

## ACTION OF AMINES, HYDRAZINES AND HYDROXYLAMINE HYDROCHLORIDE ON 3-BENZYLIDENE-5-(2-METHYL-1-NAPHTHYL)-2 (3H) FURANONE

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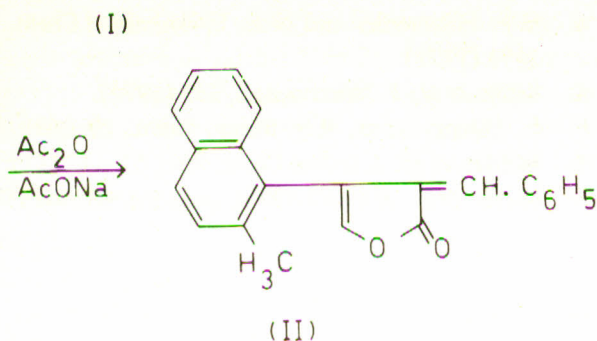
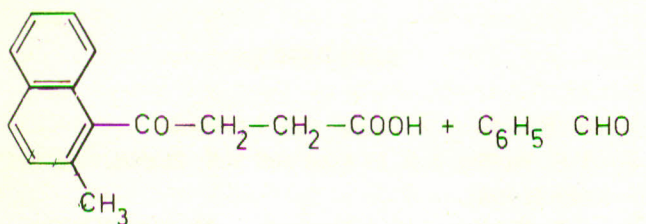
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3-(2-Methyl-1-naphthoyl) propionic acid (I) reacted with benzaldehyde, benzyl methyl ketone,  $\text{Br}_2/\text{POCl}_3/\text{CHCl}_3$ ,  $\text{NH}_2\text{OH}\cdot\text{HCl}$  and semicarbazide to give furanone, pyrrolidinone, acrylic acid, oxazinone or semicarbazone derivatives. The reaction of furanone (II) with amines, ammonium acetate, hydrazines and  $\text{NH}_2\text{OH}\cdot\text{HCl}$  has been investigated.

### INTRODUCTION

Recently [1-4] we reported the condensation of 3-(2-methyl-1-naphthoyl) propionic acid (I) with benzaldehyde to give 3-benzylidene-5-(2-methyl-1-naphthyl)-2(3H) furanone (II).



The IR spectrum of (II) showed a carbonyl stretching frequency at  $1780\text{ cm}^{-1}$  attributable to  $\gamma$ -lactone and  $\nu_{\text{C}=\text{C}}$  of  $\alpha,\beta$ -unsaturated ketone at  $1605\text{ cm}^{-1}$ . The acid (I) was cyclised by treatment with acetic anhydride to give 5-(2-methyl-1-naphthyl)-2(3H) furanone (III). Condensation of (III) with benzaldehyde in the presence of  $\text{Ac}_2\text{O}$  and  $\text{AcONa}$  gave (II). Hydrazinolysis of (III) in boiling n-butanol gave 3-(2-methyl-1-naphthoyl)propionic acid hydrazide

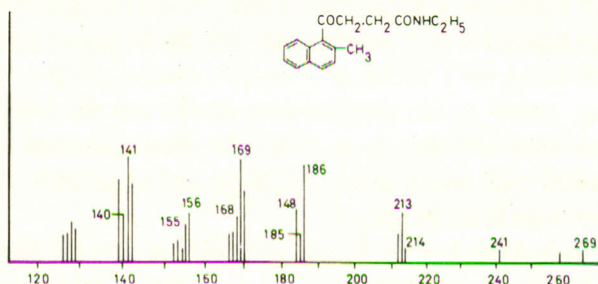
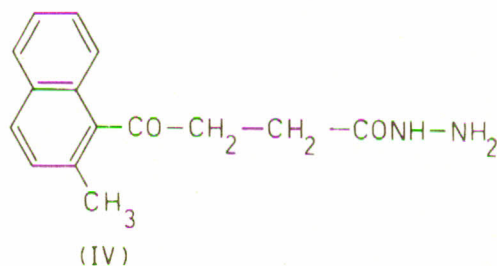
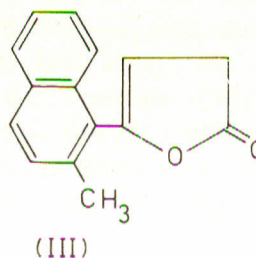


Fig. 1

(IV) and 6-(2-methyl-1-naphthyl)-2,3,4,5-tetrahydropyridazin-3-one (V) [2,4]

The IR spectrum of (IV) showed strong absorption bands at  $1640$  and  $1680\text{ cm}^{-1}$  attributable to  $\nu_{\text{max}}$  of two



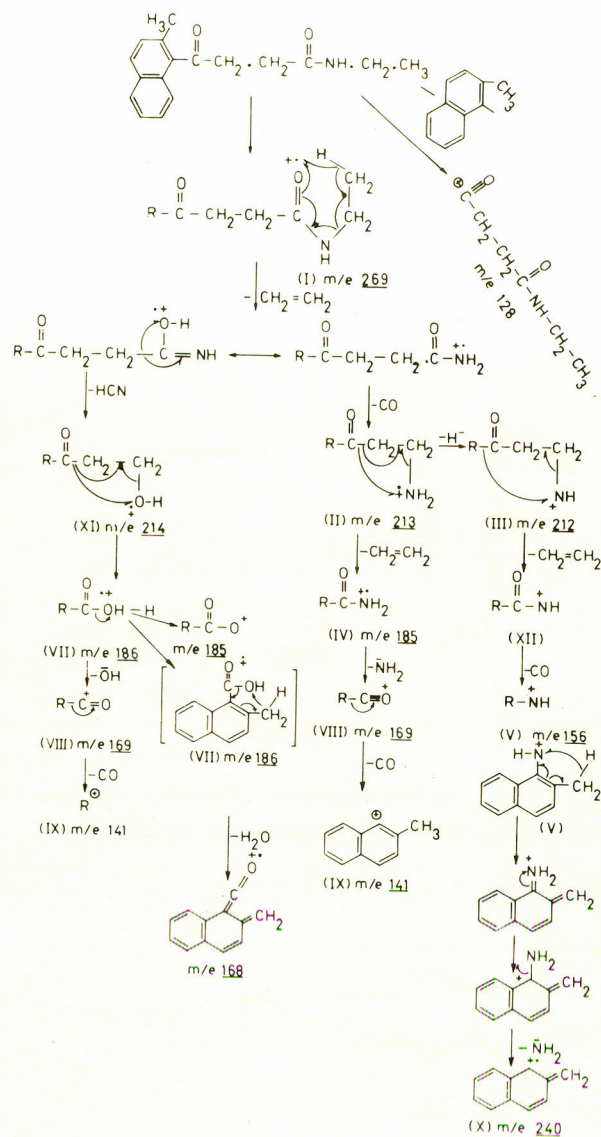
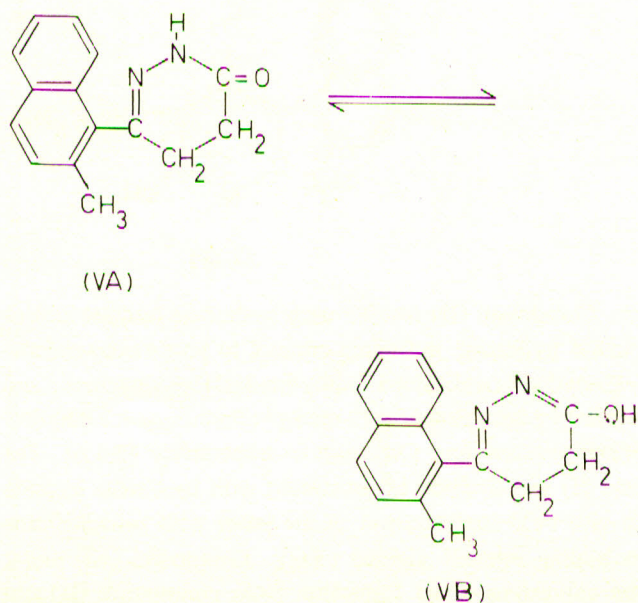
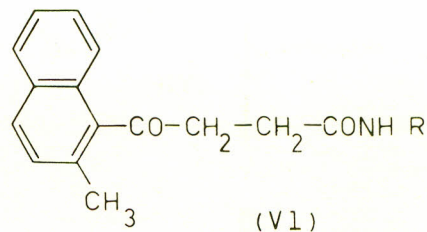


Chart 1



carbonyl groups and a broad band centered at  $3400\text{ cm}^{-1}$  due to  $\nu\text{NH}$ . The IR spectrum of (V) showed  $\nu\text{CO}$  (cyclic amide) at  $1675\text{ cm}^{-1}$ ,  $\nu\text{NH}$  and OH in the range  $3500 - 3200\text{ cm}^{-1}$  and  $\nu_{\text{C}=\text{N}}$  at  $1620\text{ cm}^{-1}$ . Aminolysis of compound (III) by ethylamine and benzylamine yielded *N*-alkyl-3-(2-methyl-1-naphthoyl) propamides (VIa and b) [2,4].



a,  $\text{R} = \text{C}_2\text{H}_5$ ; b,  $\text{R} = \text{CH}_2\text{C}_6\text{H}_5$

Treatment of 3-(2-methyl-1-naphthoyl)propionylchloride with ethylamine and/or benzylamine afforded (VIa and b) respectively. The mass spectrum of compound VIa is depicted in Fig. 1 and explained in Chart 1.

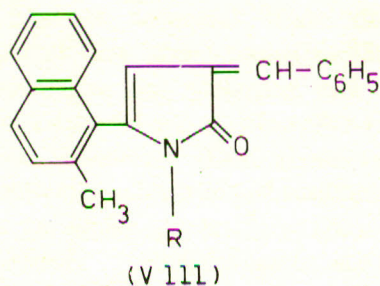
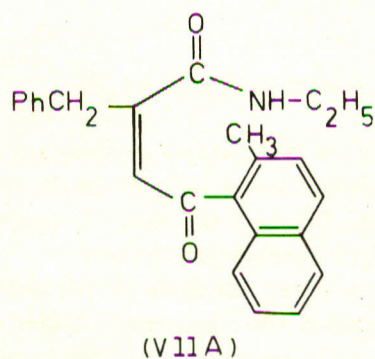
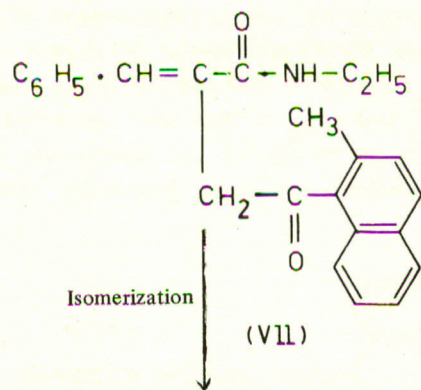
It has been found that furanone (II) underwent ring fission on treatment with ethylamine in boiling ethanol and gave *N*-ethyl-2-benzylidene-3-(2-methyl-1-naphthoyl)propamide (VII) [2,4] which could be isomerised to the thermodynamically stable compound *N*-ethyl-2-benzyl-3-(2-methyl-1-naphthoyl)prop-2-enoic acid (VIIa). Furanone (II) reacted with *p*-toluidine and *o*-chloroaniline in the presence of AcOH/AcONa to give 1-*p*-tolyl-3-benzylidene-5-(2-methyl-1-naphthyl) 2(3H) pyrrolinone and 1-*o*-chlorophenyl-3-benzylidene-5-(2-methyl-1-naphthyl)2(3H) pyrrolinone (VIII a and b) respectively. On the other hand, when the reaction was carried out in the presence of boiling alcohol, the cinnamide derivative (IXa) together with the compound (VIIIc) was obtained [2,5]. Furanone (II) reacted with ammonium acetate in boiling xylene or benzene to give 3-benzylidene-5-(2-methyl-1-naphthyl)2(3H) pyrrolinone (VIIIc) [2,5].

The IR spectrum of (VII) showed strong absorption bands at  $1685, 1640\text{ cm}^{-1}$  due to  $\nu_{\text{max}}$  of two carbonyl grouping and  $\nu\text{NH}$  at  $3200\text{ cm}^{-1}$ . The UV spectrum of compound (VII) in ethanol had  $\lambda_{\text{max}}$  251 nm ( $\epsilon$  13,000),  $\lambda_{\text{max}}$  282 nm ( $\epsilon$  26,000) and  $\lambda_{\text{max}}$  345 nm ( $\epsilon$  5,330) indicated that an extra carbon-carbon double bond must be conjugated with the carbonyl chromophore. (Fig. 2, Chart 2). IR spectrum of (VIII) showed absorption bands at  $1700\text{ cm}^{-1}$  ( $\nu_{\text{C}=\text{O}}$  of unsaturated lactam) and  $1615\text{ cm}^{-1}$  ( $\nu_{\text{C}=\text{O}}$ ).

The IR spectrum of VIIIc showed strong absorption bands at  $3400, 3200\text{ cm}^{-1}$  ( $\nu\text{NH}$  or OH),  $1695\text{ cm}^{-1}$  ( $\nu_{\text{C}=\text{O}}$ ) and  $1615\text{ cm}^{-1}$  ( $\nu_{\text{C}=\text{C}}$ ).

As a point of interest, in this investigation, the hitherto





a, R = C<sub>6</sub>H<sub>4</sub>-CH<sub>3</sub>-p; b, R = C<sub>6</sub>H<sub>4</sub>-Cl-o; c, R = H

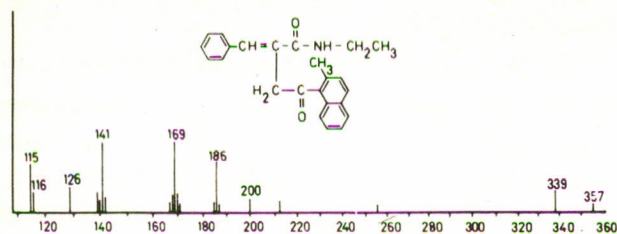
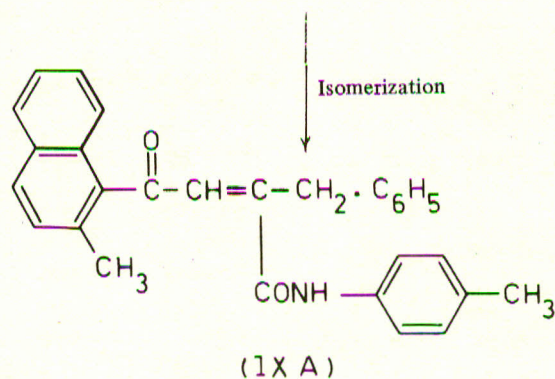
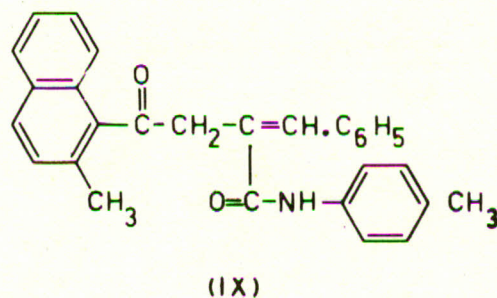


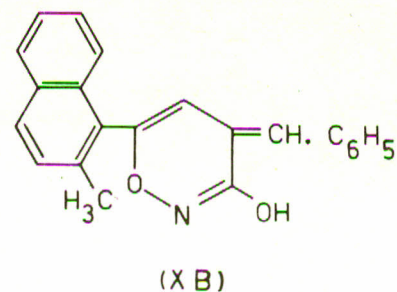
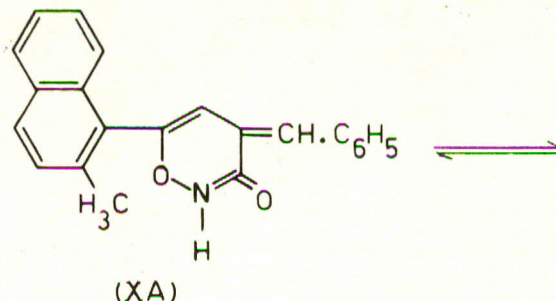
Fig. 2.

unknown reaction of NH<sub>2</sub>-OH.HCl with compound (II) was discovered. Compound (II) reacted with H<sub>2</sub>NOH.HCl in boiling pyridine and yielded 4-benzylidene-6-(2-methyl-1-naphthyl)2,4-dihydro-1,2-oxazin-3-one (X).

The IR spectrum of (X) showed strong absorption bands at 1680, 1620 and 3350 cm<sup>-1</sup> attributable to ν<sub>C=O</sub>,



ν<sub>C=N</sub> and ν<sub>OH</sub> or NH respectively. Thus the oxazinone (X) must exist in lactam-lactim tautomeric equilibrium.



Compound (II) reacted with hydrazine hydrate and/or phenyl hydrazine in boiling ethanol to give 4-benzylidene-6-(2-methyl-1-naphthyl)4,5-dihydro-3(2H)-pyridazinone and 2-phenyl-4-benzylidene-6-(2-methyl-1-naphthyl)4,5-dihydro-3(2H)-pyridazinone (XIa and b) respectively [2,4,6]. The acid (I) was cyclised by treatment with hydrazine hydrate to give (V), condensation of the latter with benzaldehyde in boiling ethanol yielded (XIa). Compound (XI) under the conditions of its formation from compounds (II) and



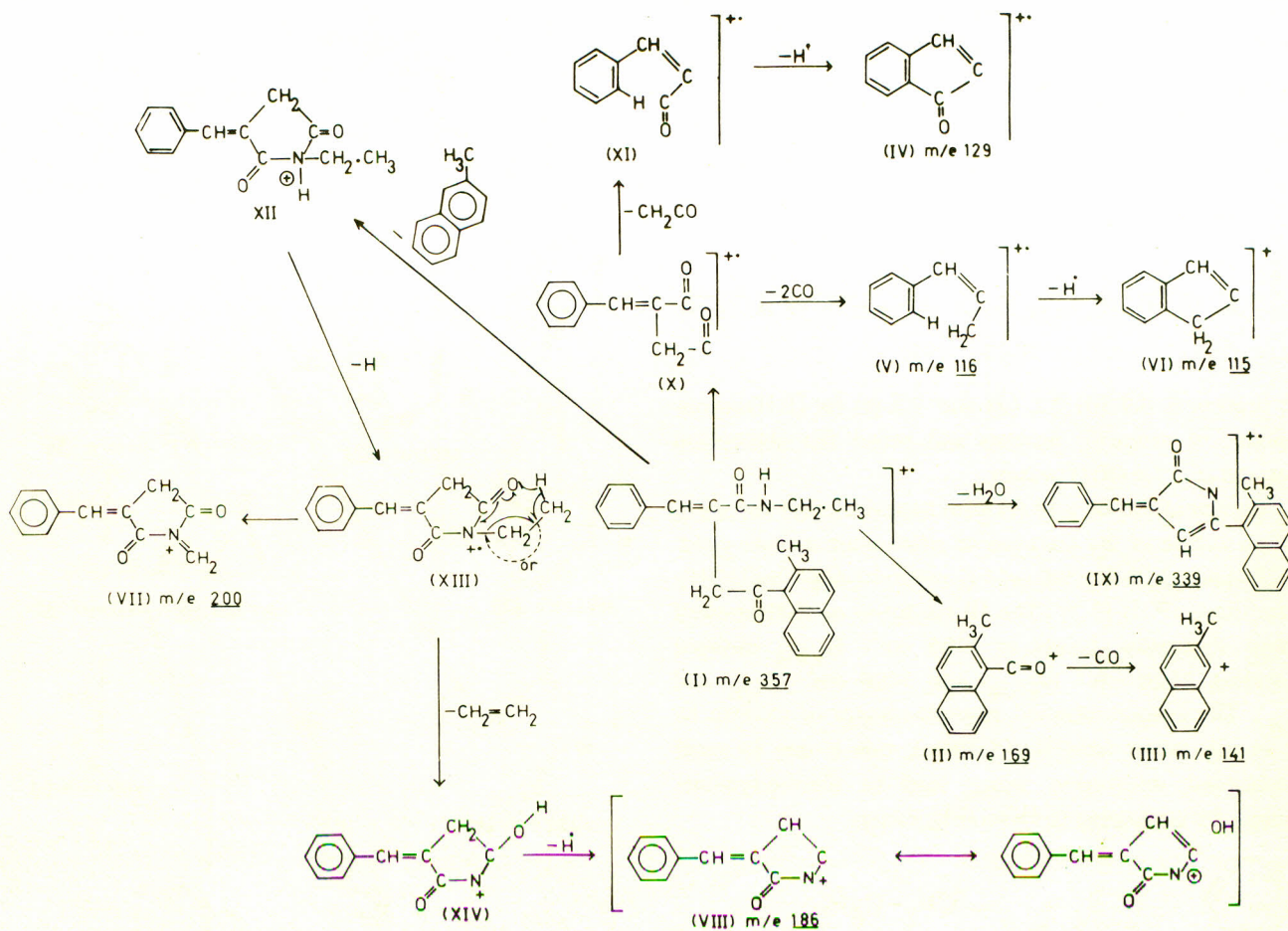
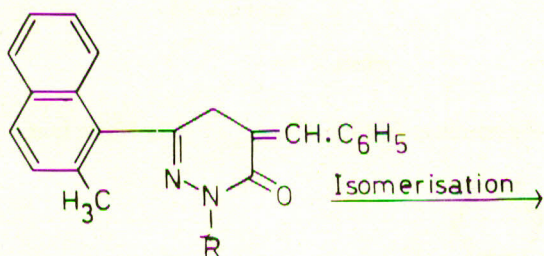
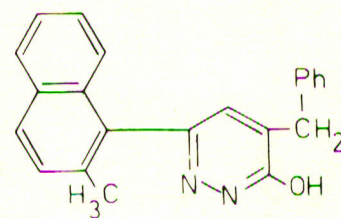
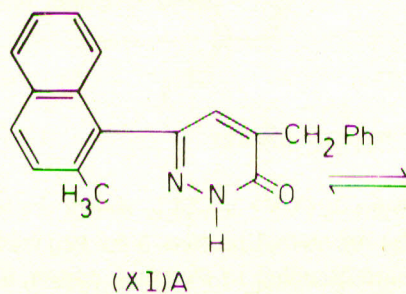


Chart 2

(V) could be isomerised to the thermodynamically more stable 3-benzyl-5-(2-methyl-1-naphthyl)3(2H)pyridazinone (XIa) which has an aromatic tautomer (XIb).



(X1)  
a, R = H; b, R = C<sub>6</sub>H<sub>5</sub>



(X1)B

The IR spectrum of (XIa) showed the carbonyl stretching frequency of cyclic amides at 1705 cm<sup>-1</sup>, the NH and OH stretching frequency at 3200 and 3400 cm<sup>-1</sup> and C=N or ν<sub>C=C</sub> at 1630 cm<sup>-1</sup>. The UV spectrum of (XIa) in ethanol had λ<sub>max</sub> 252 nm (ε 2,300), λ<sub>max</sub> 281 nm (ε 4,880), λ<sub>max</sub> 315 nm (ε 730), λ<sub>max</sub> 255 nm (ε 870), which is consistent with the proposed structure (the two bands at λ<sub>max</sub> 252 and 355 nm, evidently stand for pyridazine nucleus).

The NMR spectrum of XIa in CDCl<sub>3</sub> shows the following signals, at δ 2.1 (s) for (3H) corresponding to CH<sub>3</sub>, δ 3.2 (s) (2H) corresponding to CH<sub>2</sub> protons, δ 6.2 (s) olefi-



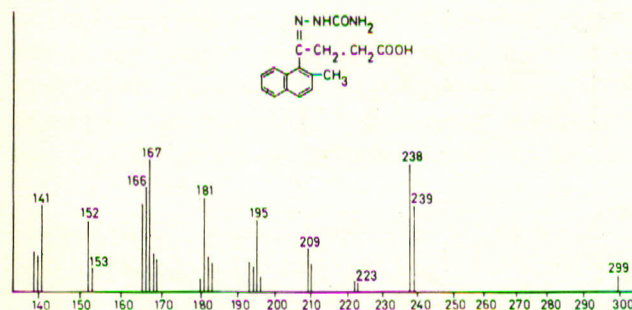
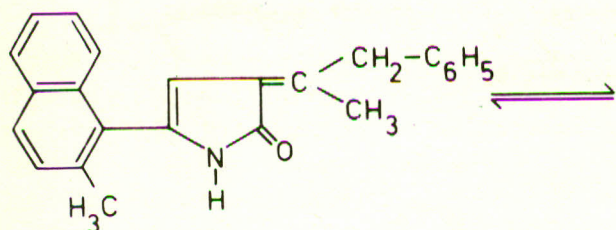


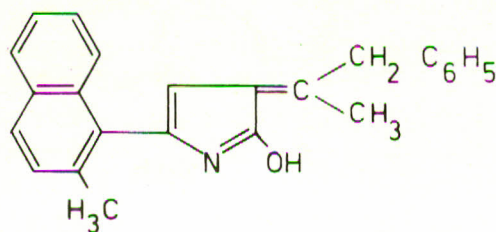
Fig. 3

nic proton,  $\delta$  6.9 (d), 7.1 (d) and 7.3 (s) for (1H) corresponding to aromatic protons and broad flat absorption centered at  $\delta$  7.9 (NH) proton.

Condensation of acid (I) with benzyl methyl ketone in boiling xylene in the presence of ammonium acetate yielded 3-(1-phenyl-2-propylidene)-5-(2-methyl-1-naphthyl)-2(3H)-pyrrolinone (XII) [1,2]. The IR spectrum of (XII) showed strong absorption bands at  $1705\text{ cm}^{-1}$  ( $\nu_{\text{C=O}}$  unfused  $\gamma$ -lactam),  $1620\text{ cm}^{-1}$  ( $\nu_{\text{C=N}}$ ) and  $3400\text{ cm}^{-1}$  ( $\nu_{\text{OH}}$  and  $\nu_{\text{NH}}$ ). The authors claimed stereochemistry of (XII) to be (z) configuration (*cis*- $\text{CH}_3/\text{CONH}$ -), this is due to small non-bonded interaction which lead to thermodynamic stability by comparing to (E)-configuration.



(XII)A



(XII)B

Treatment of acid (I) with bromine and  $\text{POCl}_3$  in chloroform gave 3-(2-methyl-1-naphthoyl)acrylic acid (XIII). The IR spectrum of the acid (XIII) showed strong absorption bands at  $1685$  and  $1670\text{ cm}^{-1}$  attributable to  $\nu_{\text{max}}$  of two carbonyl groups. The reaction of acid (I) with  $\text{NH}_2\text{OH}\cdot\text{HCl}$  in boiling pyridine [7], yielded 3-(2-methyl-1-naphthyl)-2,3-dihydro-1,2-oxazin-6-one (XIV). The IR spectrum of (XIV) showed strong absorption bands at  $1685\text{ cm}^{-1}$  ( $\nu_{\text{C=O}}$  of six-membered lactone unsaturated in

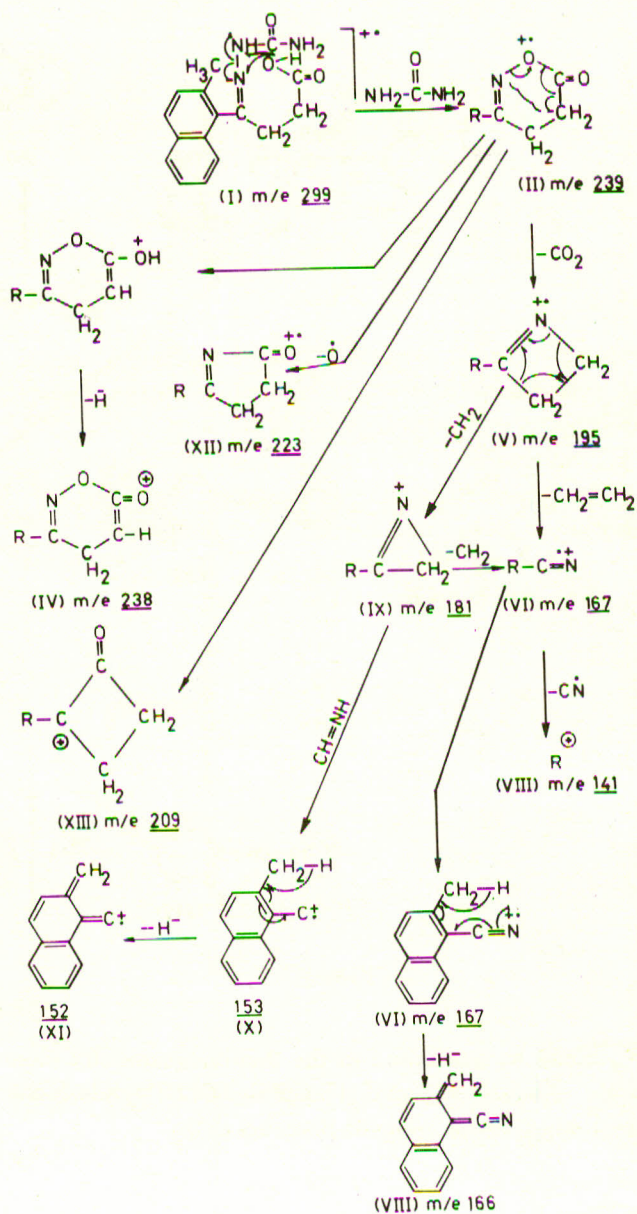
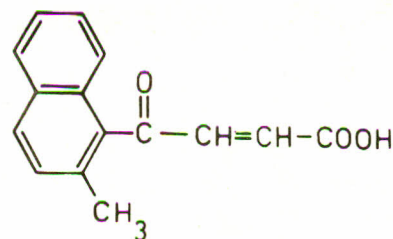


Chart 3.

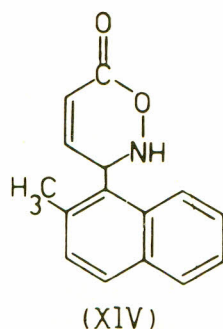
the  $\alpha$  and  $\beta$ -position),  $1620\text{ cm}^{-1}$  ( $\nu_{\text{C=C}}$ ). The NMR spec-



(XIII)

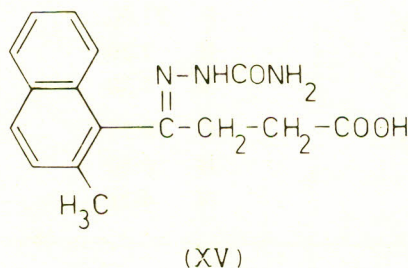
trum of (XIV) in  $\text{CDCl}_3$  shown the following signals:  $\delta$  2.2 (s) for methyl protons,  $\delta$  4.3 (m) methine proton,  $\delta$  5.9 (d) corresponding to  $\beta$ -olefinic proton,  $\delta$  6.2 (d) for  $\alpha$ -olefinic





proton, broadened absorption at  $\delta$  6.5 for NH-proton and two doublets at 7 and 7.2 corresponding to 6H (aromatic), this is agreed well with the proposed structure.

Treatment of the acid (I) with semicarbazide in boiling pyridine yielded the semicarbazone derivative (XV). The IR spectrum of (XV) showed bands at 1720, 1640, 1620 and 3200  $\text{cm}^{-1}$  attributable to  $\nu_{\text{C}=\text{O}}$  (acid),  $\nu_{\text{C}=\text{O}}$  (amide),  $\nu_{\text{C}=\text{N}}$  and  $\nu_{\text{NH}}$  respectively (Fig. 3, Chart 3).



## EXPERIMENTAL

The IR spectra in KBr were recorded on a Unicam SP 1200 spectrophotometer. UV spectra were recorded on a Unicam SP 8000 UV recording spectrophotometer. NMR spectra were recorded on a Varian Chart S-60T spectrophotometer. All m.ps. reported are uncorrected.

**Synthesis of 3-(2-Methyl-1-naphthoyl) Propionic Acid (I).** To a mixture of 10 g (0.1 mole) succinic anhydride, 42g anhydrous aluminium chloride and 40 ml acetylene tetrachloride, a solution of 14.2 g (0.1 mole) of 2-methylnaphthalene in 30 ml acetylene tetrachloride was added gradually. After stirring for 20 hr, the reaction mixture was allowed to stand overnight, the reaction mixture was then decomposed (dil HCl) and the solvent was removed by steam distillation. Then oil obtained washed several times with water, then crystallized from benzene giving (I) as a white crystalline solid (Table 1).

**Synthesis of 3-Benzylidene-5-(2-methyl-1-naphthyl)-2(3H)furanone (II):** A mixture of 5.3 g (0.05 mole) benzaldehyde, 12.1 g (0.05 mole) acid (I), 4.6 g (0.05 mole) of freshly fused  $\text{CH}_3\text{COONa}$  and 16 ml of  $\text{Ac}_2\text{O}$  was heated on hot plate until complete solution was obtained. The

reaction mixture was then transferred to a steam-bath and heating was continued until crystals separated. The reaction mixture was then poured into water, the solid product obtained was filtered off, washed with water and finally recrystallised from ethanol giving (II) as a yellow solid (Table 1).

**Synthesis of 5-(2-Methyl-1-naphthyl)-2(3H)furanone (III).** A mixture of 12.1 g (0.05 mole) of acid (I), 4.6 g (0.05 mole) of freshly fused sodium acetate and 16 ml of  $\text{Ac}_2\text{O}$  was heated on a hot plate until complete solution was obtained, then transferred to a steam bath and heating was continued until crystals separated. The reaction mixture worked up as in case of (II), the solid obtained recrystallised from benzene giving (III) as a pale brown solid (Table 1).

**Reaction of Furanone (III) with Hydrazine:** Formation of 3-(2-Methyl-1-naphthoyl)propionic Acid Hydrazide (IV) and 6-(2-Methyl-1-naphthyl)2,3,4,5-tetrahydropyridazine-3-one (V). A mixture of (III) (0.01 mole) and hydrazine (0.02 mole) was refluxed in n-butanol for 8 hr. After evaporating most of the solvent, the solid fractionally crystallized from boiling ethanol, and on recrystallization from ethanol (V) was obtained as a colourless crystals (Table 1).

The ethanol-insoluble part was crystallized from n-butanol to give (IV) as brown crystals (Table 1).

**Reaction of Furanone (III) with Ethyl and/or Benzylamine:** Formation of *N*-alkyl-3-(2-methyl-1-naphthoyl)propamide (VIa and b). A solution of (III) (0.01 mole) and primary amine namely, ethylamine and benzylamine (0.03 mole) in 50 ml ethanol was heated under reflux for 6 hr. The product that separated on cooling was crystallized from benzene-ethanol to give *N*-alkyl-3-(2-methyl-1-naphthoyl)propamide (VIa and VIb), (Table 1).

**Reaction of 3-(2-Methyl-1-naphthoyl)propionyl chloride with Ethyl and Benzyl Amines:** Formation of VIa and VIb. An ethereal solution of the acid chloride of (I) was added to an ethereal solution of ethylamine or benzylamine and then heated on a steam bath for 1 hr, evaporated most of the ether layer and poured into water, the solid separated was crystallized from benzene-ethanol and was identified to be VIa and VIb by m.p. and m.m.p. determinations.

**Reaction of 3-Benzylidene-5-(2-methyl-1-naphthyl)-2(3H) Furanone (II) with Ethyl Amine:** Formation of (VII). A solution of (II) (0.01 mole) and ethyl amine (0.03 mole) in 50 ml ethanol was heated under reflux for 6 hr. The product that separated on cooling was crystallized from suitable solvent giving (VII) as a pale yellow solid (Table 2).

**Reaction of Furanone (II) with *p*-Toluidine and/or *o*-Chloroaniline in  $\text{AcOH}/\text{AcONa}$ :** Formation of 1-*p*-Tolyl-



Table 1. Physical data of compounds (I–IV).

Compound	M.p. (°C)	Yield (%)	Formula	Analysis (%)		
				Found	Required	
I	168	70	$C_{15}H_{14}O_3$	C	74.52	74.4
				H	6.90	7.10
II	185	75	$C_{22}H_{17}O_2$	C	87.24	87.13
				H	5.57	5.60
III	211	80	$C_{15}H_{13}O_2$	C	79.80	80.00
				H	5.90	5.80
IV	252	30	$C_{15}H_{16}N_2O_2$	C	70.21	70.13
				H	6.32	6.25
				N	10.72	10.93
V	215	40	$C_{15}H_{14}N_2O$	C	75.40	75.60
				H	5.80	5.90
				N	12.00	11.80
VIa	250	80	$C_{17}H_{19}NO_2$	C	75.60	75.80
				H	7.20	7.06
				N	5.30	5.20
VIb	260	85	$C_{22}H_{21}NO_2$	C	79.73	79.76
				H	6.20	6.10
				N	4.21	4.22

Table 2. Characteristic of compounds (VII–XI).

Compound	M.p. (°C)	Solvent yield (%)	Formula	Analysis (%)		
				Found	Required	
VII	122	B/P 60	$C_{24}H_{23}NO_2$	C	80.50	80.70
				H	6.30	6.40
				N	3.80	3.90
VIIIa	127	B/P 70	$C_{29}H_{23}NO$	C	86.50	86.80
				H	5.90	5.70
				N	3.50	3.40
VIIIb	210	E 80	$C_{28}H_{20}ClNO$	C	79.62	79.71
				H	4.65	4.74
				N	3.42	3.32
VIIIc	210	B 70	$C_{22}H_{17}NO$	C	84.70	84.90
				H	5.60	5.50
				N	4.60	4.50
IX	218	E 50	$C_{29}H_{25}NO_2$	C	83.10	83.05
				H	5.89	5.96
				N	3.25	3.34
X	205	T 60	$C_{22}H_{17}NO_2$	C	80.72	80.73
				H	5.20	5.20
				N	4.29	4.28



(Table 2 contd.)

XIa	155	B/P 65	$C_{22}H_{18}N_2O$	C	81.00	81.00
				H	5.60	5.52
				N	8.50	8.60
XIb	130	B 60	$C_{28}H_{22}N_2O$	C	83.28	83.58
				H	5.50	5.47
				N	6.50	6.97

B benzene, B/P benzene–light petrol (60–80°), E ethanol, T toluene.

Table 3. Characterisation of compounds (XII–XV).

Compound	M.p. (°C)	Yield (%)	Formula	Analysis (%)		
				Found	Required	
XII	130	70	$C_{24}H_{21}NO$	C	84.86	84.96
				H	6.23	6.16
				N	4.21	4.13
XIII	135	60	$C_{15}H_{12}O_3$	C	74.90	75.00
				H	5.10	5.00
XIV	173	65	$C_{15}H_{13}NO_2$	C	74.95	75.31
				H	5.51	5.44
				N	5.85	5.86
XV	212	65	$C_{16}N_{17}N_3O_3$	C	64.32	64.20
				H	6.00	5.70
				N	14.32	14.04

3-benzylidene-5-(2-methyl-1-naphthyl) 2(3H)pyrrolinone and 1-*o*-Chlorophenyl-3-benzylidene-5-(2-methyl-1-naphthyl)2(3H)pyrrolinone (VIIIa and VIIIb). A mixture of (II) (0.01 mole), *p*-toluidine or *o*-chloroaniline (0.01 mole) and sodium acetate (0.02 mole) in 50 ml acetic acid was heated under reflux for 10 hr. The mixture was poured into ice-water and the solid separated was crystallized from the proper solvent to give (VIIIa and VIIIb) (Table 2).

**Reaction of Furanone (II) with *p*-Toluidine in Alcohol:** Formation of (VIIIa) and (IX). A solution of (II) (0.01 mole) and *p*-toluidine (0.03 mole) in 60 ml ethanol was heated under reflux for 10 hr. After evaporating most of solvent, the solid separated fractionally crystallized from boiling ethanol, and on recrystallization from ethanol or benzene (IX) was obtained as an orange crystals (Table 2).

The ethanol-insoluble part was crystallized from benzene–light petrol (60–80°) and was identified to be (VIIIa) by m.p. and m.m.p. determination, yield 25%.

**Reaction of Ammonium Acetate with (II):** Formation of 3-Benzylidene-5-(2-methyl-1-naphthyl)2(3H)pyrrolinone (VIIIc). A solution of (II) (0.01 mole) in benzene or xylene

(40 ml) was treated with ammonium acetate (10 g). The reaction mixture was refluxed for 8 hr. Cooled and poured into ice. The solid residue was crystallized from suitable solvent giving (VIIIc) as a pale green solid (Table 2).

**Reaction of Hydroxylamine Hydrochloride with (II):** Formation of 4-Benzylidene-6-(2-methyl-1-naphthyl)2,4-dihydro-1,2-oxazin-3-one (X). A mixture of (II) (0.01 mole) and of hydroxylamine hydrochloride in pyridine (0.02 mole, 100 ml) was heated under reflux for 8 hr. The cooled reaction mixture was acidified with ice-cold dil HCl. The residue that separated was crystallized from the proper solvent to give (X) as grey crystals (Table 2).

**Reactions of Hydrazines with (II):** Formation of (XIa and b). A solution of (II) (0.01 mole) in ethanol was treated with hydrazine hydrate or phenyl hydrazine (0.01 mole). The reaction mixture was refluxed for 4 hr. The solid separated after concentrating the solvent was crystallized from proper solvent and gave XI and b respectively (Table 2).

**Action of Benzaldehyde on (V):** Formation of (XIa). To a mixture of (V) (0.01 mole) and benzaldehyde (0.01



mole) in presence of few drops of piperidine was added 150 ml ethanol. The reaction mixture was heated under reflux for 7 hr. The solid that separated upon concentrating the reaction mixture was filtered off and crystallized from benzene – light petrol (60–80°) and was identified to be (XIa) by m.p. and m.m.p. determination, yield 60%.

*Synthesis of 3-(1-Phenyl-2-propylidene)-5-(2-methyl-1-naphthyl)2(3H)pyrrolinone.* A mixture of acid (I) (0.01 mole), ammonium acetate (3g, 0.04 mole) and benzyl methyl ketone (0.01 mole) was refluxed in acetic anhydride or xylene (40 ml) for 12 hr. The reaction mixture was poured into ice; the solid separated after treatment with light petrol was crystallized from benzene giving (XII) as a green solid (Table 3).

*Dehydrogenation of (I):* Formation of 3-(2-Methyl-1-naphthoyl) Acrylic Acid (XIII). A (1M) solution of bromine in chloroform (20 ml) was added to a cold stirred solution of (0.01 mole) of (I) in (20 ml) chloroform and (2 ml) POCl<sub>3</sub> during 15 min. The reaction mixture was warmed on a steam bath until evolution of hydrogen bromide had ceased (about 2 hr). The reaction mixture was concentrated by evaporation, then poured upon ice-cold water. The solid that separated was filtered, dried and crystallized from benzene – light petrol (60–80°) to give (XIII) as orange crystals (Table 3).

*Reaction of (I) with Hydroxylamine Hydrochloride and*

*Semicarbazide:* Formation of 3-(2-Methyl-1-naphthyl)2,3-Dihydro-2-Oxazin-6-one (XIV) and 3-(2-Methyl-1-naphthoyl) Propionic Acid Semicarbazone (XV). A mixture of (I) (0.01 mole) and hydroxylamine hydrochloride or semicarbazide (0.02 mole) in pyridine (80 ml) was heated under reflux for 6 hr. The cooled reaction mixture was poured upon ice then acidified with dil HCl. The solid that separated was crystallized from benzene and methanol to give XIV and XV respectively (Table 3).

#### REFERENCES

1. M. Abdalla and A. Essawy, *Indian. J. Chem.*, **15B**, 523 (1977).
2. A. Sammour, M. Abdalla and A. Essawy, *Pakistan J. Sci. Res.*, **26** (1974).
3. A. Sammour, A.F.M. Fahmy, M. Abdalla and A.A. Hamed; *Egyptian J. Chem.*, **18**, 6 (1975).
4. A. Essawy and M. Abdalla, **7th International Congress on Heterocyclic Chemistry**, Chem. Dept. University of South Florida, Tampa, Florida, 13 August (1979).
5. A. Essawy and A. Hamed, *Indian. J. Chem.*, **15B** 694 (1977).
6. Campell and Khanna, *J. Chem. Soc.*, 338 (1949).
7. A. Sammour and M.A. El Hashash, *J. Prakt. Chem.*, 314906 (1972).