Simple and one-pot synthesis of new heterocyclic compounds in three-component reactions between isoquinoline or phenanthridine and acetylenic esters in the presence of N-heterocycles or 1,3-dicarbonyl compounds

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Abstract
A new class of enamino esters has been isolated in excellent yields from the 1:1:1 addition reaction between phenanthridine or isoquinoline and acetylenic esters such as ethyl propiolate or dialkyl acetylenedicarboxylates in the presence of heterocyclic NH compounds (succinimide, indole, 2-methylindole, 2-benzoxazolinone, 6-chlorobenzoxazolinone, carbazole and 3,6-dibromocarbazole) or 1,3-dicarbonyl compounds like 1,3-dimethylbarbituric acid, acetylacetone and dibenzoylethane.

Keywords: Enamino esters, phenanthridine, isoquinoline, ethyl propiolate, heterocyclic NH or 1,3-dicarbonyl compounds

Introduction
The development of simple synthetic routes for widely used organic compounds from readily available reagents is one of the major tasks in organic synthesis. Phenanthridines are important core structures found in a variety of natural products and other biologically important molecules with a wide range of biological activities and applications including antibacterial, antiprotozoal, anticancer, antimicrobial, anti-inflammatory, antiviral, antioxidant and also with applications as drugs, DNA targeting agents, dyes, and probes. Isoquinoline is also present in various natural products such as cryptaustoline and cryptowoline. They are known to exhibit various biological activities such as antileukaemic, tubulin polymerization inhibitory and anti-tumour activities. As previously reported, reaction between phenanthridine and two mol equivalents of dimethyl acetylenedicarboxylate, leads to the
formation of a new ring fused to the phenanthridine. In the current work, we now describe an efficient synthesis of a new class of enamino esters (see Scheme 1).

\[
\begin{align*}
\text{1} + R_1\text{-C} &\equiv \text{C}-R_2 + Y+H \xrightarrow{\text{CH}_2\text{Cl}_2} \text{4} \\
4(a-c): R_1 = H, R_2 = \text{CO}_2\text{Et} & \\
4(d,e): R_1 = R_2 = \text{CO}_2\text{Me} & \\
4(f,g): R_1 = R_2 = \text{CO}_2\text{Et} & \\
4(h): R_1 = R_2 = \text{CO}_2\text{Bu}^1 & \\
\text{5} + \text{MeO}_2\text{C} &\equiv \text{C}-\text{C}_2\text{O}_2\text{Me} + Y+H \xrightarrow{\text{CH}_2\text{Cl}_2} \text{8} \\
\end{align*}
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Scheme 1
Results and Discussion

An efficient synthesis of a new class of enamino esters from reaction between phenanthridine 1 or isoquinoline 5 and activated acetylenic esters 2 or 6 as a Michael acceptor\textsuperscript{39-46} was undertaken in the presence of heterocyclic NH compounds (succinimide, indole and 2-methylindole, 2-benzoazolinone, 6-chlorobenzoazolinone, carbazole and 3,6-dibromocarbazole) or 1,3-dicarbonyl compounds such as 1,3-dimethylbarbituric acid, acetylacetone and dibenzoylethane, at ambient temperature. Reactions were carried out by first mixing the phenanthridine or isoquinoline and heterocyclic NH or 1,3-dicarbonyl compounds and then the acetylenic ester was added slowly. The reactions proceeded smoothly in CH\textsubscript{2}Cl\textsubscript{2} and then the whole reaction mixture solidified into yellow solid within a few hours. The \textsuperscript{1}H and \textsuperscript{13}C NMR spectra of the crude products clearly indicated the formation of enamino esters 4a-h and 8i,j. No product other than 4a-h and 8i,j could be detected by NMR spectroscopy. The structures of compounds 4a-h and 8i,j were confirmed by elemental analyses, mass, IR, \textsuperscript{1}H NMR and \textsuperscript{13}C NMR spectra. The \textsuperscript{1}H NMR spectrum of 4a exhibited signals for the methyl (\(\delta = 1.29, 3\text{H}, \text{t}, \text{J}_{HH} = 7.2\) Hz, OCH\textsubscript{2}CH\textsubscript{3}), methylene (\(\delta = 4.19, 2\text{H}, \text{q}, \text{J}_{HH} = 7.2\) Hz, OCH\textsubscript{2}CH\textsubscript{3}), and olefinic (\(\delta = 5.58\) and 8.36, 2d, \(\text{J}_{HH} = 13.6\) Hz, CH=CH-OCH\textsubscript{2}CH\textsubscript{3}) protons, along with multiplets at \(\delta = 7.13-8.09\) ppm for the aromatic protons. The NCHN moieties at \(\delta = 9.03-9.33\) ppm in compounds 4a-e are deshielded due to the anisotropic effect of a benzene ring of phenanthridine.\textsuperscript{49} The \textsuperscript{13}C NMR spectrum of 4a showed 22 distinct resonances in agreement with the proposed structure. In addition, product 4a displayed \textsuperscript{13}C NMR resonances at \(\delta = 94.06\) ppm, 115.84 and 120.72 ppm, respectively for the NCHN, N-CH=CH-CO\textsubscript{2}CH\textsubscript{2}CH\textsubscript{3}, and N-CH=CH-CO\textsubscript{2}CH\textsubscript{2}CH\textsubscript{3} units. The carbonyl group resonances in the \textsuperscript{13}C NMR spectra of 4a appeared at \(\delta = 174.78\) and 177.16 ppm. The \textsuperscript{1}H and \textsuperscript{13}C NMR spectra of compounds 4b-h and 8i,j are similar to those of 4a. Assignment of the configuration (Z or E) in compounds 4a-h and 8i,j was decided on the basis of the chemical shift of the olefinic proton.\textsuperscript{34,47-48} With respect to the same employed conditions (effect of same solvent and temperature in our reactions) it seems that, the structural effect of reactants is an important factor for assignment of the configuration (Z or E) (see Scheme 2).

Briefly, we have developed a new method to access a novel class of heterocyclic derivatives. The present method has the advantage that, not only is the reaction performed under neutral conditions, but also the substances can be mixed without any activation or modifications. It seems that, this procedure is easy and simple approach for synthesis of heterocyclic derivatives.
**Experimental Section**

**General.** Melting points and IR spectra of all compounds were measured on an Electrothermal 9100 apparatus and a Shimadzu IR-460 spectrometer, respectively. The $^1$H, $^{13}$C, and $^{31}$P NMR spectra were obtained with a BRUKER DRX-500 AVANCE instrument using CDCl$_3$ as applied solvent and TMS as internal standard at 500.1, 125.8, and 202.4 MHz respectively. In addition, the mass spectra were recorded on a GCMS-QP5050A mass spectrometer operating at an ionization potential of 70 eV. Elemental analysis for C, H and N were performed using a Heraeus CHN-O-Rapid analyzer. Activated acetylenic esters, phenanthridine, isoquinoline, succinimide, indole, 2-methylindole, 2-benzoxazolinone, 6-chloro- benzoxazolinone, carbazole, 3,6-dibromocarbazole, 1,3-dimethylbarbituric acid, acetylacetone and dibenzoylethane were purchased from Fluka, (Buchs, Switzerland) and used without further purification.
General synthetic procedure, exemplified by (E)-ethyl 3-(6-(2,5-dioxopyrrolidin-1-yI)phenanthridine-5(6H)-yl)acrylate 4a. To a magnetically stirred solution of phenanthridine (0.18 g, 1 mmol) and succinimide (0.09 g, 1 mmol) in CH₂Cl₂ (10 mL) was added, dropwise, a mixture of ethyl propiolate (1 mmol) in CH₂Cl₂ (5 mL) at -10 °C over 10 min. After a few hours stirring at ambient temperature, the whole reaction mixture solidified into a brown solid, the solvent was then removed under reduced pressure and product washed with cold diethyl ether (2 × 5 mL). Then the product was recrystallized from a mixture of acetonitrile and acetone.

Brown powder, yield 92%, 0.35 g mp 100-102 °C, IR (νmax, cm⁻¹): 1707 and 1773 (C=O). ¹H NMR (500.1 MHz, CDCl₃), δH 1.29 (3H, t, JHH = 7.2 Hz, OCH₂CH₃), 2.75 (4H, s, 2CH₂), 4.19 (2H, q, JHH = 7.2 Hz, OCH₂CH₃), 5.85 (1H, d, JHH = 13.6 Hz, N-CH=CH-CO₂CH₂CH₃), 7.13-8.09 (8H, m, 8CH phenanthridine), 8.60 (1H, d, JHH = 13.6 Hz, N-CH=CH-CO₂CH₂CH₃), 9.03 (1H, s, NCHN). ¹³C NMR (125.8 MHz, CDCl₃), δC 13.38 (OCH₂CH₃), 26.87 (OCH₂CH₃), 28.51 (2CH₂), 94.06 (NCHN), 115.84 (1C, N-CH=CH-CO₂CH₂CH₃), 110.18, 113.14, 119.12, 119.75 122.16 and 122.53 (12C, phenanthridine), 177.48 (C=O, ester), 177.16 (2C=O, succinimide). Anal. Caled for C₂₂H₂₀N₂O₄: C, 70.18; H, 5.36; N, 7.44%, Found: C, 70.21; H, 5.31; N, 7.60%.

Ethyl (E)-3-(6-(1H-indole-1-yl)phenanthridine-5(6H)-yl)acrylate 4b. Yellow powder, yield 93%, 0.37 g mp 138-140 °C, IR (νmax, cm⁻¹): 1718 (C=O). ¹H NMR (500.1 MHz, CDCl₃), δH 1.18 (3H, t, JHH = 7.1 Hz, OCH₂CH₃), 4.19 (2H, q, JHH = 7.1 Hz, OCH₂CH₃), 5.45 (1H, d, JHH = 16.6 Hz, N-CH=CH-CO₂CH₂CH₃), 6.25 (1H, d, JHH = 3.5 Hz, N-CH=CH, indole), 6.31 (1H, d, JHH = 3.5 Hz, N-CH=CH, indole), 7.14-8.03 (12H, m, 12CH), 8.30 (1H, d, JHH = 16.6 Hz, N-CH=CH-CO₂CH₂CH₃), 9.24 (1H, s, NCHN). ¹³C NMR (125.8 MHz, CDCl₃), δC 13.43 (OCH₂CH₃), 25.71 (OCH₂CH₃), 63.76 (NCHN), 96.41 (1C, N-CH=CH-CO₂CH₂CH₃), 111.25, 113.30, 120.19 and 120.29 (4C, indole), 121.85, 122.56 and 122.53 (3C, phenanthridine), 123.26 and 124.30 (2C, indole), 125.68, 126.35 126.93,127.29, 127.78, 129.65 and 130.15 (7C, phenanthridine), 137.14 (1C, indole), 137.43 and 139.18 (2C, phenanthridine), 153.09 (1C, indole), 115.84 (1C, N-CH=CH-CO₂CH₂CH₃), 1707 and 1773 (C=O). MS, m/z (%): 364 (M-Et and H, 13), 278 (C₁₈H₁₆NO₂, 100), 204 (M-C₈H₈N, CO₂Et and H, 48), 179 (C₁₃H₁₀N, 60), 97 (C₅H₈O₂, 26). Anal. Caled for C₂₆H₂₉N₂O₄: C, 79.15; H, 5.62; N, 7.10%, Found: C, 78.98; H, 5.64; N, 7.18%.

Ethyl (E)-3-(6-(2-methyl-1H-indole-1-yl)phenanthridine-5(6H)-yl)acrylate 4c. Brown powder, yield 92%, 0.38 g; mp 136-138 °C, IR (νmax, cm⁻¹): 1723 (C=O). ¹H NMR (500.1 MHz, CDCl₃), δH 1.25 (3H, t, JHH = 7.2 Hz, OCH₂CH₃), 2.39 (3H, s, CH₃), 4.18 (2H, q, JHH = 7.2 Hz, OCH₂CH₃), 5.40 (1H, d, JHH = 13.3 Hz, N-CH=CH-CO₂CH₂CH₃), 6.22 (1H, s, indole), 7.08-8.63 (12H, m, 12CH), 8.70 (1H, d, JHH = 13.3 Hz, N-CH=CH-CO₂CH₂CH₃), 9.30 (1H, NCHN). ¹³C NMR (125.8 MHz, CDCl₃), δC 14.03 (OCH₂CH₃), 26.80 (OCH₂CH₃), 59.41 (NCHN), 97.66 (1C, N-CH=CH-CO₂CH₂CH₃), 110.18, 113.14, 119.12, 119.75 122.16 and 123.30 (6C, indole), 123.80 and 125.95 (2C, phenanthridine), 126.03 (N-CH=CH-CO₂CH₂CH₃), 126.81, 126.93, 127.47, 127.70, 129.92 and 131.11 (8C, phenanthridine), 135.18 (1C, indole),
137.23 and 138.10 (2C, phenantridine), 152.43 (1C, indole), 174.64 (C=O, ester). MS, m/z (%) = 408 (M⁺, 3), 379 (M-CH₂CH₃, 7), 335 (M-CO₂CH₂CH₃, 39), 179 (C₁₃H₉N, 37), 130 (C₆H₄N, 100). Anal. Calcd for C₂₇H₂₄N₂O₂ (408.19): C, 79.37; H, 5.93; N, 6.86 %; Found: C, 79.42; H, 6.02; N, 6.91 %.

**Dimethyl 2-(6-(2-oxobenz[d]oxazol-3(2H)-yl)phenanthridine-5(6H)-yl)fumarate 4d.** Yellow powder, yield 94%, 0.43 g mp 112-114 °C, IR (νmax, cm⁻¹): 1722 and 1783 (C=O). ¹H NMR (500.1 MHz, CDCl₃), δH 3.71 and 3.88 (6H, 2s, 2OCH₃), 7.08 (1H, s, N-C=CH-CO₂CH₃), 7.12-8.63 (12H arom, m, 12CH), 9.35 (1H, s, NCHN). ¹³C NMR (125.8 MHz, CDCl₃), δC 52.69 and 53.70 (2OCH₃), 109.62 (NCHN), 110.41 (N-C=CH-CO₂CH₃), 109.07, 112.98 and 121.71 (3C, benzoazole), 121.91, 122.33 and 123.40 (3C, phenantridine), 123.82 (1C, benzoazole), 124.06 (N-C=CH-CO₂CH₃), 124.17, 125.96, 127.79, 128.94, 129.00 and 129.19 (6C, phenantridine), 142.23 (1C, benzoazole), 154.32 (1C, benzoazole, N-C=O), 165.96 and 162.73 (2C=O, ester). Anal. Calcd for C₂₆H₂₈N₂O₆ (456.16): C, 68.40; H, 4.42; N, 6.14 %; Found: C, 68.49; H, 4.50; N, 6.23 %.

**Diethyl 2-(6-(6-chloro-2-oxobenz[d]oxazol-3(2H)-yl)phenanthridine-5(6H)-yl)fumarate 4f.** Yellow powder, yield 92%, 0.45 g mp 126-128 °C, IR (νmax, cm⁻¹): 1712 and 1773 (C=O). ¹H NMR (500.1 MHz, CDCl₃), δH 6.30 (1H, s, N-C=CH-CO₂CH₃), 7.09-8.55 (11H arom, m, 11CH), 9.33 (1H, s, NCHN). ¹³C NMR (125.8 MHz, CDCl₃), δC 52.48 and 53.50 (2OCH₃), 76.69 (NCHN), 110.10 (N-C=CH-CO₂CH₃), 110.95, 114.23 and 122.70 (3C, benzoazole), 122.91, 122.86 and 124.24 (3C, phenantridine), 123.06 (1C, benzoazole), 123.92 (N-C=CH-CO₂CH₃), 125.47, 127.42, 127.66, 128.13, 128.45, 128.92, 129.14 and 129.25 (7C, phenantridine), 131.67 (1C, benzoazole), 132.20 and 132.61 (2C, phenantridine), 142.75 (1C, benzoazole), 152.38 (1C, benzoazole, N-C=O), 161.43 and 162.33 (2C=O, ester). Anal. Calcd for C₂₆H₁₉ClN₂O₆ (490.65): C, 63.59; H, 3.90; N, 5.71 %; Found: C, 63.67; H, 3.98; N, 5.81 %.

**Diethyl 2-(6-(9H-carbazole-9-yl)phenanthridine-5(6H)-yl)fumarate 4g.** Yellow powder, yield 95%, 0.64 g mp 181-183 °C, IR (νmax, cm⁻¹): 1716 and 1756 (C=O). ¹H NMR
(500.1 MHz, CDCl3), δH 0.82 and 1.03(6H, 2t, 3JHH = 7.1 Hz, 2OCH2CH3), 3.56 and 3.84 (4H, 2m, 2ABX3system, 2OCH2CH3), 6.38 (1H, s, N-C=CH-CO2CH2CH3), 7.99-8.10 (14Haro, m, 14CH), 8.13 (1H, s, NCHN). 13C NMR (125.8 MHz, CDCl3), 13.53 and 13.74 (2OCH2CH3), 60.96 and 61.91 (2OCH2CH3), 69.53 (NCHN), 112.23 (1C, N-C=CH-CO2CH2CH3), 113.44, 115.73, 121.19 and 122.59 (5C, phenanthridine), 122.94, 123.09, 123.30, 123.69 and 123.86 (5C, carbazole), 124.41 (1C, N-C=CH-CO2CH2CH3), 124.70, 125.25, 125.30 and 127.82 (4C, phenanthridine), 129.08, 129.18 and 129.32 (3C, carbazole), 130.38, 130.56 and 131.41 (3C, phenanthridine), 138.41, 140.90, 140.99 and 141.49 (4C, carbazole), 163.30 and 163.41 (2C=O, ester). Anal. Calcd for C33H36Br2N2O4 (674.20): C, 58.74; H, 3.89; N, 4.15%, Found: C, 58.88; H, 3.92; N, 4.30%.

Di-tert-butyl 2- (6-(1,3-dimethyl-2,4,6-trioxo-hexahydropyrimidin-5-yl)phenanthridine-5(6H)-yl)fumarate 4h. Pink powder, yield 96%, 0.54 g mp 135-137 ºC, IR (νmax, cm⁻¹): 1681 and 1740 (C=O). 1H NMR (500.1 MHz, CDCl3), δH 1.30 and 1.57 (18H, 2s, 2OC(CH3)3), 2.73 and 3.38 (6H, 2s, 2NCH3), 4.69 (1H, d, 3JHH = 7.4 Hz, NCHCH), 5.01 (1H, d, 3JHH = 7.4 Hz, O=C-CH=C-O), 5.64 (1H, s, N-C=CH-CO2C(CH3)3), 6.66-7.80 (8Haro, m, 8CH). 13C NMR (125.8 MHz, CDCl3) δC 26.71 and 26.99 (6C, 2CO2C(CH3)3), 27.02 and 27.94 (2C, 2NCH3), 65.79 (1C, NCHCH), 67.05 (1C=O=CH=CH-C=O), 73.20 (1C, N-C=CH-CO2C(CH3)3), 81.62 and 81.78 (2C, 2CO2C(CH3)3), 111.65, 116.09, 118.11, 121.00 and 121.06 (6C, phenanthridine), 124.14 (1C, N-C=CH-CO2C(CH3)3), 124.39, 125.99, 128.79, 129.20, 129.67 and 143.55 (6C, phenanthridine), 149.49 (1C, N-CO-N), 166.45 and 167.04 (2C, 2C=O-C), 167.21 and 169.93 (2C, 2C=O, ester). Anal. Calcd for C31H35N3O7 (561.27): C, 66.28; H, 6.28; N, 7.48%, Found: C, 65.96; H, 6.37; N, 7.58%.

Dimethyl 2-(1-(2,4-dioxopentan-3-yl)isoquinolin-2(1H)-yl)maleate 8i. Brown powder, yield 93%, 0.35 g mp 141-143 ºC, IR (νmax, cm⁻¹): 1620 and 1731 (C=O). 1H NMR (500.1 MHz, CDCl3), δH 1.79 and 2.23 (6H, 2s, 2CH3), 3.60 and 3.80 (6H, 2s, 2OCH3), 3.90 (1H, d, 3JHH = 10.3 Hz, O=C-CH=C-O), 5.24 (1H, s, N-C=CH-CO2CH3), 5.54 (1H, d, 3JHH = 10.3 Hz, NCHCH), 6.12 (1H, d, 3JHH = 7.6 Hz, N-CH=CH, isoquinoline), 6.61 (1H, d, 3JHH = 7.6 Hz, N-CH=CH, isoquinoline), 7.04-7.40 (4Haro, m, 4CH, isoquinoline). 13C NMR (125.8 MHz, CDCl3) δC 51.79 and 53.73 (2OCH3), 59.41 (NCHCH), 69.90 (O=C-CH=C-O), 95.23 (N-C=CH-CO2CH3), 123.14 (N-C=CH-CO2CH3), 124.78, 125.70, 126.17, 127.22, 127.94, 128.22, 128.76 and 130.11 (8C, isoquinoline), 163.90 and 165.13 (2C=O, ester), 198.82 and 199.61 (2C=O). MS, m/z (%) = 228 (C14H14N2O2, 66), 208 (C13H12NO2, 47), 143 (C6H7O4, 14), 129 (C9H7N, 46), 99 (C5H3O2, 10), 59 (CO2CH3, 22). Anal. Calcd for C20H21NO6 (371.16): C, 64.66; H, 5.70; N, 3.77%, Found: C, 64.71; H, 5.63; N, 3.81%.

Dimethyl 2-(1-(1,3-dioxo-1,3-diphenylpropan-2-yl)isoquinolin-2(1H)-yl)maleate 8j. Brown powder, yield 91 %, 0.45g mp 156-158 ºC, IR (νmax, cm⁻¹): 1618 and 1736 (C=O). 1H NMR (500.1 MHz, CDCl3), δH 3.62 and 3.78 (6H, 2s, 2OCH3), 5.75 (1H, d, 3JHH = 9.9 Hz, O=C-CH=C-O), 5.86 (1H, s, N-C=CH-CO2CH3), 6.03 (1H, d, 3JHH = 9.9 Hz, NCHCH), 6.19 (1H, d, 3JHH = 7.5 Hz, N-CH=CH, isoquinoline), 6.56 (1H, d, 3JHH = 7.5 Hz, N-CH=CH, isoquinoline), 7.10-8.45 (14Haro, m, 14CH, isoquinoline and 2C6H3), 13C NMR (125.8 MHz, CDCl3) δC 51.27 and
52.81 (2OCH₃), 58.32 (NCHCH), 63.12 (O=C-CH-C=O), 97.11 (N-C=CH-CO₂CH₃), 124.09 (N-C=CH-CO₂CH₃), 125.90, 126.70 and 126.84, (3C, isoquinoline), 127.16 (1C, 2C₆H₅), 128.22 and 128.67 (2C, isoquinoline), 128.80 and 128.84 (2C, 2C₆H₅), 130.64 (1C, isoquinoline), 164.75 and 166.80 (2C=O, ester), 191.81 and 191.90 (2C=O). MS, m/z %= 180 (C₁₂H₆NO, 12), 143 (C₆H₇O₄, 54), 129 (C₆H₇N, 100), 128 (C₅H₄O₄, 29), 102 (C₇H₂O, 51), 97 (C₄H₂O₃, 9), 76 (C₆H₄, 15), 59 (CO₂CH₃, 27). Anal. Caled for C₃₀H₂₅NO₆ (495.20): C, 72.70; H, 5.09; N, 2.83 %, Found: C, 72.85; H, 4.94; N, 2.97 %.

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References