Chemistry of Heterocyclic Compounds, Vol. 31, No. 3, 1995

REACTION OF 1,2,3,4-TETRAHYDROPYRIMIDO[1,2-a]INDOL-10-YLMETHYLENIMINIUM SALTS WITH CERTAIN NUCLEOPHILIC AGENTS

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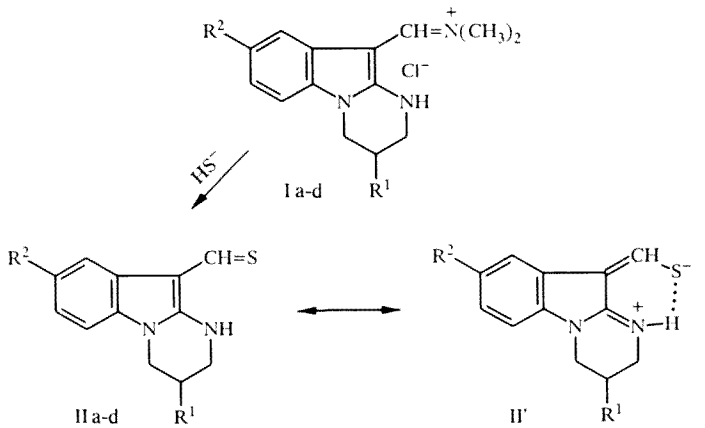
The reaction of iminium salts of 1,2,3,4-tetrahydropyrimido[1,2-a]indole-10-carbaldehydes with sodium hydrosulfide, amines, and nitroalkanes leads to the formation of the corresponding thioaldehydes, azomethines, and nitrovinyl derivatives, existing predominantly in the enamidine form.

We showed previously that tetrahydropyrimidoindoles (I) having a methyleniminium grouping in position 10, prepared by the reaction of acylarylpyrazolidines with a Vilsmeier–Haack complex, react readily with several nucleophilic reagents [1-3]. We have broadened this reaction in view of the importance of condensed indole derivatives with heteroatom substituents.

In a short communication [2] we considered the synthesis of thioaldehydes (IIa, b) of several pyrimidoindoles formed by the reaction of sodium hydrosulfide in water or in chloroform* with the iminium salts (I). In the present work we have extended the series of initial salts (I) (see Table 1).

Since the iminium salts (I) are markedly sensitive to the nucleophile strength, the process of thiolysis in water goes fairly rapidly. Only traces of hydrolysis products were detected, the formation of the formyl derivative requires time or heating [5]. The use of chloroform as solvent is preferable for preparative thiolysis for this reason.



I, II a $R^1 = R^2 = H$; b $R^1 = CH_3$, $R^2 = H$; c $R^1 = CH_3$, $R^2 = Br$; d $R^1 = H$, $R^2 = CH_3$

*Carrying out the thiolysis of the iminium salt (Ia) in DMSO leads to the formation of a crystalline solvate of thioaldehyde (IIa) with DMSO of composition 3:1 (40% yield). A similar solvate was isolated in the synthesis of indole-3-thioaldehyde [4].

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 0009-3122/95/3103-0330$12.50

TABLE 1. Characteristics of the Compounds Synthesized

Compound	Empirical formula	mp, °C	Method of synthesis	M^+	UV spectrum		PMR spectrum, δ, ppm in $CDCl_3$	Yield, %
					λ_{max}, nm	log ε		
1	2	3	4	5	6	7	8	9
II a	$C_{12}H_{12}N_2S$	138...141	A	216	218, 288, 400	4,42, 4,33, 4,32	2.23 (2H, m, 3-H); 3.45-3.80 (4H, m, 2-H, 4-H); 7.10 (3H, m, Ar); 7.45 (1H, m, 9-H); 10.25 (1H, s, CHS); 10.80 (1H, s, NH)	57
II b	$C_{13}H_{14}N_2S$	162...164	A, B	230	218, 288, 400	4,40, 4,31, 4,30	1.12 (3H, d, 3-CH_3); 2.20 (1H, m, 3-H); 2.85-4.05 (4H, m, 2-H, 4-H); 6.90 (3H, m, Ar); 7.30 (1H, m, 9-H); 10.05 (1H, s, CHS); 10.60 (1H, s, NH)	63 (A), 65 (B)
II c	$C_{13}H_{13}BrN_2S$	200...202	A	308*	226, 296, 400	4,26, 4,21, 4,20	1.21 (3H, d, 3-CH_3); 2.35 (1H, m, 3-H); 2.80-4.0 (4H, m, 2-H, 4-H); 7.05 (2H, m, Ar); 7.61 (1H, m, 9-H); 10.25 (1H, s, CHS); 10.70 (1H, s, NH)	42
II d	$C_{13}H_{14}N_2S$	145...146	A	230	220, 290, 400	4,41, 4,32, 4,30	2.20-2.55 (2H, m, 3-H); 2.35 (3H, s, 8-CH_3); 3.25-4.00 (4H, m, 2-H, 4-H); 6.85 (3H, m, Ar); 10.20 (1H, s, CHS); 10.80 (1H, s, NH)	25*[2]
II e	$C_{10}H_{10}N_2S$	170...171	A	190	234, 286, 400	4,42, 4,28, 4,39	3.45 (3H, s, NCH_3); 7.32 (4H, m, Ar); 9.15 (2H, br.s, NH_2); 10.11 (1H, s, CHS)	40*[3]
III a	$C_{18}H_{17}N_3$	184...186	A', B'	–	243, 282, 376	3,97, 3,86, 4,02	2.15 (2H, m, 3-H); 3.55-4.20 (4H, M, 2-H, 4-H); 7.00-7.60 (10H, m, Ar + NH); 8.80 (1H, s, CHN)	22
III b	$C_{20}H_{21}N_3$	174	B'	–	282, 374	4,18 4,29	1.12 (3H, d, 3-CH_3, J = 7 Hz); 2.35 (1H, m, 3-H); 2.32 (3H, s, p-CH_3); 2.80-4.30 (4H, m, 2-H, 4-H); 6.90-7.70 (8H, m, Ar); 8.06 (1H, s, NH); 8.76 (1H, s, CHN)	83
III c	$C_{19}H_{19}N_3O$	129...131	A'	–	285, 372	3,89, 4,45	2.11 (2H, m, 3-H); 3.40-4.05 (4H, m, 2-H, 4-H); 3.90 (3H, s, OCH_3); 6.90-7.75 (8H, m, Ar); 8.65 (1H, s, CHN)	37
III d	$C_{18}H_{16}BrN_3$	147...148	A', B'	355*	270, 325, 380	4,25, 3,92, 4,40	2.18 (2H, m, 3-H); 3.46 (2H, m, 4-H); 3.90 (2H, m, 2-H); 6.94-7.44 (9H, m, Ar + NH); 8.60 (1H, s, CHN)	41
III e	$C_{19}H_{19}N_4O_2$	209...210	B'	–	239, 300, 474	4,43, 4,14, 4,50	1.13 (3H, d, 3-CH_3, J = 7 Hz); 2.35 (1H, m, 3-H); 3.00-4.30 (4H, m, 2-H, 4-H); 7.10 (4H, m, Ar); 7.80 (1H, s, NH); 7.42; 8.20 (4H, AB system, p-NO_2Ph, J = 9 Hz); 8.96 (1H, s, CHN)	75
III f	$C_{21}H_{18}N_4$	247...248	B'	–	241, 282, 407	4,41, 4,22, 4,43	2.32 (2H, m, 3-H); 3.45-3.80 (2H, m, 4-H); 3.95-4.20 (2H, m, 2-H); 7.00-8.16 (9H, m, Ar); 8.85 (1H, s, 2-H Qu); 8.95 (1H, s, CHN)	24
III g	$C_{15}H_{14}N_4S$	150...151	B'	–	239, 284, 414	4,32, 4,15, 4,53	2.25 (2H, m, 3-H); 3.45 (2H, m, 4-H); 4.05 (2H, m, 2-H); 6.85-7.70 (5H, m, Ar); 7.90 (1H, d, 4-H Tz); 8.95 (1H, s, CHN)	28

TABLE 1 (continued)

1	2	3	4	5	6	7	8	9
Va	$C_{19}H_{19}N_3$	128...129	A',B'	289	247, 325, 375, 392	4,52, 4,32, 2,76, 2,56	2.20 (2H, m, 3-H); 3.40-4.20 (4H, m, 2-H, 4-H); 4.80 (2H, s, CH_2Ph); 6.65 (1H, br.s, NH); 7.10-7.90 (8H, m, Ar); 8.80 (1H, s, CHN)	85
Vb	$C_{20}H_{21}N_3$	123...124	B'	303	250, 376, 460	4,35, 4,20, 2,30	1.20 (3H, d, 3-CH_3); 2.90-4.00 (5H, m, 2-H, 3-H, 4-H); 4.60 (2H, s, CH_2Ph); 6.65-7.90 (10H, m, Ar + NH); 8.50 (1H, s, CHN)	80
Vc	$C_{16}H_{21}N_3$	172...173	B'	–	245, 287, 317, 377	4,31, 4,21, 4,19, 3,50	1.32 (9H, s, t-Bu); 2.06 (2H, m, 3-H); 3.55 (2H, m, 4-H); 3.85 (2H, m, 2-H); 6.82-7.32 (4H, m, Ar); 7.89 (1H, br.s, NH); 8.08 (1H, s, CHN)	94
Vd	$C_{17}H_{23}N_3$	161...163	B'	269	244, 288, 317, 377	4,27, 4,22, 4,20, 3,55	1.19 (12H, s + d, t-Bu + 3-CH_3); 2.30 (1H, m, 3-H); 2.95-4.05 (4H, m, 2-H, 4-H); 6.00 (1H, br.s, NH); 6.55-7.90 (4H, m, Ar); 8.12 (1H, s, CHN)	86
Ve	$C_{14}H_{17}N_3$	120...122	B'	227	262, 327, 460	4,24, 4,15, 2,50	1.12 (3H, d, 3-CH_3); 2.95-4.05 (5H, m, 2-H, 3-H, 4-H); 3.32 (3H, s, NCH_3); 6.90-7.50 (5H, m, Ar + NH); 8.35 (1H, s, CHN)	62
VIa *4	$C_{13}H_{13}N_3O_2$	240 (decomp.)	–	243	–	–	2.16 (2H, m, 3-H); 3.32 (2H, m, 4-H); 3.79 (2H, m, 2-H); 6.95-7.50 (4H, m, Ar); 7.47; 8.41 (2H, 2 d, CH=CH, J = 12.5 Hz); 8.72 (1H, s, NH)	55
VIb	$C_{14}H_{15}N_3O_2$	237...240 (decomp.)	–	257	476	4,32	1.12 (3H, d, 3-CH_3, J = 7 Hz); 2.32 (1H, m, 3-H); 3.04-4.25 (4H, m, 2-H, 4-H); 7.10-7.63 (4H, m, Ar); 7.56; 8.48 (2H, 2, d, CH=CH, J=12.5 Hz); 8.82 (1H, s, NH)	62
VIc	$C_{14}H_{15}N_3O_2$	219	–	–	505	4,56	2.17 (2H, m, 3-H); 2.41 (3H, s, CH_3–C=C); 3.42 (2H, m, 4-H); 4.05 (2H, m, 2-H); 6.95- 7.40 (4H, m, Ar); 8.15 (1H, s, NH); 8.39 (1H, s, CH=C)	54
VId	$C_{15}H_{17}N_3O_2$	236	–	271	277, 305, 490	3,96, 4,04, 4,08	1.20 (3H, t, $\underline{CH_3}CH_2$); 2.18 (2H, m, 3-H); 3.55 (2H, m, 4-H); 4.05 (4H, m, 2-H, $\underline{CH_2}CH_3$); 6.85-7.55 (5H, m, Ar + CH=C); 8.15 (1H, br.s, NH)	33

*M^+ is given for the ^{81}Br isotope.

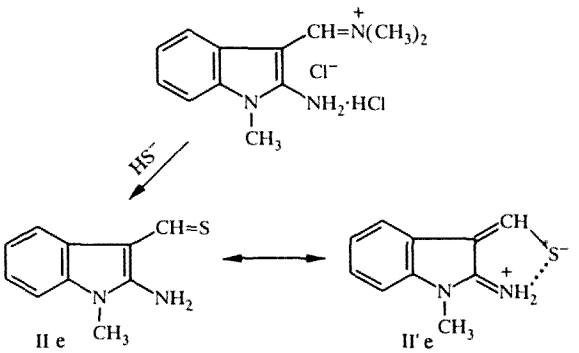
*2Yield of compound is given calculated on the initial hydrazide.

*3Yield of compound is given calculated on 2-amino-1-methylindole hydrochloride.

*4The PMR spectra of compounds (VI) were taken in DMF-D_7.

The compounds obtained (IIa-d) (see Table 1) have a signal for the thioaldehyde proton at ~10 ppm in the PMR spectra and a diffuse absorption band for the salt-forming NH group (2600-3200 cm^{-1}) in the IR spectra. The existence of an intramolecular hydrogen bond was demonstrated by measuring the IR spectra in chloroform solution with dilution. The mass spectra of these compounds, containing fragment peaks with m/z $[M-CHS]^+$ and $[M-S]^+$, confirmed the presence of the thioaldehyde group. The relative stability of the derivatives obtained, by analogy with known examples of thioaldehydes of the heterocyclic series [6], suggests the existence of the substances to be predominantly in the mesoionic form (II'). In reality the UV spectra of these compounds (λ_{max} 290, 400 nm) were closely similar to the spectrum of the corresponding 10-oxo analog having the structure of a cis α-iminoketone [7].

A similar result was obtained on carrying out thiolysis of the iminium salt of 2-amino-3-formyl-1-methylindole synthesized by the formylation of the appropriate indole according to Vilsmeier–Haack [5]. The thioaldehyde (IIe) possesses somewhat lower stability than compounds (IIa-d).



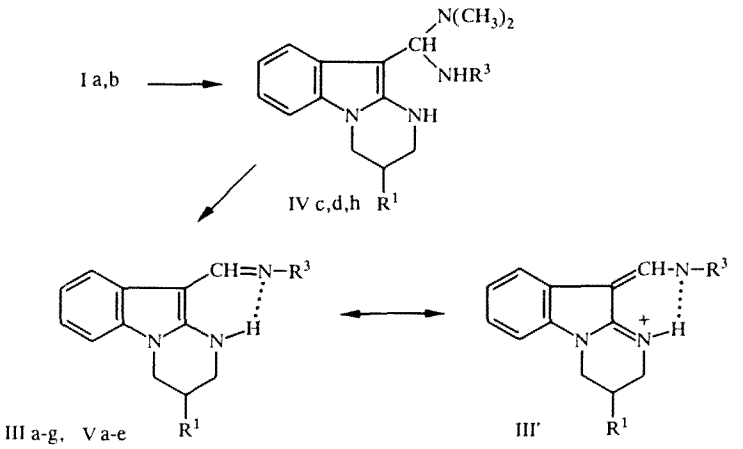
We noted previously [1] that the reaction of iminium salt (Ib) with p-toluidine and p-nitroaniline leads to the corresponding Schiff's base (III). We showed that Schiff's bases may also be obtained with other aromatic amines such as aniline, p-anisidine, and p-bromoaniline, viz. compounds (IIIa, c, d) respectively. A change in the conditions of isolating the reaction products (omitting the column chromatography) enabled us to identify the intermediate products of this conversion in two cases (with p-anisidine and p-bromoaniline). These are the corresponding animals (IVc, d). Signals were observed in the PMR spectra of these compounds for protons of the dimethylamino group and of aniline simultaneously and also a signal for the aminal proton at ~8.20 ppm. There were absorption bands for a salt-forming amino group (2800-3200 cm^{-1}) in the IR spectra. The imines (IIIc, d) are formed on chromatographing compounds (IVc, d) on a column of aluminum oxide.

Condensation of the iminium salt (Ia) with amines of the heterocyclic series, such as 3-aminoquinoline and 2-aminothiazole, proceeds in a similar manner and leads to the methylenimines (IIIf, g). The aminals (IV) were not isolated in these cases.

The reaction was common to aliphatic and aliphatic-aromatic amines. Thus on reacting the salt (Ia) with benzylamine the aminal hydrochloride (IVh) was isolated as in the previous cases and was converted into the corresponding imine (Va) on chromatography on aluminum oxide.

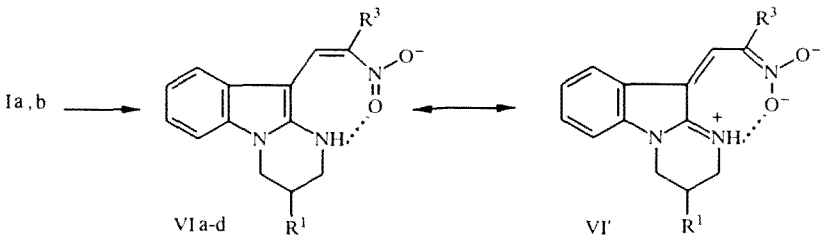
The corresponding imines (Vc-e) (see Table 1) were isolated from the reaction of iminium salts with such low-boiling liphatic amines as t-butylamine and methylamine. Aminals (IV) were not isolated in these cases.

On the basis of the fact that the UV spectra of the derivatives of aliphatic amines (Va-e) are close to the spectra of 2-amino-3-formylindoles (λ_{max} 259, 324 nm) [5], it may be assumed that they exist predominantly in the indole form. The azomethines of the aromatic series (IIIa-e) have UV spectra of a different character which is close to the spectrum of the thioaldehydes (II), i.e. they are probably in the enamidine form. According to the data of IR spectroscopy, the structure of these compounds is also stabilized by an intramolecular hydrogen bond. The absence of an effect from the substituent in the para position of the phenyl nucleus of the aniline fragment of imines (IIIa-e) on the stability of this hydrogen bond may be explained by conjugation in the quasi-aromatic structure generated. The displacement of the absorption band of the NH group on forming the hydrogen bond was ~50 cm^{-1} for all the azomethines (IIIa-e).



III a R^1 = H, R^3 = Ph; b R^1 = CH$_3$, R^3 = C$_6$H$_4$CH$_3$-*p*; c R^1 = H, R^3 = C$_6$H$_4$OCH$_3$-*p*; d R^1 = H, R^3 = -C$_6$H$_4$Br-*p*; e R^1 = CH$_3$, R^3 = C$_6$H$_4$NO$_2$-*p*; f R^1 = H, R^3 = 3-quinolyl; g R^1 = H, R^3 = 2-thiozolyl; IV c R^1 = H, R^3 = C$_6$H$_4$OCH$_3$-*p*; d R^1 = H, R^3 = C$_6$H$_4$Br-*p*; h R^1 = H, R^3 = CH$_2$Ph; V a R^1 = H, R^3 = CH$_2$Ph; b R^1 = CH$_3$, R^3 = CH$_2$Ph; c R^1 = H, R^3 = *t*-Bu; d R^1 = CH$_3$, R^3 = *t*-Bu; e R^1 = R^3 = CH$_3$

The nitrovinyl derivatives (VIa-d) (see Table 1) were obtained in good yield on reacting the iminium salts (I) with nitroalkanes in the presence of sodium ethylate. These were intensely colored compounds (λ_{max} 475-490 nm). In their IR spectra, apart from the expected absorption bands of the nitro group and the C=N bond, absorption bands were present in dilute solutions of compounds (IVa, b) corresponding to the vibrations of free and bonded OH groups (3600, 3200-3400 cm^{-1}), which suggests the existence of a prototropic transfer to an oxygen atom of the nitro group.



VI a R^1 = R^3 = H; b R^1 = CH$_3$, R^3 = H; c R^1 = H, R^3 = CH$_3$; d R^1 = H, R^3 = Et

This transfer was absent from the spectrum of compound (VId) which has an alkyl substituent at the vinyl group, probably for steric reasons.

The possibility of forming an enamidine system of bonds and its relative stability are characteristic properties of 2-aminoindole derivatives having a substituent at position 3 with conjugated multiple bonds.

EXPERIMENTAL

The UV spectra of compounds were taken on a Cary 40 instrument in alcohol, and the IR spectra on UR 20 or Specord IR 75 instruments in Nujol mulls or chloroform solution. The PMR spectra were measured on a Tesla BS 476A (60 MHz) instrument, and the mass spectra on a Varian MAT 111 (70 eV) instrument with insertion of samples into the ion source. Preparative chromatography was carried out on columns of silica gel (40/100) or neutral aluminum oxide (100/250). The constants and yields of the compounds obtained are given in Table 1.

The data of elemental analysis for C, H, and N and/or of mass spectrometry (M^+) agreed with calculated values.

3-Methyl-1,2,3,4-tetrahydropyrimido[1,2-a]indol-10-ylmethylenedimethyliminium chloride (Ib) was synthesized by the method of [5]. The remaining iminium salts (I) were obtained analogously.

1,2,3,4-Tetrahydropyrimido[1,2-a]indol-10-ylmethylenedimethyliminium Chloride (Ia) $C_{14}H_{19}ClN_3$. Yield was 63% of mp 254°C. PMR spectrum (DMSO-D_6): 2.33 (2H, m, 3-H); 2.40-3.50 (4H, m, 2-H, 4-H); 3.51 (6H, s, NMe_2); 7.40 (4H, m, Ar); 8.70 (1H, s, CHN^+); 9.85 ppm (1H, s, NH).

8-Bromo-3-methyl-1,2,3,4-tetrahydropyrimido[1,2-a]indol-10-ylmethylenedimethyliminium Chloride (Ic) $C_{15}H_{21}BrClN_3$. Yield was 66% of mp 220°C (with decomposition). PMR spectrum (DMSO-D_6): 1.05 (3H, d, 3-CH_3); 3.80 (4H, m, 2-H); 4.15 (1H, m, 3-H); 7.40 (2H, m, Ar); 7.75 (1H, s, 9-H); 8.80 (1H, S, CHN^+); 9.90 ppm (1H, s, NH).

1,2,3,4-Tetrahydropyrimido[1,2-a]indole-10-thials (IIa-e). A. The iminium salt (I) (3.5 mmole) was stirred with anhydrous sodium hydrosulfide (3.5 mmole) in absolute chloroform (5 ml) for 2-3 h at room temperature. The mixture was washed three times with water, the chloroform evaporated, and the residue recrystallized from a mixture of benzene and hexane.

B. Sodium hydrosulfide (3.5 mmole) was added to a solution of the iminium salt (I) (3.5 mmole) in distilled water (5 ml), the mixture stirred for 5 min, then chloroform (5 ml) added. The chloroform layer was separated, the solvent evaporated, and the residue recrystallized.

10-Dimethylamino-(p-anisidino)methyl-1,2,3,4-tetrahydropyrimido[1,2-a]indole Hydrochloride (IVc) $C_{21}H_{26}N_4O \cdot HCl$. A solution of iminium salt (Ia) (0.66 g; 2.5 mmole) and p-anisidine (0.4 g; 3.25 mmole) in dry pyridine (10 ml) was boiled for 2 h. The pyridine was distilled off in vacuum, the residue washed with water and with ether, dried, and recrystallized from methanol. Compound (IVc) (0.73 g; 75%) was obtained having mp 230°C (with decomposition). PMR spectrum (CD_3OD): 2.16 (2H, m, 3-H), 3.45-4.20 (4H, m, 2-H, 4-H); 4.00 (3H, s, OCH_3); 4.35 (6H, s, NMe_2); 7.00-7.70 (8H, m, Ar); 8.50 (1H, s, 10-CH); 9.00 ppm (1H, br.s, NH).

10-Dimethylamino-(p-bromoanilino)methyl-1,2,3,4-tetrahydropyrimidino[1,2-a]indole (IVd) $C_{20}H_{23}BrN_4 \cdot HCl$. A solution of iminium salt (Ia) (0.66 g; 2.5 mmole) and p-bromoaniline (0.56 g; 3.25 mmole) in dry pyridine (10 ml) was boiled for 1 h. The pyridine was distilled off in vacuum, the residue washed with water, and recrystallized from methanol. Compound (IVd) 1 g: 92%) was obtained of mp 220°C (with decomposition). PMR spectrum (CD_3OD): 2.12 (2H, m, 3-H); 3.40-4.10 (4H, m, 2-H, 4-H); 4.40 (6H, s, NMe_2); 7.00-7.80 (8H, m, Ar); 8.40 (1H, s, 10-H); 9.10 ppm (1H, br.s, NH).

10-Dimethylamino(benzylamino)methyl-1,2,3,4-tetrahydropyrimido[1,2-a]indole Hydrochloride (IVh) $C_{21}H_{26}N_4 \cdot HCl$. A solution of iminium salt (Ia) (0.66 g; 2.5 mmole) and benzylamine (0.35 g; 3.25 mmole) in dry triethylamine (6 ml) was boiled for 2 h. The triethylamine was distilled off in vacuum, the residue washed with water, and recrystallized from methanol. Compound (IVh) (0.85 g: 88%) of mp 250°C (with decomposition) was obtained. PMR spectrum (DMSO-D_6): 2.21 (2H, m, 3-H); 3.20-4.05 (4H, m, 2-H, 4-H); 4.20 (6H, s, NMe_2); 4.90 (2H, s, CH_2Ph); 7.01-7.90 (9H, m, Ar); 8.10 (1H, s, 10-CH); 9.20 ppm (1H, br.s, NH).

1,2,3,4-Tetrahydropyrimido[1,2-a]indol-10-ylmethylenimines (III), (Va, b). A. A solution of aminal (IV) in chloroform was chromatographed on a column of aluminum oxide. The solvent was distilled off in vacuum.

B. A mixture of the iminium salt (I) (1 mmole) and the substituted aniline (1.3 mmole) in dry pyridine (5 ml) [isopropyl alcohol for compounds (IIIf, g)] was boiled for 2 h. The solvent was distilled off in vacuum, the residue washed with water, dried, and chromatographed on a column of aluminum oxide in chloroform. The solvent was distilled off in vacuum.

1,2,3,4-Tetrahydropyrimido[1,2-a]indol-10-ylmethylenimines (Vc-e) were synthesized by method B using a 2-3 fold excess of t-butylamine for compounds (Vc, d) or a 15-fold excess of methylamine for compound (Ve).

10-(2-Nitrovinyl)-1,2,3,4-tetrahydropyrimido[1,2-a]indoles (VIa-d). Nitromethane (4 mmole) and 2N sodium ethylate in alcohol solution (2 ml) were added to a solution of the iminium salt (I) (2.5 mmole) in dry pyridine (10 ml). The mixture was stirred for 2 h at room temperature, the pyridine distilled off in vacuum, the residue filtered off, and washed with water. After recrystallization from DMF, the product was washed with water, and dried.

The authors are grateful to the Russian Fund for Fundamental Investigations for financing the investigations in the area of heterocyclic chemistry (Project Code 93-03-4593).

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