benzofuro[2,3-*e*][1,2,4]triazin-3(2*H*)-ones **2a–2i**, derivatives of a new fused ring system. Compounds **2** were transformed by hydrolytic splitting to the corresponding 1-aryl-5-(2-hydroxyphenyl)-6-azauracils **3a–3i**.

Key words: Fused Heterocycles; Diazo compounds; Hydrazones; Benzofurotriazines; 6-Azauracils.

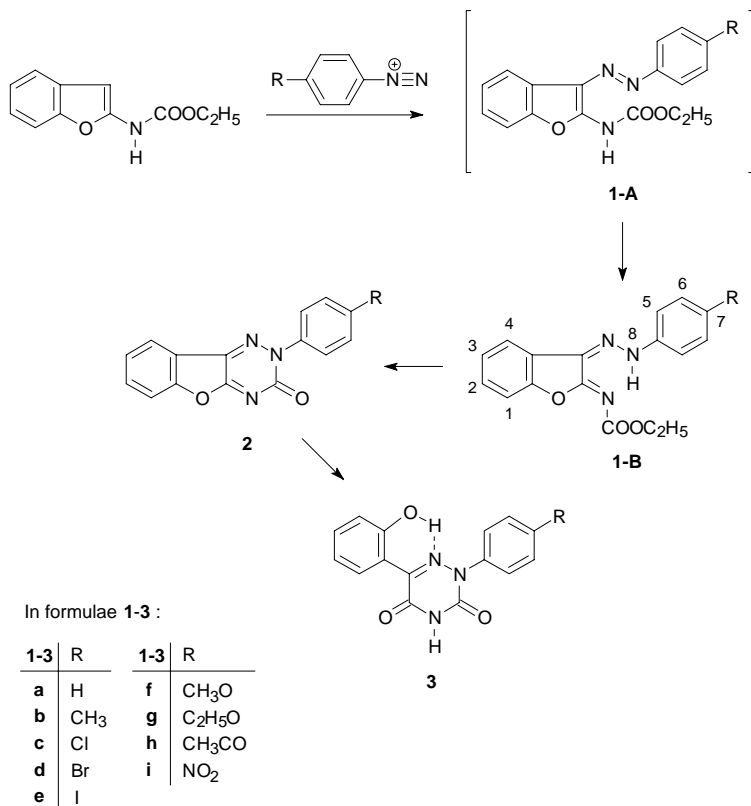
Cyclization of *N*-arylcaramates substituted in *ortho* position with azo or tautomeric hydrazono group is well-tried synthesis of condensed 1,2,4-triazines. In this manner, series of benzo[1,2,4]triazines^{2–7}, pyrazolo[3,4-*e*][1,2,4]triazine^{8,9}, [1,2,4]triazino[4,5-*a*]benzimidazole^{10–13} and [1,2,4]triazinoindeole^{14,15} derivatives, were advantageously prepared. In this paper we utilized this synthetic way for the preparation of [1]benzofuro[2,3-*e*][1,2,4]triazine derivatives which have not been described so far.

By coupling of diazonium salts with ethyl *N*-(2-benzo[*b*]furan-2-yl)carbamate in pyridine, corresponding ethyl *N*-(3-arylazobenzo[*b*]furan-2-yl)carbamates **1-A** or their hydrazono-tautomeric form **1-B** (Scheme 1) were prepared in good yields. In addition to the phenyl derivative **1a**, a series of *para* substituted derivatives with +*I* effect (**1b**), +*M* effect (**1c–1g**) and –*M* effect (**1h**, **1i**) were prepared in this way. As expected, compounds **1h** and **1i** were obtained in the highest yields.

The azo–hydrazone tautomerism with phenyl derivative **1a** was studied by IR spectroscopy in tetrachloromethane solution. For this purpose, compound **1a** containing la-

* Part XXIV in the series Cyclization Reactions of Hydrazones; Part XXIII: see ref.¹.

belled nitrogen in position 8 was prepared from aniline- ^{15}N . It was found that the unlabelled derivative shows in the region of N–H stretching vibrations an absorption maximum at $3\,206.4\text{ cm}^{-1}$ while the compound labelled with the isotope ^{15}N has the absorption maximum shifted to $3\,199.5\text{ cm}^{-1}$. This shift is in accordance with the formula $\tilde{\nu} = 1/2\pi(K/\mu)^{1/2}$. In view of these facts we conclude that the products of coupling exist in tetrachloromethane solution in the hydrazone form **1-B**. This hydrazone form of **1-B** was confirmed unambiguously by ^1H , ^{13}C and ^{15}N NMR spectroscopy. With this compound, the NMR signals were assigned using 2D NMR. The determined spin–spin coupling constants were as follows: 7.2 Hz for the interaction of hydrogen atoms in CH_3 and CH_2 groups and 7.9 or 1.08 Hz for interaction between aromatic hydrogen



SCHEME 1

atoms in *ortho* or *meta* position of the benzo[*b*]furan system. In the ^{15}N NMR spectrum of compound **1a** with the labelled atom ^{15}N at position 8, the chemical shift shows unambiguously that the compound exists in the hydrazone form **1-B**. This is in accordance with the above mentioned IR spectral results.

TABLE I
Characteristic data of compounds **1**

| Compound | M.p., °C Yield, % | Formula M.w. | Calculated/Found | | | $\nu(\text{C}=\text{O})$ cm^{-1} | λ_{max} nm | log ϵ |
|-----------|----------------------|---|------------------|------|-------|--|------------------------------|----------------|
| | | | % C | % H | % N | | | |
| 1a | 97–99 | C ₁₇ H ₁₅ N ₃ O ₃ | 66.01 | 4.89 | 13.59 | 1 737.2 | 201.1 | 4.31 |
| | 83 | 309.3 | 66.30 | 4.68 | 13.63 | 1 683.3 | 262.4 | 4.22 |
| | | | | | | | 420.8 | 4.22 |
| 1b | 124–126 | C ₁₈ H ₁₇ N ₃ O ₃ | 66.86 | 5.30 | 13.00 | 1 730.7 | 201.7 | 4.41 |
| | 79 | 323.3 | 66.65 | 5.34 | 13.13 | 1 683.8 | 266.2 | 4.23 |
| | | | | | | | 428.4 | 4.24 |
| 1c | 158–160 | C ₁₇ H ₁₄ ClN ₃ O ₃ | 59.37 | 4.10 | 12.22 | 1 732.5 | 201.7 | 4.37 |
| | 81 | 343.7 | 59.13 | 3.89 | 12.21 | 1 692.6 | 263.2 | 4.21 |
| | | | | | | | 419.2 | 4.26 |
| 1d | 138–140 | C ₁₇ H ₁₄ BrN ₃ O ₃ | 52.59 | 3.63 | 10.82 | 1 737.9 | 201.7 | 4.43 |
| | 84 | 388.3 | 52.53 | 3.67 | 10.84 | 1 692.1 | 263.2 | 4.25 |
| | | | | | | | 420.2 | 4.32 |
| 1e | 167–170 | C ₁₇ H ₁₄ IN ₃ O ₃ | 46.91 | 3.24 | 9.65 | 1 726.8 | 201.1 | 4.45 |
| | 78 | 435.3 | 46.93 | 3.13 | 9.71 | 1 686.0 | 261.6 | 4.24 |
| | | | | | | | 422.4 | 4.33 |
| 1f | 118–120 | C ₁₈ H ₁₇ N ₃ O ₄ | 63.71 | 5.05 | 12.38 | 1 738.5 | 210.9 | 4.77 |
| | 64 | 339.3 | 64.45 | 4.95 | 12.54 | 1 678.9 | 270.4 | 4.21 |
| | | | | | | | 448.8 | 4.15 |
| 1g | 87–90 | C ₁₉ H ₁₉ N ₃ O ₄ | 64.58 | 5.42 | 11.89 | 1 732.8 | 201.1 | 4.42 |
| | 66 | 353.4 | 64.86 | 5.28 | 12.07 | 1 672.0 | 268.1 | 4.14 |
| | | | | | | | 414.4 | 4.07 |
| 1h | 150–152 | C ₁₉ H ₁₄ N ₃ O ₄ | 64.95 | 4.88 | 11.96 | 1 736.9 | 201.9 | 4.51 |
| | 85 | 351.3 | 64.69 | 5.05 | 12.10 | 1 672.9 | 260.8 | 4.26 |
| | | | | | | | 287.2 | 3.97 |
| | | | | | | 424.1 | 4.51 | |
| 1i | 233–235 | C ₁₇ H ₁₄ N ₄ O ₅ | 57.62 | 3.98 | 15.81 | 1 736.6 | 201.5 | 4.47 |
| | 89 | 354.3 | 57.35 | 3.83 | 16.07 | 1 698.2 | 257.6 | 4.13 |
| | | | | | | | 434.4 | 4.53 |

Heating at a higher temperature, for example boiling in decalin, leads to cyclization of all hydrazono carbamates **1** and gives corresponding 2-aryl[1]benzofuro[2,3-*e*]-[1,2,4]triazin-3(2*H*)-ones (**2a–2i**). Cyclization of derivatives substituted by the groups with *-M* effect proceeds more slowly than in other cases; however, 60-min boiling in decaline is quite sufficient for nearly quantitative cyclization. Thermal cyclization of compound **1a** is well perceptible in its mass spectrum where both its molecular ion peak *m/z* 309.3 and the molecular peak of product **2a**, *m/z* 263.3, can be found. In the mass spectrum a distinct peak corresponding to fragment C₆H₅N (*m/z* 91.1) is present, which arose by splitting of the azo group formed by tautomeric transformation.

TABLE II
Characteristic data of compounds **2**

| Compound | M.p., °C Yield, % | Formula M.w. | Calculated/Found | | | $\nu(\text{C=O})$ cm ⁻¹ | λ_{max} nm | log ϵ |
|-----------|----------------------|--|------------------|------|-------|---------------------------------------|------------------------------|----------------|
| | | | % C | % H | % N | | | |
| 2a | 210–212 | C ₁₅ H ₉ N ₃ O ₂ | 68.43 | 3.45 | 15.96 | 1 724.7 | 375.2 | 4.01 |
| | 89 | 263.3 | 68.44 | 3.28 | 16.13 | 1 672.9 | | |
| 2b | 195–197 | C ₁₆ H ₁₁ N ₃ O ₂ | 69.30 | 4.00 | 15.16 | 1 698.9 | 379.7 | 4.01 |
| | 88 | 277.3 | 69.04 | 3.80 | 15.16 | 1 664.4 | | |
| 2c | 231–233 | C ₁₅ H ₈ ClN ₃ O ₂ | 60.59 | 2.71 | 14.11 | 1 692.5 | 376.3 | 4.02 |
| | 87 | 297.7 | 60.32 | 2.45 | 14.04 | 1 665.9 | | |
| 2d | 243–245 | C ₁₅ H ₈ BrN ₃ O ₂ | 52.65 | 2.35 | 12.28 | 1 690.2 | 379.2 | 4.02 |
| | 89 | 342.3 | 52.23 | 2.35 | 12.16 | 1 667.1 | | |
| 2e | 266–268 | C ₁₅ H ₈ IN ₃ O ₂ | 46.62 | 2.07 | 10.79 | 1 688.9 | 380.6 | 4.04 |
| | 90 | 389.3 | 46.29 | 2.02 | 10.70 | 1 666.2 | | |
| 2f | 251–253 | C ₁₆ H ₁₁ N ₃ O ₃ | 65.32 | 3.81 | 14.33 | 1 696.7 | 390.2 | 4.17 |
| | 90 | 293.3 | 65.50 | 3.77 | 14.26 | 1 667.1 | | |
| 2g | 229–231 | C ₁₇ H ₁₃ N ₃ O ₃ | 66.45 | 2.23 | 13.68 | 1 683.9 | 391.6 | 4.17 |
| | 89 | 307.3 | 66.11 | 2.25 | 13.53 | 1 666.4 | | |
| 2h | 275–277 | C ₁₇ H ₁₁ N ₃ O ₃ | 66.88 | 3.63 | 13.77 | 1 684.6 | 379.8 | 4.04 |
| | 78 | 305.3 | 66.51 | 3.53 | 13.69 | 1 665.5 | | |
| 2i | 328–330 | C ₁₅ H ₈ N ₄ O ₄ | 58.44 | 2.62 | 18.18 | 1 696.4 | 381.6 | 4.07 |
| | 79 | 308.3 | 58.01 | 2.47 | 18.03 | 1 667.6 | | |

TABLE III
 Characteristic data of compounds **3**

| Compound | M.p., °C Yield, % | Formula M.w. | Calculated/Found | | | $\nu(\text{C}=\text{O})$ cm^{-1} | λ_{max} nm | log ϵ |
|-----------|----------------------|--|------------------|------|-------|--|------------------------------|----------------|
| | | | % C | % H | % N | | | |
| 3a | 261–263 | $\text{C}_{15}\text{H}_{11}\text{N}_3\text{O}_3$ | 64.06 | 3.91 | 14.95 | 1 724.4 | 202.7 | 4.53 |
| | 90 | 281.3 | 64.01 | 4.01 | 14.78 | 1 687.9 | 242.5 | 4.26 |
| | | | | | | | 324.5 | 4.05 |
| 3b | 252–254 | $\text{C}_{16}\text{H}_{13}\text{N}_3\text{O}_3$ | 65.08 | 4.44 | 14.23 | 1 731.1 | 205.5 | 4.46 |
| | 93 | 295.3 | 65.26 | 4.71 | 14.01 | 1 692.6 | 239.6 | 4.01 |
| | | | | | | | 316.9 | 3.82 |
| 3c | 249–251 | $\text{C}_{15}\text{H}_{10}\text{ClN}_3\text{O}_3$ | 57.14 | 3.19 | 13.33 | 1 733.3 | 203.6 | 4.43 |
| | 92 | 315.7 | 56.86 | 2.95 | 12.96 | 1 696.1 | 241.6 | 3.96 |
| | | | | | | | 325.1 | 3.77 |
| 3d | 251–253 | $\text{C}_{15}\text{H}_{10}\text{BrN}_3\text{O}_3$ | 50.00 | 2.78 | 11.67 | 1 733.3 | 203.2 | 4.7 |
| | 89 | 360.3 | 50.11 | 2.63 | 11.65 | 1 692.2 | 245.1 | 4.26 |
| | | | | | | | 313.6 | 4.05 |
| 3e | 273–275 | $\text{C}_{15}\text{H}_{10}\text{IN}_3\text{O}_3$ | 44.25 | 2.48 | 10.32 | 1 710.3 | 204.2 | 4.67 |
| | 91 | 407.7 | 44.18 | 2.28 | 10.31 | 1 693.2 | 249.2 | 4.28 |
| | | | | | | | 316.1 | 4.04 |
| 3f | 247–249 | $\text{C}_{16}\text{H}_{13}\text{N}_3\text{O}_4$ | 61.73 | 4.21 | 13.51 | 1 721.4 | 204.1 | 4.38 |
| | 90 | 311.3 | 61.83 | 4.16 | 13.26 | 1 693.9 | 271.8 | 4.03 |
| | | | | | | | 297.6 | 3.86 |
| 3g | 249–251 | $\text{C}_{17}\text{H}_{15}\text{N}_3\text{O}_4$ | 62.76 | 4.65 | 12.92 | 1 722.7 | 202.5 | 4.4 |
| | 92 | 325.3 | 62.56 | 4.98 | 13.05 | 1 698.4 | 233.1 | 4.01 |
| | | | | | | | 327.2 | 3.73 |
| 3h | 265–267 | $\text{C}_{17}\text{H}_{13}\text{N}_3\text{O}_4$ | 63.16 | 4.05 | 13.01 | 1 732.9 | 203.1 | 4.61 |
| | 90 | 323.1 | 63.23 | 4.21 | 12.96 | 1 707.1 | 270.6 | 4.23 |
| | | | | | | 1 681.6 | 297.6 | 4.15 |
| 3i | 258–260 | $\text{C}_{15}\text{H}_{10}\text{N}_4\text{O}_5$ | 55.21 | 3.07 | 17.18 | 1 735.5 | 203.6 | 4.56 |
| | 88 | 326.3 | 55.45 | 3.19 | 17.11 | 1 697.6 | 241.6 | 4.25 |
| | | | | | | | 343.2 | 4.01 |

TABLE IV
NMR spectral data of compounds 1–3

| Compound | Nucleus | Chemical shifts |
|-----------------------|-----------------|---|
| 1a^a | ¹ H | 1.41 t, 3 H (CH ₃); 4.38 q, 2 H (CH ₂); 7.09 m, 1 H (H-7); 7.15 d, 1 H ((H-1); 7.22 t, 1 H (H-3); 7.30 t, 1 H (H-2); 7.34–7.42 m, 4 H (H-5,6); 7.71 d, 1 H (H-4); 12.25 bs, 1 H (NH) |
| | ¹³ C | 14.26 (CH ₃); 63.14 (CH ₂); 111.05 (C-1); 114.97 (C-5); 119.19 (C-4); 123.02; 124.07 (C-7); 124.29; 124.57 (C-3); 128.54 (C-2); 129.41 (C-6); 142.34; 153.26; 154.33; 158.40 |
| | ¹⁵ N | –205.8 brs (CH ₃ NO ₂ int) |
| 1b | ¹ H | 1.34 t, 3 H (CH ₃); 2.40 s, 3 H (CH ₃ -arom.); 4.29 q, 2 H (CH ₂); 7.34–7.46 m, 4 H; 7.56 d, 1 H; 7.72 d, 2 H; 8.19 d, 1 H; 11.50 brs, 1 H (NH) |
| 1c | ¹ H | 1.32 t, 3 H (CH ₃); 4.29 q, 2 H (CH ₂); 7.38 t, 1 H; 7.46 t, 1 H; 7.55 m, 3 H; 7.79 d, 2 H; 8.17 d, 1 H; 11.62 brs, 1 H (NH) |
| 1d | ¹ H | 1.34 t, 3 H (CH ₃); 4.30 q, 2 H (CH ₂); 7.40 t, 1 H; 7.41–7.52 m, 2 H; 7.70–7.81 m, 4 H; 8.16 d, 1 H; 11.67 brs, 1 H (NH) |
| 1e | ¹ H | 1.32 t, 3 H (CH ₃); 4.31 q, 2 H (CH ₂); 7.39 t, 1 H; 7.46 t, 1 H; 7.50–7.57 m, 3 H; 7.85 d, 2 H; 8.15 d, 1 H; 11.60 brs, 1 H (NH) |
| 1f | ¹ H | 1.33 t, 3 H (CH ₃); 3.89 s, 3 H (CH ₃ O); 4.29 q, 2 H (CH ₂); 7.14 d, 2 H; 7.42 m, 2 H; 7.62 d, 1 H; 7.92 d, 2 H; 8.27 d, 1 H; 11.30 brs, 1 H (NH) |
| 1g | ¹ H | 1.33 t, 3 H (CH ₃); 1.41 t, 3 H (CH ₃); 4.16 q, 2 H (CH ₂ O); 4.28 q, 2 H (CH ₂); 7.12 d, 2 H; 7.41 m, 2 H; 7.62 d, 1 H; 7.90 d, 2 H; 8.26 d, 1 H; 11.32 brs, 1 H (NH) |
| 1h | ¹ H | 1.33 t, 3 H (CH ₃); 2.60 s, 3 H (CH ₃ CO); 4.32 q, 2 H (CH ₂); 7.38 t, 1 H; 7.44 d, 1 H; 7.52 t, 1 H; 7.67 d, 2 H; 8.05 d, 2 H; 8.10–8.25 brs, 1 H; 11.77 brs, 1 H (NH) |
| 1i | ¹ H | 1.32 t, 3 H (CH ₃); 4.32 q, 2 H (CH ₂); 7.41 m, 2 H; 7.54 t, 1 H; 7.73 d, 2 H; 8.00–8.20 brs, 1 H; 8.32 d, 2 H; 11.86 brs, 1 H (NH) |
| 2a | ¹ H | 7.52–7.78 m, 8 H; 8.05 d, 1 H |
| 2b | ¹ H | 2.44 s, 3 H (CH ₃); 7.41 d, 2 H; 7.53 m, 3 H; 7.77 q, 2 H; 8.05 d, 1 H |
| 2c | ¹ H | 7.55 t, 1 H; 7.64 d, 2 H; 7.75 t, 1 H; 7.76 t, 1 H; 7.80 d, 2 H; 8.06 d, 1 H |
| 2d | ¹ H | 7.54 t, 1 H; 7.65 d, 2 H; 7.76 t, 1 H; 7.78 t, 1 H; 7.81 d, 2 H; 8.06 d, 1 H |
| 2e | ¹ H | 7.50 d, 2 H; 7.55 t, 1 H; 7.77 m, 2 H; 7.98 d, 2 H; 8.06 d, 1 H |
| 2f | ¹ H | 3.88 s, 3 H (CH ₃ O); 7.14 d, 2 H; 7.52–7.60 m, 3 H; 7.78 q, 2 H; 8.05 d, 1 H |
| 2g | ¹ H | 1.42 t, 3 H (CH ₃); 4.15 q, 2 H (CH ₂ O); 7.12 d, 2 H; 7.53–7.57 m, 3 H; 7.76 q, 2 H; 8.05 d, 1 H |
| 2h | ¹ H | 2.70 s, 3 H (CH ₃ CO); 7.56 m, 1 H; 7.79 q, 2 H; 7.86 d, 2 H; 8.08 d, 1 H; 8.18 d, 2 H |
| 2i | ¹ H | 7.57 m, 1 H; 7.80 q, 2 H; 8.03 d, 2 H; 8.09 d, 1 H; 8.48 d, 2 H |
| 3a | ¹ H | 6.89–6.94 m, 2 H; 7.30–7.37 m, 2 H; 7.43 t, 1 H; 7.50–7.55 m, 2 H; 7.59–7.62 m, 2 H; 9.70 s, 1 H (OH); 12.45 s, 1 H (NH) |
| 3b | ¹ H | 2.39 s, 3 H (CH ₃); 6.88–6.94 m, 2 H; 7.31–7.35 m, 4 H; 7.47 d, 2 H; 9.66 s, 1 H (OH); 12.41 s, 1 H (NH) |
| 3c | ¹ H | 6.90–6.94 m, 2 H; 7.32–7.36 m, 2 H; 7.63–7.68 m, 4 H; 9.71 s, 1 H (OH); 12.48 s, 1 H (NH) |

TABLE IV
(Continued)

| Compound | Nucleus | Chemical shifts |
|-----------|----------------|--|
| 3d | ¹ H | 6.90–6.93 m, 2 H; 7.32–7.35 m, 2 H; 7.65 A ₂ B ₂ pattern, 4 H ($\delta_A = 7.58$, $\delta_B = 7.72$, $J = 9$ Hz); 9.71 s, 1 H (OH); 12.48 s, 1 H (NH) |
| 3e | ¹ H | 6.90–6.93 m, 2 H; 7.33 t, 2 H; 7.43 d, 2 H; 7.88 d, 2 H; 9.68 s, 1 H (OH); 12.48 s, 1 H (NH) |
| 3f | ¹ H | 3.83 s, 3 H (CH ₃ O); 6.88–6.93 m, 2 H; 7.06 d, 2 H; 7.34 q, 2 H; 7.50 d, 2 H; 9.74 s, 1 H (OH); 12.39 s, 1 H (NH) |
| 3g | ¹ H | 1.38 t, 3 H (CH ₃); 4.10 q, 2 H (CH ₂ O); 6.87–6.92 m, 2 H; 7.04 d, 2 H; 7.34 q, 2 H; 7.47 d, 2 H; 9.79 s, 1 H (OH); 12.36 s, 1 H (NH) |
| 3h | ¹ H | 2.65 s, 3 H (CH ₃ CO); 6.92–6.95 m, 2 H; 7.36 q, 2 H; 7.79 d, 2 H; 8.10 d, 2 H; 9.77 s, 1 H (OH); 12.50 s, 1 H (NH) |
| 3i | ¹ H | 6.92 t, 2 H; 7.36 q, 2 H; 7.96 d, 2 H; 8.39 d, 2 H; 9.87 s, 1 H (OH); 12.59 s, 1 H (NH) |

^a Measured in CDCl₃.

Compounds **2** are derivatives of a new heterocyclic system. They are yellow crystalline substances, insoluble in water, well soluble in organic solvents, which exhibit yellow-green fluorescence at 366 nm. In the mass spectrum of compound **2a**, distinct peaks of molecular ion m/z 263.3 and of a fragment C₉H₄N₃O₂ (m/z 186.2) were observed which arose by splitting of phenyl from **2a**. The stability of compounds **2** towards hydrolytic splitting might be of interest. Contrary to 1,2,4-triazine analogues which contain fused isocoumarin ring¹⁷, compounds **2** are very susceptible to hydrolytic splitting. The hydrolysis takes place slowly already during standing of a solution of these compounds in aqueous ethanol at room temperature. That is why we preferred

TABLE V
The mass spectra of compounds **1a**, **2a** and **3a**

| Compound | m/z (relative abundance, %) |
|-----------|---|
| 1a | 309.3 (100, M ⁺), 263.3 (68, C ₁₅ H ₉ N ₃ O ₂ ⁺), 91.1 (51, C ₆ H ₅ N ⁺), 77.1 (100, C ₆ H ₅ ⁺) |
| 2a | 263.3 (100, M ⁺), 186.2 (61, C ₉ H ₄ N ₃ O ₂ ⁺), 77.1 (41, C ₆ H ₅ ⁺) |
| 3a | 281.3 (90, M ⁺), 210.3 (38, C ₁₃ H ₁₀ N ₂ O ⁺), 91.1 (100, C ₆ H ₅ N ⁺) |

measuring UV spectra of compounds **2a–2i** in benzene. Contrary to analogues derivatives of pyrazolo[3,4-*e*][1,2,4]triazine⁹ or 1,2,4-triazino[4,5-*a*]benzimidazole¹⁸, the triazine ring in compounds **2** is stable, hydrolytic splitting thus gave corresponding 1-aryl-5-(2-hydroxyphenyl)-6-azauracils **3a–3i** (Scheme 1). In the mass spectrum of compound **3a**, both molecular ion peak m/z 281.3 and the peak of fragment C₁₃H₁₀N₂O (m/z 210.3) were observed, which probably arose by partial destruction of 1,2,4-triazine ring, and also the peak of fragment C₆H₅N (m/z 91.1) mentioned above.

EXPERIMENTAL

The melting points were determined on a Boetius stage and are uncorrected. The infrared spectra were measured in KBr disks and scanned on an ATI Unicam Genesis FTIR instrument. Electronic absorption spectra were measured on a UV-VIS PU 8700 spectrophotometer (Philips) in ethanol (compounds **1** and **3**) and in benzene (compound **2**), concentration $5.0 \cdot 10^{-5}$ mol dm⁻³. The NMR spectra were measured in (CD₃)₂SO on a Bruker AMX-360 spectrometer (360 MHz); the chemical shifts are reported in ppm. Elemental analyses were performed with an EA 1108 Elemental Analyser (Fison Instrument). Mass spectra were measured on a ZAB-SEQ mass spectrometer (VG Organic, Manchester, U.K.) using electron impact (EI) ionization. The characteristic data of compounds **1–3** are given in Tables I–III, their NMR spectra in Table IV, and mass spectra of compounds **1a–3a** in Table V.

Ethyl *N*-(3-Arylhydrazono-2,3-dihydrobenzo[*b*]furan-2-ylidene)carbamates **1a–1i**. General Procedure

A solution of corresponding aromatic amine (6.00 mmol) in a mixture of ice water (30 ml) and 37% hydrochloric acid (3.6 ml) was diazotized with a solution of sodium nitrite (6.00 mmol) in ice water (10 ml). The mixture was stirred in ice bath for 15 min and then added portionwise to a solution of ethyl *N*-(benzo[*b*]furan-2-yl)carbamate¹⁹ (6.04 mmol) in pyridine (60 ml), which was cooled to 0–5 °C. The mixture was left to stand at 0–5 °C for 24 h and then slowly diluted with ice water to total volume of 900 ml. The next day the precipitated pale orange solid was collected by suction, washed with water, dried, and for analysis recrystallized from ethanol. For further details, see Tables I, IV and V.

2-Aryl[1]benzofuro[2,3-*e*][1,2,4]triazin-3(2*H*)-ones **2a–2i**. General Procedure

A solution of corresponding hydrazonocarbamate **1** (4.00 mmol) in boiling decalin (40 ml) was refluxed for 1 h and then cooled. After several hours, a yellow solid was collected by suction, washed with hexane and dried at 110 °C in vacuum. Sample for analysis was obtained by recrystallization from benzene. For further details, see Tables II, IV and V.

2-Aryl-6-(2-hydroxyphenyl)-1,2,4-triazine-3(2*H*),5(4*H*)-diones **3a–3i**. General Procedure

A solution of a compound **2** (0.2 mmol) in a mixture of pyridine (10 ml) and water (10 ml) was refluxed for 1 h and then evaporated to dryness. The residue was mixed with water (5 ml), collected by suction, washed with water and dried. Sample for analysis was obtained by recrystallization from ethanol. For further details, see Tables III–V.

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