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One-Pot Synthesis of 1,8-Dioxodecahydroacridines Catalyzed by Carbon-Doped MoO_3

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Abstract—An efficient catalytic system has been developed on the basis of carbon-doped MoO_3 (CPM-3) for the synthesis of 1,8-dioxodecahydroacridines by condensation of dimedone, aromatic aldehydes, and anilines in ethanol–water (3:1) under ultrasonication. The effects of addition of polyethylene glycol (PEG-400) and carbon (0, 1, 2, and 3 wt %) as substrates were investigated systematically to get the desired carbon-doped MoO_3 material. The carbon source was prepared from the plant *Acacia arabica*, and the obtained samples were calcined at 500°C. The catalytic material was characterized by a number of sophisticated techniques such as XRD, SEM-EDS, FT-IR, TEM, NH_3-TPD, and BET. The present protocol has several advantages, including the use of a non-corrosive, nontoxic, inexpensive, and recyclable catalytic material.

Keywords: carbon substrate, MoO_3, PEG-400, acridines, nanostructure, heterogeneous catalysis

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INTRODUCTION

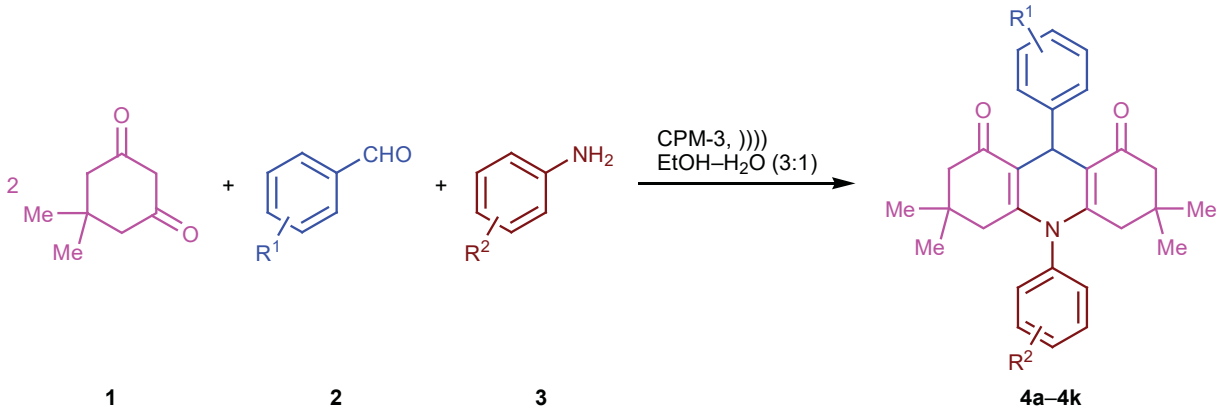
Heterocyclic compounds are important in the fields of medicinal chemistry, organic chemistry, biochemistry, and other areas of science [1]. Heterocyclic compounds exhibit antibacterial, antiviral, antifungal, anti-inflammatory, and antitumor activities [2–9]. Nitrogen-containing heterocycles constitute an important class for organic chemistry, and much research efforts aimed at synthesizing new heterocyclic compounds have contributed to the development of various synthetic protocols and found more applications in chemical sciences [10–14]. Nitrogen-containing heterocyclic compounds possess physiological and pharmacological properties and are constituents of many biologically important molecules, including vitamins, nucleic acids, pharmaceuticals, antibiotics, dyes, agrochemicals, etc. [15–20].

The use of ultrasonic irradiation in organic synthesis provides improved yields and is characterized by experimental simplicity, environmental friendliness, high efficiency [21–24], high crystallinity of the final products, and shortened reaction time [25–27].

1,8-Dioxodecahydroacridines and their derivatives can be regarded as polyfunctionalized 1,4-dihydropyridines. In recent years, 1,4-dihydropyridines and their derivatives have attracted strong interest for the treatment of cardiovascular diseases, such as angina pectoris [28] and hypertension [29]. Acridine derivatives have been used to synthesize labeled conjugates with medicines, peptides, proteins, and nucleic acids, which exhibit antitumor and DNA-binding properties [30–32]. They also exhibit antitumor, antitubercular, antimalarial, antibacterial, antihypertensive, fungicidal, anticancer, anti-inflammatory, and antidiabetic activities [33–41] and are used as dyes and photoinitiators [42–45], as well as semiconductors in materials science and luminescent agents in spectroscopy [46, 47].

Literature survey showed that there are various procedures for the synthesis of 1,8-dioxodecahydroacridines by condensation of dimedone, aldehydes, and aniline or ammonium acetate in the presence of several catalysts such as silica-bonded *S*-sulfonic acid (SBSSA) [48], $Zn(OAC)_2 \cdot H_2O$ or L-proline [49], Amberlyst-15 [50–54], benzyl(triethyl)ammonium chloride (BTEAC) [55], proline [56], ZnO nanoparticles [57], $CeCl_3 \cdot 7H_2O$ [58], nano-Fe_3O_4 [59], silica-bonded *N*-propylsulfamic acid (SBNPSA) [60], microwave irradiation [61, 62], PMA-SiO_2 [63], and *p*-dodecylbenzenesulfonic acid (DBSA) [64]. These methods suffer from one or more disadvantages such as long reaction time, low yield, the use of volatile

Scheme 1.



1 **2** **3** **4a–4k**

$R^1 = R^2 = H$ (**a**), $R^1 = 3\text{-}NO_2$, $R^2 = H$ (**b**), $R^1 = 4\text{-}Cl$, $R^2 = H$ (**c**), $R^1 = 4\text{-}Me$, $R^2 = H$ (**d**), $R^1 = 2\text{-}Cl$, $R^2 = H$ (**e**), $R^1 = 3\text{-}OH$, $R^2 = 4\text{-}Me$ (**f**), $R^1 = 4\text{-}OMe$, $R^2 = H$ (**g**), $R^1 = 3\text{-}NO_2$, $R^2 = 4\text{-}Me$ (**h**), $R^1 = 4\text{-}Cl$, $R^2 = 4\text{-}Me$ (**i**), $R^1 = R^2 = 4\text{-}Me$ (**j**), $R^1 = 2\text{-}Cl$, $R^2 = 4\text{-}Me$ (**k**).

solvents, and harsh reaction condition. Hence, we have developed a simple and efficient catalytic system for the synthesis of 1,8-dioxodecahydroacridine derivatives.

In last decade, rapid progress has been made in the field of transition metal oxides. In particular, molybdenum oxide (MoO_3) exhibited high selectivity in the synthesis of heterocyclic compounds [65–71]. Our research focused on heterogeneous catalysis and synthesis of biologically active organic compounds [72, 73]. In continuation of these studies, now we were interested in finding a simple and efficient catalytic system using carbon-doped MoO_3 (CPM-3). It was utilized for the synthesis of 1,8-dioxodecahydroacridine derivatives by condensation of dimedone, aldehydes, and aromatic amines in ethanol–water (3:1) under ultrasonication (Scheme 1).

RESULTS AND DISCUSSION

Initially, we studied the condensation of dimedone (2 mmol), benzaldehyde (1 mmol), and aniline (2 mmol) in the presence of CPM-3 as a model reaction. Various solvents such as benzene, chloroform, methanol, and acetonitrile were tried, but low to moderate yields were obtained. When ethanol and water were used, the results were satisfactory. A mixture of ethanol and water at a ratio of 1:1 provided moderate yields, whereas a good yield was achieved using ethanol–water at a ratio 3:1 without ultrasonication; however, the reaction time was longer. The best result (good to excellent yield) was observed in EtOH–H_2O (3:1) under ultrasonic irradiation (Table 1).

After screening of solvents, we studied various catalysts. The reactions in the absence of a catalyst

Table 1. Synthesis of 1,8-dioxodecahydroacridine **4a** in the presence of CPM-3 in different solvents[a]

Entry no.	Solvent	Reaction time, min	Yield of **4a**,[b] %
1	Benzene	60	35
2	Methylene chloride	60	40
3	Methanol	60	56
4	Acetonitrile	60	76
5	Water	60	72
6	Ethanol	60	77
7	Ethanol–water (1:1)	30	79
8	Ethanol–water (3:1)	30	82
9	**Ethanol–water (3:1), ultrasonication**	**10**	**91**

[a] Dimedone (2 mmol), benzaldehyde (1 mmol), aniline (1 mmol), catalyst (0.1 g), solvent (20 mL).

[b] Isolated yield.

Table 2. Synthesis of 1,8-dioxodecahydroacridine **4a** in the presence of different catalysts[a]

Entry no.	Catalyst	Reaction time, min	Yield, of **4a**,[b] %
1	No catalyst	300	51
2	Ultrasonication without a catalyst	120	62
3	CM-0	30	65
4	CPM-1	30	84
5	CPM-2	20	85
6	CPM-3	10	91

[a] Dimedone (2 mmol), benzaldehyde (1 mmol), aniline (1 mmol), catalyst (0.1 g), EtOH/H_2O (3:1, 20 mL), ultrasonication.
[b] Isolated yield.

Table 3. Synthesis of 1,8-dioxodecahydroacridine derivatives **4a**–**4k** under the optimized conditions[a]

Compound no.	R^1	R^2	Reaction time, min	Yield,[b] %	mp, °C
4a	H	H	10	91	254–255 [50]
4b	3-NO_2	H	30	94	285–287 [51]
4c	4-Cl	H	20	74	271–272 [51]
4d	4-Me	H	30	87	290–292 [52]
4e	2-Cl	H	30	81	180–182 [52]
4f	3-OH	4-Me	25	82	281–283 [54]
4g	4-OMe	H	30	93	222–224 [50]
4h	3-NO_2	4-Me	30	77	150–151 [50]
4i	4-Cl	4-Me	25	74	220–221 [50]
4j	4-Me	4-Me	30	87	194–196 [53]
4k	2-Cl	4-Me	20	81	188–190 [54]

[a] Dimedone (2 mmol), substituted benzaldehyde (1 mmol), aniline or 4-methylaniline (1 mmol), CPM-3 catalyst (0.1 g), EtOH/H_2O (3:1, 20 mL), ultrasonication.
[b] Isolated yield.

with and without ultrasonication gave poor yields, while the other catalysts like CM-0, CPM-1, and CPM-2 showed lower activity under the same conditions. As follows from the data in Table 2, CPM-3 showed the highest catalytic activity. This may be due to the small particle size and high porosity of the catalyst (see SEM, TEM, and BET surface analysis data below), which are related to the amount of carbon added.

Under the developed conditions, good to excellent yields (74–93%) were obtained with aldehydes containing both electron-withdrawing and electron-donating groups in the aromatic ring. Also, both aniline and *p*-toluidine equally underwent the excellent conversion. The structure of all products was confirmed by FT-IR, ^{1}H NMR, and mass spectra (Table 3).

The reusability of the catalyst was tested using the same model reaction under the optimized conditions. After completion of the reaction, the catalyst was easily separated from the reaction mixture by simple filtration, washed with *n*-hexane, and dried at 80°C. The isolated catalyst was then used for the next run.

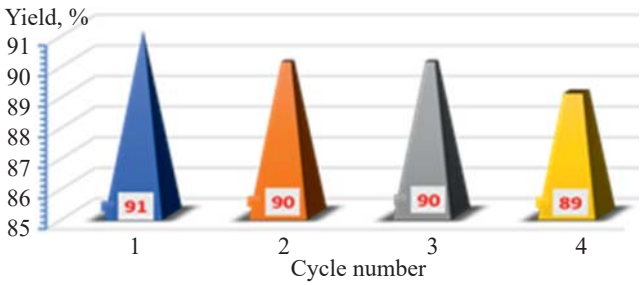


Fig. 1. Reusability of CPM-3 catalyst.

It was found that CPM-3 could be reused at least 4 times without significant loss in catalytic activity (Fig. 1) [72, 74].

Catalyst characterization. Catalyst characterization is one of the crucial aspects of catalyst design since it gives information about crystallinity, surface structure, nature of active sites, particle size and morphology, acidity, basicity, and other characteristic features. The prepared samples were characterized by various sophisticated techniques.

Figure 2 shows the XRD patterns of carbon-doped MoO_3 with addition of PEG-400, namely CPM-0, CPM-1, CPM-2, and CPM-3 samples. Highly intense and sharp peaks at 2θ = 23.36, 25.75, 27.30, 33.61, and 38.96 were observed due to the planes (110), (040), (021), (111), and (060) corresponding to the orthorhombic crystal symmetry. All XRD peaks exactly matched literature values for MoO_3 peaks from the JCPDS card 76-1003 [75] with the crystal lattice parameters a = 3.9628, b = 13.8550, and c = 3.6964 Å. The strong and sharp peaks suggest that the examined samples are crystalline. The peak observed in Fig. 2d at 2θ = 30.0° for the plane (421) corresponds to the cubic crystal symmetry of carbon [76]. The average particle size of the powder were calculated using the Debye–Scherrer formula; it was estimated around 9–10 nm [77, 78].

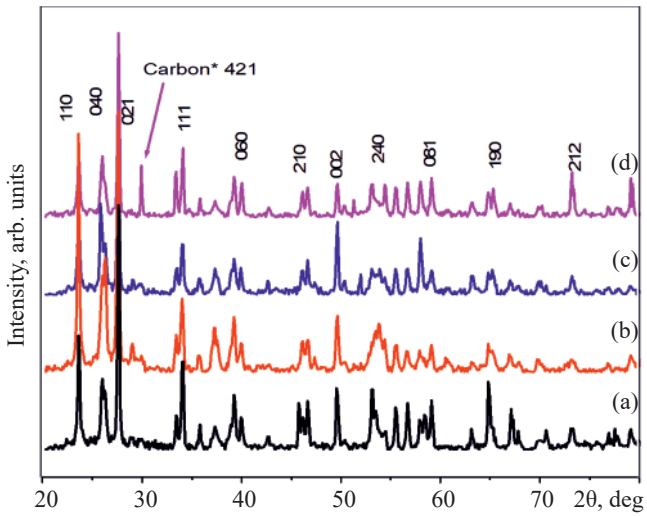


Fig. 2. XRD patterns of carbon-doped MoO_3 with addition of PEG-400: (a) CPM-0, (b) CPM-1, (c) CPM-2, and (d) CPM-3.

The surface morphology and elemental composition of the catalyst were studied by scanning electron microscopy (SEM) in combination with energy-dispersive X-ray spectroscopy (EDS). Figure 3 shows the morphology of CPM-3. It is clearly seen that the sample is characterized by porous surface with small

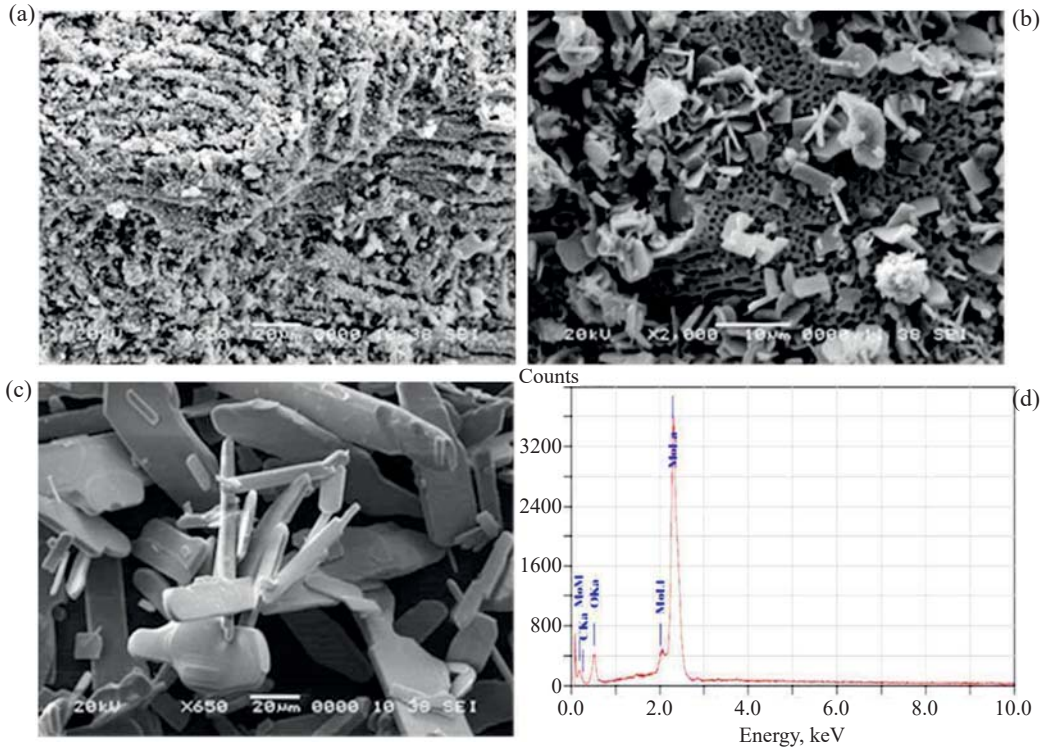


Fig. 3. (a–c) SEM and (b) EDS patterns of CPM-3.

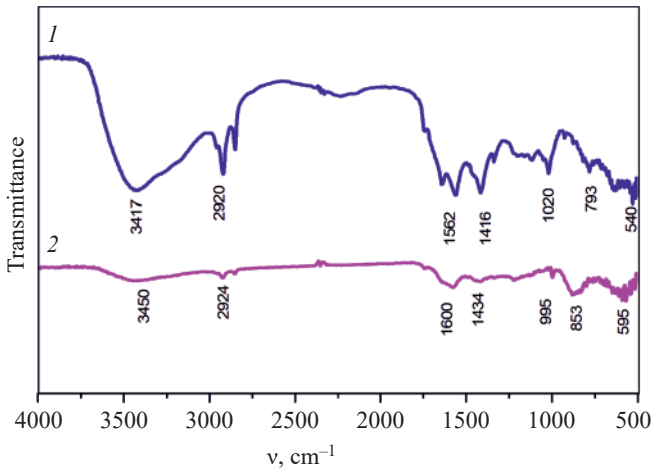


Fig. 4. FT-IR spectra of (*1*) CPM-0 and (*2*) CPM-3.

particle size. The elemental composition of CPM-3 was determined by EDS (Fig. 3d). The observed Mo/C/O atomic ratio is fairly close to the expected bulk ratio, indicating a good distribution of the metal species over the sample. Furthermore, it is seen that the minimum stoichiometric ratio of required elements in CPM-3 is maintained.

Figure 4 shows the FT-IR spectra of CPM-0 and CPM-3, respectively. A sharp band in the range 540–595 cm^{-1} was assigned to the Mo–Mo bond, and the Mo=O terminal double bond gave rise to a band around 793–853 cm^{-1}. Absorption bands at 1410–1600 cm^{-1} were attributed to C–C and C=C vibrations, and the band around 2926 cm^{-1} may appear due to CH_2 and or C(OH) stretching modes [79]. The broad band around 3417–3450 cm^{-1} was assigned to O–H stretching modes of the adsorbed water [80, 81].

The TEM image of CPM-3 shown in Fig. 5 clearly indicates the presence of highly crystalline nanorods of MoO_3 with a particle size of 10.42 to 18.57 nm. It follows from the selected area electron diffraction (SAED) pattern that the obtained *d* and *hkl* values correspond to the orthorhombic crystal structure of MoO_3. These values are also consistent with those observed in the XRD pattern [82, 83].

The ammonia temperature programmed desorption (NH_3-TPD) provides information about the total concentration and strength of acidic sites present in a material. The results of NH_3-TPD analysis of CPM-3 are shown in Fig. 6. A broad ammonia desorption profile in the range 400–550°C suggests the presence of a large number of acid sites with moderate strength. In addition to this broad peak, one low-temperature desorption peak is also observed in the range 200–350°C. Clearly, ammonia desorption in two regions indicates that the material has both Lewis and Brønsted acidic sites.

The first low-temperature desorption peak observed at 262°C is mainly due to Lewis sites present in the CPM-3 sample. Likewise, the broad desorption peak observed around 519°C corresponds to Brønsted acidic sites [84–86]. It is frequently postulated that ammonia may reduce the surface of oxides; therefore, the high-

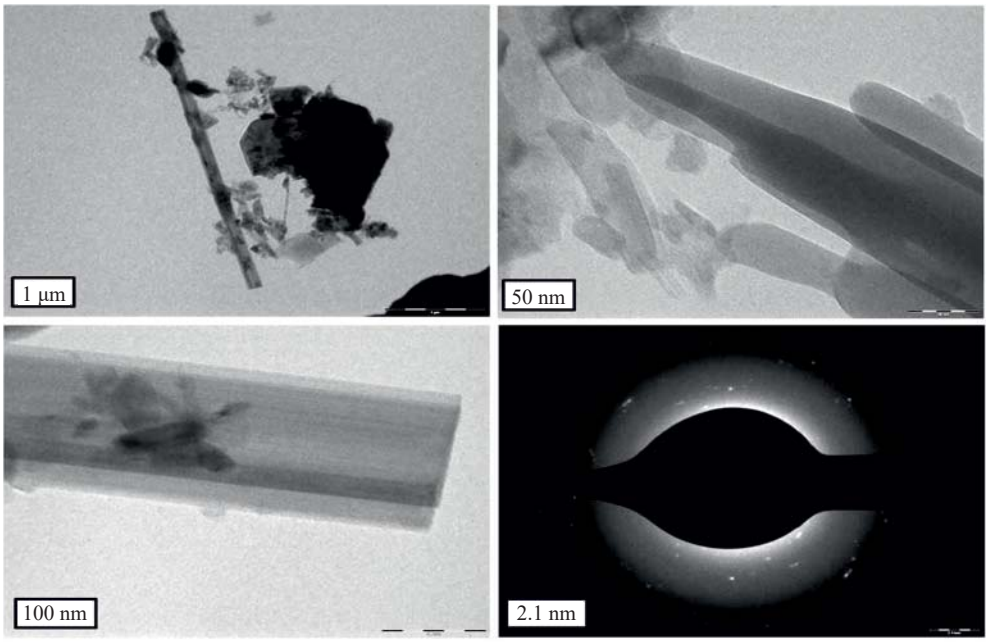


Fig. 5. TEM images of CPM-3.

temperature peak found for the MoO_3 sample could be indicative of strong acid sites created by reduction during the TPD process [87].

The surface area of CPM-3 was determined by the Brunauer–Emmett–Teller method (Fig. 7) based on the nitrogen adsorption/desorption isotherm. The single-point surface area was estimated at 2.5042 m^2/g (p/p_0), and the BET surface area, at 2.7629 m^2/g [88]. It means that prepared material has a large surface area and hence it could be expected to show high catalytic activity.

The average pore diameter of CPM-3 is 14.10 nm, which indicates increased pore size and surface area. Similar results were obtained from the XRD (12.02 nm) and TEM data (10.42 to 18.57 nm).

Similarly, the BJH (Barrett, Joyner, and Halenda) pore volume of CPM-3 is 3.56×10^{-3} cm^3/g (Fig. 8). The decrease in the average pore diameter is due to the formation of porous surface which is also observed in SEM images [89].

EXPERIMENTAL

Reagent-grade chemicals, namely ammonium heptamolybdate, oxalic acid, and ammonia (Ranbaxy Fine Chemicals), as well as poly(ethylene glycol)-400 (PEG-400) (Qualigens Fine Chemicals), were used without further purification. X-Ray diffraction analysis (XRD) of calcined samples was carried out with a Philips X-ray diffractometer (Cu K_α radiation, λ 1.54 Å; 2θ range 20–80°). Surface morphology study and elemental analysis were carried out using scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM/EDS) using a JEOL JED 2300 (LA) instrument. The IR spectra were recorded on a Shimadzu FTIR/4100 (Japan) spectrometer in the range 4000–500 cm^{-1}. The ^1H and ^{13}C NMR spectra were recorded on a Varian Mercury 300 spectrometer at 300 and 75 MHz, respectively, using TMS as internal standard. The microscopic nanostructure and particle size were determined using a Philips CM-200 transmission electron microscope at 200 kV (L = 600, l = 0.0025 nm), and the selected area electron diffraction (SAED) patterns of the prepared samples were produced to get general information about the obtained crystals. Temperature-programmed desorption (NH_3-TPD) measurements were carried out on a Micromeritics ChemiSorb 2750 instrument (Chemisoft TPx V1.02 software). A 100-mg sample was pretreated at 150°C in a helium flow of 25 cm^3/min for 1 h. Gaseous ammonia was then added to the helium environment,

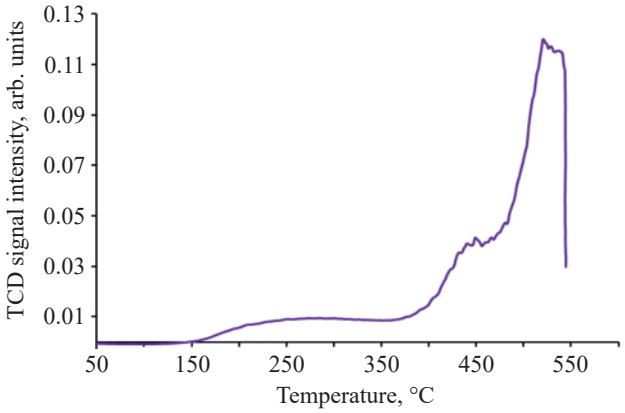


Fig. 6. NH_3-TPD plot for CPM-3.

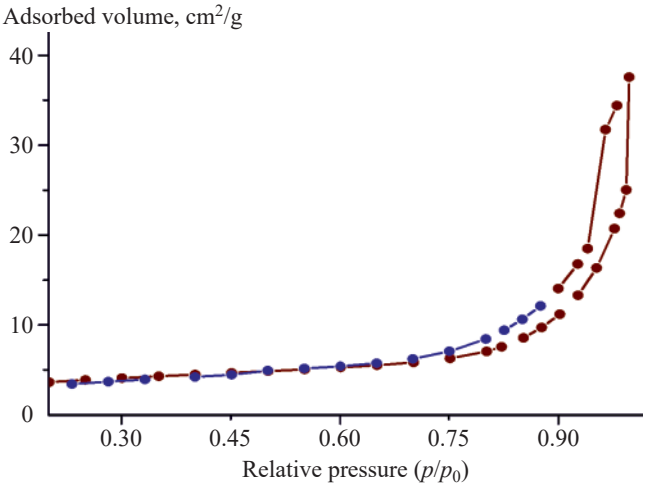


Fig. 7. Nitrogen adsorption/desorption isotherm of CPM-3.

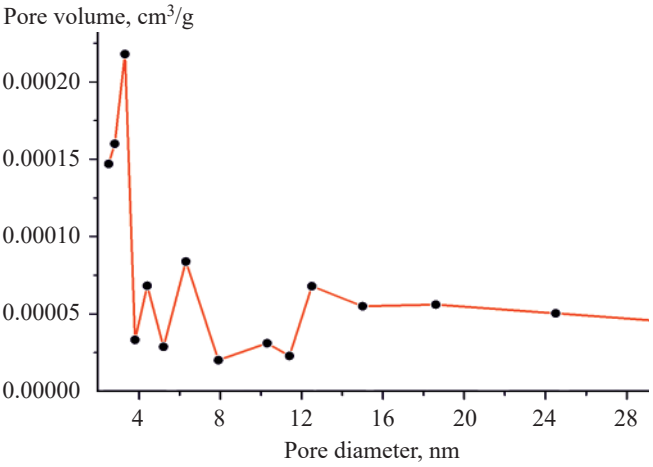


Fig. 8. Pore volume distribution of CPM-3.

and the sample was saturated for 30 min at 150°C. The helium gas was flushed for stable baseline, the sample was cooled to room temperature, and TPD measurements were performed from 50 to 500°C at a rate of 10 deg/min. The surface area of samples was charac-

terized by the BET method by measuring the adsorption of nitrogen at 77 K with a Micromeritics ASAP 2010 instrument.

Preparation of carbon-doped MoO_3 with PEG-400. A series of modified samples, viz. CPM-0, CPM-1, CPM-2, and CPM-3, were prepared by addition of carbon powder to a solution containing PEG-400 on MoO_3 by simple impregnation method. Solutions of ammonium heptamolybdate (0.2 M), oxalic acid (0.2 M), and PEG-400 (0.5 M) were mixed together, and finely powdered carbon (0, 1, 2, 3 wt %) was added to the mixture. Excess water was evaporated with continuous stirring, and the residue was dried at 110°C for 12 h and then calcined at 500°C for 2 h in air atmosphere. The carbon used for doping was prepared from the natural plant source *Acacia arabica* according to the procedure reported previously [74, 90].

General procedure for the synthesis of 1,8-dioxodecahydroacridine derivatives 4a–4k. A mixture of dimedone (2 mmol), benzaldehyde (1 mmol), aniline (1 mmol), and catalyst (0.1 g) in ethanol–water (3:1, 20 mL) was placed in a single-neck round-bottom, which was immersed in a water bath of an ultrasonic cleaner and exposed to high-intensity ultrasonic irradiation (600 W, 20 kHz) for a required time. The progress of the reaction was monitored by TLC using petroleum ether–ethyl acetate as eluent. After completion of the reaction, the mixture was heated to dissolve the product, and the catalyst was separated from the reaction mixture by simple filtration. The filtrate was evaporated, and the crude product was recrystallized from ethanol.

3,3,6,6-Tetramethyl-9,10-diphenyl-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4a). White solid, mp 254–255°C. IR spectrum (KBr), ν, cm^{-1}: 2953, 2870, 1662, 1488, 1366, 1201, 1151, 955, 833, 740, 698. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.54–7.42 m (5H), 7.32–7.09 m (5H), 4.74 s (1H), 2.45 d (4H), 2.18 d (4H), 1.09 s (6H), 0.98 s (6H). ^{13}C NMR spectrum (75 MHz, $CDCl_3$), δ_C, ppm: 27.1, 33.9, 40.2, 51.4, 109.0, 117.1, 120.3, 131.0, 140.8, 152.4, 196.0. Mass spectrum: *m*/*z* 426.24 $[M - H]^+$. $C_{29}H_{31}NO_2$.

3,3,6,6-Tetramethyl-9-(3-nitrophenyl)-10-phenyl-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4b). Yellow solid, mp 285–287°C. IR spectrum (KBr), ν, cm^{-1}: 3292, 2953, 1589, 1367, 1261, 1201, 1145, 987, 817, 691, 574. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 8.38 s (1H), 7.48 s (1H), 7.33 s (2H), 6.72–6.59 m (5H), 4.72 s (1H), 2.44 d (4H), 2.20 d (4H), 1.08 s (6H), 0.99 s (6H).

9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4c). Yellow solid, mp 271–272°C. IR spectrum (KBr), ν, cm^{-1}: 3202, 2953, 1653, 1472, 1356, 1198, 1102, 997, 859, 711, 658, 699, 574. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 8.15 s (2H), 7.85 s (2H), 7.26–7.16 m (5H), 4.71 s (1H), 2.46 d (4H), 2.21 d (4H), 1.10 s (6H), 0.98 s (6H).

3,3,6,6-Tetramethyl-9-(4-methylphenyl)-10-phenyl-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4d). Green solid, mp 290–292°C. IR spectrum (KBr), ν, cm^{-1}: 3321, 2963, 1716, 1610, 1472, 1378, 1240, 1134, 1071, 987, 859, 754, 648, 699, 574. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 8.52 m (1H), 7.85 m (1H), 7.79 m (2H), 7.26–7.00 m (5H), 4.71 s (1H), 2.45 d (4H), 2.24 d (4H), 2.41 s (3H), 1.09 s (6H), 0.99 s (6H).

9-(2-Chlorophenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4e). Yellow solid, mp 188–190°C. IR spectrum (KBr), ν, cm^{-1}: 3397, 2953, 1726, 1610, 1472, 1378, 1230, 1198, 1134, 1071, 987, 849, 743, 658, 699, 574. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.47 m (1H), 7.37–7.25 m (3H), 7.13–7.08 m (5H), 4.87 s (1H), 2.45 d (4H), 2.33 d (4H), 1.11 s (6H), 0.94 s (6H).

9-(3-Hydroxyphenyl)-3,3,6,6-tetramethyl-10-(4-methylphenyl)-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4f). White solid, mp 281–182°C. IR spectrum (KBr), ν, cm^{-1}: 3312, 2963, 1663, 1526, 1463, 1356, 1198, 1134, 997, 817, 732, 691, 699, 574. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.42–7.29 m (4H), 5.37 s (1H), 4.83 s (1H), 2.50 d (4H), 2.18 d (4H), 2.37 s (3H), 1.11 s (6H), 0.99 s (6H).

9-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4g). Light yellow solid, mp 222–224°C. IR spectrum (KBr), ν, cm^{-1}: 3309, 2956, 1662, 1595, 1452, 1361, 1199, 1141, 1003, 840, 742, 698, 572, 524. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.30 m (2H), 7.18–7.23 m (2H), 7.16–7.07 m (5H), 4.75 s (1H), 2.46 d (4H), 2.21 d (4H), 3.29 s (3H), 1.10 s (6H), 0.99 s (6H).

3,3,6,6-Tetramethyl-10-(4-methylphenyl)-9-(3-nitrophenyl)-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4h). White solid, mp 150–151°C. IR spectrum (KBr), ν, cm^{-1}: 3312, 2963, 1663, 1515, 1356, 1198, 1145, 997, 807, 699, 574. ^{1}H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.40–7.25 m (8H), 4.83 s (1H), 2.50 d (4H), 2.22 d (4H), 2.50 s (3H), 1.11 s (6H), 0.99 s (6H).

9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-methylphenyl)-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4i). Yellow–green solid, mp 220–221°C. IR spectrum (KBr), ν, cm^{-1}: 3312, 2953, 1663, 1472, 1356, 1198, 1092, 997, 849, 711, 606, 699, 521. ^1H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.85 s (2H), 7.45 s (2H), 7.26–7.15 m (4H), 4.71 s (1H), 2.46 d (4H), 2.21 d (4H), 2.51 s (3H), 1.10 s (6H), 0.99 s (6H).

3,3,6,6-Tetramethyl-9,10-bis(4-methylphenyl)-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4j). Yellow–green solid, mp 194–196°C. IR spectrum (KBr), ν, cm^{-1}: 3202, 2963, 1663, 1463, 1356, 1198, 1102, 997, 882, 700, 658, 699, 574. ^1H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.98–7.00 m (8H), 4.71 s (1H), 2.45 d (4H), 2.24 d (4H), 2.49 s (6H), 1.09 s (6H), 0.99 s (6H).

9-(2-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-methylphenyl)-3,4,6,7,9,10-tetrahydroacridine-1,8(2*H*,5*H*)-dione (4k). Yellow solid, mp 188–190°C. IR spectrum (KBr), ν, cm^{-1}: 3397, 2963, 1663, 1472, 1356, 1198, 1134, 1007, 839, 732, 617, 699, 574. ^1H NMR spectrum (300 MHz, $CDCl_3$), δ, ppm: 7.37 m (1H), 7.34–7.25 m (3H), 7.16–7.06 m (4H), 4.87 s (1H), 2.45 d (4H), 2.30 d (4H), 2.51 s (3H), 1.11 s (6H), 0.94 s (6H).

CONCLUSIONS

A new methodology has been proposed for the preparation of 1,8 dioxodecahydroacridine derivatives from dimedone, benzaldehydes, and anilines in the presence of carbon-doped MoO_3 in $EtOH/H_2O$ (3:1) solvent system under ultrasonication. The effect of the concentration of carbon substrate in PEG-400/MoO_3 was successfully evaluated. The catalyst was recovered and reused at least four times without any noticeable loss of reactivity. The advantages of the proposed method include mild reaction conditions, experimental simplicity, short reaction time, and high yield.

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CONFLICT OF INTEREST

The authors declare the absence of conflict of interest.

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