

Synthesis of Benzo[c]acridine Derivatives: Reaction of N-Arylidenenaphthalen-1-amine with 5,5-Dimethyl-1,3-cyclohexadione

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Synthesis of 10,10-dimethyl-7-aryl-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one derivatives was accomplished in high yields *via* the reaction of N-arylidenenaphthalen-1-amine with 5,5-dimethyl-1,3-cyclohexadione in aqueous medium catalyzed by triethylbenzyl ammonium chloride. The structures were established from spectroscopic data and melting points.

Key Words: Synthesis, Benzo[c]acridine derivatives, Triethylbenzyl ammonium chloride.

INTRODUCTION

1,4-Dihydropyridine (DHP) and its derivatives were widely prescribed as calcium β -blocker and used for the treatment of hypertension and heart defibrillation^{1,2}. The chemical modifications carried on the DHP ring such as variation of different substituents³ or heteroatoms⁴ have allowed expansion of the structure activity relationship and afforded some insight into the molecular interactions at the receptor level. Recently there are many methods available for synthesis of tricyclic compounds containing the 1,4-dihydropyridines, such as acridine derivatives, from aldehyde, dimedone and ammonium acetate *via* traditional heating in organic solvents⁵ or in water catalyzed by triethylbenzylammonium chloride (TEBAC)⁶ or improved under microwave irradiation⁷ or by green chemistry in ionic liquids⁸. Because of the toxic and volatile nature of many organic solvents, ionic liquids are emerging as effective solvents for 'green' processes. However, the high cost of most conventional room temperature ionic liquids and

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apprehension about their toxicity led us to explore other clean method. The use of water as a solvent in organic reactions in place of commonly used organic solvents as reported in the application of green chemistry⁹⁻¹⁶ also prompted us to use it in our work for the synthesis of potential bioactive compounds. As part of our current studies on the development of new routes to heterocyclic systems¹⁷⁻¹⁹, we now report an efficient and clean synthetic route to 10,10-dimethyl-7-aryl-7,9,10,11-tetrahydro-9*H*-benzo[c]acridin-8-one derivatives in aqueous medium catalyzed by TEBAC by the reaction of *N*-arylidenenaphthalen-1-amine with 5,5-dimethyl-1,3-cyclohexadione.

EXPERIMENTAL

Melting points were determined in open capillaries or XT-4 microscope melting-point detector (Beijing Keyi Electro-optic Instrument Plant) and are uncorrected. IR spectra were recorded on a TENSOR 27 spectrometer in KBr. ¹H NMR spectra were obtained for solution in DMSO-*d*₆ with Me₄Si as internal standard using a Bruker-400 spectrometer. Elemental analyses were carried out using Carlo Erba 1110 analyzer. MS were determined on Agilent-6890/5973N -GL/MS spectrometer.

A suspension of a mixture of *N*-arylidenenaphthalen-1-amine (**1**) (2 mmol), 5,5-dimethyl-1,3-cyclohexadione (**2**) (2 mmol) and TEBAC (0.1 g) was stirred in water (10 mL) at 100 °C for 12-18 h. The solid products formed was collected by filtration, washed with water and recrystallized from DMF and water to give pure 10,10-dimethyl-7-aryl-7,9,10,11-tetrahydro-9*H*-benzo[c]acridin-8-one derivatives (**3**).

10,10-Dimethyl-7-(3-nitrophenyl)-7,9,10,11-tetrahydro-9*H*-benzo[c]acridin-8-one (3a): m.p. 267-269 °C. IR: (KBr, ν_{\max} , cm⁻¹) 3307, 2955, 1671, 1588, 1531, 1518, 1497, 1384, 1345, 1261, 1150, 1092, 1060, 809, 759, 727; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz) 0.99 (s, 3H, CH₃), 1.11 (s, 3H, CH₃), 2.06 (d, *J* = 16.4 Hz, 1H, C¹¹-H), 2.58 (d, *J* = 16.4 Hz, 1H, C¹¹-H), 2.70 (d, *J* = 16.8 Hz, 1H, C⁹-H), 2.75 (d, *J* = 16.8 Hz, 1H, C⁹-H), 5.43 (s, 1H, CH), 7.33 (d, *J* = 8.4 Hz, 1H, ArH), 7.49-7.54 (m, 3H, ArH), 7.58-7.62 (m, 1H, ArH), 7.72 (d, *J* = 7.6 Hz, 1H, ArH), 7.84 (d, *J* = 7.6 Hz, 1H, ArH), 7.95-8.07 (m, 2H, ArH), 8.49 (d, *J* = 8.8 Hz, 1H, ArH), 9.42 (s, 1H, NH); ¹³C NMR δ (DMSO-*d*₆, 400 MHz): 26.9, 29.4, 32.3, 39.9, 40.4, 50.3, 106.7, 119.9, 121.5, 122.37, 123.3, 126.1, 126.2, 127.7, 127.9, 128.4, 128.5, 129.2, 130.6, 130.7, 130.9, 132.7, 149.5, 152.4, 193.7. MS (EI): *m/z* 398 (M⁺, 1), 366 (100), 337 (35), 310 (27), 231 (20); Anal. (%) calcd. for C₂₅H₂₂N₂O₃: C 75.36, H 5.57, N 7.03; found C 75.28, H 5.78, N 7.21.

10,10-Dimethyl-7-(4-dimethylaminophenyl)-7,9,10,11-tetrahydro-9*H*-benzo[c]acridin-8-one (3b): m.p. 276-278 °C (Lit.²¹ 278-279 °C). IR (KBr, ν_{\max} , cm⁻¹) 3293, 2952, 1683, 1589, 1518, 1510, 1388, 1340, 1259, 1147, 1124, 1057, 946, 831, 807, 758; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz)

1.02 (s, 3H, CH₃), 1.08 (s, 3H, CH₃), 2.08 (d, $J = 16.4$ Hz, 1H, C¹¹-H), 2.58 (d, $J = 16.4$ Hz, 1H, C¹¹-H), 2.68 (d, $J = 17.6$ Hz, 1H, C⁹-H), 2.74 (d, $J = 17.6$ Hz, 1H, C⁹-H), 2.76 (s, 6H, 2NCH₃), 5.06 (s, 1H, CH), 6.53 (d, $J = 8.8$ Hz, 2H, ArH), 7.02 (d, $J = 8.8$ Hz, 2H, ArH), 7.24 (d, $J = 8.4$ Hz, 1H, ArH), 7.44 (d, $J = 8.4$ Hz, 1H, ArH), 7.45~7.58 (m, 2H, ArH), 7.81 (d, $J = 7.6$ Hz, 1H, ArH), 8.44 (d, $J = 8.8$ Hz, 1H, ArH), 9.18 (s, 1H, NH).

10,10-Dimethyl-7-(3,4-dimethoxyphenyl)-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3c): m.p. 246-247 °C. IR: (KBr, ν_{\max} , cm⁻¹) 3340, 2930, 1670, 1591, 1517, 1510, 1471, 1384, 1260, 1232, 1141, 1033, 859, 766; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz) 1.02 (s, 3H, CH₃), 1.10 (s, 3H, CH₃), 2.08 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.26 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.66 (d, $J = 16.8$ Hz, 1H, C⁹-H), 2.74 (d, $J = 16.8$ Hz, 1H, C⁹-H), 3.63 (s, 3H, CH₃O), 3.66 (s, 3H, CH₃O), 5.13 (s, 1H, CH), 6.66 (dd, $J = 8.0$ Hz, $J' = 2.0$ Hz, 1H, ArH), 6.74 (d, $J = 8.0$ Hz, 1H, ArH), 6.90 (d, $J = 2.0$ Hz, 1H, ArH), 7.32 (d, $J = 8.4$ Hz, 1H, ArH), 7.45-7.56 (m, 3H, ArH), 7.82 (d, $J = 8.0$ Hz, 1H, ArH), 8.44 (d, $J = 8.4$ Hz, 1H, ArH), 9.25 (s, 1H, NH); ¹³C NMR δ (DMSO-*d*₆, 400 MHz): 26.8, 29.6, 32.3, 40.0, 40.5, 50.5, 55.6, 55.7, 107.6, 111.5, 112.0, 119.1, 121.4 (2C), 122.4, 122.8, 125.8, 125.9, 128.1, 128.3, 130.6, 132.4, 141.7, 147.2, 148.6, 151.9, 193.8. MS (EI): *m/z* 413 (M⁺, 26), 411 (100), 394 (10), 324 (15), 296 (9), 253 (8); Anal. (%) calcd. for C₂₇H₂₇NO₃: C 78.42, H 6.58, N 3.39; found C 78.45, H 6.71, N 3.52.

7-(4-Bromophenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3d): m.p. 278-280 °C (Lit.²² 276-278 °C). IR (KBr, ν_{\max} , cm⁻¹) 3305, 2953, 1670, 1588, 1517, 1498, 1383, 1260, 1150, 1067, 1010, 848, 805, 755; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz) 0.98 (s, 3H, CH₃), 1.06 (s, 3H, CH₃), 2.05 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.25 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.66 (d, $J = 16.4$ Hz, 1H, C⁹-H), 2.72 (d, $J = 16.4$ Hz, 1H, C⁹-H), 5.21 (s, 1H, CH), 7.17 (d, $J = 8.0$ Hz, 2H, ArH), 7.25 (d, $J = 8.0$ Hz, 1H, ArH), 7.37 (d, $J = 8.4$ Hz, 2H, ArH), 7.37 (d, $J = 8.0$ Hz, 1H, ArH), 7.46~7.60 (m, 3H, ArH), 7.83 (d, $J = 8.4$ Hz, 1H, ArH), 8.47 (d, $J = 8.4$ Hz, 1H, ArH), 9.30 (s, 1H, NH).

7-(2,4-Dichlorophenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3e): m.p. 280-282 °C. IR (KBr, ν_{\max} , cm⁻¹) 3311, 2953, 1681, 1589, 1518, 1499, 1470, 1391, 1262, 1148, 1100, 1043, 858, 811, 761, 733; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz) 1.02 (s, 3H, CH₃), 1.10 (s, 3H, CH₃), 2.02 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.24 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.63 (d, $J = 16.4$ Hz, 1H, C⁹-H), 2.73 (d, $J = 16.4$ Hz, 1H, C⁹-H), 5.71 (s, 1H, CH), 7.20 (d, $J = 8.4$ Hz, 1H, ArH), 7.26-7.28 (m, 2H, ArH), 7.45 (d, $J = 8.0$ Hz, 1H, ArH), 7.49-7.61 (m, 3H, ArH), 7.81 (d, $J = 7.6$ Hz, 1H, ArH), 8.47 (d, $J = 8.4$ Hz, 1H, ArH), 9.30 (s, 1H, NH); ¹³C NMR δ (DMSO-*d*₆, 400 MHz): 27.1, 29.4, 32.3, 37.2, 40.4, 50.3, 106.7, 120.0, 121.5, 122.4, 123.2, 126.1, 126.2, 126.8, 127.8, 128.4, 128.5, 130.6, 131.1,

131.6, 131.9, 132.7, 145.5, 152.7, 193.5. MS (EI): m/z 422 (M^+ , 2), 384 (100), 368 (10), 300 (9), 264 (12); Anal. (%) calcd. for $C_{25}H_{21}Cl_2NO$: C 71.10, H 5.01, N 3.32; found C 70.91, H 5.14, N 3.50.

10,10-Dimethyl-7-(3,4-methylenedioxyphenyl)-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3f): m.p. 283-285 °C (Lit.²¹ 282-283 °C). IR (KBr, ν_{max} , cm^{-1}) 3328, 2947, 1683, 1584, 1517, 1497, 1391, 1357, 1284, 1261, 1249, 1230, 1151, 1036, 938, 808, 793, 774, 751; 1H NMR δ_H (DMSO- d_6 , 400 MHz) 1.01 (s, 3H, CH_3), 1.08 (s, 3H, CH_3), 2.07 (d, $J = 16.8$ Hz, 1H, C^{11} -H), 2.24 (d, $J = 16.8$ Hz, 1H, C^{11} -H), 2.65 (d, $J = 16.8$ Hz, 1H, C^9 -H), 2.74 (d, $J = 16.8$ Hz, 1H, C^9 -H), 5.13 (s, 1H, CH), 5.88 (d, $J = 10.8$ Hz, 2H, CH_2), 6.66 (dd, $J = 8.0$ Hz, $J' = 1.6$ Hz, 1H, ArH), 6.70 (d, $J = 8.0$ Hz, 2H, ArH), 6.75 (d, $J = 1.6$ Hz, 2H, ArH), 7.28 (d, $J = 8.4$ Hz, 1H, ArH), 7.45-7.60 (m, 3H, ArH), 7.82 (d, $J = 7.6$ Hz, 1H, ArH), 8.45 (d, $J = 8.4$ Hz, 1H, ArH), 9.24 (s, 1H, NH).

10,10-Dimethyl-7-(4-methoxyphenyl)-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3g): m.p. 257-258 °C. (Lit.²² 260-262 °C). IR (KBr, ν_{max} , cm^{-1}): 3308, 2946, 2882, 1684, 1594, 1518, 1495, 1420, 1382, 1256, 1170, 1149, 1032, 838, 811, 765, 746 (w); 1H NMR δ_H (DMSO- d_6 , 400 MHz) 1.00 (s, 3H, CH_3), 1.08 (s, 3H, CH_3), 2.04 (d, $J = 16.0$ Hz, 1H, C^{11} -H), 2.24 (d, $J = 16.0$ Hz, 1H, C^{11} -H), 2.65 (d, $J = 16.4$ Hz, 1H, C^9 -H), 2.73 (d, $J = 16.4$ Hz, 1H, C^9 -H), 3.64 (s, 3H, CH_3O), 5.14 (s, 1H, CH), 6.73 (d, $J = 8.4$ Hz, 2H, ArH), 7.12 (d, $J = 8.4$ Hz, 2H, ArH), 7.25 (d, $J = 8.0$ Hz, 1H, ArH), 7.44-7.60 (m, 3H, ArH), 7.81 (d, $J = 8.0$ Hz, 1H, ArH), 8.45 (d, $J = 8.4$ Hz, 1H, ArH), 9.23 (s, 1H, NH).

10,10-Dimethyl-7-(4-hydroxyphenyl)-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3h): m.p. 312-315 °C. IR (KBr, ν_{max} , cm^{-1}) 3298, 2957, 2923, 1675, 1569, 1514, 1493, 1466, 1389, 1248, 1167, 1148, 1115, 1062, 842, 792, 754; 1H NMR δ_H (DMSO- d_6 , 400 MHz) 1.00 (s, 3H, CH_3), 1.08 (s, 3H, CH_3), 2.04 (d, $J = 16.0$ Hz, 1H, C^{11} -H), 2.23 (d, $J = 16.0$ Hz, 1H, C^{11} -H), 2.64 (d, $J = 16.8$ Hz, 1H, C^9 -H), 2.72 (d, $J = 16.8$ Hz, 1H, C^9 -H), 5.08 (s, 1H, CH), 6.55 (d, $J = 8.4$ Hz, 2H, ArH), 6.99 (d, $J = 8.4$ Hz, 2H, ArH), 7.24 (d, $J = 8.4$ Hz, 1H, ArH), 7.44-7.58 (m, 3H, ArH), 7.81 (d, $J = 7.6$ Hz, 1H, ArH), 8.44 (d, $J = 8.4$ Hz, 1H, ArH), 9.08 (s, 1H, OH), 9.20 (s, 1H, NH). ^{13}C NMR δ (DMSO- d_6 , 400 MHz): 26.9, 29.5, 32.3, 39.6, 41.2, 50.5, 107.9, 114.9, 121.4, 121.7, 122.4, 122.7, 125.7, 125.9, 128.2 (2C), 128.3, 130.7, 132.4, 139.5, 151.7, 155.5, 193.7. MS (EI): m/z 368 (M^+ -1, 2), 329 (3), 275 (100), 247 (10), 219 (53), 191 (72); Anal. (%) calcd. for $C_{25}H_{23}NO_2$: C 81.27, H 6.27, N 3.79; found C 81.01, H 6.33, N 3.86.

7-(3,4-Dichlorophenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (3i): m.p. 284-286 °C. IR: (KBr, ν_{max} , cm^{-1}): 3310, 2952, 1671, 1592, 1518, 1498, 1469, 1381, 1262, 1151, 1126, 1060, 1029, 955, 884, 810, 761; 1H NMR δ_H (DMSO- d_6 , 400 MHz) 1.00 (s, 3H, CH_3),

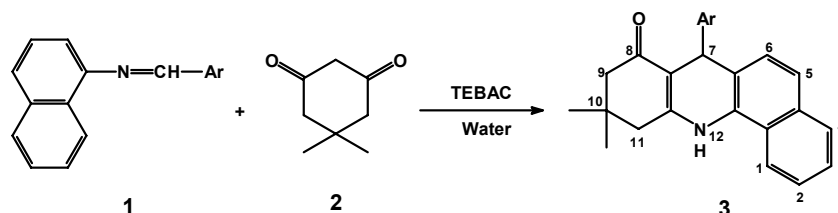
1.09 (s, 3H, CH₃), 2.07 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.26 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.67 (d, $J = 17.2$ Hz, 1H, C⁹-H), 2.76 (d, $J = 17.2$ Hz, 1H, C⁹-H), 5.28 (s, 1H, CH), 7.17 (dd, $J = 8.4$ Hz, $J' = 2.0$ Hz, 1H, ArH), 7.30 (d, $J = 8.4$ Hz, 1H, ArH), 7.44-7.62 (m, 5H, ArH), 7.84 (d, $J = 8.0$ Hz, 1H, ArH), 8.48 (d, $J = 8.8$ Hz, 1H, ArH), 9.34 (s, 1H, NH); ¹³C NMR δ (DMSO-*d*₆, 400 MHz): 26.9, 29.4, 32.3, 39.9, 40.4, 50.3, 106.7, 119.8, 121.5, 122.4, 123.3, 126.1, 126.2, 127.8, 127.9, 128.4, 128.5, 129.2, 130.6, 130.7, 130.9, 132.7, 149.5, 152.4, 193.7. MS (EI): m/z 422 (M⁺, 20), 419 (100), 390 (33), 363 (61), 300 (70), 264 (60); Anal. (%) calcd. for C₂₅H₂₁Cl₂NO: C 71.10, H 5.01, N 3.32; found C 70.87, H 5.23, N 3.36.

7-(4-Chlorophenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-9H-benzo[*c*]acridin-8-one (3j): m.p. 265-266 °C; (Lit.²² 267~269 °C). IR (KBr, ν_{\max} , cm⁻¹): 3307, 2954, 1670, 1589, 1518, 1384, 1261, 1151, 1089, 1060, 1014, 850, 806, 756; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz) 0.98 (s, 3H, CH₃), 1.08 (s, 3H, CH₃), 2.05 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.25 (d, $J = 16.0$ Hz, 1H, C¹¹-H), 2.66 (d, $J = 16.8$ Hz, 1H, C⁹-H), 2.73 (d, $J = 16.8$ Hz, 1H, C⁹-H), 5.23 (s, 1H, CH), 7.18-7.28 (m, 5H, ArH), 7.46-7.53 (m, 2H, ArH), 7.57-7.60 (m, 1H, ArH), 7.83 (d, $J = 8.4$ Hz, 1H, ArH), 8.47 (d, $J = 8.4$ Hz, 1H, ArH), 9.31 (s, 1H, NH).

7-(2-Chlorophenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-9H-benzo[*c*]acridin-8-one (3k): m.p. 275-278 °C. (Lit.²² 273-275 °C). IR (KBr, ν_{\max} , cm⁻¹): 3306, 2949, 2865, 1671, 1588, 1518, 1498, 1383, 1260, 1151, 1089, 1014, 745; ¹H NMR δ_{H} (DMSO-*d*₆, 400 MHz) 1.04 (s, 3H, CH₃), 1.10 (s, 3H, CH₃), 2.02 (d, $J = 16.4$ Hz, 1H, C¹¹-H), 2.24 (d, $J = 16.4$ Hz, 1H, C¹¹-H), 2.69 (d, $J = 16.8$ Hz, 1H, C⁹-H), 2.77 (d, $J = 16.8$ Hz, 1H, C⁹-H), 5.74 (s, 1H, CH), 7.07-7.60 (m, 8H, ArH), 7.80 (d, $J = 8.0$ Hz, 1H, ArH), 8.47 (d, $J = 8.4$ Hz, 1H, ArH), 9.26 (s, 1H, NH).

RESULTS AND DISCUSSION

When the reaction of N-arylidene-naphthalen-1-amine (**1**)²⁰ and 5,5-dimethyl-1,3-cyclohexadione (**2**) was performed in water in the presence of TEBAC at 100 °C, high yields of 10,10-dimethyl-7-aryl-7,9,10,11-tetrahydro-9H-benzo[*c*]acridin-8-one derivatives were obtained (**Scheme-I**).



Scheme-I

In order to apply this reaction to a library synthesis, various kinds of N-arylidenenaphthalen-1-amine and 5,5-dimethyl-1,3-cyclohexadione were subjected to give the corresponding 10,10-dimethyl-7-aryl-7,9,10,11-tetrahydro-9*H*-benzo[c]acridin-8-one (**3**) and representative examples are shown in Table-1. All of the N-arylidene-naphthalen-1-amine gave expected products in quantitative yields and purity. The isolated **3** were completely characterized by IR, ¹H NMR and elemental analyses. The melting points of the known compounds were in agreement with those of literature reported. The IR spectra for **3a** exhibited sharp bands at 3307 cm⁻¹ (NH), 1671 cm⁻¹ ν(C=O). The ¹H NMR spectrum of **3a** exhibited two singlets identified methyl group (0.99 and 1.11), four doublets identified methylene (2.06, 2.58, 2.70 and 2.75), a singlet identified methine (5.91) and along with multiplets (7.33-8.50) for aromatic protons. The NH proton resonance at 9.42 disappeared after addition of D₂O to the DMSO-*d*₆ solution of **3a**.

TABLE-1
RESULTS ON THE REACTION OF **1** AND **2** IN WATER AT 100 °C

Entry	Ar	Products	Time (h)	Yield (%)
1	3-NO ₂ C ₆ H ₄	3a	12	95
2	4-(CH ₃) ₂ NC ₆ H ₄	3b	18	85
3	3,4-(CH ₃ O) ₂ C ₆ H ₃	3c	18	93
4	4-BrC ₆ H ₄	3d	12	91
5	2,4-Cl ₂ C ₆ H ₃	3e	12	92
6	3,4-OCH ₂ OC ₆ H ₃	3f	18	89
7	4-CH ₃ OC ₆ H ₄	3g	18	89
8	4-OHC ₆ H ₄	3h	18	92
9	3,4-Cl ₂ C ₆ H ₃	3i	12	90
10	4-ClC ₆ H ₄	3j	12	93
11	2-ClC ₆ H ₄	3k	12	98

Finally, the reuse of the water and TEBAC was studied. At completion monitored by TLC, the reaction mixture was allowed to cool to room temperature, the solid products was isolated by filtration and filtrate was used directly. Investigations by using **1a** and **2** as model substrates showed that successive reuse of the recovery water and TEBAC. A summary of the reuse of water and TEBAC is shown in Table-2. Even in the fourth round the yield of the product **3** is fairly high.

In conclusion, an efficient green method for the synthesis of 10,10-dimethyl-7-aryl-7,9,10,11-tetrahydro-9*H*-benzo[c]acridin-8-one derivatives by condensation of N-arylidenenaphthalen-1-amine and 5,5-dimethyl-1,3-cyclohexadione was achieved. Compared to other methods^{14,15} this method has the advantages of good yields, mild reaction conditions, easy work-up, inexpensive reagents and environmentally friendly procedure.

TABLE-2
STUDY ON THE REUSE OF WATER AND TEBAC^a

Entry	Temperature (°C)	Reaction time (h)	Yield (%) ^b
1	100	12	95
2	100	12	95
3	100	12	94
4	100	12	90

^aReaction condition: 10 mL water and TEBAC, 2 mmol **1a** and 2 mmol **2**.^bIsolated yields.

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