THE STRUCTURE OF PRODUCTS OF COUPLING OF ARENEDIAZONIUM SALTS WITH 3-AMINOCROTONONITRILE DERIVATIVES

Mohamed Hilmy ELNAGDI[a], Kamal Usef SADEK[b], Nadia Mohamed TAHA[a]
and Youssef Mahfous YASSIN[a]

[a] *Chemistry Department, Cairo University, Giza, A.R. Egypt and*
[b] *Chemistry Department, Minia University, Minia, A.R. Egypt*

The structure of products of coupling arenediazonium salts with 3-aminocrotononitrile derivatives is reported. The spectral data as well as the chemical behaviour of coupling products indicated that it is the hydrazone in (E)-form.

Some time ago we reported a new synthesis of pyridazines by cyclization of products of coupling *Ib* with benzenediazonium chloride[1]. Although this synthetic approach has been extensively utilized for synthesis of polyfunctionally substituted pyridazines[2-11], no trial to establish the exact structure of the products of coupling *I* with arenediazonium salts has been made. Further the mechanism of conversion of the coupling products into pyridazines has not been clarified. In the present article we report the results of our investigation aiming to establish both the structure of coupling of *Ia — Id* with arenediazonium salts and the mechanism of their conversion into pyridazines. Thus, it has been found that *Ia — Id* coupled with arenediazonium salts to yield products that may be formulated as the hydrazone *II* or the 3(2*H*)-pyridazinimines *III*. The hydrazone structure *II* is readily established for products of coupling of *Ib* with arenediazonium salts as the IR spectra of all prepared products revealed C≡N absorption which should be absent if these are the cyclic imines *III* (Table II). In addition ¹H NMR of these products revealed two ester functions (Table III). Compounds *IIa*, *IIb*, and *IIe* have been synthesized earlier by us[1,6]. Although IR spectra cannot be utilized for establishing the structure of other products of coupling with arenediazonium salts, the chemical behaviour of all prepared coupling products revealed that they are the acyclic hydrazones *II*. Thus, the products of coupling of *Ic, Id* are isomerized on reflux in acetic acid––hydrochloric acid mixture for a short period. Although these isomerization products seemed to be the 3(2*H*)-pyridazinimines *IIIk — IIIo* one can also argue that these are the (Z)-isomer of *II*. In order to eliminate this possibility products *IIe — IIi* are also isomerized on reflux in acetic acid–hydrochloric acid mixture to yield products that are considered to be the 3(2*H*)-pyridazinimines *III* and not the (Z)-hydrazone *IV*

based on IR spectra which revealed the absence of C≡N absorption in the spectra of isomerization products of *IIe−IIh*. The UV spectra of these 3($2H$)-pyridazinimides was found very similar to the UV spectra of the products of isomerising *IIk−IIo* in acetic acid–hydrochloric acid mixture. Thus, the 3($2H$)-pyridazinimine structure was established for these products. Compounds *IIIa, IIIe* have been synthesized earlier by cyclization of *IIa, IIe* with acetic acid[1,8]. Compound *IIIa* has also been synthesized via reaction of malononitrile with phenylhydrazonomesoxalonitriles[8]. Attempted cyclization of *IIi* under the same conditions afforded the 3($2H$)-pyridazinone *V*.
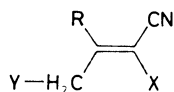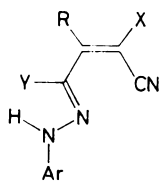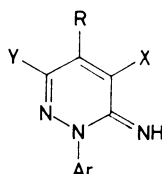

Ia, X = CN ; Y = $CO_2C_2H_5$; R = NH_2
Ib, X = Y = $CO_2C_2H_5$; R = NH_2
Ic, X = Y = CN ; R = NH_2
Id, X = Y = CN ; R = C_6H_5

II   *III*

In formulae *II* and *III* :
a, X = CN ; Y = $CO_2C_2H_5$; Ar = C_6H_5 ; R = NH_2
b, X = CN ; Y = $CO_2C_2H_5$; Ar = $C_6H_4CH_3$-*p* ; R = NH_2
c, X = CN ; Y = $CO_2C_2H_5$; Ar = C_6H_4Cl-*o* ; R = NH_2
d, X = CN ; Y = $CO_2C_2H_5$; Ar = C_6H_4Cl-*p* ; R = NH_2
e, X = Y = $CO_2C_2H_5$; Ar = C_6H_5 ; R = NH_2
f, X = Y = $CO_2C_2H_5$; Ar = $C_6H_4CH_3$-*p* ; R = NH_2
g, X = Y = $CO_2C_2H_5$; Ar = $C_6H_4OCH_3$-*p* ; R = NH_2
h, X = Y = $CO_2C_2H_5$; Ar = C_6H_4Cl-*o* ; R = NH_2
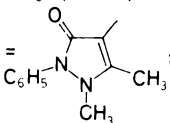
i, X = Y = $CO_2C_2H_5$; Ar = ; R = NH_2

j, X = Y = CN ; Ar = C_6H_5 ; R = NH_2
k, X = Y = CN ; Ar = $C_6H_4CH_3$-*p* ; R = NH_2
l, X = Y = CN ; Ar = $C_6H_4OCH_3$-*p* ; R = NH_2
m, X = Y = CN ; Ar = C_6H_5 ; R = C_6H_5
n, X = Y = CN ; Ar = $C_6H_4CH_3$-*p* ; R = C_6H_5
o, X = Y = CN ; Ar = $C_6H_4OCH_3$-*p* ; R = C_6H_5

In contrast, attempted isomerization of *IIa—IIc* under similar conditions failed. The extra stability of these products can be rationalized by assuming extra stabilisation of the (E)-hydrazone formed in these compounds by hydrogen bonding. This rationalization seems more logical than assuming that these products are realy 3(2H)-pyridazinimines as has been recently suggested by Mittelbach[11]. In order to provide further evidence to support the belief that products of coupling *Ia* and *Ic* are *IIa—IId* and *IIj—IIm*, ³C NMR spectra of *IIb* and *IIk* were run and inspected. ^{13}C NMR spectra revealed two C≡N signals in the spectrum of *IIb* and three C≡N signals in the spectrum of *IIk*. In addition a signal at $\delta \sim 80$ was observed which is assigned to C-4 of *IIb* and C-2 of *IIk*. This signal is hard to be interpreted in terms of the cyclic structure *III*. It is worth to mention that arylhydrazone C-2 in both *IIb* and *IIk* appeared at a somewhat lower field than that expected for a phenylhydrazine derivative (c. 114 ppm). This deshielding is attributed to involvement of nitrogen lone pair in resonance across the C=N linkage which decreases its availability for resonance with benzene ring. For assignment of the particular atoms in ^{13}C NMR spectrum of *IIb* and *IIk* see Fig. 1.

Refluxing *IIa—IIc* in acetic acid–hydrochloric acid mixture for a long time afforded the amino acids *VIa—VIc*. It is believed that *IIa—IIc* are first hydrolyzed into *VIIa—VIIc* which then can be isomerized into the (Z)-hydrazone *VIII*. This latter hydrazones then cyclized into the final isolable *VIa—VId*. Compound *IIj* gave the isomeric amino acid *IX* on treatment with acetic acid in presence of concentrated hydrochloric acid. This indicates that the functional group at C-5 flanked by two amino groups is more readily attacked by hydroxide ion than that at C-3 (Scheme 1).
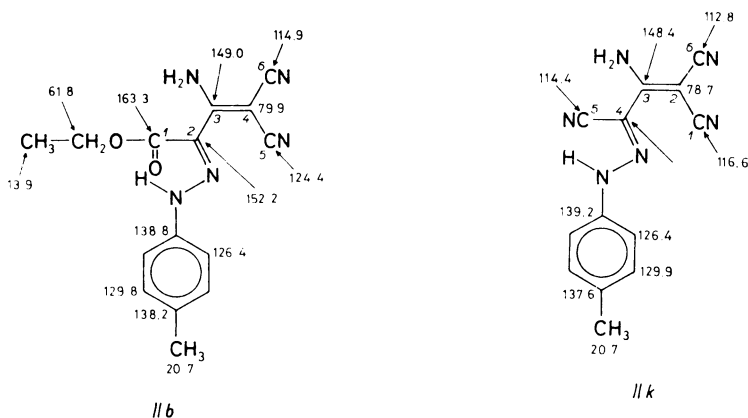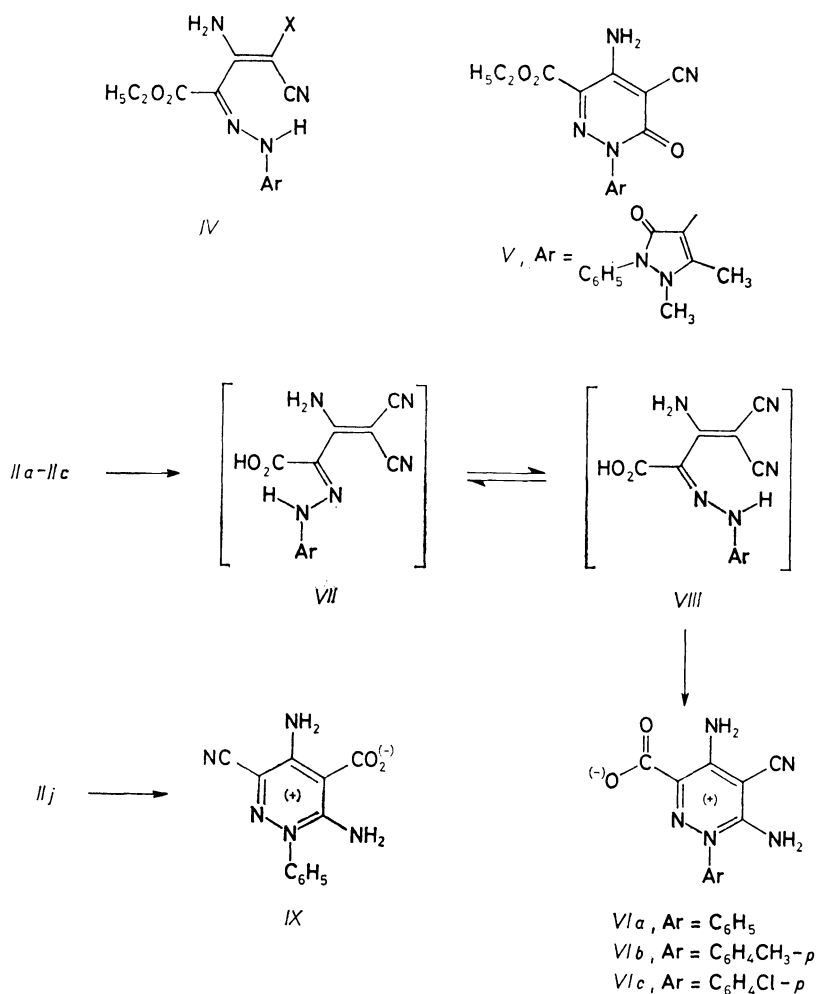

Fig. 1

Assignment of ^{13}C NMR spectra of compounds *IIb* and *IIk*

SCHEME 1

In contrast to previously reported formation of X on refluxing IIe in the presence of acetic anhydride[1], compounds IIe, IIb gave the acetylamino derivatives XIa, XIb. In no case we were able to isolate X. Compound X could be produced only from the 3(2H)-pyridazinone.

Compounds $IIa-IIc$ reacted with hydrazine hydrate to yield products that may be formulated as XII or $XIII$. Structure $XIII$ was readily established for the products based on its conversion into XIV by reflux in AcOH–HCl mixture. If the reaction products were XII then the acid VI would be produced.

In conclusion it may be stated that $Ia - Id$ give always (E)-hydrazones on coupling with arenediazonium salts and not the (Z)-hydrazones as has been previously suggested[6]. These hydrazones need to be isomerized into (Z)-hydrazones to cyclize.
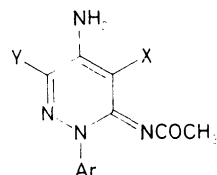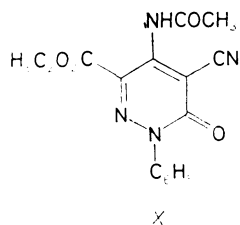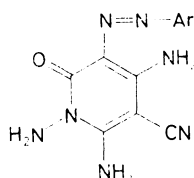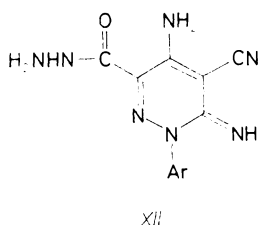
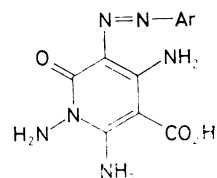This isomerization becomes difficult when an ester function is placed in a position that stabilizes the (E)-hydrazone form while no such effect is present on the stability of the (Z)-form. Cyclization in acid media enhance pyridazine formation while basic reagents as hydrazines react with other functions in the molecule.

EXPERIMENTAL

All melting points are uncorrected. IR spectra were recorded (KBr disc) on a Shimadzu-408 and Perkin–Elmer spectrometer. ^1H NMR spectra were measured in $(CD_3)_2SO$ on a Jeol-PMX--60 spectrometer at 60 MHz and Varian EM-390 spectrometer at 90 MHz using tetramethylsilane as internal standard and chemical shifts are expressed as δ (ppm). Microanalytical data (C, H, N) were obtained from the Microanalytical data unit at Cairo University.

Coupling of $Ia - Id$ with Arenediazonium Chloride

An ice cold solution of 0·01 mol of arenediazonium chlorides (prepared from 0·01 mol of aromatic amine and the appropriate amount of sodium nitrite and hydrochloric acid) was added to a solution of $Ia - Id$ (0·01 mol) in ethanol (100 ml) containing sodium acetate (5·0 g). The precipitate was collected by filtration and crystallized from the proper solvent (see Table I).

TABLE I

List of the newly synthesized compounds

Compound	Cryst. solvent	M.p., °C	Yield %	Formulae (M.w.)	Calculated/Found		
					% C	% H	% N
IIa	ethanol	202[a]	83	$C_{14}H_{13}N_5O_2$ (283·3)	59·4 59·5	4·6 4·5	24·7 24·5
IIb	ethanol	247	88	$C_{15}H_{15}N_5O_2$ (297·3)	60·6 60·6	5·1 5·2	23·6 23·5
IIc	ethanol	237	83	$C_{15}H_{15}N_5O_3$ (313·3)	57·5 57·4	4·8 4·9	22·4 22·5
IId	ethanol	263	80	$C_{14}H_{12}ClN_5O_2$[b] (317·8)	52·9 52·8	3·8 4·0	22·0 22·2
IIe	ethanol	110	80	$C_{16}H_{18}N_4O_4$ (330·3)	58·2 58·2	5·5 5·5	17·0 16·9
IIf	ethanol	130	75	$C_{17}H_{20}N_4O_4$ (344·4)	59·3 59·3	5·9 5·9	16·3 16·3
IIg	ethanol	112	85	$C_{17}H_{20}N_4O_5$ (360·4)	56·7 56·7	5·6 5·7	15·5 15·9
IIh	ethanol	135	82	$C_{16}H_{17}ClN_4O_4$[c] (364·8)	52·7 52·8	4·7 4·7	15·3 15·2
IIi	ethanol	137	92	$C_{21}H_{24}N_6O_5$ (440·5)	57·3 56·9	5·5 5·5	19·1 18·7
IIj	ethanol– –dioxane	216	72	$C_{12}H_8N_6$ (236·2)	61·0 61·2	3·4 3·5	35·6 35·6
IIk	ethanol– –dioxane	190	87	$C_{13}H_{10}N_6$ (250·3)	62·4 62·4	4·0 4·2	33·6 33·5
IIl	ethanol	205	80	$C_{13}H_{10}N_6O$ (266·3)	58·6 58·6	3·8 3·9	31·6 31·3
IIm	ethanol	143	75	$C_{18}H_{11}N_5$ (297·3)	72·7 72·3	3·7 3·7	23·6 23·6
IIn	ethanol	192	72	$C_{19}H_{13}N_5$ (311·4)	73·3 73·5	4·2 4·3	22·5 22·5
IIo	ethanol	170	73	$C_{19}H_{13}N_5O$ (327·4)	69·7 69·9	4·0 4·0	21·4 21·4
IIIe	ethanol	220	60	$C_{16}H_{18}N_4O_4$ (330·3)	58·2 58·3	5·5 5·5	17·0 17·0
IIIf	ethanol– –water	188	60	$C_{17}H_{20}N_4O_4$ (344·3)	59·3 59·6	5·9 5·8	16·3 16·2

TABLE I
(*Continued*)

Compound	Cryst. solvent	M.p., °C	Yield %	Formulae (M.w.)	Calculated/Found		
					% C	% H	% N
IIIg	ethanol–water	>250	62	$C_{17}H_{20}N_4O_5$ (360·4)	56·7 56·3	5·6 5·5	15·5 15·5
IIIh	ethanol–water	145	65	$C_{16}H_{17}ClN_4O_4$[d] (364·8)	52·7 52·6	4·7 4·5	21·3 21·2
IIIk	ethanol	225	65	$C_{13}H_{10}N_6$ (250·3)	62·4 62·5	4·0 4·3	33·6 33·6
IIIm	ethanol	218	60	$C_{18}H_{11}N_6$ (297·3)	72·7 72·5	3·7 3·8	23·6 23·7
IIIn	ethanol	180	62	$C_{19}H_{13}N_5$ (311·4)	73·3 73·3	4·2 4·4	22·5 22·5
IIIo	ethanol	150	62	$C_{19}H_{13}N_5O$ (327·4)	69·7 69·9	4·0 4·0	21·4 21·4
V	DMF–ethanol	284	84	$C_{19}H_{18}N_6O_4$ (394·4)	57·7 57·9	4·5 4·6	21·0 21·3
VIa	acetic acid	>300	70	$C_{12}H_9N_5O_2$ (255·2)	56·5 56·6	3·6 3·6	27·4 27·4
VIb	acetic acid	>300	70	$C_{13}H_{11}N_5O_2$ (269·3)	58·0 58·0	4·1 4·2	26·0 26·0
VIc	acetic acid	>300	70	$C_{12}H_8ClN_5O_2$[e] (289·7)	49·8 49·5	2·8 2·8	24·2 24·2
IX	ethanol	218	65	$C_{12}H_9N_5O_2$ (255·2)	56·5 56·4	3·6 3·8	27·4 27·5
XIa	ethanol	196	72	$C_{18}H_{20}N_4O_5$ (372·4)	58·1 58·2	5·4 5·2	15·0 15·3
XIb	acetone	222	72	$C_{15}H_{12}N_6O_2$ (308·8)	58·4 58·3	3·9 3·9	27·4 27·5
XIIIa	ethanol	162	70	$C_{12}H_{11}N_7O$ (269·3)	53·5 53·8	4·1 4·0	36·4 36·5
XIIIb	ethanol	210	60	$C_{13}H_{13}N_5O$ (283·3)	55·1 55·2	4·6 4·6	34·6 34·5
XIIIc	ethanol–water	240	60	$C_{12}H_{10}ClN_7O$[f] (303·7)	47·5 47·6	3·3 3·5	32·2 32·2
XIV	ethanol	>250	65	$C_{12}H_{12}N_6O$ (288·3)	50·0 49·8	4·2 4·1	29·2 29·3

[a] Misprinted 270°C in ref.[6]; [b] calculated 11·2% Cl, found 11·0%; [c] calculated 9·7% Cl, found 9·5%; [d] calculated 9·7% Cl, found 9·8%; [e] calculated 12·2% Cl, found 12·0%; [f] calculated 11·7% Cl, found 11·6%.

TABLE II

Selected IR data for compounds listed in Table I

Compound	$\tilde{v}_{max}$, cm^{-1}
IIa	3 380, 3 320, 3 240 (NH and NH$_2$); 2 210, 2 200 (CN); 1 710 (C=O); 1 670 (C=N)
IIb	3 390, 3 300, 3 220 (NH and NH$_2$); 2 210, 2 200 (CN); 1 710 (C=O); 1 640 (C=N)
IIc	3 390, 3 300, 3 220 (NH and NH$_2$); 2 220, 2 200 (CN); 1 715 (C=O); 1 640 (C=N)
IId	3 380, 3 300, 3 220, 3 100 (NH and NH$_2$); 2 220, 2 190 (CN); 1 715 (C=O)
IIe	3 500 — 3 200 (NH and NH$_2$); 2 220 (CN); 1 730, 1 720 (C=O)
IIf	3 400, 3 300, 3 200 (NH and NH$_2$); 2 220 (CN); 1 720, 1 680 (C=O); 1 640 (C=N)
IIg	3 400, 3 250, 3 200 (NH and NH$_2$); 2 200 (CN); 1 680, 1 670 (C=O); 1 630 (C=N)
IIh	3 400, 3 270, 3 200 (NH and NH$_2$); 1 690, 1 680 (C=O); 1 620 (C=N)
IIi	3 420 — 3 100 (NH and NH$_2$); 1 720 — 1 690 (C=O); 1 660 (ring C=O); 1 625 (C=N)
IIj	3 450, 3 350 (NH and NH$_2$); 2 210, 2 200 (CN); 1 660 (C=N)
IIk	3 400, 3 350 (NH and NH$_2$); 2 220, 2 200 (CN); 1 650 (C=N)
IIl	3 450, 3 300 (NH and NH$_2$); 2 220, 2 200 (CN); 1 660 (C=N)
IIm	3 350 (NH); 2 200, 2 190 (CN); 1 625 (C=N)
IIn	3 450 (NH); 2 210, 2 200 (CN); 1 630 (C=N)
IIo	3 450 (NH); 2 220, 2 200 (CN); 1 630 (C=N)
IIIe	3 260 — 3 200 (NH and NH$_2$); 1 700 (C=O); 1 660 — 1 630 (C=N)
IIIf	3 450 — 3 200 (br, NH and NH$_2$); 1 730 — 1 710 (C=O groups)
IIIg	3 450 — 3 200 (br, NH and NHα); 1 720 — 1 700 (br, C=O groups)
IIIh	3 400 — 3 300 (br, NH and NH$_2$); 1 730 — 1 710 (C=O groups)
IIIk	3 450, 3 350 (NH and NH$_2$); 2 200 (CN); 1 660 (exocyclic NH)
IIIm	3 420 (NH); 2 200 (br, CN); 1 670 (exocyclic C=NH)
IIIn	3 350 (NH); 2 220 (br, CN)
IIIo	3 350 (NH); 2 210 (br, CN); 1 670 (exocyclic C=NH)
V	3 400, 3 200 (NH$_2$); 2 200 (CN); 1 710 (ester C=O); 1 680 (pyridazine CO); 1 640 (pyrazolin C=O)
VIa	3 500 — 3 200 (br, NH$_2$); 2 200 (CN); 1 660 (C=O)

TABLE II
(*Continued*)

Compound	$\tilde{\nu}_{max}$, cm^{-1}
VIb	3 450—3 200 (br, NH_2); 2 200 (CN); 1 670 (C=O)
VIc	3 450—3 200 (br, NH_2); 2 210 (CN); 1 660 (C=O)
IX	3 450, 3 300, 3 150 (NH_2); 2 200 (CN); 1 690 (C=O)
XIa	3 250 (NH_2); 1 730, 1 710, 1 680 (C=O groups)
XIb	3 400, 3 300, 3 200 (NH_2); 2 200 (CN); 1 720 (C=O)
XIIIa	3 400, 3 300 (NH_2); 2 200 (CN); 1 670 (ring CO)
XIIIb	3 420, 3 340, 3 300 (NH_2); 2 200 (CN); 1 660 (ring CO)
XIIIc	3 400—3 100 (NH_2); 2 200 (CN); 1 650 (ring CO)
XIV	3 450—3 120 (OH and NH_2); 1 670—1 650 (br, C=O groups)

Reaction of *IIa—IIo* and *XIIIa* with Acetic Acid-Hydrochloric Acid Mixture

A solution of the appropriate *IIa—IIo* or *XIIIa* (3·0 g) in acetic acid (20 ml) and hydrochloric acid (37%; 3·0 ml) is heated under reflux for 4 h in case of *IIe—IIo, XIIIa* or 12 h in case of *IIa—IIc*. The reaction mixture is then allowed to cool and poured onto water (80 ml) containing sodium hydrogencarbonate (5·0 g). The solid product, so formed, is collected by filtration and crystallized from the proper solvent (see Table I).

Reaction of *IIe, III* with Acetic Anhydride

A solution of the appropriate *IIe, III* (3·0 g) in acetic anhydride (20 ml) is heated under reflux for 7 h, evaporated in vacuo and then poured onto water. The solid product, so formed, is collected by filtration and crystallized from the proper solvent (see Table I).

Reaction of Compounds *IIa—IIc* with Hydrazine Hydrate

A solution of the appropriate *IIa—IIc* (0·01 mol) in ethanol (50 ml) is treated with hydrazine hydrate (0·5 g, 0·01 mol). The reaction mixture was heated under reflux for 2 h. The precipitate formed during reflux is collected by filtration and crystallized from the proper solvent (see Table I).

T<small>ABLE</small> III

^{1}H NMR data for compounds listed in Table I

Compound	δ, ppm
IIa	1·3 t, 3 H (CH$_3$); 4·3 q, 2 H (CH$_2$); 7·0—8·4 m, 7 H (5 × H-arom. and NH$_2$); 9·0 br, 1 H (NH)
IIb	1·3 t, 3 H (CH$_3$); 2·3 s, 3 H (CH$_3$); 4·2 q, 2 H (CH$_2$); 7·0—8·5 m, 6 H (4 × H-arom. and NH$_2$); 9·0 br, 1 H (NH)
IIe	1·2 t, 6 H (2 × CH$_3$); 4·2 q, 4 H (2 × CH$_2$); 6·5 br, 2 H (NH$_2$); 7·2—8·0 m, 5 H (H-arom.); 10·9 br, 1 H (NH)
IIg	1·2 t, 6 H (2 × CH$_3$); 3·3 s, 3 H (OCH$_3$); 3·8 m, 4 H (2 × CH$_2$); 6·2—6·6 m, 4 H (H-arom.); 8·0 br, 2 H (NH$_2$); 9·3 s, 1 H (NH)
IIi	1·3 t, 6 H (2 × CH$_3$); 2·2 s, 3 H (CH$_3$); 3·2 s, 3 H (CH$_3$); 4·3 q, 4 H (2 × CH$_2$); 7·2—7·5 m, 5 H (H-arom.); 8·3 bs, 2 H (NH$_2$); 9·3 s, 1 H (NH)
IIk	2·3 s, 3 H (CH$_3$); 7·2—7·6 m, 4 H (H-arom.); 8·3 br, 2 H (NH$_2$); 9·2 s, 1 H (NH)
IIl	7·2—7·6 m, 5 H (H-arom.); 8·3 br, 2 H (NH$_2$); 9·2 s, 1 H (NH)
IIn	2·3 s, 3 H (CH$_3$); 6·9—7·8 m, 10 H (H-arom.); 9·3 br, 2 H (NH$_2$)
IIIf	1·3 t, 6 H (2 × CH$_3$); 2·2 s, 3 H (CH$_3$); 4·2 m, 4 H (2 × CH$_2$); 7·2—8·3 m, 7 H (4 × H-arom., NH and NH$_2$)
IIIh	1·3 t, 6 H (2 × CH$_3$); 4·2 m, 4 H (2 × CH$_2$); 7·2—8·4 m, 7 H (4 × H-arom., NH and NH$_2$)
IIIn	2·2 s, 3 H (CH$_3$); 7·2—8·3 m, 10 H (9 × H-arom. and NH)
V	1·3 t, 3 H (CH$_3$); 2·3 s, 3 H (CH$_3$); 3·2 s, 3 H (CH$_3$); 4·2 q, 2 H (CH$_2$); 7·2—7·5 m, 5 H (H-arom.); 8·3 br, 2 H (NH$_2$)
XIa	1·3 t, 6 H (2 × CH$_3$); 2·9 s, 3 H (COCH$_3$); 3·8 q, 4 H (2 × CH$_2$); 6·6—6·9 m, 5 H (H-arom.); 9·7 br, 2 H (NH$_2$)
XIb	2·9 s, 3 H (COCH$_3$); 3·2 s, 3 H (OCH$_3$); 6·7—7·2 m, 4 H (H-arom.); 9·2 br, 2 H (NH$_2$)

REFERENCES

1. Fahmy S. M., Abed N. M., Mohareb R. M., Elnagdi M. H.: Synthesis *1982*, 490.
2. Abdel-Galil F. M., Elnagdi M. H.: Liebigs Ann. Chem. *1987*, 477.
3. Ibrahim N. S., Abdel-Galil F. M., Kandeel Z. E., Abdel-Motaleb R. M., Elnagdi M. H.: Liebigs Ann. Chem. *1988*, (in press).
4. Sadek K. U., Fahmy S. M., Mohareb R. M., Elnagdi M. H.: J. Chem. Eng. Data *29*, 101 (1984).
5. Elnagdi M. H., Elfahham H. A., Elmoghayer M. R. H., Sadek K. U., Elgemie G. E. H.: J. Chem. Soc., Perkin Trans. 1, *1982*, 989.

6. Mohamed M. H., Ibrahim N. S., Elnagdi M. H.: Heterocycles *1987*, 899.

7. Ibrahim N. S., Abdel-Galil F. M., Abdel-Motaleb R. M., Elnagdi M. H.: Heterocycles *1986*, 1219.

8. Hafez E. A., Khalifa M. A. E., Guda S. K. A., Elnagdi M. H.: Z. Naturforsch. *35b*, 485 (1980).

9. Ibrahim N. S., Abdel-Galil F. M., Abdel-Motaleb R. M., Elnagdi M. H.: Bull. Chem. Soc. Jpn. *60*, 4486 (1987).

10. Portmann R. (Ciba-Geigy, A.-G.): Ger. Offen 2,922,227 (1979); Chem. Abstr. *92*, 199741v (1980).

11. Mittelbach M., Wagner U., Kartky C.: Liebigs Ann. Chem. *1987*, 889.