Tetrahedron Vol 39, No. 13, pp. 2231 to 2235, 1983
Printed in Great Britain.

0040-4020/83 $03.00 + 00
Pergamon Press Ltd

ADDITION OF NITRILE OXIDES TO OLEFINS. SYNTHESIS OF DIHYDROJASMONE AND STARTING MATERIAL FOR PROSTANOIDS. A NOVEL ROUTE TO PYRROLES

S. K. MUKERJI,† K. K. SHARMA‡ and K. B. G. TORSSELL*
Chemical Institute, Department of Organic Chemistry, University of Aarhus, 8000 Aarhus C, Denmark

(*Received in the UK 25 June* 1982)

Abstract—Routes to dihydrojasmone and γ-keto-aldehydes and an alternative procedure for a key intermediate in our prostaglandin synthesis are described. 2-Isoxazolines are useful intermediates for a preparation of a variety of classes of compounds. A novel route to pyrroles is established.

In connection with our studies of prostanoid synthesis it was of interest to prepare 5-formyl-2-isoxazolines (**1**) which conceivably could give 2-hydroxy-4-keto-aldehydes (**3**) on reduction with Ti^{3+}, and subsequently 4-hydroxy-cyclopentenones (**4**) on base catalyzed cyclization, eqn (1), in analogy with the reactions of other 5-acyl-2-isoxazolines.[1] Contrary to expectation, **1** could not be obtained by dipolar addition of silyl nitronates to acrolein, nor was **1** obtained by addition of nitrile oxides prepared *in situ* to acrolein. Whilst the addition of silyl nitronates to acrolein diethyl acetal proceeded in poor yield the nitrile oxides reacted in good yields to form 5-diethoxymethyl-2-isoxazolines (**2a–e**) (eqn 2) **2f** was obtained in *ca* 40% yield from the silyl ester of nitromethane and acrolein diethyl acetal after 8 days. This is in agreement with our earlier experience with the reactivities of silyl nitronates and nitrile oxides. Thus, the nitrile oxides react with unactivated double bonds, such as mono alkyl and certain dialkyl substituted olefins,[2] whereas silyl nitronates react sluggishly or not at all.[3]

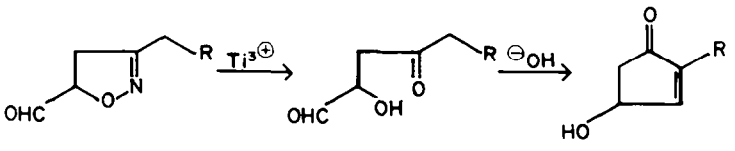
Only one formyl compound of type **1**, 3-phenyl-5-formyl-2-isoxazoline,[4] but several 5-acyl-2-isoxazolines, obtained from the reaction of vinyl ketones with silyl nitronates[1,5] or nitrile oxides,[6–8] are known. A facile cleavage according to eqn (3)[7] could explain the instability of the 5-formyl derivatives.

The present paper deals with the synthesis and some reactions of 5-diethoxymethyl-2-isoxazolines (**2**), and the synthesis of a few other substituted 2-isoxazolines by the nitrile oxide route, of particular interest for syntheses of dihydrojasmone and prostaglandins.[9] Finally a novel route to pyrroles is worked out.

Reactions of the 2-isoxazoline acetals, **2.** *Synthesis of γ-keto aldehydes.* The isoxazolines **2** (Table 1) were synthesized in 26–78% yield by 1,3-dipolar addition of acrolein diethylacetal to nitrile oxides, generated *in situ* from hydroxamic acid chlorides and triethylamine.[2,10,11] The hydrolysis of the acetals into aldehydes proceeded poorly under a variety of conditions. Small amounts, *ca* 10%, of the corresponding formyl derivatives were detected in the product mixture by ^1H NMR spectroscopy. When the acid catalyzed hydrolysis of **2a** was carried out in the presence of *o*-phenylenediamine, the quinoxaline derivative **7** was obtained, indicating that the aldehydes are cleaved easily also under acid conditions according to eqn (3).

The planned reaction (eqn 1) was therefore abandoned and reduction of **2b** and **2d**, directly with Ti^{3+} in a weakly acidic solution, was attempted. Contrary to expectation, no hydroxy derivate of type **3** was formed. The main products were the γ-keto aldehydes **5b** and **5d** (*ca* 30%). The base catalyzed cyclization of **5d** gave in our hands poor yields (see Ref. 12) of **6** but 71% yield has been reported in the literature for this step in a synthesis of dihydrojasmone (**11b**).[13] Reaction, eqn (2), may be of some interest in connection with the preparation of γ-keto aldehydes for which there exists only a limited synthetic methodology. An alternative route to dihydrojasmone is described in the next section.

Synthesis of dihydrojasmone, **11b**. The facile addition of nitrile oxides to vinyl ketones was confirmed and substantiated by the preparation of **8a** and **8b**.

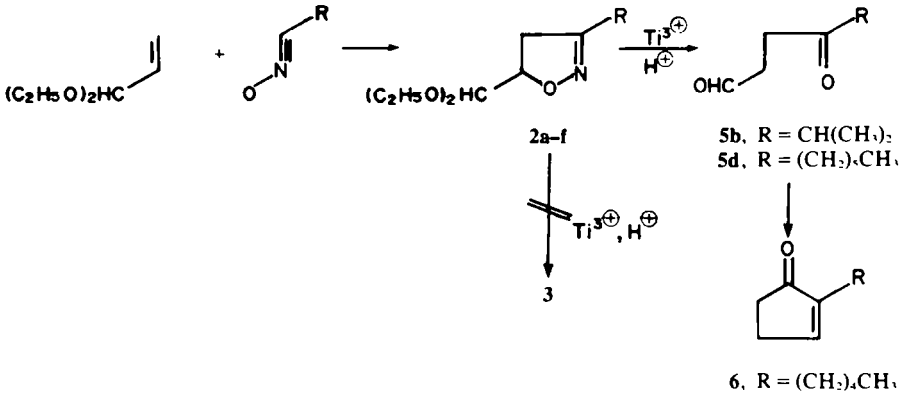


(1)

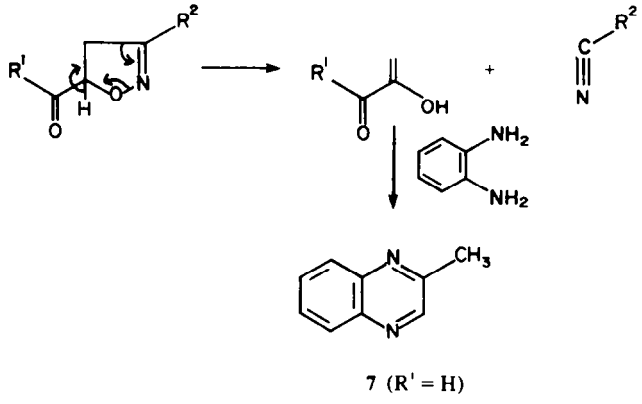
1 3 4

†Present address: The University of Rajastan, Jaipur, India.

‡Present address: Arun Chemicals and Dyes, New Delhi 110020, India.



(2)



(3)

7 (R' = H)

These were reduced by Ti^{3+} to the corresponding alka 1,4-dion-2-ols (**9a,b**) which were cyclized to the 3(2H)-furanones (**10a,b**) by heating with acetic acid and sodium acetate.[1,14] **9a** was cyclized to **11a** by treatment with aqueous sodium hydroxide.

As noted earlier,[1] the presence of metallic zinc at the Ti^{3+} reduction of 2-isoxazolines causes fission of the C–O bond. When this reduction was applied to **8a**, **12** was obtained which on cyclization gave dihydrojasmone, **11b**, and a novel, facile route to this compound can now be added to the many others previously published.[15,16]

Table 1. Isoxazolidines (**2**) from acrolein diethyl acetal and nitrile oxides

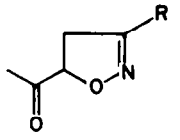
	R	Yield	bp.°C mmHg	^{1}H NMR values, δ, J Hz ($CDCl_3$)
2a	CH_3	26	68-76/ 0.05	1.18 (3H,t,J7), 1.21 (3H,t, J7), 1.97 (3H,br.s), 2.94 (2H,d,J8), 3.2-3.7 (4H,m), 4.1-4.6 (2H,m)
2b	$(CH_3)_2CH$	39	82-90/ 0.1-0.15	1.16 (6H,d,J6.6), 1.20 (3H,t,J7), 1.23 (3H,t, J7), 2.5-3.1 (3H,m), 3.4-3.9 (4H,m), 4.3-4.6 (2H, m)
2c	$(CH_2)_3CH_3$	41	120/0.1	0.93 (3H,t,J6), 1.0-2.0 (10H,m), 2.35 (2H,br.t,J8), 2.96 (2H,d,J7.6), 3.4-3.9 (4H,m), 4.2-4.7 (2H,m)
2d	$(CH_2)_5CH_3$	78	132/ 0.02	0.87 (3H,t,J6), 1.0-2.0 (14H,m), 2.35 (2H,br.t,J8), 2.94 (2H,d,J7.6), 3.4-3.9 (4H,m), 4.2-4.7 (2H,m)
2e	C_6H_5	55	140-150/ 0.15-0.20	1.13 (3H,t,J7), 1.19 (3H,t, J7), 3.1-3.9 (6H,m), 4.4-4.8 (2H,m), 7.2-7.8 (5H,m)
2f	H	38	68/0.2	1.19 (3H,t,J7), 1.23 (3H,t J7), 3.02 (2H,d,J8), 3.3-3.9 (4H,m), 4.3-4.7 (2H,m), 7.10 (1H,t,J1)

Synthesis of 3 - (ω - *methoxycarbonylheptyl*) - 5 - (3 - *methyl* - 2 - *butenoyl*) - 2 - *isoxazoline*, (**15**). This compound was of interest to us as a starting material for our prostaglandin synthesis.[1,9] The lower homologue, the 3 - (ω - methoxycarbonylhexyl) - 5 - (3 - methyl - 2 - butenoyl) - 2 - isoxazoline was prepared by the silyl nitronate procedure, but we found it more convenient to prepare **15** by the alternative less expensive route, eqn (4). By standard procedures, oleic acid was oxidized,[17] and cleaved[18] to methyl 9-oxo-nonanoate (**13**). Oximation and chlorination gave (**14**) which finally was converted *in situ* into the nitrile oxide and added to 3-methyl-1-propenyl vinyl ketone to yield **15**. The lower homologue has already been converted into a prostanoid, and by having access to **15** a route to PGE_1 is opened.[1,9]

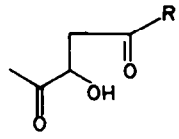
Syntheses of pyrroles. The nitrile oxides, generated *in situ*, add to allyl acetate and allyl acetamide giving the functionalized 2-isoxazolines **16a–c**. They are reduced by Ti^{3+} to the corresponding β-hydroxy ketones **17a–c**. These reactions are of interest in connection with synthesis of polyols and amino alcohols. **17b,c** can be cyclized to **18** and **19** by heating in acetic acid with sodium acetate as catalyst, which constitutes a novel pyrrole synthesis (eqn 5).

EXPERIMENTAL

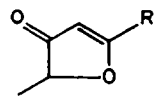
5-Diethoxymethyl-2-isoxazoline, **2f** was obtained in 30–40% yield from a equimolecular mixture of nitromethane, triethylamine, chlorotrimethylsilane and acrolein diethyl acetal in benzene:acetonitrile, 2:1. The solution was stirred at 25° for 10 days, filtered, washed with water, and



8a,b

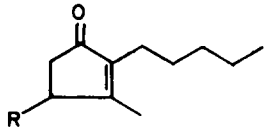


9a,b

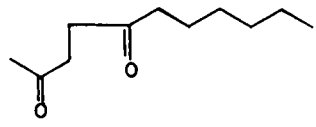


10a,b

a, R = $(CH_2)_4CH_3$ b, R = $(CH_3)_2CH$

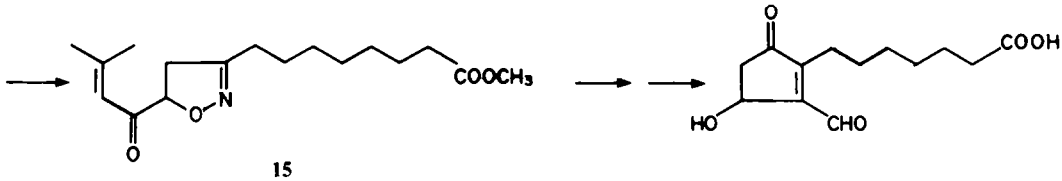


11a R = OH
11b R = H, Dihydrojasmone



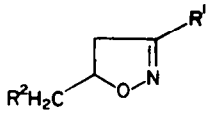
12

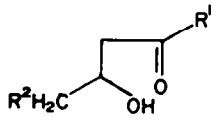
$$\text{Oleic acid} \xrightarrow{a,b,c} \underset{\textbf{13}}{OHC(CH_2)_7COOCH_3} \xrightarrow{d,e} \underset{\textbf{14}}{HON{=}CCl(CH_2)_7COOCH_3} \xrightarrow{f} \quad (4)$$



15

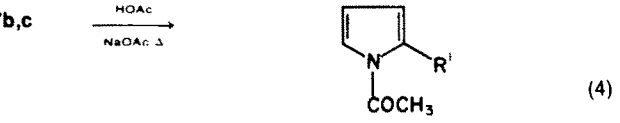
a. H_2O_2, H^+, b. CH_3OH, H^+, c. $Pb(OAc)_4$, d. NH_2OH, e. Cl_2, f. NEt_3, 3-methyl-1-propenyl vinyl ketone.





16a R' = $(CH_3)_2CH$, R^2 = OAc
16b R' = $(CH_3)_2CH$, R^2 = NHAc
16c R' = $CH_3(CH_2)_4$, R^2 = NHAc

17a R' = $(CH_3)_2CH$, R^2 = OAc
17b R' = $(CH_3)_2CH$, R^2 = NHAc
17c R' = $CH_3(CH_2)_4$, R^2 = NHAc



(4)

18 $R^1 = (CH_3)_2CH$
19 $R^1 = CH_3(CH_2)_4$

dried over sodium sulfate. Distillation gives **2f**, b.p.$_{0.1}$ 60–63°. MS ($M^+ + 1$) 174.

General procedure for preparation of 2-isoxazolines from aldoximes and alkenes. Cl_2 was passed through a soln of the aldoxime (0.03 mole) in chloroform (30 ml) at − 30–35°. The solution turned blue but after a while the blue colour faded and shifted to green (marking the end point of the chlorination). Excess Cl_2 was purged by N_2. ^1H NMR showed characteristic peaks from the CH–N=OH group at around $\delta = 7.0$ were no longer present. The hydroxamic acid chloride formed was added dropwise to a mixture of the olefin (0.04 mole) and triethylamine (0.04 mole) in chloroform (10 ml) at 10–20°. After 30 min the solution was washed with water, dried (Na_2SO_4), evaporated, and the remaining 2-isoxazoline was distilled *in vacuo* or recrystallized. The crude 2-isoxazolines are often sufficiently pure for direct further use. This procedure was used for the acetal derivatives, **2a–e**, Table 1, for the acyl derivatives **8a,b**, **15**, and the functionalized derivatives **16a–c**.

Synthesis of the γ-ketoaldehydes **5b** *and* **5d**. The pH of the aqueous acid Ti^{3+} solution (10 ml, 1 M) was adjusted to *ca* 2.0–2.5. Acetic acid (30 ml), water (20 ml), and 0.004 mole of **2b** or **2d** is added. The mixture was stirred at 25° for 3–4 days under nitrogen and decolorized slowly. After addition of water (10 ml) it was continuously extracted with chloroform. Evaporation of solvent and purification by TLC gave the γ-ketoaldehydes **5b** and **5d** in *ca* 30% yield. ^1H NMR ($CDCl_3$), **5b**: δ 1.03 (6 H, d, *J* 7 Hz), 2.58 (1 H, hept, *J* 7 Hz), 2.60 (4 H, s), 9.64 (1 H, s). **5d**: δ 0.83 (3 H, br.t), 1.0–1.9 (8 H, m), 2.23 (2 H, br.t), 2.58 (4 H, s), 9.61 (1 H, s). **5d** was cyclized[12] to **6** in 10–20% yield (spectroscopical).

2-Methylquinoxaline (**7**). **2a** (180 mg) was heated with one equivalent of *o*-phenylenediamine in formic acid (0.5 ml) at 80° for 5 min. The formic acid was evaporated *in vacuo*. Methylene chloride (3–4 ml) was added and the solution was extracted with an aqueous sodium bicarbonate solution. Purification of the product by TLC ($CHCl_3$, 5% ethyl acetate) gave **7** (30 mg, 21%). ^1H NMR ($CDCl_3$): δ 2.73 (3 H, s), 7.1–8.1 (4 H, m), 8.63 (1 H, s). MS: (M^+) 144.

8a. From methyl vinyl ketone and hexane-1-hydroximidoyl chloride. B.p. 118°/0.02 mmHg. Yield 68%. MS (M^+) 197. ^1H NMR ($CDCl_3$): δ 0.90 (3 H, br.t, *J* 6 Hz), 1.1–1.7 (8 H, m), 2.23 (3 H, s), 2.30 (2 H, t, *J* 7 Hz), 3.09 and 3.12 (2 H, AB spectrum, *J* 6.9, 10.2, 16 Hz), 4.76 (1 H, dd, *J* 6.9, 10.2 Hz).

8b. From methyl vinyl ketone and propane-2-hydroximidoyl chloride. Bp 60–62°/0.15–0.20 mmHg. Yield 49%. ^1H NMR ($CDCl_3$): δ 1.17 (6 H, d, *J* 7.2 Hz), 2.29 (3 H, s), 2.71 (1 H, hept, *J* 7.2 Hz), 3.1 (2 H, d, *J* 9 Hz), 4.79 (1 H, dd, *J* 7.3, 9.9 Hz).

9a and **9b** were obtained as oils by reduction of **8a** and **8b** with Ti^{3+} (2.5 equiv) in aqueous acetic acid solution (1:1) for 3 days under nitrogen. The pH of the acid titanous solution was first adjusted to *ca* 2.5 with solid sodium bicarbonate before addition of acetic acid. Usual workup gives **9a** and **9b** in *ca* 60–70% yield, purified by preparative TLC. ^1H NMR ($CDCl_3$) **9a**: δ 0.90 (3 H, br.t), 1.0–1.8 (8 H, m), 2.25 (3 H, s), 2.48 (2 H, br.t), 2.90 (2 H, d, *J* 5.2 Hz), 4.34 (1 H, br.t, *J* 5.2 Hz), 5.0 (1 H, br.s). **9b**: 1.10 (6 H, d, *J* 7 Hz), 2.27 (3 H, s), 2.62 (1 H, hept, *J* 7 Hz), 2.93 (2 H, d, *J* 5 Hz), 4.33 (1 H, t, *J* 5 Hz), 6.1 (1 H, br.s).

The 3(2 H)-furanones **10a** and **10b** were obtained as oils by refluxing **9a** and **9b** in acetic acid with sodium acetate as catalyst.[1] ^1H NMR ($CDCl_3$) **10a**: δ 0.90 (3 H, br.t), 1.0–1.9 (8 H, m), 1.42 (3 H, d, *J* 7.2 Hz), 2.49 (2 H, br.t), 4.46 (1 H, q, *J* 7.2 Hz), 5.36 (1 H, s), MS (M^+) 182. IR (film): 1700 (s), 1590 (s) cm^{-1}. **10b**: 1.25 (6 H, d, *J* 7 Hz), 1.45 (3 H, d, *J* 7 Hz), 2.71 (1 H, hept, *J* 7 Hz), 4.48 (1 H, q, *J* 7 Hz), 5.37 (1 H, s). MS: (M^+) 140.

2 - *Pentyl* - 3 - *methyl* - 4 - *hydroxy* - 2 - *cyclopentenone* (**11a**). **9a** (100 mg) was treated with aqueous sodium hydroxide (4 ml, 10%) for 3 h at 25°. The reaction mixture was neutralized with dil HCl and extracted with ether. Evaporation of the solvent and purification of the remainder on preparative TLC ($CHCl_3$, 1% CH_3OH) gave **11a** as an oil (40 mg, 48%). ^1H NMR ($CDCl_3$): δ 0.90 (3 H, br.t), 1.0–1.8 (6 H, m), 2.09 (3 H, s), 2.1–3.0 (5 H, m), 4.71 (1 H, br.d, *J* 6 Hz). MS: (M^+) 182.

Reduction of **8a** *to 2,5-dioxoundecane* (**12**). **8a** (975 mg) dissolved in methanol (40 ml), and diluted hydrochloric acid (25 ml, 4 M) was reduced with Zn-powder (4.5 g) which was added in small portions at intervals. Titanous chloride (0.5 ml, 1 M) was added. After 18 h the solution was extracted with methylene chloride giving 1.13 g of crude **12** which was sufficiently pure for the subsequent cyclization to dihydrojasmone. ^1H NMR ($CDCl_3$): δ 0.90 (3 H, br.t), 1.0–1.9 (8 H, m), 2.11 (3 H, s), 2.37 (3 H, br.t), 2.58 (4 H, s).

Dihydrojasmone (**11b**) was synthesized by refluxing the crude **12** for 4 h in 3% ethanolic sodium hydroxide.[19] The yield of **11b** was 62%, b.p. 120–122°/12 mmHg. The spectroscopic data of **11b** agree with those in the literature.

14 was prepared according to standard procedures (eqn 4).[17,18]

15 was prepared in 84% yield from **14** and 3-methyl-1-propenyl vinyl ketone[20] according to the general procedure. ^1H NMR ($CDCl_3$): δ 1.1–1.8 (10 H, m), 1.97 (3 H, s), 2.17 (3 H, s), 2.1–2.5 (4 H, m), 3.15 (2 H, d, *J* 9.4 Hz), 3.66 (3 H, s), 4.85 (1 H, dd, *J* 5.3 and 3.9 Hz), 6.45 (1 H, br.s). MS: (M^+) 309. IR (film): 1740 (s), 1635 (s), 1615 (s).

3 - *Isopropyl* - 5 - *acetoxymethyl* - 2 - *isoxazoline*, **16a**, b.p.$_{0.1}$ 90–94°, yield 67%. ^1H NMR (CCl_4): δ 1.16 (6 H, d, *J* 6.8 Hz), 2.01 (3 H, s), 2.4–3.1 (3 H, m), 3.99 (2 H, d, *J* 5.0 Hz), 4.58 (1 H, m). MS: 186 ($M + 1$)$^+$.

3 - *Isopropyl* - 5 - *acetylaminomethyl* - 2 - *isoxazoline*, **16b**, b.p.$_{0.1}$ 154°, yield 19%. ^1H NMR (CCl_4): δ 1.14 (6 H, d, *J* 6.8 Hz), 1.91 (3 H, s), 2.4–3.1 (3 H, m), 3.2–3.6 (2 H, m), 4.56 (1 H, m), 6.9 (1 H, br.s). MS: 185 ($M + 1$)$^+$.

3 - *Hexyl* - 5 - *acetylaminomethyl* - 2 - *isoxazoline*, **16c**, m.p. 88° (ethanol), yield 44%. ^1H NMR (CCl_4): δ 0.89 (3 H, br.t), 1.0–1.7 (8 H, m), 1.97 (3 H, s), 2.1–2.5 (2 H, m), 2.7–3.0 (2 H, m), 3.2–3.5 (2 H, m), 4.6 (1 H, m), 6.2 (1 H, br.s). MS: 227 ($M + 1$)$^+$.

16a,b,c were reduced by Ti^{3+} in aqueous acetic acid for 3 days to the corresponding hydroxyketones **17a,b,c**. They were purified on preparative TLC. (Silica, $CHCl_3$, 0.5% methanol.) It was later found that the reduction can be carried out practically quantitatively by catalytic reduction over RaNi (commercial, active, stored under H_2O) in ethanol.

1 - *Acetoxy* - 4 - *keto* - 5 - *methyl* - 2 - *hexanol*, **17a**, *liquid*. ^1H NMR ($CDCl_3$): δ 1.10 (6 H, d, *J* 6.8 Hz), 2.08 (3 H, s), 2.4–3.0 (3 H, m), 4.0–4.4 (3 H, m), 5.8 (1 H, br.s). MS: 189 ($M + 1$)$^+$.

1 - *Acetylamino* - 4 - *keto* - 5 - *methyl* - 2 - *hexanol*, **17b**, *liquid*. ^1H NMR ($CDCl_3$): δ 1.12 (6 H, d, *J* 6.8 Hz), 2.00

(3 H, s), 2.3–2.9 (3 H, m), 3.1–3.5 (2 H, m), 3.9–4.3 (2 H, m), 6.6 (1 H, br.s).

1 - *Acetylamino* - 4 - *keto* - 2 - *decanol*, **17c**, *liquid*. ^{1}H NMR ($CDCl_3$): δ 0.89 (3 H, br.t), 1.0–1.8 (8 H, m), 1.99 (3 H, s), 2.43 (2 H, t, *J* 7 Hz), 2.59 (2 H, d, *J* 6 Hz), 3.1–3.4 (2 H, m), 3.9–4.3 (2 H, m), 6.4 (1 H, br.s). MS: 230 $(M+1)^+$.

1-*Acetyl*-2-*isopropylpyrrole*, **18**. **17b** (90 mg) was heated with sodium acetate (190 mg) in acetic acid (1 ml) for 3.25 h at 100°. Chloroform was added and the solution was washed with water and aqueous sodium bicarbonate solution, dried and evaporated to give **18**. 60 mg, 80%. ^{1}H NMR (CCl_4): δ 1.20 (6 H, d, *J* 7 Hz), 2.49 (3 H, s), 3.5 (1 H, hept., *J* 7 Hz), 5.98 (2 H, m), 6.84 (1 H, dd, *J* 3.4, 1.6 Hz). MS: 151 (M^+).

1-*Acetyl*-2-*hexylpyrrole*, **19**, was obtained according to the same method as **18**, yield 76%. ^{1}H NMR ($CDCl_3$): δ 0.89 (3 H, br.t), 1.1–1.9 (8 H, m), 2.51 (3 H, s), 2.90 (2 H, t, *J* 7 Hz), 5.98 (1 H, br.s), 6.12 (1 H, t, *J* 3.3 Hz), 6.98 (1 H, dd, *J* 3.3, 1.6 Hz). MS: 193 (M^+).

Acknowledgement—We wish to thank DANIDA for fellowships to K.S.M. and K.K.S.

REFERENCES

[1]S. H. Anderson, N. B. Das, R. D. Jørgensen, G. Kjeldsen, J. S. Knudsen, S. C. Sharma and K. B. G. Torssell, *Acta Chem. Scand.* **B36**, 1 (1982).
[2]C. Grundmann and P. Grünanger, *The Nitrile Oxides*, Springer Verlag, Berlin (1971).
[3]K. B. G. Torssell and O. Zeuthen, *Acta Chem. Scand.* **B32**, 118 (1978).
[4]G. Stagno d'Alcontres and G. De Giacomo, *Atti. Soc. Peloritana* **5**, 159 (1958/59).
[5]S. C. Sharma and K. B. G. Torssell, *Acta Chem. Scand.* **B33**, 379 (1979).
[6]A. Quilico and P. Grünanger, *Rend. Ist. Lombardi Sci. Lettere* **88**, 990 (1955).
[7]G. Bianchi, R. Gandolfi and P. Grünanger, *J. Heter. Chem.* **5**, 49 (1968); G. Bianchi, A. Gamba-Invernizzi and R. Gandolfi, *J. Chem. Soc. Perkin I* 1757 (1974).
[8]G. Bianchi and P. Grünanger, *Chem. Ind. (Milan)* **46**, 1187 (1964).
[9]N. B. Das and K. B. G. Torssell, *Tetrahedron* **39**, 2227 (1983).
[10]R. Huisgen and W. Mack, *Tetrahedron Letters* 583 (1961).
[11]G. Casnati and A. Ricca, *Tetrahedron Letters* 327 (1967).
[12]G. W. K. Cavill, B. S. Goodrich and D. G. Laing, *Austral. J. Chem.* **23**, 83 (1970).
[13]K. Oshima, H. Yamamoto and H. Nozaki, *J. Am. Chem. Soc.* **95**, 4446 (1977).
[14]S. H. Andersen, K. K. Sharma and K. B. G. Torssell, *Tetrahedron* **39**, 2241 (1983).
[15]R. A. Ellison, *Synthesis* 379 (1973).
[16]T. L. Ho, *Synth. Commun.* **4**, 265 (1974).
[17]*Organic Syntheses Coll.* **IV**, 317 (1963).
[18]L. Nowak, G. Baan, J. Marosfalvi and C. Szantay, *Chem. Ber.* **113**, 2939 (1980).
[19]H. Hunsdiecker, *Chem. Ber.* **75**, 455 (1942).
[20]G. Kjeldsen, J. S. Knudsen, L. S. Ravn-Petersen and K. B. G. Torssell, *Tetrahedron* **39**, 2237 (1983).