Synthesis of Ethyl 2-Amino-4-benzoyl-5-oxo-5,6-dihydro-4*H*-pyrano[3,2-*c*]quinoline-3-carboxylates by a One-pot, Three-Component Reaction in the Presence of TPAB

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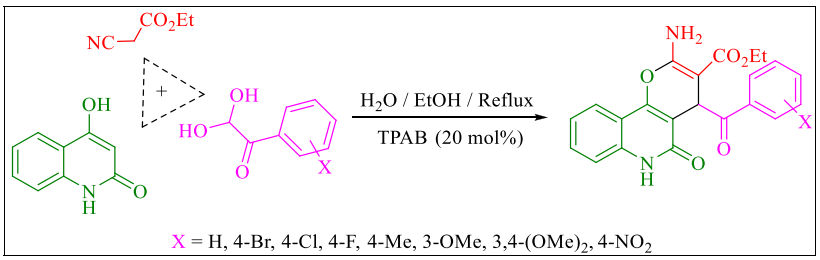
Received July 17, 2018
DOI 10.1002/jhet.3404
Published online 00 Month 2018 in Wiley Online Library (wileyonlinelibrary.com).



In this research, in order to synthesize a series of ethyl 2-amino-4-benzoyl-5-oxo-5,6-dihydro-4*H*-pyrano[3,2-*c*]quinoline-3-carboxylates, a green and an efficient method is proposed through one-pot three-component reaction of substituted arylglyoxals, ethyl cyanoacetate, and 4-hydroxyquinolin-2(1*H*)-one in the presence of terapropylammonium bromide as a catalyst in good yields. All synthesized new substances were characterized by FTIR, ^{1}H-NMR, and ^{13}C-NMR spectral data and elemental analysis.

J. Heterocyclic Chem., **00,** 00 (2018).

INTRODUCTION

Multicomponent reactions has gained remarkable attention in the field of chemical biology and organic synthesis in as much as numerous advantages such as possibility of several bonds formation in one-step process in addition to significant benefits like convergent nature, extraction, and purification steps and reducing waste production, the superior atom economy, fast, efficient, and time-saving manner without the isolation of any intermediate reduction of work ups, and easy automation and straightforward experimental techniques that resulted in the green transformations [1–6].

Tetrapropylammonium bromide (TPAB) is a phase-transfer catalyst that has been used in the preparation of high-silica zeolites of the ZSM type such as ZSM-5 and has many other applications in various catalytic processes [7,8]. TPAB is low in price, easily accessible catalyst, environmentally friendly, simple utilization, noncorrosive, and reusable.

Water is the ideal natural green solvent and an alternate for organic solvents, due to its nonflammable, nontoxic, and uniquely redox-stable and inexpensive characteristics [9].

Pyran derivatives are widely used as agrochemical and drug as a result of their unique chemical [10], biological [11], and physical properties [12].

Substantial characteristics of pyran such as antimicrobial [13], anticancer [14], antidyslipidemic [15], antihyperglycemic [16], anti-HIV [17], antioxidant [18], antiallergic [19], xanthine oxidase inhibitory [20], acetylcholinesterase and butyrylcholinesterase inhibitory [21], and Src kinase inhibitory activities [22] in addition to its presence in nature, making pyran derivatives specific structures.

The presence of pyranoquinoline moiety in many alkaloids has attracted serious attention. For instance, antitumor [23,24], antimicrobial [25], anti-inflammatory [26], antiallergic [27], antimalarial [28] inhibition of calcium signaling [29], and platelet aggregation [30] are some of the unique properties, in behalf of this attraction (Fig. 1). Moreover, some intermediates of pyranoquinolines are used in manufacturing of azo dyes which are applied in dyeing of natural and synthetic fibers [31].

In continuation of our interest in the synthesis of new and various heterocyclic compounds [32–40], an efficient protocol for the synthesis of pyrano[3,2-*c*]quinolines by multicomponent condensation of arylglyoxals, ethyl cyanoacetate, and 4-hydroxyquinolin-2(1*H*)-one in the presence of TPAB under reflux conditions was reported.

RESULTS AND DISCUSSION

According to our initial studies, the reaction of 3,4-dimethoxyphenylglyoxal monohydrate (**1g**), ethyl cyanoacetate (**2**), and 4-hydroxyquinolin-2(1*H*)-one (**3**) was chosen as a model reaction (Scheme 1) by stirring the mixture at 50°C; however, the expected product formation was not achieved even after 12 h. The reaction was carried out using different amounts of catalysts and also solvent systems as mentioned in Table 1.

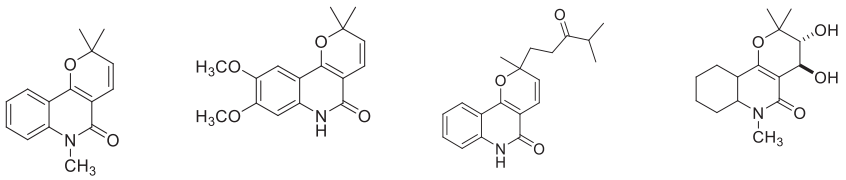


Figure 1. Drug molecules having pyranoquinoline moieties.

Scheme 1. The model reaction for synthesis of **4g**. [Color figure can be viewed at wileyonlinelibrary.com]

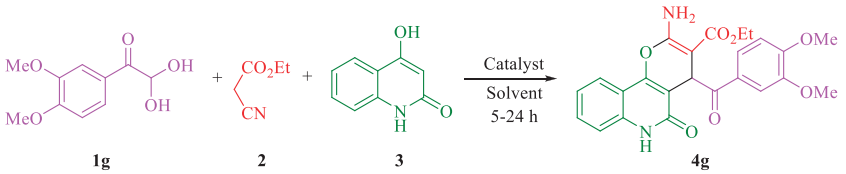


Table 1

Optimization of the reaction conditions.

Entry	Solvent	Conditions	Catalyst (mol%)	Time (h)	Yield (%)
1	H_2O/EtOH (1:1)	40–50	TPAB (20)	12	-
2	H_2O	Reflux	TPAB (20)	24	40
3	EtOH	Reflux	TPAB (20)	24	35
4	**H_2O/EtOH (1:1)**	**Reflux**	**TPAB (20)**	**5**	**91**
5	H_2O/EtOH (1:1)	Reflux	TPAB (15)	10	85
6	H_2O/EtOH (1:2)	Reflux	TPAB (25)	8	88
7	Acetone	60	TPAB (20)	10	25
8	DMF	Reflux	TPAB (20)	14	-
9	AcOH	Reflux	TPAB (20)	15	-
10	H_2O/EtOH (1:1)	Reflux	Sulfanilic acid (20)	24	-
11	H_2O/EtOH (1:1)	Reflux	E_3N (20)	15	20
12	H_2O/EtOH (1:1)	Reflux	-	12	25
13	H_2O/EtOH (1:1)	Reflux	TEACB (20)	24	58
14	H_2O/EtOH (1:1)	Reflux	*L*-proline (20)	10	67
15	H_2O/EtOH (1:1)	Reflux	*L*-cysteine (20)	12	-
16	H_2O/EtOH (1:1)	Reflux	*p*-TSA (20)	18	-

Bold values show the best reaction conditions in terms of yield. DMF, dimethylformamide; TPAB, tetrapropylammonium bromide.

The highest yield of 91% was obtained using 20 mol% of TPAB as a catalyst in H_2O/EtOH (1:1) at 5 h reaction time (Table 1, entry 4). In order to find out the best solvent system for this reaction, we accomplished the trail reaction using various solvents such as EtOH, H_2O, H_2O/EtOH (1:1), H_2O/EtOH (1:2), acetone, dimethylformamide, and AcOH as shown in Table 1. Among all these solvents, H_2O/EtOH (1:1) was proved to be the best solvent system in terms of yield (Table 1, entry 4).

Furthermore, the effect of different catalysts on the reaction was conducted through different catalytic systems including sulfanilic acid, *p*-toluenesulfonic acid, *L*-proline, *L*-cysteine, triethylamine, and different amounts of TPAB ranging from 15 to 25 mol%. The best results were obtained using 20 mol% of catalyst in H_2O/EtOH (1:1). Also, increase in the amount of catalyst did not improve the yield.

In the next step and after optimization of the reaction conditions, the scope of the reaction was verified with different arylglyoxals to form the desired pyranoquinoline derivatives **4a**–**h**. The reaction conditions and the yields of products are listed in Table 2.

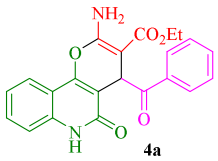
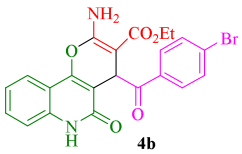
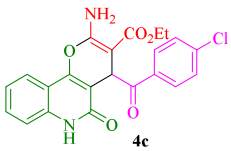
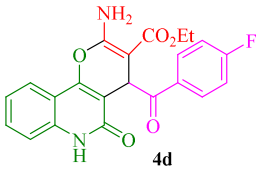
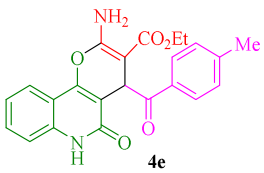
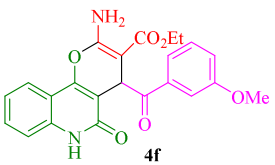
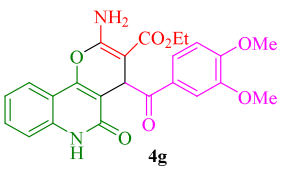
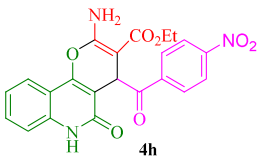
All arylglyoxal monohydrates **1a**–**h** were prepared from the corresponding acetophenones by oxidation procedure with selenium dioxide *via* the literature procedure as shown in Scheme 2 [41].

The one-pot three-component reaction of arylglyoxal monohydrates **1a**–**h**, ethyl cyanoacetate (**2**), and 4-hydroxyquinolin-2(1*H*)-one (**3**) in H_2O/EtOH (1:1) in the presence of TPAB (20 mol%) as a catalyst afforded the corresponding 2-amino-pyrano[3,2-*c*]quinolines in 80–91% yield as shown in Scheme 3.

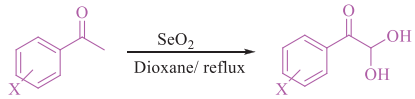
The proposed mechanism of the reaction is shown in Scheme 4. Mechanism includes dehydration of the

Table 2

Synthesis of pyranoquinoline derivatives.

Entry	Pyrano[3,2-*c*]quinoline	Reaction time (h)	Yield (%)	Mp (°C)	Color
1	 **4a**	6	88	227–229	White
2	 **4b**	5	83	239–241	Yellow
3	 **4c**	6	86	244–246	White
4	 **4d**	5	87	245–247	White
5	 **4e**	5	85	240–242	White
6	 **4f**	6	80	228–230	White
7	 **4g**	5	91	232–234	Cream
8	 **4h**	4	81	225–227	Brown

Scheme 2. Preparation of arylglyoxal monohydrates. [Color figure can be viewed at wileyonlinelibrary.com]



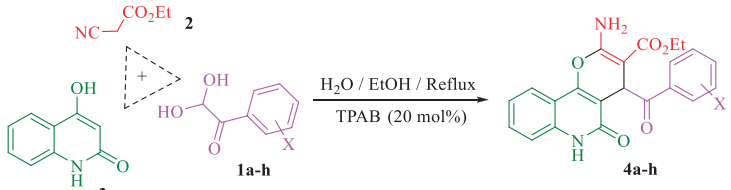
X = H, 4-Br, 4-Cl, 4-F, 4-Me, 3-OMe, 3,4-$(OMe)_2$, 4-NO_2

arylglyoxal **1a–h** by the catalyst following by a *Knoevenagel* condensation occurring between ethyl cyanoacetate (**2**) and arylglyoxal that is facilitated by TPAB *via* C–H activation on ethyl cyanoacetate which reacts with 4-hydroxyquinolin-2(1*H*)-one (**3**) to give the intermediate [**A**] that undergoes heterocyclization and tautomerization to obtain the desired products.

The structures of ethyl-2-amino-4-benzoyl-5-oxo-5,6-dihydro-4*H*-pyrano[3,2-*c*]quinoline-3-carboxylate **4a–h** were characterized by FTIR, ^{1}H-NMR, and ^{13}C-NMR spectral data. The characteristic singlet peaks at around δ = 11.65–11.78 and δ = 7.86–7.96 ppm are related to the NH group and NH_2 of the pyranoquinoline moiety, respectively, which were present in all new products. In the ^{13}C-NMR spectra of the products **4a–h**, signals located around δ = 161.4–198.6 ppm were attributed to different carbonyl groups. In the obtained FTIR spectra, the characteristic bonds at 3387–3407, 3288–3302, and 1669–1682 cm^{-1} could be assigned to the vibrations of NH, NH_2, and different carbonyl groups, respectively.

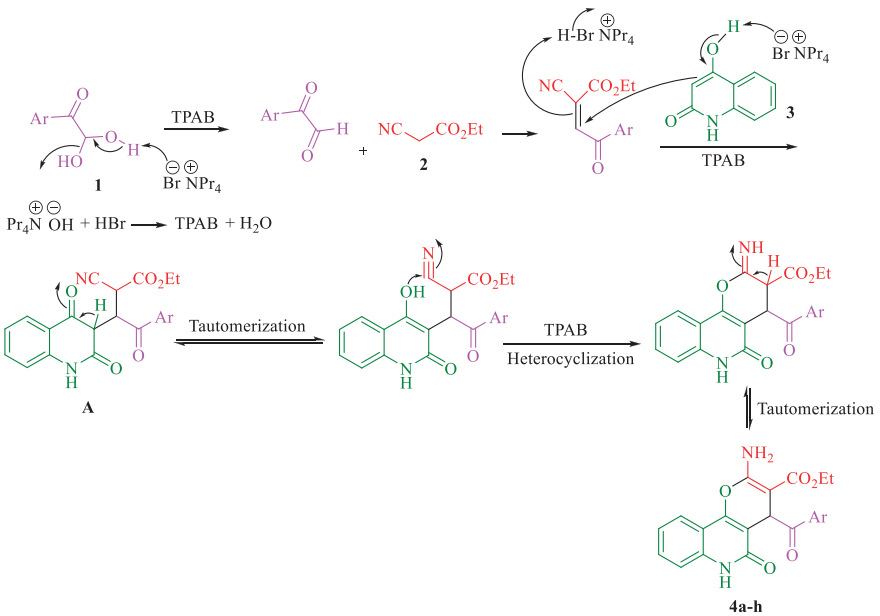
In another study, reusability of the catalyst was investigated upon the reaction of 3,4-dimethoxyphenylglyoxal monohydrate and ethyl cyanoacetate with 4-hydroxyquinolin-2(1*H*)-one under reflux condition to provide compound **4g**. After completion of the reaction, the obtained product **4g** was filtered out, followed by rising them with water, eventually, the filtrate was extracted with $CHCl_3$ and aqueous phase was separated. The water was evaporated to give TPAB as white crystals, which may be recrystallized from Et_2O. It was observed that the catalytic activity of TPAB was restored for four runs.

Scheme 3. One-pot synthesis of substituted 2-amino-pyrano[3,2-*c*]quinolines. [Color figure can be viewed at wileyonlinelibrary.com]



X = H, 4-Br, 4-Cl, 4-F, 4-Me, 3-OMe, 3,4-$(OMe)_2$, 4-NO_2

Scheme 4. Proposed mechanism for the synthesis of 2-amino-pyrano[3,2-*c*]quinolines. [Color figure can be viewed at wileyonlinelibrary.com]



CONCLUSION

In brief, we have reported a new one-pot three-component synthesis of a series of ethyl 2-amino-4-benzoyl-5-oxo-5,6-dihydro-4*H*-pyrano[3,2-*c*]quinoline-3-carboxylate derivatives **4a**–**h** in the presence of TPAB as a catalyst. The proposed method has got some considerable advantages such as using an inexpensive and recoverable catalyst, a simple workup process, using water as a green solvent, short reaction times, and high yields.

EXPERIMENTAL

Melting points were determined on a digital melting point apparatus (Electrothermal, Cole-Parmer, Staffordshire, UK) and reported uncorrected. Infrared spectra were recorded on a Thermo Nicolet (Nexus 670) FTIR spectrometer (Thermo Fisher Scientific, Waltham, MA), using KBr disks. ^{1}H and ^{13}C nuclear magnetic resonance (NMR) spectra were recorded with a Bruker spectrometer at 300 and 75 MHz, respectively (Bruker, Billerica, MA). The spectra were measured in DMSO-d_6 using tetramethylsilane as the internal standard. Elemental analyses were performed using a Leco Analyzer 932 (Leco Corp., St. Joseph, MI).

General procedure for synthesis of new pyranoquinoline derivatives 4a–h. The arylglyoxal monohydrates (1 mmol) was dissolved in H_2O/EtOH (1:1) (6 mL), and then, ethyl cyanoacetate (1 mmol), catalyst (TPAB) (20 mol%), and 4-hydroxyquinolin-2(1*H*)-one (1 mmol) were added to the reaction mixture. The reaction mixture was heated at reflux for the appropriate time as indicated in Table 2. The reaction completion was monitored by thin-layer chromatography using (EtOAc/hexane/MeOH, 1:1:1) as eluent. The precipitate was filtered and washed with (chloroform/hexane 1:2) to give the desired products as white to brown powders in 80–91% yields.

Ethyl 2-amino-4-benzoyl-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4a). Yield, 88%; white powder; mp 227–229°C. FTIR (ν_{max}, cm^{-1}): 3408, 3300, 2971, 2843, 1676, 1523, 1375, 1272, 1218, 1084, 747. ^{1}H-NMR δ (ppm): 11.73 (s, exchanged by D_2O addition, 1H, NH), 8.16 (d, *J* = 6.6 Hz, 2H, ArH), 7.94 (t, *J* = 7.8 Hz, 1H, ArH), 7.93 (s, exchanged by D_2O addition, 2H, NH_2), 7.60–7.54 (m, 3H, ArH), 7.58 (d, *J* = 7.8 Hz, 2H, ArH), 7.37–7.30 (m, 1H, ArH), 5.63 (s, CH, 1H), 3.75 (bs, 2H, CH_2), 0.68 (bs, 3H, CH_3). ^{13}C-NMR δ (ppm): 193.8, 167.9, 161.5, 160.6, 152.8, 138.2, 137.9, 132.8, 130.7, 130.6, 129.5, 128.4, 123.5, 116.8, 115.0, 114.9, 112.5, 109.9, 73.8, 14.7. *Anal*. Calcd for $C_{22}H_{18}N_2O_5$: C, 67.69; H, 4.65; N, 7.18. Found: C, 67.58; H, 4.79; N, 7.01%.

Ethyl 2-amino-4-(4-bromobenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4b). Yield, 83%; yellow powder; mp 239–241°C. FTIR (ν_{max}, cm^{-1}): 3401, 3292, 2964, 2847, 1667, 1525, 1377, 1271, 1081, 755 cm^{-1}. ^{1}H-NMR δ (ppm): 11.74 (s, exchanged by D_2O addition, 1H, NH), 7.94 (d, *J* = 8.4 Hz, 2H, ArH), 7.93–7.90 (m, 1H, ArH), 7.92 (s, exchanged by D_2O addition, 2H, NH_2), 7.75 (d, *J* = 8.4 Hz, 2H, ArH), 7.66 (d, *J* = 8.4 Hz, 1H, ArH), 7.59 (t, *J* = 8.1 Hz, 1H, ArH), 7.47 (d, *J* = 8.1 Hz, 1H, ArH), 5.57 (s, 1H, CH), 3.79 (q, *J* = 6.6 Hz, 2H, CH_2), 0.71 (t, *J* = 6.9 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 197.7, 176.1, 167.9, 161.5, 160.5, 152.7, 140.5, 139.9, 138.2, 137.1, 132.7, 132.5, 130.4, 127.1, 123.6, 112.4, 109.6, 105.9, 73.7, 14.7. *Anal*. Calcd for $C_{22}H_{17}BrN_2O_5$: C, 56.31; H, 3.65; N, 5.97. Found: C, 56.20; H, 3.74; N, 5.87%.

Ethyl 2-amino-4-(4-chlorobenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4c). Yield, 86%; white powder; mp 244–246°C. FTIR (ν_{max}, cm^{-1}): 3404, 3294, 2957, 2839, 1670, 1593, 1526, 1378, 1271, 1083, 757. ^{1}H-NMR δ (ppm): 11.75 (s, exchanged by D_2O addition, 1H, NH), 8.18 (d, *J* = 7.8 Hz, 2H, ArH), 7.95–7.92 (m, 1H, ArH), 7.92 (s, exchanged by D_2O addition, 2H, NH_2), 7.60 (d, *J* = 8.7 Hz, 2H, ArH), 7.59–7.52 (m, 1H, ArH), 7.35 (d, *J* = 9.3 Hz, 1H, ArH), 7.31 (t, *J* = 7.2 Hz, 1H, ArH), 5.58 (s, 1H, CH), 3.78 (q, *J* = 6.6 Hz, 2H, CH_2), 0.70 (t, *J* = 6.6 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 196.6, 167.9, 161.5, 160.5, 152.7, 138.2, 136.7, 132.4, 129.7, 127.5, 123.5, 121.6, 121.5, 116.9, 115.0, 114.8, 112.4, 109.6, 73.7, 14.8. *Anal*. Calcd for $C_{22}H_{17}ClN_2O_5$: C, 62.20; H, 4.03; N, 6.59. Found: C, 62.33; H, 3.91; N, 6.41%.

Ethyl 2-amino-4-(4-fluorobenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4d). Yield, 87%; white powder; mp 245–247°C. FTIR (ν_{max}, cm^{-1}): 3407, 3302, 2971, 2850, 1670, 1513, 1378, 1273, 1221, 1083, 757. ^{1}H-NMR δ (ppm): 11.65 (s, exchanged by D_2O addition, 1H, NH), 8.24 (d, *J* = 7.8 Hz, 2H, ArH), 7.91 (s, exchanged by D_2O addition, 2H, NH_2), 7.70 (d, *J* = 8.7 Hz, 2H, ArH), 7.57 (d, *J* = 7.2 Hz, 1H, ArH), 7.47 (d, *J* = 8.1 Hz, 1H, ArH), 7.36 (t, *J* = 7.2 Hz, 1H, ArH), 7.31–7.12 (m, 1H, ArH), 5.60 (s, 1H, CH), 3.79 (q, *J* = 7.2 Hz, 2H, CH_2), 0.70 (t, *J* = 8.7 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 194.2, 167.9, 163.9, 162.8, 161.5, 160.6, 152.7, 139.7, 138.2, 134.7, 131.4, 123.2, 122.0, 121.9, 116.3, 114.6, 112.4, 109.6, 73.8, 14.7. *Anal*. Calcd for $C_{22}H_{17}FN_2O_5$: C, 64.70; H, 4.20; N, 6.86. Found: C, 64.85; H, 4.11; N, 6.70%.

Ethyl 2-amino-4-(4-methylbenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4e). Yield, 85%; white powder; mp 240–242°C. FTIR (ν_{max}, cm^{-1}): 3387, 3288, 2947, 2843, 1666, 1532, 1377, 1268, 1179, 1082, 1022, 748. ^{1}H-NMR δ (ppm): 11.72 (s, exchanged by D_2O addition, 1H, NH), 8.07 (d, *J* = 8.4 Hz, 2H, ArH),

7.93 (t, J = 6.9 Hz, 1H, ArH), 7.89 (s, exchanged by D_2O addition, 2H, NH_2), 7.58 (t, J = 8.1 Hz, 1H, ArH), 7.35 (d, J = 8.1 Hz, 1H, ArH), 7.32 (d, J = 7.2 Hz, 2H, ArH), 7.31–7.27 (m, 1H, ArH), 5.60 (s, 1H, CH), 3.79 (q, J = 7.2 Hz, 2H, CH_2), 0.75 (t, J = 6.9 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 192.0, 167.9, 161.4, 160.7, 152.8, 143.5, 138.2, 138.1, 135.2, 130.9, 130.8, 128.7, 128.1, 123.5, 116.9, 114.9, 112.5, 109.9, 73.9, 21.6, 14.7. *Anal*. Calcd for $C_{23}H_{20}N_2O_5$: C, 68.31; H, 4.98; N, 6.93. Found: C, 68.52; H, 4.83; N, 6.87%.

Ethyl 2-amino-4-(3-methoxybenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4f). Yield, 80%; white powder; mp 228–230°C. FTIR (ν_{max}, cm^{-1}): 3409, 3300, 2960, 2837, 1682, 1592, 1522, 1374, 1260, 1085, 759. ^{1}H-NMR δ (ppm): 11.75 (s, exchanged by D_2O addition, 1H, NH), 7.94 (t, J = 7.8 Hz, 1H, ArH), 7.91 (s, exchanged by D_2O addition, 2H, NH_2), 7.79 (d, J = 7.8 Hz, 1H, ArH), 7.61 (s, 1H, ArH), 7.57 (t, J = 7.6 Hz, 1H, ArH), 7.47 (t, J = 7.8 Hz, 1H, ArH), 7.38–7.28 (m, 2H, ArH), 7.24–7.19 (m, 1H, ArH), 5.59 (s, 1H, CH), 3.82 (s, 3H, OCH_3), 3.80 (q, J = 6.9 Hz, 2H, CH_2), 0.73 (t, J = 6.9 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 198.6, 167.9, 161.5, 160.6, 159.4, 152.8, 139.3, 139.2, 138.2, 132.8, 130.6, 128.5, 123.2, 121.0, 119.8, 114.9, 112.5, 109.8, 73.9, 54.9, 14.7. *Anal*. Calcd for $C_{23}H_{20}N_2O_6$: C, 65.71; H, 4.80; N, 6.66. Found: C, 65.66; H, 4.70; N, 6.53%.

Ethyl 2-amino-4-(3,4-dimethoxybenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4g). Yield, 91%; cream powder; mp 232–234°C. FTIR (ν_{max}, cm^{-1}): 3390, 3293, 2946, 2843, 1669, 1517, 1377, 1250, 1160, 1083, 1025, 763. ^{1}H-NMR δ (ppm): 11.71 (s, exchanged by D_2O addition, 1H, NH), 7.93–7.87 (m, 2H, ArH) 7.86 (s, exchanged by D_2O addition, 2H, NH_2), 7.63 (s, 1H, ArH), 7.58–7.55 (m, 1H, ArH), 7.36–7.28 (m, 2H, ArH), 7.13–7.10 (m, 2H, ArH), 5.61 (s, 1H, CH), 3.86 (s, 3H, OCH_3), 3.85 (q, J = 8.1 Hz, 2H, CH_2), 3.80 (s, 3H, OCH_3), 0.82 (t, J = 8.1 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 193.6, 167.9, 161.4, 160.8, 152.9, 148.4, 138.2, 130.3, 126.1, 123.4, 123.2, 116.8, 114.8, 113.1, 112.6, 110.9, 110.0, 109.9, 74.1, 55.3, 55.1, 14.9. *Anal*. Calcd for $C_{24}H_{22}N_2O_7$: C, 64.00; H, 4.92; N, 6.22. Found: C, 64.20; H, 4.89; N, 6.12%.

Ethyl 2-amino-4-(4-nitrobenzoyl)-5-oxo-5,6-dihydro-4H-pyrano[3,2-c]quinoline-3-carboxylate (4h). Yield, 81%; brown powder; mp 225–227°C. FTIR (ν_{max}, cm^{-1}): 3405, 3299, 2975, 2856, 1520, 1681, 1345, 1281, 1223, 1090, 756. ^{1}H-NMR δ (ppm): 11.78 (s, exchanged by D_2O addition, 1H, NH), 8.39 (d, J = 7.5 Hz, 2H, ArH), 8.38–8.33 (m, 1H, ArH), 7.96 (s, exchanged by D_2O addition, 2H, NH_2), 7.94–7.90 (m, 1H, ArH), 7.62–7.56 (m, 2H, ArH), 7.38–7.28 (m, 2H, ArH), 5.62 (s, CH, 1H), 3.74 (q, J = 7.8 Hz, 2H, CH_2), 0.64 (t, J = 7.2 Hz, 3H, CH_3). ^{13}C-NMR δ (ppm): 194.2, 167.9, 163.9, 162.8, 161.5, 160.6, 152.7, 139.7, 138.2, 134.7, 131.4, 123.2, 122.0, 121.9, 116.3, 114.6, 112.4, 109.6, 73.8, 14.9. *Anal*. Calcd for $C_{22}H_{17}N_3O_7$: C, 60.69; H, 3.94; N, 9.65. Found: C, 60.83; H, 4.09; N, 9.55%.

Acknowledgment. The authors gratefully acknowledge the financial support from Urmia University.

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