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Synthesis of Novel Bis-Enaminones by $KHSO_4$-Assisted Facile Michael Addition-Elimination Reaction of 3-(Dimethylamino)-1-phenylprop-2-en-1-ones with Diamines in Water[#]

Asem Satyapati Devi[1], Philippe Helissey[2], Jai Narain Vishwakarma[1*]
[1]*Organic Research Lab.*, *Department of Chemistry*, *St. Anthony's College*, *Shillong*, *India*
[2]*Laboratoire de Chimie Thérapeutique*, *Faculte des Sciences Pharmaceutiques et Biologiques*, *Université Paris Descartes*, *Paris*, *France*
E-mail: *jnvishwakarma@rediffmail.com*
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Abstract

3-(Dimethylamino)-1-phenylprop-2-en-1-ones (formylated acetophenones) **1** reacted with aliphatic diamines in water assisted by $KHSO_4$ to give bis-enaminones **2a-h** in good yields. Compound **1** also reacted with *o*-phenylenediamine under similar conditions to produce bis-enaminones **3** instead of benzodiazepines **4** in excellent yields.

Keywords: Enaminone, Bis-Enaminone, Formylated Acetophenone, Michael Addition-Elimination, Formylation, Dimethylformamide-Dimethylacetal

1. Introduction

Formylated products obtained by reacting active proton compounds with dimethylformamide-dimethylacetal (DMF-DMA) have proved to be very useful intermediates in the formation and modification of heterocyclic compounds [1]. Keeping in view the synthetic potential of 3-(dimethylamino)-1-phenylprop-2-en-1-ones (formylated acetophenones) [2-12], we recently reported an efficient method for the formylation of active proton compounds [13]. We subsequently developed a facile synthetic strategy for the synthesis of enaminones by reacting formylated acetophenones with primary amines [14]. These enaminones were then transformed into tetrahydropyrimidines [15] and bis-tetrahydropyrimidines [16].

In view of the environmental concerns, carrying out organic reactions in water has attracted considerable attention [17-29]. We have, from our laboratory, recently reported [30] a facile general reaction of formylated acetophenones with primary amines in water.

In connection with our synthetic studies on enaminones, we required bis-enaminones derived from acetophenones. Our literature survey at this stage revealed that bis-enaminones derived from acetophenones have received very little attention [31] and hence examination of their biological properties and synthetic potential has remained unexplored. Prompted by the above facts, we herein report a facile general strategy for the reaction of formylated acetophenones with primary diamines assisted by $KHSO_4$ in water that lead to the formation of novel bis-enaminones in excellent yields.

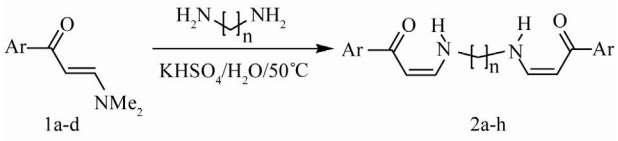
2. Results and Discussion

Thus, when formylated acetophenone **1a** was reacted with ethylenediamine in water in the presence of $KHSO_4$, bis-enaminone **2a** was obtained in 88% yields. The structure of **2a** was established as 1,2-bis-[3-oxo-3-phenylpropenylamino]ethane on the basis of spectral and analytical data. Thus, the IR spectra of **2a** showed peaks at 1581, 1638, 3249, 3388 cm^{-1} due carbonyl and NH groups. In the NMR spectra of **2a**, the NCH_2 protons resonated as multiplets at 3.44 - 3.46 ppm. The α-vinylic proton appeared as a doublet at 5.73 ppm (J = 7.2 Hz), while the β-vinylic proton gave a double-doublet at 6.88 ppm (J = 7.2, 12.4 Hz) due to its coupling with α-vinylic as well as NH protons. On D_2O shake, the signal at 5.73

[#]This paper is dedicated to Rev Fr Dr. I Warpakma, SDB on his 50th birth anniversary.

ppm remained unchanged whereas the signal at 6.88 was reduced to a doublet (J = 7.2 Hz). The aromatic protons appeared in their usual range. The NH proton resonated at 10.35 ppm indicating its hydrogen bonded state. The low coupling constant of the vinylic protons and the appearance of the NH signal at low fields confirm the Z-configuration of the enaminones.

The reaction of **1a** with 1,4-diaminobutane gave the expected products **2b** in 84% yield under identical conditions. Other formylated acetophenones **1b-e** behaved identically with aliphatic diamines forming the envisaged products in good yields (**Scheme 1**). The structures of bis-enaminones **2b-h** were also established with the help of spectral and analytical data and in all cases the enaminone moieties were found to exist exclusively in Z-form.

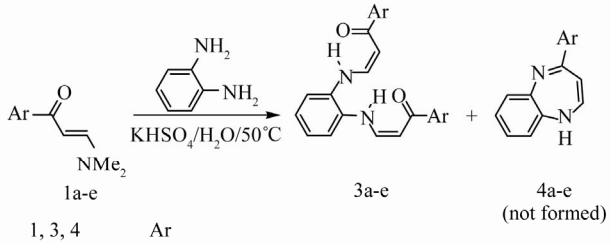


1a-d 2a-h

1	R	2	Ar	n
a	C_6H_5	a	C_6H_5	2
b	4-MeC_6H_4	b	C_6H_5	4
c	4-ClC_6H_4	c	4-MeC_6H_4	2
d	4-$MeOC_6H_4$	d	4-MeC_6H_4	4
		e	4-ClC_6H_4	2
		f	4-ClC_6H_4	4
		g	4-$MeOC_6H_4$	2
		h	4-$MeOC_6H_4$	4

Scheme 1

Subsequently, we planned to examine the reactions of *o*-phenylenediamine with **1a** envisaging the formation of benzodiazepines of the type **4 (Scheme 2**). However, when the reaction was carried out, the product isolated in 94% yield was found to be bis-enaminone **3a**, the structure of which was well established with the help of spectral and analytical data. Thus, the ^{1}H NMR spectra of **3a** showed a doublet at 6.14 ppm (J = 7.8 Hz) for the vinylic proton at C-α whereas the vinylic proton at C-β, which is expected to appear as double-doublet was obscured by the aromatic proton signals between 7.39 - 7.52 ppm. Other aromatic protons resonated in their usual range. The N-H protons in this case appeared as doublet at 12.21 ppm indicating that these are hydrogen bonded with the carbonyl group. This appearance of N-H proton at 12.21 ppm and the low coupling constant of vinylic proton at C-α confirm Z-configuration of the enaminone moieties. **1b-e** also behaved identically with o-phenylene diamine under similar conditions giving bis-enaminones **3b-e.** In none of these cases, product **4** was formed even with varying stoichiometry of **1** and o-phenylene diamine.



1a-e 3a-e 4a-e (not formed)

1, 3, 4	Ar
a	C_6H_5
b	4-McC_6H_4
c	4-ClC_6H_4
d	4-$MeOC_6H_4$
e	4-$NO_2C_6H_4$

Scheme 2

3. Experimental Section

Melting points were recorded by open capillary method and are uncorrected. The IR spectra were recorded on a Perkin-Elmer 983 spectrometer. ^{1}H NMR and ^{13}C NMR spectra were recorded on Bruker ACF-300 spectrometer. The chemical shifts (δ ppm) and the coupling constants (Hz) are reported in the standard fashion with reference to TMS as internal reference. FAB-mass spectra (MS) were measured on JEOL 3SX 102/DA-6000 mass spectrometer using argon as the carrier and m-nitro-benzylalcohol as the matrix. Elemental analyses were performed on a Vario-EL III instrument. Formylated acetophenones **1a-e** were synthesized by our previously reported procedure [13].

3.1. Reaction of 3-Dimethylamino-1-Arylpropenone 1 with Aliphatic Diamines

General Procedure. To a mixture of **1** (2 mmol) and aliphatic diamine (1 mmol) in 5 ml water was added $KHSO_4$ (2 mmol) in one lot and the resulting mixture was stirred at 50˚C - 60˚C for 1-2 hours. After the completion of the reaction (tlc), the reaction mixture was cooled and the precipitated product was collected by filtration. The product **2** thus obtained was found to be practically pure, which however was chromatographed (silica gel, ethyl acetate).

1,2-Bis-[3-oxo-3-phenylpropenylamino]ethane (2a). Pale yellow solid in 88% yield, mp 140˚C - 141˚C (lit. [31] 142˚C); IR (KBr): 1581, 1638, 3249, 3388 cm^{-1};

^{1}HNMR ($CDCl_3$): δ 3.44 - 3.46 (m, 4H), 5.73 (d, 2H, J = 7.2 Hz), 6.88 (dd, 2H, J = 7.2, 12.4 Hz), 7.39 - 7.46 (m, 6H), 7.85 - 7.87 (m, 4H), 10.35 (br m, 2H); ^{13}CNMR ($CDCl_3$): δ 50.2, 91.2, 127.1, 128.3, 131.1, 139.4, 154.4, 190.5. MS: m/z 321 (MH^+).

1,4-Bis-[3-oxo-3-phenylpropenylamino]butane (2b). Pale yellow solid in 84% yield, mp 121˚C - 122˚C; IR (KBr): 1582, 1633, 3268 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 1.53 - 1.71 (m, 4H), 3.31 - 3.33 (m, 4H), 5.71 (d, 2H, J = 7.6 Hz), 6.94 (dd, 2H, J = 7.6, 12.8 Hz), 7.39 - 7.45 (m, 6H), 7.86 - 7.87 (m, 4H), 10.38 (br m, 2H); ^{13}CNMR ($CDCl_3$): δ 28.2, 48.8, 90.3, 127.0, 128.2, 130.9, 139.6, 154.2, 190.0. MS: m/z 349 (MH^+).

Anal. Calcd for $C_{22}H_{24}N_2O_2$: C, 75.83; H, 6.94; N, 8.04. Found: C, 76.01; H, 6.88; N, 8.09%.

1,2-Bis-[3-oxo-3-(4-methylphenyl)propenylamino]ethane (2c). Pale yellow solid in 85% yield, mp 192˚C - 193˚C; IR (KBr): 1579, 1641, 3278 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 2.39 (s, 6H), 3.43 - 3.49 (m, 4H), 5.71 (d, 2H, J = 7.6 Hz), 6.85 (dd, 2H, J = 7.6, 12.4 Hz), 7.21 (d, 4H, J = 8 Hz), 7.76 (d, 4H, J = 8 Hz), 10.31 (br m, 2H). ^{13}CNMR ($CDCl_3$): δ 21.4, 50.2, 91.1, 127.2, 128.9, 136.8, 141.5, 154.1, 190.3. MS: m/z 349 (MH^+).

Anal. Calcd for $C_{22}H_{24}N_2O_2$: C, 75.83; H, 6.94; N, 8.04. Found: C, 75.70; H, 6.89; N, 8.10%.

1,4-Bis-[3-oxo-3-(4-methylphenyl)propenylamino]butane (2d). Pale yellow solid in 91% yield, mp 163˚C - 164˚C; IR (KBr): 1579, 1608, 1636, 3285, 3435 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 1.63 - 1.70 (m, 4H), 2.38 (s, 6H), 3.30 - 3.31 (m, 4H), 5.69 (d, 2H, J = 7.2 Hz), 6.91 (dd, 2H, J = 7.2, 12.8 Hz), 7.21 (d, 4H, J = 8 Hz), 7.77 (d, 4H, J = 8 Hz), 10.33 (br, s, 2H); ^{13}CNMR ($CDCl_3$): δ 21.4, 28.2, 48.8, 90.2, 127.1, 128.9, 137.0, 141.3, 153.9, 189.9; MS: m/z 377 (MH^+), 378 ($MH^+ + 1$).

Anal. Calcd for $C_{24}H_{28}N_2O_2$: C, 76.56; H, 7.50; N, 7.44. Found: C, 76.72; H, 7.43; N, 7.61%.

1,2-Bis-[3-oxo-3-(4-chlorophenyl)propenylamino]ethane (2e). Pale yellow solid in 89% yield, mp 200˚C - 201˚C; IR (KBr): 1578, 1629, 3257, 3395 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 3.45 - 3.47 (m, 4H), 5.68 (d, 2H, J = 7.6 Hz), 6.89 (dd, 2H, J = 7.6, 12.4 Hz), 7.38 (d, 4H, J = 8.4 Hz), 7.79 (d, 4H, J = 8.4 Hz), 10.35 (br, s, 2H); ^{13}CNMR ($CDCl_3$): δ 49.9, 90.6, 128.3, 128.4, 132.1, 137.3, 154.7, 186.5. MS: m/z 389 (MH^+).

Anal. Calcd for $C_{20}H_{18}Cl_2N_2O_2$: C, 61.71; H, 4.66; N, 7.20. Found: C, 61.52; H, 4.61; N, 7.11%.

1,4-Bis-[3-oxo-3-(4-chlorophenyl)propenylamino]butane (2f). Pale yellow solid in 93% yield, mp 176˚C -177˚C; IR (KBr): 1578, 1634, 3289, 3428 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 1.71 (br, s, 4H), 3.32 - 3.33 (m, 4H), 5.66 (d, 2H, J = 7.2 Hz), 6.95 (dd, 2H, J = 7.6, 12.8 Hz), 7.37 (d, 4H, J = 8.4 Hz), 7.79 (d, 4H, J = 8.4 Hz), 10.37 - 10.40 (br, m, 2H); ^{13}CNMR ($CDCl_3$): δ 28.1, 48.9, 90.7, 128.4, 128.4, 137.0, 138.0, 154.5, 188.5; MS: m/z 419 (MH^+), 417 (MH^+).

Anal. Calcd for $C_{22}H_{22}Cl_2N_2O_2$: C, 63.32; H, 5.31; N, 6.71. Found: C, 63.11; H, 5.37; N, 6.65%.

1,2-Bis-[3-oxo-3-(4-methoxyphenyl)propenylamino]ethane (2g). Pale yellow solid in 88% yield, mp 184˚C -185˚C; IR (KBr): 1582, 1601, 1645, 3293, 3433 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 3.41 - 3.43 (m, 4H), 3.85 (s, 6H), 5.68 (d, 2H, J =7.6 Hz), 6.84 (dd, 2H, J = 7.6, 12.4 Hz), 6.90 (d, 4H, J = 8.4 Hz), 7.85 (d, 4H, J = 8.4 Hz), 10.21 (br, s, 2H); MS: m/z 381 (MH^+).

Anal. Calcd for $C_{22}H_{24}N_2O_4$: C, 69.46; H, 6.36; N, 7.36. Found: C, 69.31; H, 6.43; N, 7.28%.

1,4-Bis-[3-oxo-3-(4-methoxyphenyl)propenylamino]butane (2h). Pale yellow solid in 94% yield, mp 161˚C - 162˚C; IR (KBr): 1582, 1600, 1636, 3292, 3433 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 1.69 (br, s, 4H), 3.29 - 3.30 (br, m, 4H), 3.85 (s, 6H), 5.66 (d, 2H, J = 7.2 Hz), 6.87 - 6.92 (m, 6H), 7.85 (d, 4H, J = 8.8 Hz), 10.26 - 10.29 (br m, 2H); ^{13}CNMR ($CDCl_3$): δ 28.2, 48.8, 55.3, 89.8, 113.4, 128.9, 132.4, 153.7, 161.9, 189.2; MS: m/z 409 (MH^+).

Anal. Calcd for $C_{24}H_{28}N_2O_4$: C, 70.57; H, 6.91; N, 6.86. Found: C, 70.80; H, 6.95; N, 6.81%.

3.2. Reaction of 3-Dimethylamino-1-Arylpropenone 1 with 1,2-Diaminobenzene

General Procedure. To a mixture of **1** (2 mmol) and aromatic diamine (1 mmol) in 5 ml water was added $KHSO_4$ (2 mmol) in one lot and the resulting mixture was stirred at 50˚C - 60˚C for 1.5 - 4 hours. After the completion of the reaction (tlc), the reaction mixture was cooled and the precipitated product was collected by filtration. The product **3** thus obtained was found to be practically pure, which however was chromatographed (silica gel, ethyl acetate).

1,2-Bis-[3-oxo-3-phenylpropenylamino]benzene (3a). Yellow solid, 94% yield, mp 300˚C; IR (KBr): 1597, 1625, 1639, 3422 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 6.14 (d, 2H, J = 7.8 Hz), 7.14 -7.22 (m, 4H), 7.39 - 7.52 (m, 8H), 7.95 - 7.98 (m, 4H), 12.20 (d, 2H, J = 11.4 Hz); ^{13}CNMR ($CDCl_3$): δ 95.5, 119.0, 125.1, 127.5, 128.3, 131.5, 132.1, 139.1, 146.3, 191.2; MS: m/z 369 (MH^+).

Anal. Calcd for $C_{24}H_{20}N_2O_2$: C, 78.24; H, 5.47; N, 7.60. Found: C, 78.02; H, 5.42; N, 7.66%.

1,2-Bis-[3-oxo-3-(4-methylphenyl)propenylamino]benzene (3b). Yellow solid, 88% yield, mp ›300˚C; IR (KBr): 1609, 1634, 3432 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 2.40 (s, 6H), 6.12 (d, 2H, J = 7.8 Hz), 7.12 - 7.25 (m, 8H), 7.39 (dd, 2H, J = 7.8, 11.4 Hz), 7.87 (d, 4H, J = 8.1 Hz), 12.16 (d, 2H, J = 11.4 Hz); ^{13}CNMR ($CDCl_3$): δ 21.5, 95.4, 118.8, 125.0, 127.6, 129.0, 132.1, 136.5, 142.0, 146.0, 191.0; MS: m/z 397 (MH^+).

Anal. Calcd for $C_{26}H_{24}N_2O_2$: C, 78.76; H, 6.10; N, 7.07. Found: C, 78.97; H, 6.05; N, 7.12%.

1,2-Bis-[3-oxo-3-(4-chlorophenyl)propenylamino]benzene (3c). Yellow solid, 93% yield, mp ›300˚C; IR (KBr): 1627, 3421 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 6.06 (d, 2H, *J* = 8.1 Hz), 7.12 - 7.19 (m, 4H), 7.36 -7.43 (m, 6H), 7.85 - 7.88 (m, 4H), 12.17 (d, 2H, J = 11.4 Hz); ^{13}CNMR ($CDCl_3$): *δ* 95.1, 118.9, 125.4, 128.6, 128.9, 131.9, 137.4, 137.8, 146.7, 189.8; MS: m/z 439 (MH^+), 437 (MH^+).

Anal. Calcd for $C_{24}H_{18}Cl_2N_2O_2$: C, 65.91; H, 4.15; N, 6.41. Found: C, 65.70; H, 4.09; N, 6.47%.

1,2-Bis-[3-oxo-3-(4-methoxyphenyl)propenylamino]benzene (3d). Yellow solid in 84% yield, mp ›300˚C; IR (KBr): 1601, 1624, 1636, 3430 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ 3.83 (s, 6H), 6.07 (d, 2H, *J* = 7.8 Hz), 6.89 (d, 4H, J = 8.7 Hz), 7.08 - 7.16 (m, 4H), 7.34 (dd, 2H, J = 7.8, 11.4 Hz), 7.92 (d, 4H, J = 8.7 Hz), 12.09 (d, 2H, J = 11.4 Hz); ^{13}CNMR ($CDCl_3$): *δ* 55.3, 95.2, 113.5, 118.7, 124.9, 129.6, 131.9, 132.1, 145.7, 162.4, 190.2; MS: m/z 429 (MH^+).

Anal. Calcd for $C_{26}H_{24}N_2O_4$: C, 72.88; H, 5.65; N, 6.54. Found: C, 72.69; H, 5.70; N, 6.48%.

1,2-Bis-[3-oxo-3-(4-nitrophenyl)propenylamino]benzene (3e). Red solid in 95% yield, mp 90˚C; IR (KBr): 1618, 1719, 3079, 3423 cm^{-1}; ^{1}HNMR ($CDCl_3$): *δ* 6.03 (d, 2H, J = 7.5 Hz), 6.80-6.88 (m, 2H), 6.98-7.10 (m, 2H), 7.54 (dd, 2H, J = 7.5, 12.3 Hz), 8.04 (d, 4H, *J* = 9 Hz), 8.27 (d, 4H, *J* =9 Hz), 12.23 - 12.32 (m, 2H); ^{13}CNMR ($CDCl_3$): *δ* 93.2, 117.0, 119.6, 123.1, 125.5, 127.6, 131.1, 136.7, 148.1, 187.4; MS: m/z 457 (M^+–1), 458 (M^+).

Anal. Calcd for $C_{24}H_{18}N_4O_6$: C, 62.88; H, 3.96; N, 12.22. Found: C, 63.10; H, 3.91; N, 12.15%.

4. Conclusions

In conclusion, we have developed facile environment-friendly strategy for the synthesis of hitherto unknown bis-enaminones from 3-(dimethylamino)-1-phenylprop-2-en-1-ones. Mild reaction conditions, easy work-up, excellent yields and water being used as solvent make this protocol very useful.

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