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A facile route to *N*-acetyl α,β-unsaturated γ-lactam derivatives using ethyl acetamidocyanoacetate and dialkyl acetylenedicarboxylate in the presence of triphenylphosphine

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Abstract

The three component reaction of ethyl acetamidocyanoacetate with dialkyl acetylenedicarboxylates and triphenylphosphine leads to phosphorus ylides in good yields. These ylides undergo a 1,2-proton shift, loss of triphenylphosphine, and subsequent intramolecular amidation leads to the formation of *N*-acetyl α,β-unsaturated γ-lactams.

Keywords: Ethyl acetamidocyanoacetate; Dialkyl acetylenedicarboxylate; Phosphorus ylides; *N*-Acetyl α,β-unsaturated γ-lactam

Five membered cyclic lactams have been used successfully in routes to various alkaloids[1–3] and are suitable precursors for unusual γ-amino acids such as statine and its analogues.[4,5] There are also many examples of pyrroline-containing natural products with interesting pharmacological activities, such as antihypertensive,[6] antitumor, and antibiotic activities.[7,8] 3-Pyrrolin-2-ones are also important structural units of the related indolocarbazole alkaloids (+)-staurosporine[9] and (+)-K252a,[10] which are strong kinase inhibitors widely used as molecular tools. On the other hand, 3,4-diaryl- and 1,3,4-triaryl-3-pyrrolin-2-ones, have been shown to be a potential new type of selective COX2 inhibitor.[11,12] Moreover, the α,β-unsaturated γ-butyrolactam moiety can be utilized as a Michael acceptor for a variety of nucleophiles.[13] Therefore, the synthesis of 3-pyrrolin-2-ones is currently receiving considerable attention.[14]

As part of our current studies on the development of new routes to heterocyclic systems,[15–17] we report on the reaction between ethyl acetamidocyanoacetate **1** and dialkyl acetylenedicarboxylates **2** in the presence of triphenylphosphine. Thus, the reaction of a strong CH-acid such as **1** with an acetylenic ester **2** in the presence of triphenylphosphine leads to a stable phosphorus ylide **3**. These ylides are converted into *N*-acetyl α,β-unsaturated γ-lactam derivatives **4** in boiling toluene[18] (Scheme 1).

On the basis of the chemistry of trivalent phosphorus nucleophiles,[19–21] it is reasonable to assume that *N*-acetyl α,β-unsaturated γ-lactam **4** results from the initial addition of triphenylphosphine to the acetylenic ester and subsequent protonation of the reactive 1:1 adduct by **1**, followed by the attack of the conjugate base of **1** on vinyltriphenylphosphonium cation **5** to generate phosphorane **3** which in turn is converted into betaine **6** by a 1,2-proton transfer. Loss of triphenylphosphine and cyclization of betaine **6** leads to compound **4** (Scheme 2).

The structures of compounds **3a–c** were deduced from their elemental analysis and their MS, IR, ^{1}H, ^{13}C, and ^{31}P NMR spectra. Ylide **3a** could have been formed as a pair of diastereomers, but according to the NMR analysis only one diastereomer appeared to have been formed. However, we did not succeed in determining the stereochemistry of ylide **3a**.

The ^{1}H and ^{13}C NMR spectra of **3b** were consistent with the presence of two isomers. The ylide moiety is strongly

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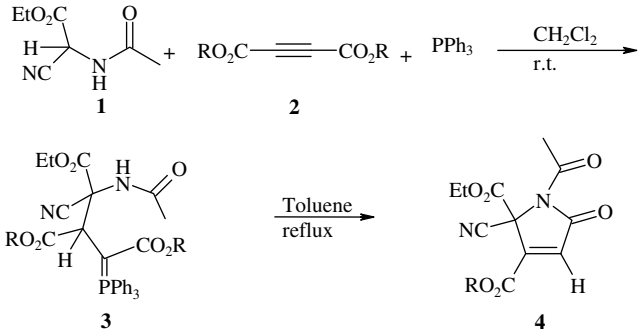
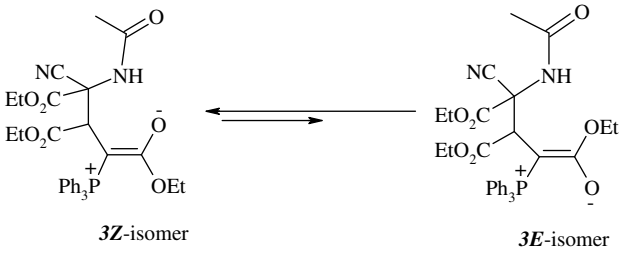


2-4	R
a	Me
b	Et
c	*t*-Bu

Scheme 1.

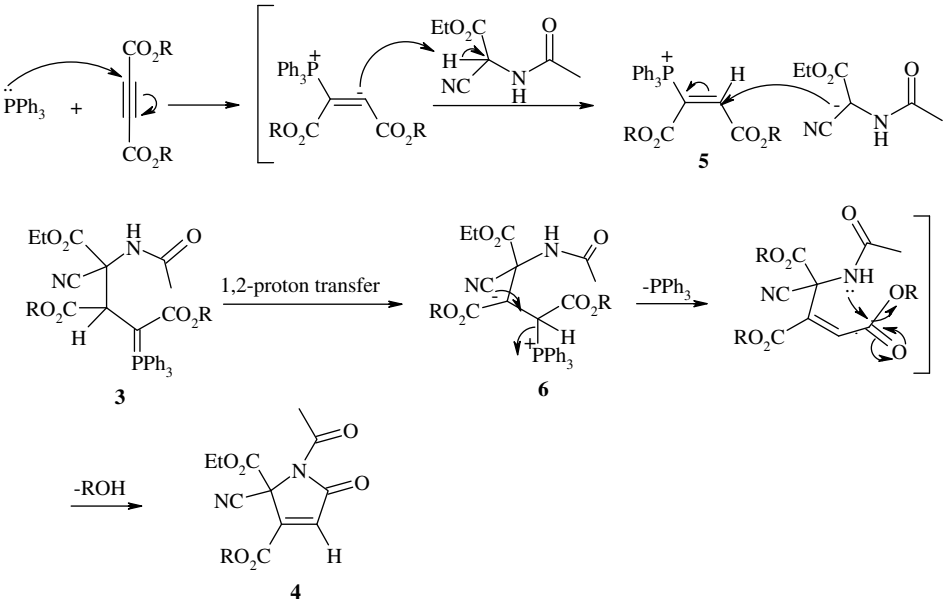


Scheme 3.

conjugated with the adjacent carbonyl group, and the rotation about the partial double bond in isomers (*E*)-**3b** and (*Z*)-**3b** is slow on the NMR time scale at ambient temperature. Assignment of the (*Z*)-configuration to the major geometric isomer is based on the ^{1}H chemical shift of the OR moiety, which is expected to be shielded as a result of the anisotropic effect of the phenyl groups (Scheme 3).

The ^{1}H NMR spectrum of **3b** at room temperature (25 °C) exhibited two pairs of sharp singlets at δ 2.00 and δ 2.03 ppm with an intensity of 42:58 for the methyl groups of the *E*- and *Z*-isomer, respectively. Increasing the temperature resulted in coalescence of their acetyl resonances. At 60 °C, a fairly broad singlet was observed for the acetyl group. This observation was attributed to the temperature dependent equilibrium between the geometrical (rotational) isomers. From the spectrum it was seen that the methine proton appeared as a pair of symmetrical doublets with unequal intensities at δ 3.18 ppm ($^{3}J_{PH}$ 18.5 Hz) and δ 3.28 ppm ($^{3}J_{PH}$ 18.5 Hz) at 25 °C. Raising the temperature also led to the gradual collapse of these resonances to a single doublet. The NH proton was observed as a pair of singlets at δ 9.63 and δ 10.17 ppm. The ^{31}P NMR spectrum of **3b** exhibited two singlets at δ 25.68 and δ 26.28 ppm. The ^{13}C NMR of **3b** exhibited two doublets of unequal intensities at δ 40.09 ($^{1}J_{PC}$ 124.0 Hz, P=C) for the *E*-isomer and at δ 41.27 ($^{1}J_{PC}$ 124.0 Hz, P=C) for the *Z*-isomer and two doublets at δ 49.71 ($^{2}J_{PC}$ 13.4 Hz), and δ 50.70 ($^{2}J_{PC}$ 13.9 Hz) for the methine carbons of the *E*- and *Z*-isomers, respectively.

The ^{1}H NMR of **4a** exhibited a triplet at δ 1.36 ppm ($^{3}J_{HH}$ 7.3 Hz) and a multiplet at δ 4.14–4.23 ppm for the ethoxy group, a singlet at δ 3.82 ppm for the methoxy group and a singlet at δ 6.95 ppm for the olefinic proton. The ^{13}C NMR spectrum of **4a** displayed twelve distinct resonances in agreement with the lactam structure. The ^{1}H and ^{13}C NMR spectra of **4b** and **4c** were similar to those of **4a** except for the ester groups, which exhibited characteristic resonances with appropriate chemical shifts (see Ref. 18).



Scheme 2.

The structural assignments made on the basis of the ^{1}H and ^{13}C NMR spectra of compounds **4b** and **4c** were supported by the measurement of their IR spectra. The carbonyl regions of the spectra exhibited distinct absorption bands for each compound. The mass spectra of **4b–4c** displayed molecular ion peaks and other fragmentations involved with the loss of the ester moiety.

In conclusion, the method presented here carries the advantage of being performed under neutral conditions and requires no activation or modification of the reactants. We anticipate that the reactions described herein represent a simple process in the synthesis of polyfunctionalized α,β-unsaturated γ-lactams of interest.

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18. *General procedure for the preparation of compound* **3**. To a magnetically stirred solution of 0.52 g of triphenylphosphine (2 mmol) and 0.34 g of ethyl acetamidocyanoacetate (2 mmol) in 5 ml of dry CH_2Cl_2 was added, dropwise, a mixture of 0.28 g of dimethyl acetylenedicarboxylate (2 mmol) in 3 ml of dry CH_2Cl_2 at −5 °C over 10 min. The reaction mixture was then allowed to warm to room temperature and stirred for 24 h. The solvent was removed under reduced pressure and the residue was purified by silica gel (Merck Silica Gel 60, 70–230 mesh) column chromatography using hexane/ethyl acetate (1:5) as eluent. The solvent was removed to afford product **3a** as a white powder, yield 1.1 g (80%), mp 193–195 °C. IR (KBr) (ν_{max}, cm^{-1}): 3474 (NH), 2378 (CN), 1766, 1736 (C=O), 1692 (C=C); ^{1}H NMR (500.1 MHz, $CDCl_3$): δ_H 1.03 (3H, t, $^{3}J_{HH}$ 7 Hz, CH_3), 2.06 (3H, s, CH_3), 3.04 (3H, s, OCH_3), 3.29 (1H, d, $^{3}J_{PH}$ 18.5 Hz, CH), 3.69 (3H, s, OCH_3), 3.99–4.04 (2H, m, OCH_2), 7.45–7.77 (15H, m, arom), 9.63 (1H, s, NH); ^{13}C NMR (125.8 MHz, $CDCl_3$): δ_C 13.70 (CH_3), 23.24 (CH_3), 41.57 (d, $^{1}J_{PC}$ 123.9 Hz, P=C), 44.64 (OCH_3), 50.60 (d, $^{2}J_{PC}$ 14.0 Hz, CH), 52.3 (OCH_3), 62.64 (OCH_2), 62.97 (d, $^{3}J_{PC}$ 4.0 Hz, quaternary carbon), 116.75 (CN), 125.78 (d, $^{1}J_{PC}$ 93.5 Hz, C_{ipso}), 128.75 (d, $^{3}J_{PC}$ 12.3 Hz, C_{meta}), 132.39 (C_{para}), 134.14 (d, $^{2}J_{PC}$ 8.3 Hz, C_{ortho}), 164.55 (C=O, amide), 170.08 (C=O, ester), 170.91 (d, $^{2}J_{PC}$ 5.3 Hz, C=O ester), 172.42 (d, $^{3}J_{PC}$ 12.2 Hz, C=O ester); ^{31}P NMR (202.5 MHz, $CDCl_3$): δ_P 26.24; MS (EI), *m/z* (%): 574 (1), 278 (5), 262 (2), 239 (30), 221 (91), 175 (44), 57 (100), 43 (21). Anal. Calcd for $C_{31}H_{31}N_2O_7P$ (574.57): C, 64.80; H, 5.44; N, 4.88. Found: C, 64.86; H, 5.49; N, 4.83.

Compound **3b**: White powder, mp 164–166 °C, yield 85%, IR (KBr) (ν_{max}, cm^{-1}): 3459 (NH),1762, 1740 (C=O), 1681 (C=C); ^{1}H NMR (500.1 MHz, $CDCl_3$): (**3b-*Z***, 58%) δ_H 0.4 (3H, t, $^{3}J_{HH}$ 7.1 Hz, CH_3), 1.01 (3H, t, $^{3}J_{HH}$ 7.1 Hz, CH_3), 1.24 (3H, t, $^{3}J_{HH}$ 7.2 Hz, CH_3), 2.03 (3H, s, CH_3), 3.28 (1H, d, $^{3}J_{PH}$ 18.5 Hz, CH), 3.45–3.53 (2H, m, OCH_2), 3.98–4.02 (2H, m, OCH_2), 4.20–4.26 (2H, m, OCH_2), 7.45–7.76 (15H, m, arom), 9.63 (1H, s, NH); (**3b-*E***, 42%): δ_H 0.34 (3H, t, $^{3}J_{HH}$ 7.1 Hz, CH_3), 1.09 (3H, t, $^{3}J_{HH}$ 7.1 Hz, CH_3), 1.25 (3H, t, $^{3}J_{HH}$ 7.3 Hz, CH_3), 2.00 (3H, s, CH_3), 3.18 (1H, d, $^{3}J_{PH}$ 18.5 Hz, CH), 3.67–3.73 (2H, m, OCH_2), 3.90–3.97 (2H, m, OCH_2), 4.08–4.12 (2H, m, OCH_2), 7.45–7.76 (15H, m, arom), 10.17 (1H, s, NH); ^{13}C NMR (125.8 MHz, $CDCl_3$) (**3b-*Z***): δ_C 13.66, 13.75, and 14.18 ($3CH_3$), 23.21 (CH_3), 41.27 (d, $^{1}J_{PC}$ 124.0 Hz, P=C), 50.70 (d, $^{2}J_{PC}$ 13.9 Hz, CH), 58.56, 61.45, and 62.51 ($3OCH_2$), 63.00 (d, $^{3}J_{PC}$ 3.8 Hz, quaternary carbon) 116.82 (CN), 126.03 (d, $^{1}J_{PC}$ 93.5 Hz, C_{ipso}), 128.64 (d, $^{3}J_{PC}$ 12.4 Hz, C_{meta}), 132.34 (C_{para}), 134.23 (d, $^{2}J_{PC}$ 8.2 Hz, C_{ortho}), 164.47 (C=O, amide), 169.99 (C=O, ester), 170.45 (d, $^{2}J_{PC}$ 5.5 Hz, C=O, ester), 172.02 (d, $^{2}J_{PC}$ 12.3 Hz, C=O, ester); (**3b-*E***): δ_C 13.53, 13.61, and 14.14 ($3CH_3$), 22.77(CH_3), 40.09 (d, $^{1}J_{PC}$ 125.3 Hz, P=C), 49.71 (d, $^{2}J_{PC}$ 13.4 Hz, CH), 58.67, 61.58, and 62.95 ($3OCH_2$), 61.36 (d, $^{3}J_{PC}$ 6.0 Hz, quaternary carbon), 116.96 (C≡N), 126.51 (d, $^{1}J_{PC}$ 93.1 Hz, C_{ipso}), 128.69 (d, $^{3}J_{PC}$ 12.5 Hz, C_{meta}), 132.49 (C_{para}), 132.94 (d, $^{2}J_{PC}$ 8.1 Hz, C_{ortho}), 165.03(C=O, amide), 169.94 (C=O, ester), 170.00 (d, $^{2}J_{PC}$ 5.0 Hz, C=O, ester), 171.95 (d, $^{3}J_{PC}$ 12.1 Hz, C=O, ester); ^{31}P NMR (202.5 MHz, $CDCl_3$): δ_P 25.68, 26.28; MS (EI), *m/z* (%): 602 (1), 434 (4), 262 (8), 184 (16), 152 (24), 77 (88), 43 (100). Anal. Calcd for $C_{33}H_{35}N_2O_7P$ (602.63): C, 65.77; H, 5.85; N, 4.65. Found: C, 65.82; H, 5.89; N, 4.69.

Compound **3c**: White powder, mp 120–123 °C, yield 80%; IR (KBr) (ν_{max}, cm^{-1}): 3437 (NH), 2356 (CN), 1776, 1732 (C=O), 1689 (C=C); ^{1}H NMR (500.1 MHz, $CDCl_3$): δ_H 0.88 (9H, s, CMe_3), 1.01 (3H, t, $^{3}J_{HH}$ 7.1 Hz, CH_3), 1.46 (9H, s, CMe_3), 2.08 (3H, s, CH_3), 3.13 (1H, d, $^{3}J_{PH}$ 18.8 Hz, CH), 3.94–4.06 (2H, m, OCH_2), 7.23–7.92 (15H, m, arom), 9.95 (1H, s, NH); ^{13}C NMR(125.8 MHz, $CDCl_3$): δ_C 13.65 (CH_3), 23.15 (CH_3), 28.08 and 28.12 ($2CMe_3$), 41.01 (d, $^{1}J_{PC}$ 124.7 Hz, P=C), 51.67 (d, $^{2}J_{PC}$ 14.1 Hz, CH), 62.30 (OCH_2), 63.02 (d, $^{3}J_{PC}$ 3.6 Hz, quaternary carbon), 78.45 and 81.92 ($2OCMe_3$), 116.80 (CN), 125.8 (d, $^{1}J_{PC}$ 93.5 Hz, C_{ipso}), 128.42 (d, $^{3}J_{PC}$ 12.3 Hz, C_{meta}), 132.13 (C_{para}), 134.77 (d, $^{2}J_{PC}$ 8.5 Hz, C_{ortho}), 164.49 (C=O, amide), 169.78 (d, $^{2}J_{PC}$ 6.0 Hz, C=O ester), 169.84 (C=O, ester), 171.77 (d, $^{3}J_{PC}$ 11.82 Hz, C=O ester); ^{31}P NMR (202.5 MHz, $CDCl_3$): δ_P 25.85; MS (EI), *m/z* (%): 658 (1), 332 (4), 295 (34), 262 (5), 105 (4), 57 (48), 44 (100). Anal. Calcd for $C_{37}H_{43}N_2O_7P$ (658.73): C, 67.46; H, 6.58; N, 4.25. Found: C, 67.51; H, 6.53; N, 4.21.

Preparation of 2-ethyl-3-methyl 1-acetyl-2-cyano-5-oxo-2,5-dihydro-1H-pyrrole-2,3- dicarboxylate **4a**. Compound **3a** was refluxed in toluene for 96 h. The solvent was removed under reduced pressure and the residue was purified by silica gel (Merck, Silica Gel, 230–400 mesh) column chromatography using hexane/ethyl acetate (3:1) as eluent. The solvent was removed under reduced pressure and **4a**

was obtained as a yellow oil, yield (35%); IR (KBr) (ν_{max}, cm^{-1}): 2371 (CN), 1787, 1725 (C=O); ^1H NMR (500.1 MHz, $CDCl_3$): δ_H 1.36 (3H, t, $^3J_{HH}$ 7.3 Hz, CH_3), 2.54 (3H, s, CH_3), 3.82 (3H, s, OCH_3), 4.14–4.23 (2H, m, OCH_2), 6.95 (1H, s, CH); ^{13}C NMR (125.8 MHz, $CDCl_3$): δ_C 13.73 (CH_3), 24.43 (CH_3), 52.47 (OCH_3) 64.36 (OCH_2), 82.30 (quaternary carbon), 112.29 (CN), 134.64 and 146.54 (olefinic carbons), 158.75 and 162.87 (2C=O, imide), 164.76 and 168.42 (2C=O, ester); MS (EI), *m*/*z* (%): 280 (2), 265 (3), 223 (2), 181(2.5), 151 (2.4), 59 (2), 43 (5). Anal. Calcd for $C_{12}H_{12}N_2O_6$ (280.24): C, 51.43; H, 4.32; N, 9.99. Found: C, 51.49; H, 4.36; N, 10.06.

Compound **4b**:Yellow oil, yield 45%, IR (KBr) (ν_{max}, cm^{-1}): 2375 (CN), 1781, 1730 (C=O); ^1H NMR (500.1 MHz, $CDCl_3$): δ_H 1.36 (3H, t, $^3J_{HH}$ 7.1 Hz, CH_3), 1.37 (3H, t, $^3J_{HH}$ 7.1 Hz, CH_3), 2.57 (3H, s, CH_3), 4.33–4.41 (4H, m, $2OCH_2$), 6.98 (1H, s, CH); ^{13}C NMR (125.8 MHz, $CDCl_3$); δ_C 13.75 and 13.89 ($2CH_3$), 24.32 (CH_3), 63.34 and 65.10 ($2OCH_2$), 82.40 (quaternary carbon), 111.51 (CN), 134.64 and 145.24 (olefinic carbons), 158.77 and 161.67 (2C=O, imide), 165.04 and 168.43 (2C=O, ester); MS (EI), *m*/*z* (%): 294 (3), 249 (4), 180 (72), 152 (81), 134 (30), 57 (10), 43 (100). Anal. Calcd for $C_{13}H_{14}N_2O_6$ (294.27): C, 53.06; H, 4.80; N, 9.52. Found: C, 53.09; H, 4.85; N, 9.58.Compound **4c**: Yellow oil, yield 45% IR (KBr) (ν_{max}, cm^{-1}): 2370 (CN), 1785, 1725 (C=O); ^1H NMR (500.1 MHz, $CDCl_3$): δ_H 1.35 (3H, t, $^3J_{HH}$ 7.1 Hz, CH_3), 1.54 (9H, s, CMe_3), 2.55 (3H, s, CH_3), 4.29 (1H, ABX_3, dq, $^2J_{HH}$ 10.6 Hz, $^3J_{HH}$ 7.1 Hz, OCH), 4.40 (1H, ABX_3, dq, $^2J_{HH}$ 10.6 Hz, $^3J_{HH}$ 7.1 Hz, OCH), 6.92(1H, s, CH); ^{13}C NMR (125.8 MHz, $CDCl_3$): δ_C 13.75 (CH_3), 24.33 (CH_3), 27.83 (CMe_3), 64.97 (OCH_2), 86 ($OCMe_3$), 111.65 (CN), 82.60 (quaternary carbon), 134.17 and 146.78 (olefinic carbons), 157.62 and 161.71 (2C=O, imide), 165.21 and 168.43 (2C=O ester); MS (EI), *m*/*z* (%), 322 (2), 239 (8), 194 (90), 180 (98), 152 (98), 134 (48), 57 (71), 43 (100). Anal. Calcd for $C_{15}H_{18}N_2O_6$ (322.32): C, 55.90; H, 5.63; N, 8.69. Found: C, 55.94; H, 5.59; N, 8.67.

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