SYNTHESIS OF SOME 2-ARYL-6-AMINO-2,3-DIHYDRO-1,2,4-BENZO-TRIAZIN-3-ONES*

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Acylation of 2,4-diaminazobenzene and its derivatives *Ia—Ih* with ethyl chloroformate gives high yields of the corresponding 2,4-bis(ethoxycarbonylamino)azobenzenes *IIa—IIh* which, on boiling in acid medium, are cyclized with simultaneous splitting off of the second ethoxycarbonyl group to give the mesomeric 6-amino-2-aryl-3-oxo-2,3-dihydro-1,2,4-benzotriazinium cations *IIIa—IIIh*; the latter have been transformed into the free bases *IVa—IVh* or their tautomers *Va—Vh*. Constitution of the prepared compounds has been studied by IR spectroscopy. Influence of medium on valence vibrations of the carbamate NH groups in the compounds *II* has also been studied.

In one of the previous papers[1] a simple preparation method of 2-aryl-2,3,4,6-tetra-hydro-1,2,4-benzotriazine-3,6-diones was described. In the present work this method was used for preparation of analogous 6-amino derivatives which can be considered vinylogs of 1-aryl-6-azacytosines.

The starting material was 2,4-diaminoazobenzene and its derivatives *Ia—Ih* prepared by azo coupling of diazonium salts with 1,3-diaminobenzene. The compounds *I* underwent smooth acylation with ethyl chloroformate in pyridine medium to give the corresponding 2,4-bis(ethoxycarbonylamino) derivatives *IIa—IIh*.

Structure of the latter compounds was confirmed by IR spectra showing intensive absorption bands in the region of N—H valence vibrations ($3433-3347$ cm^{-1}), that of carbonyl group (1741 cm^{-1}), C=N and C=C bonds ($1611-1400$ cm^{-1}) and C—O bonds (the most intensive spectral band at 1201 cm^{-1}). Except for the N—H valence vibrations, position of these bands is independent of the ring substituents. Presence of these substituents made itself felt by a small number of new bands of medium intensity in spectrum of *IIc* (1114 cm^{-1}), *IId* ($844, 1138$ and 1500 cm^{-1}), *IIe* (1009 cm^{-1}), *IIf* (1005 cm^{-1}), *IIg* (1003 cm^{-1}) and *IIh* ($1034, 1143, 1504$ and 1600 cm^{-1}). In contrast to solutions in tetrachloromethane, IR spectrum of the compound *IIa* measured in solid state (Nujol) shows isolated bands of the two carbonyl groups of carbamate groups. The effect is obviously due to intermolecular hydro-

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gen bonds between the carbamate CO and NH groups which results in the decrease of carbonyl vibration by 40 cm^{-1} and that of N—H valence vibration by as much as 111 cm^{-1}. Table I gives the wave-numbers of the N—H valence vibrations of the azocarbamates *II* measured in tetrachloromethane solutions. The band at lower wave-number values was assigned to the N—H valence vibration, the respective hydrogen being involved in intramolecular hydrogen bond to azo group. Substituents in benzene ring have distinct effect on this vibration in accord with mechanism of substituents influence on this intramolecular hydrogen bond through the electron-donor azo group. Increasing electron-donor nature of the substituents increases strength of this hydrogen bond, the wave-number of the valence vibration being shifted to lower values (and *vice versa*). The ring substituents have very small and opposite effect on wave-number of the N—H valence vibration at the position 4.

TABLE I

2,4-Bis(ethoxycarbonylamino)azobenzenes *II*

Compound	Formula (m.wt.)	Calculated/Found			M.p., °C (ethanol)	ν(N—H)^a, cm^{-1}	
		% C	% H	% N		free	bonded
IIa	$C_{18}H_{20}N_4O_4$ (356·4)	60·66 60·80	5·66 5·71	15·72 15·59	134—135	3 433	3 399
IIb	$C_{19}H_{22}N_4O_4$ (370·4)	61·61 61·60	5·99 6·08	15·13 15·09	125—127	3 432·5	3 397
IIc	$C_{19}H_{22}N_4O_4$ (370·4)	61·61 61·38	5·99 6·00	15·13 15·17	123—124	3 433·5	3 398
IId	$C_{18}H_{19}FN_4O_4$ (374·4)	57·75 58·02	5·12 5·12	14·97 15·19	144—145	3 432·5	3 402
IIe	$C_{18}H_{19}ClN_4O_4$ (390·8)	55·32 55·25	4·90 4·94	14·34 14·43	160—161	3 432	3 402·5
IIf	$C_{18}H_{19}BrN_4O_4$ (435·3)	49·57 49·39	4·40 4·31	12·87 12·76	155—157	3 432	3 402
IIg	$C_{18}H_{19}IN_4O_4$ (482·3)	44·83 44·55	3·97 3·74	11·62 11·49	159—161	3 432	3 402·5
IIh	$C_{19}H_{22}N_4O_5$ (386·4)	59·06 58·93	5·74 5·81	14·50 14·39	126—128	3 433·5	3 397·5

^{a} In tetrachloromethane.

Cyclization of the azocarbamates *II* proceeds on boiling in strongly acidic medium enabling formation of mesomeric cations which are likely intermediates in the cyclization. The cyclization is, however, accompanied by hydrolytic splitting of the second carbamate group so that mesomeric 6-amino-2-aryl-3-oxo-2,3-dihydro-1,2,4-benzotriazinium cations *IIIa−IIIh* are produced. In alkaline medium the latter cations were transformed into 6-amino-2-aryl-2,3-dihydro-1,2,4-benzotriazin-3-ones *IVa* to *IVh* for which 6-imino-2,3,4,6-tetrahydro forms *Va−Vh* can also be considered (Table II). However, the problem of tautomerism could not be solved by IR and ¹H-NMR spectra. The benzotriazine *IVa* was transformed into the acetyl derivative *VIa* by acetylation with acetanhydride.

IR spectra of the compounds *IV* show a single absorption band (within 3274 to 3352 cm^{-1}) (KBr or Nujol) in the region of N—H valence vibrations, which is

TABLE II

6-Amino-2-aryl-2,3-dihydro-1,2,4-benzotriazin-3-ones (*IV*) and their 6-Imino-2,3,4,6-tetrahydro Tautomers (*V*)

Compound	Formula	Calculated/Found			Yield %
		% C	% H	% N	
IVa	$C_{13}H_{10}N_4O$ (238·2)	65·53 65·44	4·23 4·19	23·52 23·43	75·5
IVb	$C_{14}H_{12}N_4O$ (252·3)	66·65 66·53	4·79 4·55	22·21 21·99	86·4
IVc	$C_{14}H_{12}N_4O$ (252·3)	66·65 66·60	4·79 4·69	22·21 22·09	80·3
IVd	$C_{13}H_9FN_4O$ (256·2)	60·93 60·82	3·54 3·32	21·87 21·61	87·7
IVe	$C_{13}H_9ClN_4O$ (272·7)	57·26 57,01	3·33 2·92	20·55 20·26	85·0
IVf	$C_{13}H_9BrN_4O$ (317·2)	49·23 49·50	2·80 2·66	17·67 17·40	75·8
IVg	$C_{13}H_9IN_4O$ (364·2)	42·88 43·13	2·49 2·33	15·39 15·35	74·5
IVh	$C_{14}H_{12}N_4O_2$ (268·3)	62·68 62·48	4·51 4·40	20·89 20·67	74·1

obviously due to intermolecular interactions. Compared with the starting azocarbamates *II*, the bands of carbamate carbonyl vibrations are missing. The benzotriazines *IV* are weak monoacid bases forming with acids mesomeric cations *III* soluble in water. The protonation makes itself felt in IR spectrum of the compound *IIIa* by marked shift of absorption bands in the region of C=O and C=N valence vibrations towards higher wave-numbers as it is the case with a number of similar systems[2,3]. In electronic spectrum this protonation causes bathochromic and hyperchromic shift of the long-wave absorption band.
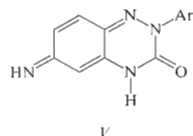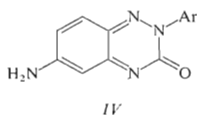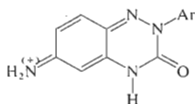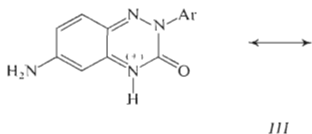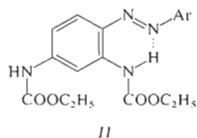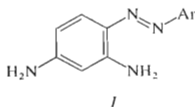
a: Ar = phenyl
b: Ar = 2-tolyl
c: Ar = 4-tolyl
d: Ar = 4-fluorophenyl

e: Ar = 4-chlorophenyl
f: Ar = 4-bromophenyl
g: Ar = 4-iodophenyl
h: Ar = 4-methoxyphenyl

Further attention was paid to influence of medium on position of absorption bands of valence vibrations of the carbamate N—H groups in the azocarbamate *IIh*. Wave-numbers of these vibrations were measured in eight solvents (Table III). Due to low solubility the measurements were not carried out in hydrocarbons. The effect of medium on position of these bands was evaluated quantitatively with the use

of cross-term[4], of Buckingham power series[5]. For free NH group the correlation equation was in the form:

$$v(NH) = 3454 \cdot 8 - 435 \cdot 6(n^2 - 1)(\varepsilon - 1)/(2n^2 + 1)(2\varepsilon + 1),$$

$$R = 0 \cdot 991; \quad F = 279 \cdot 3,$$

where ε and n are relative permittivity and refractive index of the solvent, respectively, R is the correlation coefficient, and the value of the Fisher criterion F is statistically significant for the significance level $\alpha = 0 \cdot 05$. However, this equation failed in evaluation of effect of medium on the wave-number of valence vibration of the bonded NH group. For this purpose (as in the systems characterized by rapid tautomerization[6]) the following equation was useful:

$$v(NH) = 3409 \cdot 1 - 254 \cdot 3(n^2 - 1)^2/(2n^2 + 1)^2, \quad R = 0 \cdot 975, \quad F = 115 \cdot 3$$

$$(\alpha = 0 \cdot 1)$$

The cross-term $(n^2 - 1)(\varepsilon - 1)/(2n^2 + 1)(2\varepsilon + 1)$ represents a product of the terms $(n^2 - 1)/(2n^2 + 1)$ and $(\varepsilon - 1)/(2\varepsilon + 1)$. From the viewpoint of the Onsager model these terms are proportionality constants between the reaction field of the polarized solvent molecules affecting the solute molecule and dipole moment of the latter. The term containing the refractive index characterizes the effect of the oscillating dipole of the vibrating bond, and that containing the relative permittivity characterizes the effect of the equilibrium dipole moment of the studied bond. The term $(n^2 - 1)/(2n^2 + 1)$ only reflects the electronic and atomic polarization of the solvent molecules when orientation polarization of the solvent molecules (orientation of the equilibrium dipole is changed relatively slowly) is impossible due to high velocity of the bond vibration. From the viewpoint of influence on energy levels of the vibrational transitions the primary effect is due to the reaction field of the oscillating dipole. The fact that this reaction field is formed in an already existing reaction field of the equilibrium dipole of the bond leads to a modification of the effect of this field by a factor proportional to the term $(\varepsilon - 1)/(2\varepsilon + 1)$ of the solvent. If, however, a rapid reorientation of this equilibrium bond dipole takes place as a result of a rapid intramolecular process (*e.g.* tautomerization *via* intramolecular hydrogen bond), then the contribution of the orientation polarization of polar solvent molecules to the process of formation of the reaction field becomes zero. Hence, according to the Maxwell theory, the permittivity is transformed into n^2, the term $(\varepsilon - 1)/(2\varepsilon + 1)$ becomes $(n^2 - 1)/(2n^2 + 1)$, and the term $(\varepsilon - 1)(n^2 - 1)$: $:(2\varepsilon + 1)(2n^2 + 1)$ becomes $(n^2 - 1)^2/(2n^2 + 1)^2$. Dependence of wave-number of this N—H bond vibration on mere refractive index shows thus the existence of a very rapid orientation change of equilibrium dipole of this bond, at least so during

excitation by IR radiation. In accord with the Bayliss theory[7] a similar dependence on mere refractive index is also exhibited by position of the long-wave absorption maximum of this compound in electronic spectrum (Table III). The Bayliss correlation equation had the form:

$$\lambda = 357 \cdot 5 + 165 \cdot 0(n^2 - 1)/(2n^2 + 1); \quad R = 0 \cdot 9750, \quad F = 192 \cdot 2 \quad (\alpha = 0 \cdot 1).$$

In contrast to vibrational transitions, electronic transitions from the ground to the excited state are accompanied by marked changes of both magnitude and orientation of dipole moment vector. According to the Franck–Condon principle, transition to an excited state is much faster than motions of molecules, so that electronic and atomic polarization of solvent molecules only is significant for formation of the reaction field. This fact results in position of the absorption maximum being dependent on the solvent refractive index only.

EXPERIMENTAL

The melting points were determined with a Boetius apparatus and are not corrected. The IR spectra and UV spectra were measured with a Zeiss UR-20 and a Unicam SP 1800 spectrophotometers, respectively.

TABLE III

Effect of Medium on Wave-Numbers of Valence Vibrations of Carbamate N—H Groups and on Position of Absorption Maximum in Electronic Spectrum of Compound *IIh*

Solvent	$f(\varepsilon, n^2)$	$f(n^2)$	$\nu(N-H)$, cm^{-1}		λ_{max} nm
			bonded	free	
Pentane	0·032	0·179	—	—	388·0
Hexane	0·034	0·185	—	—	388·0
Heptane	0·037	0·192	—	—	388·5
Cyclohexane	0·042	0·204	—	—	389·5
Tetrachloromethane	0·048	0·214	3 398·0	3 433·5	392·5
Tetrachloroethylene	0·053	0·228	3 396·0	3 432·5	393·5
Trichloroethylene	0·068	0·220	3 395·0	3 427·0	392·5
Carbon disulphide	0·068	0·261	3 392·0	3 423·0	400·5
Dichloromethane	0·085	0·202	3 398·5	3 417·0	392·0
1,2-Dichloroethane	0·090	0·209	3 398·5	3 415·0	393·5
Dibromomethane	0·095	0·239	3 395·0	3 413·0	399·0
Diiodomethane	0·106	0·287	3 388·0	3 409·0	405·0

2,4-Diaminobenzenes *Ia—Ig*

The respective aromatic amine (10.0 mmol) was dissolved in 25—50 ml ice water, 6.0 ml 37% HCl and 10 g ice and diazotized by addition of solution of 690 mg (10.0 mmol) $NaNO_2$ in 20 ml ice water. The diazonium salt was added to solution of 2.0 g (18.49 mmol) 1,3-diaminobenzene in 50 ml ice water. After 2 h the reaction mixture was treated with solution of 6.0 g Na_2CO_3 in 20 ml water with stirring. After several hours the products were collected by suction, washed with water and dried. The yields were within 96—99%. After recrystallization from ethanol the derivatives *Ia—Ic, Ie, If, Ih* had m.p. in agreement with those in refs[8—11]. The 4'-fluoro derivative *Id*, yellow crystals, m.p. 159—161°C; for $C_{12}H_{11}FN_4$ (230.3) calculated: 62.60% C, 4.81% H, 24.33% N; found: 62.86% C, 4.93% H, 24.52% N. The 4'-iododerivative *Ig*, m.p. 154—156°C; for $C_{12}H_{11}IN_4$ (338.2) calculated: 42.62% C, 3.28% H, 16.57% N; found: 42.39% C, 3.39% H, 16.29% N.

2,4-Bis(ethoxycarbonylamino)azobenzenes *IIa—IIh*

Solution of 5.0 mmol respective azo compound *I* in 25 ml pyridine was cooled in ice bath and treated with 3.5 ml (36.7 mmol) ethyl chloroformate added drop by drop with stirring and cooling. The mixture was stirred and cooled for another 2 h and then left to stand at room temperature 20 h. Then it was diluted with 80 ml water added portionwise so that the carbamates *II* might separate in crystalline form. After several hours the products were collected by suction, washed with water and dried. The yields were within 97 to 100%. After recrystallization from ethanol the azocarbamates *II* formed yellow needles. The UV absorption maxima of $2 . 10^{-5}$M ethanolic solution of *IIa* λ_{max} (log ε): 225 nm (4.41), 266 nm (4.11), 379 nm (4.17). IR spectrum of *IIa* (Nujol, cm^{-1}): 423 w, 532 m, 553 m, 620 m, 665 w, 690 m, 723 m, 733 w, 765 sh, 771 s, 817 sh, 828 w, 875 w, 888 m, 921 w, 988 m, 1055 s, 1065 s, 1093 m, 1113 m, 1146 w, 1202 s, 1234 s, 1250 w, 1290 s, 1322 m, 1367 w, 1380 m, 1389 w, 1406 m, 1484 m, 1503 sh, 1515 s, 1530 s, 1585 sh, 1598 s, 1700 s, 1740 s, 3322 m, 3396 m. IR spectrum of *IIa* (solution in CCl$_4$, cm^{-1}): 834 m, 867 m, 884 w, 958 m, 1005 w, 1016 m, 1056 s, 1066 sh, 1093 m, 1114 w, 1127 w, 1148 w, 1201 s, 1225 sh, 1255 m, 1315 s, 1322 sh, 1387 m, 1417 s, 1440 m, 1468 m , 1483 w. 1520 s, 1585 sh, 1598 s, 1611,s 1741 s, 3399 m, 3433 m. For further data see Table I.

6-Amino-2-phenyl-3-oxo-2,3-dihydro-1,2,4-benzotriazinium Chloride (*IIIa*)

a) Mixture of 180 mg (0.50 mmol) azo compound *IIa* and 150 ml 20% HCl was refluxed 5 h, filtered and concentrated. After cooling the separated solid was collected by suction, washed with little 20% HCl and dried. Yield 110 mg (79.2%). After repeated crystallization from 20% HCl yellow crystals, m.p. above 350°C.

b) Recrystallization of the base *IVa* from 20% HCl gave identical product. The UV absorption maxima of $2 . 10^{-5}$M *IIIa* solution in 0.01M-HCl λ_{max}, nm (log ε): 212 (4.51) 258 (4.16), 410 (4.39). IR spectrum (KBr, cm^{-1}): 685 m, 733 m, 752 m, 767 m, 821 m, 844 m, 868 m, 928 w, 950 sh, 996 sh, 1007 m, 1030 m, 1070 m, 1133 m, 1145 s, 1150 sh, 1219 w, 1243 s, 1274 m, 1283 w, 1308 sh, 1355 m, 1375 m, 1436 m, 1455 m, 1480 sh, 1490 s, 1510 sh, 1545 m, 1509 s, 1640 s, 1653 s, 1702 s, 2942 m, 3042 m, 3342 m. For $C_{13}H_{11}ClN_4O$ (274.7) calculated: 56.84% C, 4.04% H, 20.40% N; found: 56.99% C, 3.92% H, 20.11% N.

6-Amino-2-aryl-2,3-dihydro-1,2,4-benzotriazin-3-ones *IVa—IVh* and their
6-Imino-2,3,4,6-tetrahydro Tautomers *Va—Vh*

The respective azocarbamate *II* (*IIa* 2.0 mmol, *IIb—IIe* 1 mmol, *IIf—IIh* 0.5 mmol) was added

to the mixture of 10 ml acetic acid, 40 ml water and 20 ml 98% H_2SO, and the mixture was refluxed 5 h. After cooling it was filtered and neutralized (pH 7) by addition of aqueous ammonia. After several hours the separated solid was collected by suction, washed with water and dried at 110°C. The yields varied within 74 to 88%. For analysis the compounds were crystallized from a large volume of ethanol; orange crystals do not melt up to 350°C being decomposed above 320°C. The UV absorption maxima of $2 . 10^{-5}$M solution of *IVa* in ethanol, λ_{max}, nm (log ε): 227 (4·47), 269 (4·36), 389 (4·33). IR spectrum (Nujol, cm^{-1}) *IVa*: 434 m, 470 w, 485 w, 485 w, 506 m, 527 s, 612 m, 626 w, 656 m, 694 s, 729 s, 767 s, 800 w, 821 m, 828 m, 852 s, 860 w, 887 m, 910 w, 956 m, 1 000 m, 1 006 w, 1 035 w, 1 070 m, 1 140 s, 1 155 s, 1 190 w, 1 200 m, 1 245 s, 1 296 s, 1 305 s, 1 404 m, 1 426 s, 1 480 s, 1 510 s, 1 558 m, 1 601 s, 1 650 s, 1 700 m, 3 274 m, 3 308 m; *IVb*: 423 m, 437 m, 499 m, 513 s, 532 m, 563 m, 617 m, 629 m, 642 m, 696 m, 717 m, 731 s, 764 s, 790 m, 827 m, 839 m, 858 m, 952 m, 1 002 m, 1 036 w, 1 136 w, 1 152 m, 1 188 m, 1 212 s, 1 242 s, 1 273 s, 1 303 s, 1 376 s, 1 401 m, 1 445 s, 1 473 m, 1 509 s, 1 556 m, 1 575 sh, 1 600 s, 1 649 m, 1 680 m, 1 696 w, 1 755 m, 3 283 m; *IVc*: 422 m, 431 w, 460 m, 488 w, 502 w, 520 s, 587 m, 608 w, 623 m, 640 w, 663 m, 705 m, 728 m, 770 m, 785 w, 809 m, 821 m, 836 w, 855 m, 873 w, 950 m, 1 001 m, 1 054 m, 1 107 m, 1 138 s, 1 200 w, 1 217 s, 1 245 s, 1 282 s, 1 307 m, 1 360 sh, 1 380 m, 1 408 w, 1 440 s, 1 474 s, 1 483 s, 1 507 s, 1 562 s, 1 606 s, 1 723 m, 3 328 m; *IVd*: 431 w, 465 w, 485 w, 514 m, 524 m, 587 m, 610 w, 666 m, 710 w, 716 m, 768 m, 798 m, 826 m, 835 w, 853 m, 950 m, 980 w, 1 005 m, 1 088 m, 1 137 s, 1 152 m, 1 172 w, 1 194 w, 1 227 s, 1 245 sh, 1 284 m, 1 308 m, 1 366 sh, 1 375 s, 1 406 sh, 1 415 m, 1 430 sh, 1 450 sh, 1 473 s, 1 485 m, 1 505 s, 1 540 w, 1 565 m, 1 610 s, 1 657 s, 1 680 w, 3 274 m; *IVe*: 447 w, 497 m, 513 w, 552 m, 612 w, 628 w, 664 w, 705 m, 727 m, 732 m, 768 m, 805 w, 812 m, 830 m, 835 w, 845 m, 858 w, 950 m, 1 000 m, 1 088 m, 1 095 m, 1 140 s, 1 178 w, 1 198 w, 1 220 sh, 1 248 m, 1 285 m, 1 308 m, 1 378 m, 1 401 m, 1 450 sh, 1 473 s, 1 490 s, 1 513 s, 1 560 m, 1 608 s, 1 656 s, 1 680 sh, 3 352 m; *IVf*: 439 w, 462 m, 487 m, 504 w, 539 m, 613 w, 626 w, 633 w, 663 m, 702 m, 728 m, 768 m, 805 m, 833 m, 845 m, 855 m, 951 m, 998 m, 1 064 m, 1 140 s, 1 180 w, 1 200 w, 1 224 m, 1 248 s, 1 283 s, 1 308 s, 1 377 m, 1 393 m, 1 405 w, 1 453 m, 1 457 s, 1 488 s, 1 507 s, 1 520 sh, 1 561 m, 1 608 s, 1 650 s, 1 680 sh, 3 340 m; *IVg*: 438 w, 470 w, 485 w, 507 w, 536 m, 663 m, 699 w, 728 m, 769 m, 803 m, 816 w, 832 w, 843 m, 854 w, 951 m, 999 m, 1 057 w, 1 090 w, 1 138 m, 1 177 w, 1 198 m, 1 220 sh, 1 247 m, 1 283 m, 1 308 m, 1 363 sh, 1 377 m, 1 391 m, 1 404 w, 1 452 w, 1 475 s, 1 484 s, 1 508 s, 1 560 w, 1 605 s, 1 653 m, 1 676 sh, 1 690 w, 3 340 m; *IVh*: 423 m, 457 m, 516 m, 538 m, 589 m, 627 m, 641 w, 655 w, 685 m, 720 w, 733 m, 768 m, 785 w, 821 m, 837 sh, 844 m, 865 m, 945 w, 953 m, 996 m, 1 015 m, 1 030 m, 1 075 w, 1 105 m, 1 137 s, 1 149 m, 1 160 m, 1 170 m, 1 202 w, 1 245 s, 1 285 m, 1 305 s, 1 365 sh, 1 376 m, 1 450 sh, 1 474 s, 1 506 s, 1 590 sh, 1 600 s, 1 650 m, 1 679 m, 1 697 m, 3 308 m. For further data see Table II.

6-Acetylamino-2-phenyl-2,3-dihydro-1,2,4-benzotriazin-3-one (*VIa*)

Mixture of 120 mg (0·50 mmol) compound *IVa* and 10 ml acetic acid was heated until dissolution; 5 ml acetanhydride was added thereto and the mixture was refluxed 20 min. After pouring in 30 ml water and decomposition of the excess anhydride the separated solid was collected by suction, washed with water and dried. Yield 108 mg (76·5%). After recrystallization from ethanol yellow crystals, m.p. 342—344°C with decomposition. IR spectrum (KBr, cm^{-1}): 420 w, 477 w, 495 m, 535 w, 597 m, 630 w, 650 w, 695 m, 735 m, 740 w, 764 m, 783 w, 790 w, 824 m, 855 w, 880 m, 937 m, 960 w, 1 005 m, 1 109 m, 1 152 w, 1 190 w, 1 225 m, 1 245 m, 1 253 m, 1 288 m, 1 320 w, 1 368 m, 1 383 m, 1 444 s, 1 470 m, 1 504 s, 1 566 s, 1 620 s, 1 663 s, 1 710 c. For $C_{15}H_{12}N_4O_2$ (280·3) calculated: 64·27% C, 4·32% H, 19·99% N; found: 64·10% C, 4·25% H, 19·72% N.

REFERENCES

 1. Slouka J., Šrámková M., Bekárek V.: This Journal *42*, 3449 (1977).
 2. Leonard N. J., Gash V. W.: J. Amer. Chem. Soc. *76*, 2781 (1954).
 3. Leonard N. J., Middleten W. J., Thomas P. D., Choudhury D.: J. Org. Chem. *21*, 344 (1956).
 4. Bekárek V., Mikulecká A.: This Journal *43*, 2879 (1978).
 5. Buckingham A. D.: Proc. Roy. Soc. A *248*, 169 (1958).
 6. Bekárek V.: Acta Univ. Palacki. Olomuc., Fac. Rerum Natur., in press.
 7. Bayliss N. S.: J. Chem. Phys. *18*, 292 (1950).
 8. Hofmann A. W.: Ber. Deut. Chem. Ges. *10*, 216 (1877).
 9. Čepčiansky I., Slavík V., Němec L., Fingerová H., Németh I.: This Journal *33*, 100 (1968).
10. Arient J., Dvořák J.: This Journal *22*, 632 (1957).
11. Charrier G., Beretta A.: Gazz. Chim. Ital. *53*, 743 (1923).