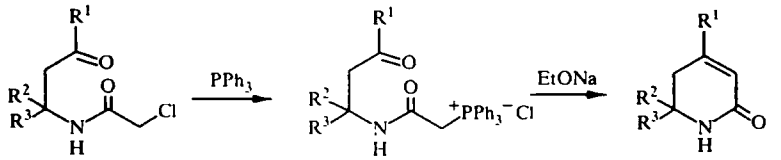
Chemistry of Heterocyclic Compounds, Vol. 34, No. 2, 1998

PREPARATION OF 5,6-DIHYDRO-2(1H)-PYRIDINONES BY AN INTRAMOLECULAR WITTIG REACTION USING N-3-OXOALKYLCHLORACETAMIDES

A. S. Fisyuk, N. V. Poendaev, and Yu. G. Bundel'

The reaction of N-(1,1-dimethyl-3-oxobutyl)chloracetamide (Ia) with triphenylphosphine gave 1-(1,1-dimethyl-3-oxobutylcarbamoylmethyl)triphenylphosphonium chloride (IIa), which cyclizes upon the action of sodium ethylate in ethanol to give 5,6-dihydro-4,6,6-trimethyl-2(1H)-pyridinone (IIIa). Analogously, N-(1,3-diphenyl-3-oxobutyl)chloracetamide (Ib) gave 5,6-dihydro-4,6-diphenyl-2(1H)-pyridinone (IIIb) without isolation of the triphenylphosphonium derivative IIb. Product IIIb is unstable in the air.



Ia, b IIa, b IIIa, b

I–III a $R^1 = R^2 = R^3 = Me$; b $R^1 = R^2 = Ph$, $R^3 = H$

Chloracetamides Ia and Ib were obtained as described in our previous work [1].

1-(1,1-Dimethyl-3-oxobutylcarbamoylmethyl)triphenylphosphonium Chloride (IIa). A solution of 3.21 g (16.7 mmoles) N-3-oxoalkylchloracetamide Ia and 4.84 g (18.4 mmoles) triphenylphosphine in 20 ml dry dioxane was heated at reflux for 16 h. The reaction mixture was cooled. The precipitate was filtered off, dried in vacuum, and recrystallized from 5:1 benzene–ethanol to give 7.60 g (65%) salt IIa. PMR spectrum at 200 MHz in $CDCl_3$ (TMS): 9.68 (1H, s, NH), 7.67-7.91 (15H, m, 3Ph), 1.28 (6H, s, $C\underline{H}_3-C-C\underline{H}_3$), 5.07 (2H, d, $^2J_{HP}$ = 14.4 Hz, $C\underline{H}_2-{}^+PPh_3$), 2.79 (2H, s, $OC-C\underline{H}_2$), 2.04 ppm (3H, s, $C\underline{H}_3-CO$).

5,6-Dihydro-4,6,6-trimethyl-2(1H)-pyridinone (IIIa). A solution of sodium ethylate obtained from 0.074 g (3.2 mmoles) sodium and 5 ml absolute ethanol was added dropwise with stirring to 1.400 g (3.2 mmoles) IIa in 20 ml absolute ethanol. The reaction mixture was stirred for 1 h. The precipitate of NaCl was then filtered off. The solvent was removed in vacuum and the residue was purified by treatment on a silica gel column using 3:1 chloroform–ethyl acetate as the eluent to give 0.125 g (90%) IIIa. PMR spectrum at 200 MHz in $CDCl_3$ (TMS): 6.33 (1H, s, NH), 5.72 (1H, m, 3-CH), 2.23 (2H, s, 5-CH_2), 1.91 (3H, s, 4-CH_3), 1.28 ppm (6H, s, 2 × 6-CH_3).

5,6-Dihydro-4,6-diphenyl-2(1H)-pyridinone (IIIb). A solution of 0.961 g (3.2 mmoles) Ib and 0.840 g (3.2 mmoles) triphenylphosphine in 5 ml absolute ethanol was heated at reflux for 4 h. After cooling to 20-25°C, a solution of sodium ethylate obtained from 0.074 g (3.2 mmoles) sodium in 5 ml ethanol was added dropwise with stirring to the reaction mixture, which was then stirred for an additional 1 h. The precipitate of NaCl was filtered off. The solvent was removed in vacuum and the residue was subjected to flash chromatography on silica gel using chloroform as the eluent to give 0.230 g (30%) IIIb. PMR spectrum at 200 MHz in $CDCl_3$ (TMS): 8.13 (1H, s, NH), 7.50-7.14 (10H, m, 2Ph), 5.53 (1H, d, ^4J = 3.8 Hz, $OC-C\underline{H}$), 3.93 (1H, m, $Ph-C\underline{H}$), 2.85 (1H, B of ABX, $^2J_{AB}$ = 16.1, $^3J_{BX}$ = 7.0 Hz, $CH_A\underline{H}_B-CHPh$), 2.68 ppm (1H, B of ABX, $^2J_{AB}$ = 16.1, $^3J_{AX}$ = 10.1 Hz, $C\underline{H}_AH_B-CHPh$).

Omsk State University, 664077 Omsk, Russia. Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 2, pp. 281-282, February, 1998. Original article submitted December 10, 1997.

 0009-3122/98/3402-0258$20.00 ©1998 Plenum Publishing Corporation

This work was carried out with the financial support of the Russian Basic Research Fund (Project 97-03-33119a).

REFERENCES

1. A. S. Fisyuk [Fyssiuk], M. A. Vorontsova, and R. S. Sagitullin, Mendeleev Commun., No. 6, 249 (1993).