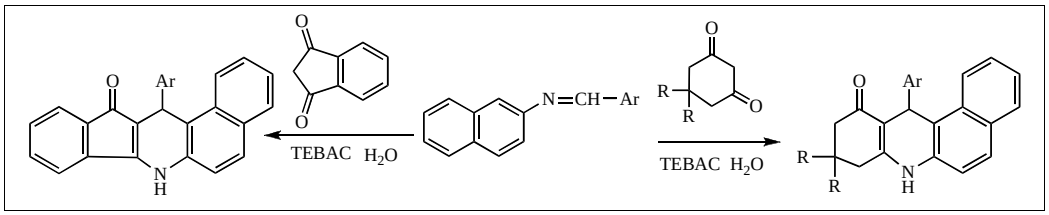
A Clean Synthesis of Polyhydroacridine and Indenoquinoline Derivatives Catalyzed by Triethylbenzylammonium Chloride in Aqueous Media

Xiang-Shan Wang*[a,b,c], Mei-Mei Zhang[a], Zhao-Sen Zeng[a], Da-Qing Shi[a,b], Shu-Jiang Tu[a,b], Xian-Yong Wei[c], Zhi-Min Zong[c]

[a]Department of Chemistry, Xuzhou Normal University, Xuzhou, Jiangsu 221116 China
[b]The Key Laboratory of Biotechnology for Medical Plant of Jiangsu Province, Xuzhou, Jiangsu 221116 China
[c]School of Chemical Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221008 China

Received October 11, 2005



An efficient and convenient synthesis of benzo[*a*]acridines and indeno[1,2-*b*]benzo[*f*]quinolines was achieved in high yields by the reaction of *N*-arylidenenaphthalen-2-amine with 1,3-dicarbonyl compounds catalyzed with triethylbenzylammmonium chloride (TEBAC) in aqueous media. The structures were established by spectroscopic data and further confirmed by X-ray analysis. This method provides several advantages such as neutral conditions, high yields and simple work-up procedure. In addition, water was chosen as a green and recyclable solvent.

J. Heterocyclic Chem., **43**, 989 (2006).

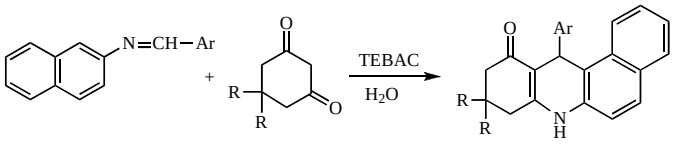
Introduction.

Nitrogen-containing heteroaromatic compounds often play important roles as the scaffolds of bioactive substances. Quinoline is one of the most popular *N*-heteroaromatics incorporated into the structure of many pharmaceuticals. It is known that many quinoline containing compounds exhibit a wide spectrum of pharmacological activities such as antiasthmatic, anti-inflammatory and antimalarial [1]. In addition, acridine derivatives containing 1,4-dihydropyridine (1,4-DHP), are reported as well-known compounds for their pharmacological profile in calcium channel modulations [2]. The chemical modifications on the DHP ring, such as different substituents [3] or heteroatoms [4], have allowed the study of the extended structure and activity relationship, and also provided some insight into the molecular interactions at the receptor level. The general method for the synthesis of acridine derivatives are in conventional organic solvents [5-7]. In order to avoid the disadvantages such as toxicity and instability that many organic solvents inherently have, we have working to find a new procedure that will be environmentally friendly, and easy to operate for the synthesis of those above-mentioned compounds. Specifically, we focused our attention on the use of water as reaction medium. They were considered very promising and attractive substitutes for volatile organic solvents and were widely used in the Green Chemistry area, since Breslow [8] demonstrated hydrophobic effects could strongly enhance the rate of some organic reactions and rediscovered the use of water as solvent in organic chemistry in 1980s. There has been growing recognition that water is an attractive media for many organic reactions resulting in less expensive, less dangerous and environment-friendly, such as Diels-Alder reactions [9], Claisen Rearrangement reactions [10], Reformatsky reactions [11] and Pinacol-coupling reactions [12]. As part of our current studies on the development of new routes to heterocyclic systems [13], we now report an efficient and clean synthetic route to the derivatives of benzo[*a*]acridine and indeno[1,2-*b*]benzo[*f*]quinoline in aqueous media catalyzed by TEBAC, through the reaction of *N*-arylidenenaphthalen-2-amine with 1,3-dicarbonyl compounds.

Results and Discussion.

When the reaction of *N*-arylidenenaphthalen-2-amine **1a-1v** and substituted 1,3-cyclohexadione **2** was performed in water in the presence of TEBAC at 100 ℃, high yields of benzo[*a*]acridine derivatives **3** were obtained (Scheme 1).

Scheme 1



1 **2** **3**

In order to apply this reaction to a library synthesis, various kinds of **1a-1u** and **2** were selected to give the corresponding acridine **3**, and representative examples are

shown in Table 1. All of the **1a-1v** gave expected products in quantitative yields and purity. The isolated acridines **3** were characterized by IR, ^{1}H NMR and elemental analyses. The analysis were in agreement with

Table 1

Results of the reaction **1** and **2** in water at 100 ºC [a].

Entry	R	Ar	Time (h)	Yields (%)[b]
3a	Me	C_6H_5	8	88
3b	Me	2,4-$Cl_2C_6H_3$	2	99
3c	Me	3,4-$Cl_2C_6H_3$	2	87
3d	Me	4-ClC_6H_4	3	98
3e	Me	3-$NO_2C_6H_4$	3	92
3f	Me	3-ClC_6H_4	2	96
3g	Me	3,4-$(CH_3O)_2C_6H_3$	10	96
3h	Me	4-OHC_6H_4	8	87
3i	Me	4-$CH_3OC_6H_4$	5	97
3j	Me	4-BrC_6H_4	4	87
3k	Me	3,4-$(CH_3)_2C_6H_3$	6	95
3l	Me	3-OH-4-$CH_3OC_6H_3$	12	99
3m	Me	4-$(CH_3)_2NC_6H_4$	10	89
3n	Me	2-ClC_6H_4	3	94
3o	Me	2-thiophenyl	5	98
3p	H	4-BrC_6H_4	10	86
3q	H	4-$CH_3OC_6H_4$	10	85
3r	H	4-FC_6H_4	8	89
3s	H	4-ClC_6H_4	8	88
3t	H	3-ClC_6H_4	8	82
3u	H	C_6H_5	12	80
3v	H	4-OHC_6H_4	12	81

[a] Reaction conditions: 10 mL water, 0.1 g TEBAC, 2 mmoles of **1** and 2 mmoles of **2**; [b] Isolated yields.

their structures. The IR spectra for **3a** exhibited sharp bands at 3236 cm^{-1} (NH). The ^{1}H NMR spectrum of **3a** exhibited a singlet identified as methine (5.79 ppm) along with multiplets (6.97–7.95 ppm) for aromatic protons. The NH proton resonance at 9.71 ppm disappeared after addition of D_2O to the DMSO-d_6 solution of **3a**.

It is interesting that the positions of the two methyl groups are different due to the conformation of the hexagonal ring: one methyl is in the pseudoaxial site while the other is in the pseudoequatorial site, so they exhibit two singlets at 0.84 and 1.03 ppm respectively, due to the different shielding degree. Furthermore the four protons of the methylene also form four double peaks at 2.03 (16.0 Hz), 2.22 (16.0 Hz), 2.40 (16.8 Hz) and 2.55 (16.8 Hz), respectively, due to the conformation of the hexagonal ring. In order to further confirm the structure of the product, X-ray analysis [14] of **3d** was carried out. The structure of **3d** is shown in Figure 1.

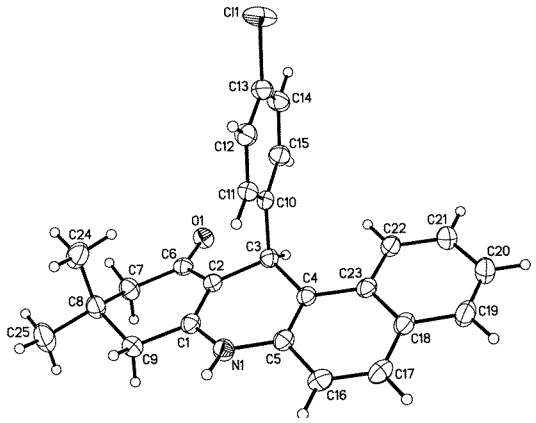


Figure 1. The crystal structure of the product **3d**.

Lastly, we studied the reuse of the water and TEBAC. After disappearance of starting materials monitered by TLC, a reaction mixture was allowed to cool down to room temperature, the solid products were isolated by filtration, and the filtrate of the water together with TEBAC could be reused directly. The yield of the product **3a** is fairly high (87%) even in the fourth cycle.

To expand the reaction scope of Schiff base with 1,3-dicarbonyl compounds, we tried the reaction of **1** with 1,3-indenedione **4**, as is expected, when the substituted 1,3-cyclohexadione **2** was replaced by 1,3-indenedione, indeno[1,2-*b*]benzo[*f*] quinoline **5** were obtained under the same reaction conditions in good yields (Scheme 2). The results are summarized in Table 2.

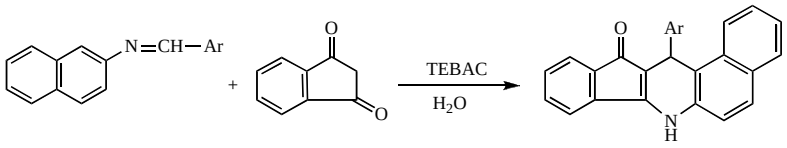
Table 2

Results of the reaction **1** and **4** in water at 100 ºC [a].

Entry	Ar	Time (h)	Yields (%)[b]
5a	2-thiophenyl	18	87
5b	4-$CH_3OC_6H_4$	18	90
5c	4-FC_6H_4	10	95
5d	3-ClC_6H_4	10	92
5e	C_6H_5	18	93
5f	4-ClC_6H_4	12	94
5g	2,4-$Cl_2C_6H_3$	12	80
5h	3,4-$Cl_2C_6H_3$	12	84
5i	4-OHC_6H_4	20	92
5j	3,4-$(CH_3)_2C_6H_3$	12	92

[a] Reaction condition: 10 mL water, 0.1 g TEBAC, 2 mmoles of **1** and 2 mmoles of **4**; [b] Isolated yields.

Scheme 2



1 **4** **5**

In conclusion, an efficient green chemistry method for the synthesis of benzo[*a*]acridine and indeno[1,2-*b*]benzo-[*f*]quinoline derivatives by condensation reaction of 1,3-dicarbonyl compounds and *N*-arylidenenaphthalen-2-amine was successfully established.

EXPERIMENTAL

Melting points were determined in open capillaries and are uncorrected. IR spectra were recorded on a TENSOR 27 spectrometer in KBr pellet. ^{1}H NMR spectra were obtained in DMSO-d_6 with Me_4Si as internal standard using a Bruker-400 spectrometer. Elemental analyses were carried out using Carlo Erba 1110 analyzer. X-ray diffraction was measured on a Rigaku Mercury diffractometer.

General Procedure for the Synthesis of Benzo[*a*]acridines (**3**).

A suspension of a mixture of *N*-arylidenenaphthalen-2-amine **1** (2 mmoles), substituted 1,3-cyclohexadione **2** (2 mmoles) and TEBAC (0.1 g) was stirred in water (10 mL) at 100 ºC for 2-12 hours. The crystalline precipitates were collected by filtration, washed with water and recrystallized from DMF and water to give pure benzo[*a*]acridines **3**.

9,9-Dimethyl-12-phenyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3a**).

This compound was obtained as pale yellow crystals, mp >300 ℃ (Lit [7] 336~337 ºC); ir (KBr): ν_{max} 3236, 3077, 3021, 2961, 1578, 1520, 1467, 1428, 1386, 1337, 1261, 1237, 1152, 1035, 816, 765, 752, 729, 698; ^{1}H nmr (DMSO-d_6): δ 0.84 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.03 (d, J = 16.0 Hz, 1H, CH), 2.22 (d, J = 16.0 Hz, 1H, CH), 2.40 (d, J = 16.8 Hz, 1H, CH), 2.55 (d, J = 16.8 Hz, 1H, CH), 5.79 (s, 1H, CH), 6.97~7.01 (m, 1H, ArH), 7.12 (t, J = 7.6 Hz, 2H, ArH), 7.22~7.25 (m, 2H, ArH), 7.29~7.33 (m, 2H, ArH), 7.40~7.44 (m, 1H, ArH), 7.77~7.81 (m, 2H, ArH), 7.95 (d, J = 8.4 Hz, 1H, ArH), 9.71 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{23}NO$: C 84.95, H 6.56, N 3.96. Found: C 84.78, H 6.54, N 4.07.

12-(2,4-Dichlorophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3b**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3244, 3078, 2956, 2932, 1580, 1557, 1521, 1467, 1428, 1382, 1259, 1245, 1184, 1150, 1099, 1035, 1044, 869, 846, 819, 787, 757, 745; ^{1}H nmr (DMSO-d_6): δ 0.87 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 1.99 (d, J = 16.4 Hz, 1H, CH), 2.22 (d, J = 16.4 Hz, 1H, CH), 2.40 (d, J = 16.4 Hz, 1H, CH), 2.59 (d, J = 16.4 Hz, 1H, CH), 5.99 (s, 1H, CH), 7.21 (dd, J = 8.4 Hz, J' = 2.4 Hz, 1H, ArH), 7.28~7.34 (m, 3H, ArH), 7.38 (d, J = 2.4 Hz, 1H, ArH), 7.42~7.46 (m, 1H, ArH), 7.79 (d, J = 8.8 Hz, 1H, ArH), 7.80 (d, J = 7.6 Hz, 1H, ArH), 8.06 (d, J = 8.4 Hz, 1H, ArH), 9.84 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{21}Cl_2NO$: C 71.10, H 5.01, N 3.32. Found C 70.92, H 5.24, N 3.13.

12-(3,4-Dichlorophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3c**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3272, 3073, 2957, 1600, 1582, 1519, 1467, 1428, 1194, 1149, 1130, 1030, 817, 781, 744; ^{1}H nmr (DMSO-d_6): δ 0.85 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.05 (d, J = 16.0 Hz, 1H, CH), 2.24 (d, J = 16.0 Hz, 1H, CH), 2.41 (d, J = 16.4 Hz, 1H, CH), 2.56 (d, J = 16.4 Hz, 1H, CH), 5.83 (s, 1H, CH), 7.14 (dd, J = 8.4 Hz, J' = 2.0 Hz, 1H, ArH), 7.32~7.35 (m, 2H, ArH), 7.39 (d, J = 8.4 Hz, 1H, ArH), 7.44 (d, J = 8.0 Hz, 1H, ArH), 7.46 (d, J = 2.0 Hz, 1H, ArH), 7.83 (d, J = 8.8 Hz, 2H, ArH), 7.91 (d, J = 8.4 Hz, 1H, ArH), 9.83 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{21}Cl_2NO$: C 71.10, H 5.01, N 3.32. Found: C 71.03, H 5.15, N 3.31.

12-(4-Chlorophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3d**)

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3257, 3076, 2956, 2935, 2873, 1635, 1578, 1540, 1519, 1500, 1488, 1468, 1429, 1387, 1286, 1258, 1248, 1190, 1150, 1035, 843, 820, 748; ^{1}H nmr (DMSO-d_6): δ 0.84 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.02 (d, J = 16.0 Hz, 1H, CH), 2.23 (d, J = 16.0 Hz, 1H, CH), 2.39 (d, J = 16.4 Hz, 1H, CH), 2.56 (d, J = 16.4 Hz, 1H, CH), 5.79 (s, 1H, CH), 7.19 (d, J = 8.4 Hz, 2H, ArH), 7.24 (d, J = 8.4 Hz, 2H, ArH), 7.30~7.34 (m, 2H, ArH), 7.40~7.44 (m, 1H, ArH), 7.79~7.82 (m, 2H, ArH), 7.90 (d, J = 8.8 Hz, 1H, ArH), 9.77 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{22}ClNO$: C 77.41, H 5.72, N 3.61. Found: C 77.29, H 5.88, N 3.56.

9,9-dimethyl-12-(3-nitrophenyl)-8,9,10,12-tetrahydro-7*H*-benzo-[*a*]acridin-11-one (**3e**)

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3257, 2958, 2871, 1637, 1581, 1522, 1501, 1468, 1383, 1347, 1299, 1255, 1224, 1154, 1121, 811, 742, 729, 687; ^{1}H nmr (DMSO-d_6): δ 0.82 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.04 (d, J = 16.0 Hz, 1H, CH), 2.25 (d, J = 16.0 Hz, 1H, CH), 2.43 (d, J = 16.8 Hz, 1H, CH), 2.54 (d, J = 16.8 Hz, 1H, CH), 5.97 (s, 1H, CH), 7.31~7.35 (m, 1H, ArH), 7.36 (d, J = 8.8 Hz, 1H, ArH), 7.40~7.46 (m, 1H, ArH), 7.47 (d, J = 8.0 Hz, 1H, ArH), 7.70 (d, J = 7.6 Hz, 1H, ArH), 7.82~7.86 (m, 2H, ArH), 7.89 (dd, J = 8.0 Hz, J' = 1.6 Hz, 1H, ArH), 7.94 (d, J = 8.8 Hz, 1H, ArH), 8.06 (s, 1H, ArH), 9.88 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{22}N_2O_3$: C 75.36, H 5.57, N 7.03. Found: C 75.28, H 5.54, N 7.14.

12-(3-Chlorophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo-[*a*]acridin-11-one (**3f**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3268, 3087, 2960, 2873, 1598, 1521, 1469, 1383, 1306, 1259, 1178, 1145, 1080, 1034, 881, 810, 794, 777, 744, 691; ^{1}H nmr (DMSO-d_6): δ 0.85 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.05 (d, J = 16.0 Hz, 1H, CH), 2.24 (d, J = 16.0 Hz, 1H, CH), 2.42 (d, J = 16.8 Hz, 1H, CH), 2.55 (d, J = 16.8 Hz, 1H, CH), 5.82 (s, 1H, CH), 7.05~7.08 (m, 1H, ArH), 7.16~7.17 (m, 2H, ArH), 7.25 (s, 1H, ArH), 7.32~7.36 (m, 2H, ArH), 7.43~7.47 (m, 1H, ArH), 7.82 (d, J = 8.8 Hz, 2H, ArH), 7.93 (d, J = 8.4 Hz, 1H, ArH), 9.79 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{22}ClNO$: C 77.41, H 5.72, N 3.61. Found: C 77.26, H 5.75, N 3.50.

12-(3,4-Dimethoxylphenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3g**)

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3292, 2937, 2899, 2828, 2602, 1583, 1512, 1469, 1418, 1381, 1339, 1266, 1234, 1137, 1029, 823,

815, 748; ^{1}H nmr (DMSO-d_6): δ 0.89 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.05 (d, J = 16.0 Hz, 1H, CH), 2.23 (d, J = 16.0 Hz, 1H, CH), 2.40 (d, J = 16.8 Hz, 1H, CH), 2.55 (d, J = 16.8 Hz, 1H, CH), 3.60 (s, 3H, CH_3O), 3.63 (s, 3H, CH_3O), 5.75 (s, 1H, CH), 6.58 (dd, J = 8.0 Hz, J' = 2.0 Hz, 1H, ArH), 6.67 (d, J = 8.0 Hz, 1H, ArH), 6.96 (d, J = 2.0 Hz, 1H, ArH), 7.30~7.33 (m, 2H, ArH), 7.41~7.45 (m, 1H, ArH), 7.77 (d, J = 8.8 Hz, 1H, ArH), 7.80 (d, J = 8.0 Hz, 1H, ArH), 8.00 (d, J = 8.8 Hz, 1H, ArH), 9.67 (s, 1H, NH).

Anal. Calcd. for $C_{27}H_{27}NO_3$: C 78.42, H 6.58, N 3.39. Found: C 78.26, H 6.57, N 3.22.

9,9-Dimethyl-12-(4-hydroxylphenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3h**).

This compound was obtained as pale yellow crystals, mp >300 ℃ (Lit. [7] 315~316 ℃) ; ir (KBr): ν_{max} 3304, 3250, 3014, 2946, 1634, 1602, 1578, 1519, 1469, 1441, 1399, 1384, 1235, 1170, 1149, 1123, 1034, 982, 837, 816, 747; ^{1}H nmr (DMSO-d_6): δ 0.86 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.02 (d, J = 16.0 Hz, 1H, CH), 2.21 (d, J = 16.0 Hz, 1H, CH), 2.38 (d, J = 16.8 Hz, 1H, CH), 2.53 (d, J = 16.8 Hz, 1H, CH), 5.68 (s, 1H, CH), 6.50 (d, J = 8.4 Hz, 2H, ArH), 7.00 (d, J = 8.4 Hz, 2H, ArH), 7.30~7.42 (m, 3H, ArH), 7.75 (d, J = 8.8 Hz, 1H, ArH), 7.79 (d, J = 7.6 Hz, 1H, ArH), 7.94 (d, J = 8.8 Hz, 1H, ArH), 9.02 (s, 1H, OH), 9.62 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{23}NO_2$: C 81.27, H 6.27, N 3.79. Found: C 81.22, H 6.31, N 3.83.

12-(4-Methoxylphenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3i**).

This compound was obtained as pale yellow crystals, mp >300 ℃ (Lit [7] 297~298 ℃) ; ir (KBr): ν_{max} 3268, 3088, 2959, 2928, 2833, 1598, 1584, 1498, 1469, 1426, 1398, 1383, 1302, 1259, 1239, 1176, 1148, 1033, 833, 811, 746; ^{1}H nmr (DMSO-d_6): δ 0.86 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.03 (d, J = 16.0 Hz, 1H, CH), 2.22 (d, J = 16.0 Hz, 1H, CH), 2.39 (d, J = 16.8 Hz, 1H, CH), 2.54 (d, J = 16.8 Hz, 1H, CH), 3.61 (s, 3H, CH_3O), 5.74 (s, 1H, CH), 6.68 (d, J = 8.4 Hz, 2H, ArH), 7.13 (d, J = 8.4 Hz, 2H, ArH), 7.28~7.32 (m, 2H, ArH), 7.39~7.43 (m, 1H, ArH), 7.77 (d, J = 8.8 Hz, 1H, ArH), 7.79 (d, J = 8.4 Hz, 1H, ArH), 7.94 (d, J = 8.4 Hz, 1H, ArH), 9.66 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{25}NO_2$: C 81.43, H 6.57, N 3.65. Found: C 81.32, H 6.48, N 3.57.

12-(4-Bromophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo-[*a*]acridin-11-one (**3j**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3263, 3086, 2960, 2927, 2873, 1595, 1519, 1493, 1468, 1428, 1418, 1383, 1259, 1238, 1186, 1169, 1147, 1011, 835, 809, 741; ^{1}H nmr (DMSO-d_6): δ 0.84 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.03 (d, J = 16.0 Hz, 1H, CH), 2.23 (d, J = 16.0 Hz, 1H, CH), 2.39 (d, J = 16.4 Hz, 1H, CH), 2.55 (d, J = 16.4 Hz, 1H, CH), 5.79 (s, 1H, CH), 7.18 (d, J = 8.4 Hz, 2H, ArH), 7.31~7.34 (m, 4H, ArH), 7.40~7.44 (m, 1H, ArH), 7.79~7.82 (m, 2H, ArH), 7.90 (d, J = 8.4 Hz, 1H, ArH), 9.76 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{22}BrNO$: C 69.45, H 5.13, N 3.24. Found: C 69.21, H 5.10, N 3.23.

9,9-Dimethyl-12-(3,4-dimethylphenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3k**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3279, 3089, 3052, 2960, 2914, 2878, 1601, 1583, 1519, 1496, 1468, 1427, 1397, 1382, 1258, 1236, 1183, 1147, 1033, 822, 745, 699; ^{1}H nmr (DMSO-d_6): δ 0.87 (s, 3H, CH_3), 1.03 (s, 3H, CH_3), 2.03~2.06 (m, 7H, CH + 2 CH_3), 2.21 (d, J = 16.0 Hz, 1H, CH), 2.40 (d, J = 16.4 Hz, 1H, CH), 2.49 (d, J = 16.8 Hz, 1H, CH), 5.71 (s, 1H, CH), 6.85 (d, J = 7.6 Hz, 1H, ArH), 6.91 (dd, J = 8.0 Hz, J' = 1.6 Hz, 1H, ArH), 7.03 (s, 1H, ArH), 7.29~7.32 (m, 2H, ArH), 7.40~7.43 (m, 1H, ArH), 7.76 (d, J = 8.4 Hz, 1H, ArH), 7.79 (d, J = 7.6 Hz, 1H, ArH), 7.96 (d, J = 8.4 Hz, 1H, ArH), 9.64 (s, 1H, NH).

Anal. Calcd. for $C_{27}H_{27}NO$: C 85.00, H 7.13, N 3.67. Found: C 85.19, H 7.24, N 3.55.

12-(3-Methoxyl-4-hydroxylphenyl)-9,9-dimethyl-8,9,10,12-tetra-hydro-7*H*-benzo[*a*]acridin-11-one (**3l**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3524, 3265, 3087, 2958, 1600, 1584, 1519, 1469, 1430, 1385, 1267, 1240, 1208, 1149, 1121, 1034, 827, 816, 747; ^{1}H nmr (DMSO-d_6): δ 0.88 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.04 (d, J = 16.0 Hz, 1H, CH), 2.22 (d, J = 16.0 Hz, 1H, CH), 2.39 (d, J = 16.8 Hz, 1H, CH), 2.54 (d, J = 16.8 Hz, 1H, CH), 3.64 (s, 3H, CH_3O), 5.97 (s, 1H, CH), 6.48 (s, 2H, ArH), 6.91 (s, 1H, ArH), 7.28~7.33 (m, 2H, ArH), 7.41~7.45 (m, 1H, ArH), 7.76 (d, J = 8.8 Hz, 1H, ArH), 7.80 (d, J = 7.6 Hz, 1H, ArH), 8.00 (d, J = 8.4 Hz, 1H, ArH), 8.60 (s, 1H, OH), 9.04 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{25}NO_3$: C 78.18, H 6.31, N 3.51. Found: C 78.00, H 6.24, N 3.63.

12-(4-Dimethylaminophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3m**).

This compound was obtained as pale yellow crystals, mp >300 ℃ (Lit [7] 346~347 ℃) ; ir (KBr): ν_{max} 3265, 3073, 2956, 2932, 2904, 1598, 1581, 1522, 1498, 1467, 1443, 1397, 1384, 1343, 1264, 1239, 1125, 820, 780, 744; ^{1}H nmr (DMSO-d_6): δ 0.88 (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.02 (d, J = 16.0 Hz, 1H, CH), 2.21 (d, J = 16.0 Hz, 1H, CH), 2.39 (d, J = 16.4 Hz, 1H, CH), 2.53 (d, J = 16.4 Hz, 1H, CH), 2.74 (s, 6H, 2 CH_3), 5.66 (s, 1H, CH), 6.48 (d, J = 8.8 Hz, 2H, ArH), 7.03 (d, J = 8.8 Hz, 2H, ArH), 7.23~7.31 (m, 2H, ArH), 7.39~7.43 (m, 1H, ArH), 7.74 (d, J = 8.8 Hz, 1H, ArH), 7.78 (d, J = 8.0 Hz, 1H, ArH), 7.96 (d, J = 8.0 Hz, 1H, ArH), 9.60 (s, 1H, NH).

Anal. Calcd. for $C_{27}H_{28}N_2O$: C 81.78, H 7.12, N 7.06. Found: C 81.69, H 7.06, N 7.00.

12-(2-Chlorophenyl)-9,9-dimethyl-8,9,10,12-tetrahydro-7*H*-benzo-[*a*]acridin-11-one (**3n**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3241, 3076, 2958, 2880, 159, 1522, 1467, 1381, 1317, 1259, 1236, 1152, 1034, 979, 815, 765, 742; ^{1}H nmr (DMSO-d_6): δ 0.87 (s, 3H, CH_3), 1.05 (s, 3H, CH_3), 1.99 (d, J = 16.0 Hz, 1H, CH), 2.22 (d, J = 16.0 Hz, 1H, CH), 2.43 (d, J = 16.8 Hz, 1H, CH), 2.59 (d, J = 16.8 Hz, 1H, CH), 6.02 (s, 1H, CH), 6.99~7.04 (m, 1H, ArH), 7.08~7.13 (m, 1H, ArH), 7.24 (dd, J = 8.0 Hz, J' = 1.2 Hz, 1H, ArH), 7.28~7.33 (m, 3H, ArH), 7.42~7.46 (m, 1H, ArH), 7.76~7.80 (m, 2H, ArH), 8.15 (d, J = 8.4 Hz, 1H, ArH), 9.79 (s, 1H, NH).

Anal. Calcd. for $C_{25}H_{22}ClNO$: C 77.41, H 5.72, N 3.61. Found: C 77.31, H 5.79, N 3.66.

9,9-Dimethyl-12-(2-thiophenyl)-8,9,10,12-tetrahydro-7*H*-benzo-[*a*]acridin-11-one (**3o**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3265, 2956, 1586, 1558, 1520, 1472, 1383, 1346, 1261, 1236, 811, 781, 775; ^{1}H nmr (DMSO-d_6): δ 0.96 (s, 3H, CH_3), 1.06 (s, 3H, CH_3), 2.12 (d, J = 16.0 Hz, 1H, CH), 2.27 (d, J = 16.0 Hz, 1H, CH), 2.42 (d, J = 16.8 Hz, 1H, CH), 2.56 (d, J = 16.8 Hz, 1H, CH), 6.12 (s, 1H, CH), 6.61 (d, J = 3.2 Hz, 1H, ArH), 6.72 (dd, J = 4.8 Hz, J' = 3.2 Hz, 1H, ArH), 7.11 (dd, J = 4.8 Hz, J' = 0.8 Hz, 1H, ArH), 7.29 (d, J = 8.8 Hz, 1H, ArH), 7.32 (t, J = 7.2 Hz, 1H, ArH), 7.47~7.51 (m, 1H, ArH), 7.81 (d, J = 8.8 Hz, 1H, ArH), 7.84 (d, J = 8.0 Hz, 1H, ArH), 8.00 (d, J = 8.4 Hz, 1H, ArH), 9.82 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{21}NOS$: C 76.84, H 5.89, N 3.90. Found: C 76.71, H 5.93, N 3.90.

12-(4-Bromophenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3p**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3264, 3086, 2946, 1683, 1599, 1581, 1518, 1494, 1467, 1427, 1397, 1384, 1336, 1317, 1286, 1237, 1191, 1140, 1124, 1010, 831, 811, 749; ^{1}H nmr (DMSO-d_6): δ 1.75~1.81 (m, 1H, CH), 1.91~ 1.97 (m, 1H, CH), 2.20~2.28 (m, 2H, 2.60~2.62 (m, 2H, CH_2), 5.82 (s, 1H, CH), 7.16 (d, J = 8.4 Hz, 2H, ArH), 7.30~7.34 (m, 4H, ArH), 7.40~7.44 (m, 1H, ArH), 7.69~7.82 (m, 2H, ArH), 7.87 (d, J = 8.8 Hz, 1H, ArH), 9.80 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{18}BrNO$: C 68.33, H 4.49, N 3.46. Found: C 68.20, H 4.52, N 3.29.

12-(4-Methoxylphenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3q**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3268, 3089, 2941, 2825, 1662, 1598, 1582, 1519, 1495, 1426, 1426, 1399, 1380, 1286, 1258, 1238, 1189, 1174, 1137, 1120, 813, 756; ^{1}H nmr (DMSO-d_6): δ 1.70~1.80 (m, 1H, CH), 1.91~1.96 (m, 1H, CH), 2.20~2.30 (m, 2H, CH_2), 2.59~2.62 (m, 2H, CH_2), 3.61 (s, 3H, CH_3O), 5.77 (s, 1H, CH), 6.68 (d, J = 8.8 Hz, 2H, ArH), 7.11 (d, J = 8.8 Hz, 2H, ArH), 7.28~7.32 (m, 2H, ArH), 7.39~7.43 (m, 1H, ArH), 7.77 (d, J = 8.8 Hz, 1H, ArH), 7.79 (d, J = 8.4 Hz, 1H, ArH), 7.91 (d, J = 8.4 Hz, 1H, ArH), 9.71 (s, 1H, NH).

Anal. Calcd. for $C_{24}H_{21}NO_2$: C 81.10, H 5.96, N 3.94. Found: C 81.23, H 5.82, N 4.01.

12-(4-Fluorophenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3r**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3264, 3087, 295, 1685, 1601, 1582, 1519, 1495, 1427, 1385, 1283, 1236, 1219, 1189, 1137, 1092, 957, 837, 815, 800, 742; ^{1}H nmr (DMSO-d_6): δ 1.73~1.81 (m, 1H, CH), 1.91~1.97 (m, 1H, CH), 2.21~2.28 (m, 2H, 2.60~2.63 (m, 2H, CH_2), 5.84 (s, 1H, CH), 6.95 (t, J = 8.8 Hz, 2H, ArH), 7.22 (dd, J = 6.0 Hz, J' = 8.4 Hz, 2H, ArH), 7.30~7.34 (m, 2H, ArH), 7.40~7.44 (m, 1H, ArH), 7.78~7.82 (m, 2H, ArH), 7.90 (d, J = 8.4 Hz, 1H, ArH), 9.78 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{18}FNO$: C 80.45, H 5.28, N 4.08. Found: C 80.28, H 5.40, N 4.05.

12-(4-Chlorophenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3s**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3265, 3087, 2950, 2888, 1684, 1600, 1581, 1519, 1494, 1467, 1428, 1385, 1317, 1286, 1237, 1191, 1139, 1124, 1088, 1014, 835, 811, 750; ^{1}H nmr (DMSO-d_6): δ 1.75~1.79 (m, 1H, CH), 1.91~ 1.95 (m, 1H, CH), 2.23~2.32 (m, 2H, CH_2), 2.60~2.63 (m, 2H, CH_2), 5.84 (s, 1H, CH), 7.19 (d, J = 8.4 Hz, 2H, ArH), 7.22 (d, J = 8.4 Hz, 2H, ArH), 7.30~7.34 (m, 2H, ArH), 7.40~7.44 (m, 1H, ArH), 7.79~7.82 (m, 2H, ArH), 7.88 (d, J = 8.4 Hz, 1H, ArH), 9.80 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{18}ClNO$: C 76.77, H 5.04, N 3.89. Found: C 76.80, H 5.14, N 3.93.

12-(3-Chlorophenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3t**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3273, 3064, 2933, 1654, 1601, 1582, 1519, 1493, 1467, 1427, 1397, 1379, 1282, 1237, 1191, 1137, 1123, 1079, 957, 817, 750, 688; ^{1}H nmr (DMSO-d_6): δ 1.72~1.80 (m, 1H, CH), 1.90~1.97 (m, 1H, CH), 2.25~2.30 (m, 2H, CH_2), 2.63~2.65 (m, 2H, CH_2), 5.86 (s, 1H, CH), 7.06~7.19 (m, 3H, ArH), 7.21~7.24 (m, 1H, ArH), 7.31~7.35 (m, 2H, ArH), 7.42~7.46 (m, 1H, ArH), 7.82 (d, J = 8.8 Hz, 2H, ArH), 7.89 (d, J = 8.8 Hz, 1H, ArH), 9.84 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{18}ClNO$: C 76.77, H 5.04, N 3.89. Found: C 76.79, H 5.05, N 3.88.

12-Phenyl-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3u**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3251, 3079, 2952, 2925, 2882, 1662, 1617, 1597, 1583, 1519, 1493, 1468, 1425, 1384, 1335, 1280, 1235, 1190, 1138, 1010, 958, 809, 744, 717, 697; ^{1}H nmr (DMSO-d_6): δ 1.73~1.80 (m, 1H, CH), 1.92~1.96 (m, 1H, CH), 2.23~2.28 (m, 2H, CH_2), 2.60~2.63 (m, 2H, CH_2), 5.83 (s, 1H, CH), 6.97~7.01 (m, 1H, ArH), 7.12 (t, J = 7.6 Hz, 2H, ArH), 7.22 (d, J = 7.6 Hz, 2H, ArH), 7.29~7.33 (m, 2H, ArH), 7.39~7.43 (m, 1H, ArH), 7.77~7.81 (m, 2H, ArH), 7.92 (d, J = 8.4 Hz, 1H, ArH), 9.77 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{19}NO$: C 84.89, H 5.89, N 4.30. Found: C 84.91, H 5.74, N 4.15.

12-(4-Hydroxylphenyl)-8,9,10,12-tetrahydro-7*H*-benzo[*a*]acridin-11-one (**3v**).

This compound was obtained as pale yellow crystals, mp >300 ℃; ir (KBr): ν_{max} 3153, 2940, 1651, 1637, 1615, 1595, 1562, 1509, 1455, 1390, 1361, 1291, 1263, 1234, 1171, 1135, 955, 868, 846, 776, 756; ^{1}H nmr (DMSO-d_6): δ 1.53~1.62 (m, 1H, CH), 1.77~1.82 (m, 1H, CH), 2.19~2.22 (m, 2H, CH_2), 2.24~2.32 (m, 2H, CH_2), 5.07 (s, 1H, CH), 6.67 (d, J = 7.6 Hz, 2H, ArH), 7.13 (d, J = 7.6 Hz, 2H, ArH), 7.41 (s, 1H, OH), 7.64~7.66 (m, 2H, ArH), 8.06~8.14 (m, 4H, ArH), 9.12 (s, 1H, NH).

Anal. Calcd. for $C_{23}H_{19}NO_2$: C 80.92, H 5.61, N 4.10. Found: C 80.78, H 5.54, N 4.12.

General Procedure for the Synthesis of Indeno[1,2-*b*]benzo[*f*]-quinolines (**5**).

The mixture of *N*-arylidenenaphthalen-2-amine **1** (2 mmol), 1,3-indenedione **4** (2 mmol) and TEBAC (0.1 g) was stirred in water (10 mL) at 100 ºC for 10-20 h. The crystalline powder formed was collected by filtration, washed with water and

recrystallized from DMF and water to give pure indeno[1,2-*b*]benzo[*f*]quinolines **5**.

13-(2-Thiophenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]quinolin-12-one (**5a**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3220, 3065, 1663, 1626, 1604, 1575, 1523, 1509, 1468, 1431, 1404, 1366, 1238, 1209, 1192, 1135, 901, 809, 654; ^{1}H nmr (DMSO-d_6): δ 6.11 (s, 1H, CH), 6.79 (dd, J = 8.0 Hz, J' = 1.6 Hz, 1H, ArH), 6.89 (d, J = 8.0 Hz, 1H, ArH), 7.16 (dd, J = 8.0Hz, J' = 1.6 Hz, 1H, ArH), 7.34~7.50 (m, 6H, ArH), 7.63 (d, J = 8.4 Hz, 1H, ArH), 7.89 (d, J = 8.4 Hz, 1H, ArH), 7.93 (d, J = 8.8 Hz, 1H, ArH), 8.08 (d, J = 8.8 Hz, 1H, ArH), 11.04 (s, 1H, NH).

Anal. Calcd. for $C_{24}H_{15}NOS$: C 78.88, H 4.14, N 3.83. Found: C 78.75, H 4.23, N 3.69.

13-(4-Methoxylphenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]-quinolin-12-one (**5b**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3218, 3098, 3065, 1660, 1627, 1605, 1574, 1533, 108, 167, 1405, 1253, 1203, 1174, 1106, 1031, 900, 821, 808, 742, 701; ^{1}H nmr (DMSO-d_6): δ 3.62 (s, 1H, CH), 5.72 (s, 1H, CH), 6.73 (d, J = 8.8 Hz, 2H, ArH), 7.14 (d, J = 8.8 Hz, 2H, ArH), 7.25 (d, J = 8.4 Hz, 1H, ArH), 7.33~7.47 (m, 4H, ArH), 7.51 (d, J = 8.4 Hz, 1H, ArH), 7.61 (d, J = 8.0 Hz, 1H, ArH), 7.86 (d, J = 8.8 Hz, 1H, ArH), 7.91 (d, J = 8.8 Hz, 2H, ArH), 11.91 (s, 1H, NH).

Anal. Calcd. for $C_{27}H_{19}NO_2$: C 83.27, H 4.92, N 3.60. Found: C 83.09, H 4.90, N 3.53.

13-(4-Fluorophenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]quinolin-12-one (**5c**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3218, 3064, 1661, 1602, 1573, 1533, 1468, 1432, 1406, 1366, 1224, 1201, 1177, 1146, 1136, 901, 830, 811, 740, 700; ^{1}H nmr (DMSO-d_6): δ 5.82 (s, 1H, CH), 7.00 (t, J = 8.8 Hz, 2H, ArH), 7.25~7.28 (m, 3H, ArH), 7.36~7.48 (m, 4H, ArH), 7.53 (d, J = 9.2 Hz, 1H, ArH), 7.63 (d, J = 7.2 Hz, 1H, ArH), 7.88 (d, J = 8.4 Hz, 2H, ArH), 7.93 (d, J = 8.8 Hz, 1H, ArH), 10.96 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{16}FNO$: C 82.74, H 4.27, N 3.71. Found: C 82.70, H 4.25, N 3.61.

13-(3-Chlorophenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]quinolin-12-one (**5d**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3277, 3062, 1660, 1625, 1601, 1590, 1568, 1508, 1468, 1430, 1401, 1366, 1234, 1196, 1178, 1133, 1081, 898, 814, 765, 743, 719, 692; ^{1}H nmr (DMSO-d_6): δ 5.85 (s, 1H, CH), 7.11~7.48 (m, 9H, ArH), 7.54 (d, J = 8.8 Hz, 1H, ArH), 7.64 (d, J = 7.2 Hz, 1H, ArH), 7.89 (d, J = 8.0 Hz, 2H, ArH), 7.96 (d, J = 8.8 Hz, 1H, ArH), 11.01 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{16}ClNO$: C 79.29, H 4.09, N 3.56. Found: C 79.13, H 3.99, N 3.57.

13-Phenyl-7,13-dihydroindeno[1,2-*b*]benzo[*f*]quinolin-12-one (**5e**)

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3218, 3053, 1660, 1626, 1603, 1575, 1532, 1468, 1431, 1405, 1366, 1263, 1136, 1070, 910, 896, 831, 806, 746, 696; ^{1}H nmr (DMSO-d_6): δ 5.78 (s, 1H, CH), 7.04 (t, J = 7.2 Hz, 1H, ArH), 7.17 (t, J = 7.6 Hz, 2H, ArH), 7.24~7.27(m, 3H, ArH), 7.34~7.47(m, 4H, ArH), 7.53 (d, J = 8.8 Hz, 1H, ArH), 7.62 (d, J = 7.2 Hz, 1H, ArH), 7.86~7.91 (m, 2H, ArH), 7.93 (d, J = 8.8 Hz, 1H, ArH), 11.94 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{17}NO$: C 86.88, H 4.77, N 3.90. Found: C 86.73, H 4.58, N 3.92.

13-(4-Chlorophenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]quinolin-12-one (**5f**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3226, 3063, 1661, 1627, 1604, 1574, 1533, 1468, 1406, 1365, 1239, 1200, 1145, 1072, 900, 809, 741, 698; ^{1}H nmr (DMSO-d_6): δ 5.82 (s, 1H, CH), 7.22~7.28(m, 5H, ArH), 7.35~7.46(m, 4H, ArH), 7.53 (d, J = 8.8 Hz, 1H, ArH), 7.63 (d, J = 6.8 Hz, 1H, ArH), 7.85~7.89 (m, 2H, ArH), 7.94 (d, J = 8.8 Hz, 1H, ArH), 10.98 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{16}ClNO$: C 79.29, H 4.09, N 3.56. Found: C 79.19, H 4.04, N 3.62.

13-(2,4-Dichlorophenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]-quinolin-12-one (**5g**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3269, 3050, 1663, 1630, 1606, 1574, 1532, 1469, 1404, 1240, 1198, 1146, 1102, 1046, 902, 860, 839, 813, 743, 700; ^{1}H nmr (DMSO-d_6): δ 6.09 (s, 1H, CH), 7.19 (dd, J = 8.4 Hz, J' = 1.6 Hz, 1H, ArH), 7.27 (d, J = 8.4 Hz, 1H, ArH), 7.37~7.68(m, 7H, ArH), 7.81 (d, J = 8.4 Hz, 1H, ArH), 7.88 (d, J = 8.4 Hz, 1H, ArH), 7.94 (d, J = 8.8 Hz, 1H, ArH), 8.11 (d, J = 8.8 Hz, 1H, ArH), 11.08 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{15}Cl_2NO$: C 72.91, H 3.53, N 3.27. Found: C 72.86, H 3.49, N 3.10.

13-(3,4-Dichlorophenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]-quinolin-12-one (**5h**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3253, 3028, 1686, 1627, 1608, 1573, 1520, 1513, 1468, 1404, 1363, 1203, 1184, 1146, 1031, 905, 812, 735, 701; ^{1}H nmr (DMSO-d_6): δ 5.88 (s, 1H, CH), 7.12 (dd, J = 8.4 Hz, J' = 2.0 Hz, 1H, ArH), 7.29 (d, J = 8.4 Hz, 1H, ArH), 7.36~7.49(m, 5H, ArH), 7.54 (d, J = 8.8 Hz, 1H, ArH), 7.56 (d, J = 2.0 Hz, 1H, ArH), 7.64 (d, J = 8.0 Hz, 1H, ArH), 7.85~7.91 (m, 2H, ArH), 8.01 (d, J = 8.8 Hz, 1H, ArH), 11.05 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{15}Cl_2NO$: C 72.91, H 3.53, N 3.27. Found: C 72.81, H 3.44, N 3.19.

13-(4-Hydroxylphenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]quinolin-12-one (**5i**).

This compound was obtained as red crystals, mp >300 ℃; ir (KBr): ν_{max} 3442, 3219, 3068, 1660, 1626, 1604, 1575, 1532, 1509, 1405, 1405, 1269, 1202, 1135, 1072, 900, 834, 812, 742, 702; ^{1}H nmr (DMSO-d_6): δ5.65 (s, 1H, CH), 6.54 (d, J = 8.4 Hz, 2H, ArH), 7.02 (d, J = 8.4 Hz, 2H, ArH), 7.25 (d, J = 8.4 Hz, 1H, ArH), 7.32~7.46 (m, 4H, ArH), 7.50 (d, J = 8.4 Hz, 1H, ArH), 7.60 (d, J = 7.6 Hz, 1H, ArH), 7.87 (d, J = 8.8 Hz, 1H, ArH), 7.89~7.92 (m, 2H, ArH), 9.13 (s, 1H, OH), 10.87 (s, 1H, NH).

Anal. Calcd. for $C_{26}H_{17}NO_2$: C 83.13, H 4.56, N 3.73. Found: C 82.97, H 4.50, N 3.84.

13-(3,4-Dimethylphenyl)-7,13-dihydroindeno[1,2-*b*]benzo[*f*]-quinolin-12-one (**5j**).

This compound was obtained as red crystals, mp >300 °C; ir (KBr): ν_{max} 3278, 3037, 2970, 1662, 1628, 1602, 1575, 1529, 1468, 1443, 1400, 1339, 1233, 1193, 1179, 1133, 901, 810, 768, 745, 710; ^{1}H nmr (DMSO-d_6): δ 2.05 (s, 3H, CH_3), 2.06 (s, 3H, CH_3), 5.68 (s, 1H, CH), 6.91 (s, 2H, ArH), 7.01 (s, 1H, ArH), 7.25 (d, J = 8.4 Hz, 1H, ArH), 7.32~7.46 (m, 4H, ArH), 7.52 (d, J = 8.8 Hz, 1H, ArH), 7.60 (d, J = 7.6 Hz, 1H, ArH), 7.86 (d, J = 8.8 Hz, 1H, ArH), 7.89~7.92 (m, 2H, ArH), 10.92 (s, 1H, NH).

Anal. Calcd. for $C_{28}H_{21}NO$: C 86.79, H 5.46, N 3.61. Found: C 86.92, H 5.51, N 3.33.

Acknowledgments.

We are grateful to the Foundation (04KJB150139) of the Education Committee of Jiangsu Province for financial support.

REFERENCES AND NOTES

[1] A. Arcadi, M. Chiarini, S. D. Giuseppe and F. Marinelli, *Synlett*, 203 (2003).

[2a] R. A. Janis, P. J. Silver and D. J. Triggle, *Adv. Drug Res.*, **16**, 30 (1987); [b] R. Shan, C. Velazquez and E. E. Knaus, *J. Med. Chem.*, **41**, 254 (2004); [c] A. F. Dulhunty, S. M. Curtis, S. Watson, L. Cengia and G. Marco, *J. Biol. Chem.*, **279**, 11853 (2004).

[3] U. Eisner and J. Kuthan, *Chem. Rev.*, **72**, 1 (1972).

[4] R. J. Chorvat and K. J. Rorig, *J. Org. Chem.*, **53**, 5779 (1988).

[5] N. Martin, M. Quinteiro, C. Seoane, L. Mora, M. Suarez, E. Ockoa, and A. Morales, *J. Heterocyclic Chem.*, **51**, 235 (1995).

[6] S. J. Tu, C. B. Miao, Y. Gao, Y. J. Feng and J. C. Feng, *Chin. J. Chem.*, **20,** 703 (2002).

[7] R. Martinez, E. Cortes, R. A. Toscano and I. Linzaga, *J. Heterocyclic Chem.*, **27**, 363 (1990).

[8a] R. Breslow and D. C. Rideout, *J. Am. Chem. Soc.*, **102**, 7816 (1980); [b] R. Breslow, *Acc. Chem. Res.*, **24**, 159 (1991).

[9] R. Breslow and U. Maitra, *Tetrahedron Lett.*, **25**, 1239 (1984).

[10a] A. A. Ponaras, *J. Org. Chem.*, **48**, 3866 (1983); [b] R. M. Coates, B. D. Rogers, S. J. Hobbs, D. R. Peck and D. P. Curran, *J. Am. Chem. Soc.*, **109**, 1160 (1987).

[11a] H. Mattes, and C. Benezra, *Tetrahedron Lett.*, **26**, 5697 (1985); [b] J. Y. Zhou, G. D. Lu and S. H. Wu, *Synth. Commun.*, **22**, 481 (1992).

[12] P. Delair and J. L. Luche, *J. Chem. Soc., Chem. Commun.*, 398 (1989).

[13a] X. S. Wang, M. M. Zhang, Z. S. Zeng, D. Q. Shi, X. Y. Wei, and Z. M. Zong, *Chem. Lett.*, **34**, 1316 (2005); [b] X. S. Wang, D. Q. Shi, Y. L. Li, H. Chen, X. Y. Wei and Z. M. Zong, *Synth. Commun.*, **35**, 97 (2005); [c] X. S. Wang, D. Q. Shi, Y. F. Zhang, S. H. Wang and S. J. Tu, *Chin. J. Org. Chem.*, **24**, 430 (2004).

[14] Crystal data for **5a**: $C_{25}H_{22}ClNO$; M = 387.89, colorless block crystals, 0.40 × 0.35 × 0.12 mm, monoclinic, space group $P2_1/n$, a = 10.4242(14), b = 11.777(2), c = 16.173(3) Å, = 91.024(5)°, V = 1985.1(5) Å^3, Z = 4, D_c = 1.298 g.cm^{-3}. $F(000)$ = 816, (Mo$K\alpha$) = 0.208 mm^{-1}. Intensity data were collected on a Rigaku Mercury diffractometer with graphite monochromated Mo$K\alpha$ radiation (λ=0.71070Å) using the ω scan mode with 3.06°<θ<25.35°. 3622 unique reflections were measured and 3006 reflections with $I>2\sigma(I)$ were used in the refinement. Structure solved by direct methods and expanded using Fourier techniques. The final cycle of full-matrix least squares technique to R = 0.0571 and wR = 0.1186.