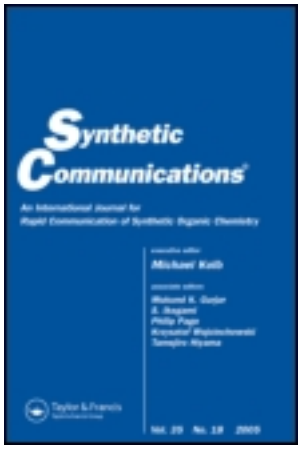

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# $NH_2SO_3H$ and $H_6P_2W_{18}O_{62} \cdot 18H_2O$-Catalyzed, Three-Component, One-Pot Synthesis of Benzo[c]acridine Derivatives

Majid M. Heravi [a], Hamideh Alinejhad [a], Fatemeh Derikvand [a], Hossein A. Oskooie [a], Bita Baghernejad [a] & Fatemeh F. Bamoharram [b]

[a] Department of Chemistry, School of Sciences, Azzahra University, Vanak, Tehran, Iran

[b] Department of Chemistry, Islamic Azad University, Mashhad Branch, Mashhad, Iran

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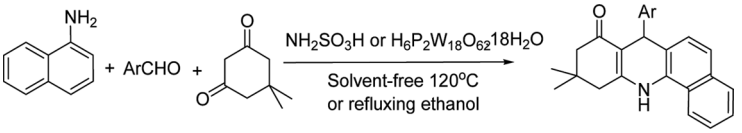
# $NH_2SO_3H$ AND $H_6P_2W_{18}O_{62} \cdot 18H_2O$-CATALYZED, THREE-COMPONENT, ONE-POT SYNTHESIS OF BENZO[*c*]ACRIDINE DERIVATIVES

**Majid M. Heravi,[1] Hamideh Alinejhad,[1] Fatemeh Derikvand,[1] Hossein A. Oskooie,[1] Bita Baghernejad,[1] and Fatemeh F. Bamoharram[2]**

[1]*Department of Chemistry, School of Sciences, Azzahra University, Vanak, Tehran, Iran*
[2]*Department of Chemistry, Islamic Azad University, Mashhad Branch, Mashhad, Iran*

## GRAPHICAL ABSTRACT




**Abstract** *We report here two simple methods for the synthesis of benzo[c]acridine derivatives from three-component, one-pot condensation of 1-naphthylamine, dimedone, and a variety of substituted aldehydes in the presence of a catalytic amount of $NH_2SO_3H$ or $H_6P_2W_{18}O_{62} \cdot 18H_2O$ under solvent-free conditions at 120°C or in refluxing ethanol.*

**Keywords** Benzo[*c*]acridine; $H_6P_2W_{18}O_{62} \cdot 18H_2O$; multicomponent one-pot reactions; $NH_2SO_3H$; solvent-free conditions

## INTRODUCTION

Solid acids can play a significant role in the development of cleaner technologies.[1] They have emerged as a substitute for conventional acidic catalysts in different areas of organic synthesis.[2–4] A large number of 1,4-dihydropyridine derivatives have pharmacological applications. They are calcium channel blockers and are widely used in the treatment of arterial hypertension and ischemic heart disease.[5] The chemical modifications of the dihydropyridine (DHP) ring and variations with different substituents[6] or heteroatoms,[7] afforded some insight into the molecular interactions at the receptor level. Acridine derivatives containing 1,4-DHP are well-known compounds for their pharmacological profile in calcium channel modulations.[8] They can be synthesized by various methods,[9–11] and recently some

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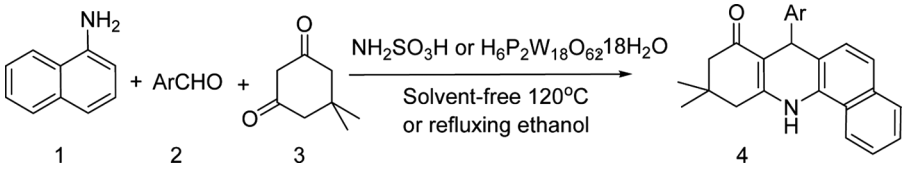
Address correspondence to Majid M. Heravi, Department of Chemistry, School of Sciences, Azzahra University, Vanak, Tehran 1234, Iran. E-mail: mmh1331@yahoo.com




**Scheme 1.** The total reaction for the synthesis of benzo[*c*]acridines.

new methods have been reported for the construction of acridine derivatives via condensing *N*-arylidenenaphthalen-1-amine with dimedone in the presence of triethylbenzyl ammonium chloride (TEBAC) or benzyltriethylammonium bromide (BTEA) in water under heating conditions.[12–14]

The use of multicomponent, reactions (MCRs) to generate remarkable and new druglike scaffolds is replete in the recent chemical literature.[15]

As a consequence of our interest in the development of new routes to heterocyclic systems from multicomponent, one-pot protocols,[16–18] herein we report two highly efficient methods for the three-component, one-pot synthesis of a series of polyhydroacridine derivatives via the reaction of 1-naphthylamine, dimedone, and a variety of aldehydes catalyzed by sulfamic acid (SA) or $H_6P_2W_{18}O_{62} \cdot 18H_2O$ as a Wells–Dawson-type heteropolyacid (HPA) in a solvent-free system or in refluxing ethanol (Scheme 1).

## RESULTS AND DISCUSSION

The procedures are very simple: 1 equiv. of 1-naphthylamine was mixed with 1 equiv. of aldehyde, 1 equiv. of dimedone, and 0.1 equiv. of $NH_2SO_3H$ or 0.01 equiv.

**Table 1.** Synthesis of benzo[*c*]acridine derivatives under solvent-free conditions

| | | | | Mp (°C) | | | |
|---|---|---|---|---|---|---|---|
| Entry | Ar- | Product | Catalyst | Observed | Reported | Time (min) | Yield (%)[a] |
| 1 | Ph | **4a** | SA | 261–262 | 260–262[19] | 110 | 70 |
| 2 | 3-$NO_2$-Ph | **4b** | SA | 269–270 | 267–269[20] | 110 | 75 |
| 3 | 2-Cl-Ph | **4c** | SA | 277–278 | 275–278[20] | 110 | 70 |
| 4 | 4-Cl-Ph | **4d** | SA | 266–267 | 265–266[20] | 100 | 70 |
| 5 | 2,4-$Cl_2$-Ph | **4e** | SA | 282–283 | 280–282[20] | 105 | 75 |
| 6 | 4-HO-Ph | **4f** | SA | 314–315 | 312–315[20] | 130 | 65 |
| 7 | 4-MeO-Ph | **4g** | SA | 257–259 | 257–258[20] | 135 | 75 |
| 8 | 4-$NMe_2$-Ph | **4h** | SA | 261–262 | 260–262[20] | 90 | 70 |
| 9 | Ph | **4a** | HPA | 269–270 | 267–269[20] | 100 | 75 |
| 10 | 3-$NO_2$-Ph | **4b** | HPA | 277–278 | 275–278[20] | 95 | 75 |
| 11 | 2-Cl-Ph | **4c** | HPA | 266–267 | 265–266[20] | 90 | 70 |
| 12 | 4-Cl-Ph | **4d** | HPA | 282–283 | 280–282[20] | 85 | 75 |
| 13 | 2,4-$Cl_2$-Ph | **4e** | HPA | 314–315 | 312–315[20] | 95 | 80 |
| 14 | 4-HO-Ph | **4f** | HPA | 257–259 | 257–258[20] | 120 | 60 |
| 15 | 4-MeO-Ph | **4g** | HPA | 261–262 | 260–262[20] | 135 | 70 |

[a]Yields were analyzed by GC.

**Table 2.** Synthesis of benzo[*c*]acridine derivatives catalyzed in refluxing ethanol

| Entry | Ar- | Catalyst | Time (min) | Yield (%)[a] |
|---|---|---|---|---|
| 1 | Ph | SA | 85 | 65 |
| 2 | 3-$NO_2$-Ph | SA | 80 | 70 |
| 3 | 4-Cl-Ph | SA | 80 | 70 |
| 4 | 2-Me-Ph | SA | 85 | 70 |
| 5 | 4-HO-Ph | SA | 95 | 60 |
| 6 | 4-MeO-Ph | SA | 90 | 70 |
| 7 | Ph | HPA | 80 | 68 |
| 8 | 3-$NO_2$-Ph | HPA | 80 | 72 |
| 9 | 4-Cl-Ph | HPA | 70 | 75 |
| 10 | 2-Me-Ph | HPA | 70 | 70 |
| 11 | 4-MeO-Ph | HPA | 70 | 60 |

[a]Yields were analyzed by GC.

**Table 3.** Recovery of the $NH_2SO_3H$ in the synthesis of 10,10-dimethyl-7-(3-nitrophenyl)-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one

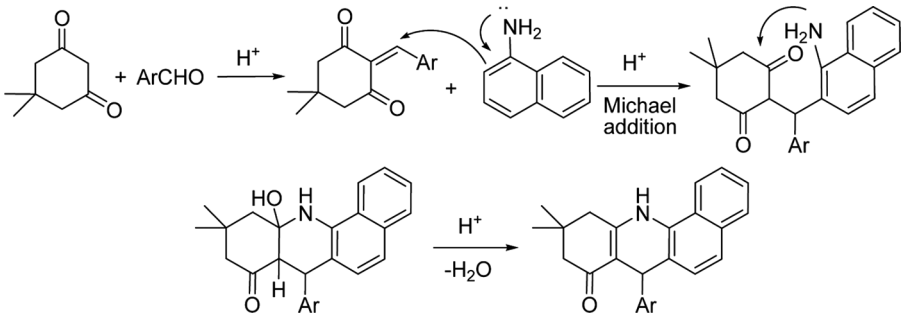
| Entry | Reaction conditions | Number of cycles | Yields (%)[a] |
|---|---|---|---|
| 1 | Solvent free, 120 °C, 80 min | 5 | 70, 68, 68, 65, 60 |
| 2 | Refluxing ethanol, 110 min | 5 | 75, 70, 67, 65, 65 |

[a]Yields were analyzed by GC.

of $H_6P_2W_{18}O_{62} \cdot 18H_2O$ and heated at 120 °C under solvent-free conditions or refluxed in ethanol (5 mL) for an appropriate time.

The scope and generality of these methods are illustrated with respect to various aromatic aldehydes, and the results are summarized in Tables 1 and 2. These one-pot procedures work well for a wide range of substrates. Various substituted aldehydes were reacted with 1-naphthylamine and dimedone to give excellent yields of the desired products. As shown in Tables 1 and 2, the electron-withdrawing or electron-donating group on the phenyl rings did not affect the reaction.

It is noteworthy to mention that the catalyst is recyclable and could be reused without significant loss of activity (Table 3).




**Scheme 2.** Mechanism for the synthesis of benzo[*c*]acridines.

A probable mechanism for this condensation reaction is patterned in Scheme 2, which begins with the condensation of dimedone and aldehyde catalyzed by Brønsted acids.

## CONCLUSIONS

In summary, we report here a high-yielding, simple, convenient, straightforward, and practical three-component, one-pot procedure for the synthesis of 7-aryl-10,10-dimethyl-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one derivatives from the condensation of 1-naphthylamine, aldehydes, and dimedone under solvent-free conditions at 120 °C or in refluxing ethanol. Using solid acids as catalyst offers advantages including simplicity of operation, easy workup, recycling of the catalyst, and good yields of products.



## EXPERIMENTAL

All the chemicals were purchased from Merck Company. Melting points were measured by using the capillary tube method with an Electrothermal 9200 apparatus. $^1$H NMR spectra were recorded on a Bruker AQS Avance 500-MHz spectrometer using tetramethylsilane (TMS) as an internal standard ($CDCl_3$ solution). Infrared (IR) spectra were recorded on the FT-IR Bruker Tensor 27 instrument. All products were known compounds and identified by comparison of their spectral and physical data with the literature description.[12,14,21] Yields were obtained using gas chromatographic (GC) analysis.

### General Procedure for the Synthesis of 7-Aryl-10,10-dimethyl-7,9,10,11-tetra-hydro-9H-benzo[c]acridin-8-one Derivatives Under Solvent-Free Conditions

A solution of an aromatic aldehyde (1 mmol), dimedone (1 mmol), 1-naphthylamine (1 mmol), and $NH_2SO_3H$ (10 mol%) or $H_6P_2W_{18}O_{62} \cdot 18H_2O$ (1 mol%) was stirred at 120 °C for an appropriate time. After completion of the reaction, which was monitored by thin-layer chromatography (TLC), boiling ethanol was added to the mixture. The catalyst was filtered off, and the mixture was cooled to room temperature. The resulting solid product was then removed by filtration and purified by column chromatography.

### Recycling of $NH_2SO_3H$

After completion of the reaction, boiling ethanol was added to the mixture, and the catalyst was recycled by simple filtration, washed with diethyl ether, dried at 80 °C, and reused for the next reaction with only a modest loss in activity. The catalyst was recovered and reused five times in the synthesis of 10,10-dimethyl-7-(3-nitrophenyl)-7,9,10,11-tetrahydro-9H-benzo[*c*] acridin-8-one, and the obtained results are summarized in Table 3.

### 10,10-Dimethyl-7-(3-nitrophenyl)-7,9,10,11-tetrahydro-9H-benzo[c]acridin-8-one (4b)

IR (KBr) ($\nu_{max}$, $cm^{-1}$): 3303, 2956, 1664, 1589, 1526, 1494, 1386, 1347, 1259, 1150, 1092, 1030, 806, 758, 730 $cm^{-1}$; $^1H$ NMR (DMSO-$d_6$, 500 MHz) $\delta_H$ (ppm): 0.98 (s, 3H, $CH_3$), 1.15 (s, 3H, $CH_3$), 2.28 (d, $J = 16.47$ Hz, 1H, $C_{11}H$), 2.36 (d, $J = 16.32$, 1H, $C_{11}$-H), 2.61 (d, $J = 16.6$ Hz, 1H, $C_9$-H), 2.65 (d, $J = 16.15$ Hz, 1H, $C_9$-H), 5.57 (s, 1H, CH), 6.89 (s, 1H, NH), 7.23 (d, $J = 8.4$ Hz, 1H, ArH), 7.4–7.43 (t, $J = 7.9$, 1H, ArH), 7.51–7.64 (m, 3H, ArH), 7.79 (d, $J = 7.7$ Hz, 1H, ArH), 7.83 (d, $J = 8.56$ Hz, 1H, ArH), 7.86 (d, $J = 7.9$, 1H, ArH), 7.98–8.1 (m, 2H, ArH); GC/MS: 398 ($M^+$).

### 10,10-Dimethyl-7-(2-chlorophenyl)-7,9,10,11-tetrahydro-9*H*-benzo[*c*]acridin-8-one (4c)

IR (KBr) ($\nu_{max}$, $cm^{-1}$): 3300, 2950, 2860, 1670, 1588, 1520, 1492, 1380, 1267, 1150, 1077, 1010, 751; $^1H$ NMR (DMSO-$d_6$, 500 MHz) $\delta_H$ (ppm): 1.03 (s, 3H, $CH_3$), 1.15 (s, 3H, $CH_3$), 2.08 (d, $J = 16.4$ Hz, 1H, $C^{11}$-H), 2.22 (d, $J = 16.4$ Hz, 1H, $C^{11}$-H), 2.58 (d, $J = 16.8$ Hz, 1H, $C^9$-H), 2.79 (d, $J = 16.8$ Hz, 1H, $C^9$-H), 5.71 (s, 1H, CH), 7.09–7.65 (m, 8H, ArH), 7.88 (d, $J = 8.0$ Hz, 1H, ArH), 8.42 (d, $J = 8.4$ Hz, 1H, ArH), 9.24 (s, 1H, NH). GC/MS: 386 ($M^+$).

### 10,10-Dimethyl-7-(4-chlorophenyl)-7,9,10,11-tetrahydro-9*H*-benzo[*c*]acridin-8-one (4d)

IR (KBr) ($\nu_{max}$, $cm^{-1}$): 3312, 2966, 1687, 1590, 1520, 1378, 1277, 1159, 1093, 1077, 1010, 859, 811, 762; $^1H$ NMR (DMSO-$d_6$, 500 MHz) $\delta_H$ (ppm): 0.99 (s, 3H, $CH_3$), 1.09 (s, 3H, $CH_3$), 2.09 (d, $J = 16.0$ Hz, 1H, $C^{11}$-H), 2.33 (d, $J = 16.0$ Hz, 1H, $C^{11}$-H), 2.76 (d, $J = 16.8$ Hz, 1H, $C^9$-H), 2.75 (d, $J = 16.8$ Hz, 1H, $C^9$-H), 5.233 (s, 1H, CH), 7.21–7.28 (m, 5H, ArH), 7.50–7.59 (m, 2H, ArH), 7.52–7.60 (m, 1H, ArH), 7.89 (d, $J = 8.4$ Hz, 1H, ArH), 8.38 (d, $J = 8.4$ Hz, 1H, ArH), 9.22 (s, 1H, NH); GC/MS: 386 ($M^+$).

### 10,10-Dimethyl-7-(2,4-dichlorophenyl)-7,9,10,11-tetrahydro-9*H*-benzo[*c*]acridin-8-one (4e)

IR (KBr) ($\nu_{max}$, $cm^{-1}$): 3321, 2966, 1681, 1589, 1521, 1487, 1471, 1388, 1260, 1155, 1102, 1057, 862, 810, 765, 730; $^1H$ NMR (DMSO-$d_6$, 500 MHz) $\delta_H$ (ppm): 1.04 (s, 3H, $CH_3$), 1.15 (s, 3H, $CH_3$), 2.08 (d, $J = 16.4$ Hz, 1H, $C^{11}$-H), 2.24 (d, $J = 16.4$ Hz, 1H, $C^{11}$-H), 2.66 (d, $J = 16.6$ Hz, 1H, $C^9$-H), 2.78 (d, $J = 16.6$ Hz, 1H, $C^9$-H), 5.80 (s, 1H, CH), 7.35 (d, $J = 8.4$ Hz, 1H, ArH), 7.25–7.35 (m, 2H, ArH), 7.52 (d, $J = 8.0$ Hz, 1H, ArH), 7.69–7.71 (m, 3H, ArH), 7.81 (d, $J = 7.6$ Hz, 1H, ArH), 8.55 (d, $J = 8.4$ Hz, 1H, ArH), 9.36 (s, 1H, NH); GC/MS: 422 ($M^+$).

### 10,10-Dimethyl-7-(4-hydroxylphenyl)-7,9,10,11-tetrahydro-9*H*-benzo[*c*]acridin-8-one (4f)

IR (KBr) ($\nu_{max}$, $cm^{-1}$): 3298, 2966, 2922, 1670, 1569, 1523, 1458, 1474, 1398, 1268, 1174, 1147, 1110, 1071, 830, 758; $^1H$ NMR (DMSO-$d_6$, 500 MHz) $\delta_H$

(ppm): 1.03 (s, 3H, $CH_3$), 1.11 (s, 3H, $CH_3$), 2.08 (d, $J = 16.0$ Hz, 1H, $C^{11}$-H), 2.33 (d, $J = 16.0$ Hz, 1H, $C^{11}$-H), 2.65 (d, $J = 16.8$ Hz, 1H, $C^9$-H), 2.77 (d, $J = 16.8$ Hz, 1H, $C^9$-H), 5.69 (s, 1H, CH), 6.78 (d, $J = 8.4$ Hz, 2H, ArH), 6.99 (d, $J = 8.4$ Hz, 2H, ArH), 7.32 (d, $J = 8.4$ Hz, 1H, ArH), 7.44–7.61 (m, 3H, ArH), 7.91 (d, $J = 7.6$ Hz, 1H, ArH), 8.49 (d, $J = 8.4$ Hz, 1H, ArH), 9.09 (s, 1H, OH), 9.207 (s, 1H, NH). GC/MS: 368 ($M^+$).

### 10,10-Dimethyl-7-(4-methoxylphenyl)-7,9,10,11-tetrahydro-9*H*-benzo[*c*]acridin-8-one (4g)

IR (KBr) ($\nu_{max}$, $cm^{-1}$): 3309, 2988, 2877, 1680, 1589, 1520, 1488, 1433, 1377, 1269, 1188, 1150, 1029, 839, 811, 765, 750; $^1$H NMR (DMSO-$d_6$, 500 MHz) $\delta_H$ (ppm): 1.01 (s, 3H, $CH_3$), 1.12 (s, 3H, $CH_3$), 2.10 (d, $J = 16.4$ Hz, 1H, $C^{11}$-H), 2.35 (d, $J = 16.4$ Hz, 1H, $C^{11}$-H), 2.78 (d, $J = 16.6$ Hz, 1H, $C^9$-H), 2.73 (d, $J = 16.6$ Hz, 1H, $C^9$-H), 3.58 (s, 3H, $CH_3O$), 5.20 (s, 1H, CH), 6.73 (d, $J = 8.4$ Hz, 2H, ArH), 7.18 (d, $J = 8.4$ Hz, 2H, ArH), 7.32 (d, $J = 8.0$ Hz, 1H, ArH), 7.49–7.58 (m, 3H, ArH), 7.66 (d, $J = 8.0$ Hz, 1H, ArH), 8.32 (d, $J = 8.4$ Hz, 1H, ArH), 9.09 (s, 1H, NH). GC/MS: 382 ($M^+$).



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