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# Molybdenum Oxide Supported on Silica ($MoO_3/SiO_2$): An Efficient and Reusable Catalyst for the Synthesis of 1,8-dioxodecahydroacridines Under Solvent-free Conditions

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**Abstract.** Silica supported molybdenum oxide ($MoO_3/SiO_2$) was found to be an efficient, eco-friendly and heterogeneous catalyst for the multicomponent reaction of aromatic aldehydes, dimedone and ammonium acetate or aromatic amines under solvent-free conditions to afford the corresponding 1,8-dioxodecahydroacridines in high yields. The catalyst can be easily recovered and reused for several times without considerable loss of activity. Furthermore, the present method offers several advantages, such as an easy experimental and work-up procedures, short reaction times and good to excellent yields.
**Key words:** Silica Supported Molybdenum Oxide, Multicomponent Reaction, solvent-free Conditions, 1,8-dioxodecahydroacridines.

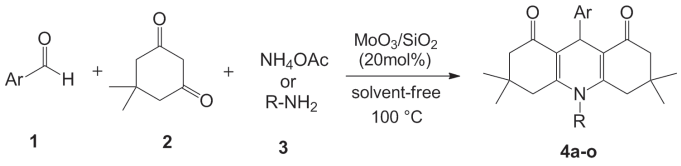
**Resumen.** Se encontró que el óxido de molibdeno soportado en sílica ($MoO_3/SiO_2$) es un catalizador eficiente y amigable con el ambiente para la reacción multicomponente de aldehídos aromáticos, dimedona y acetato de amonio o aminas aromáticas bajo condiciones libres de disolvente para producir las correspondientes 1,8-dioxodecahydroacridinas con rendimientos elevados. El catalizador puede ser recuperado fácilmente y usado por varias veces sin pérdidas de actividad considerables. Más aún, este método ofrece varias ventajas como procedimientos sencillos, tiempos cortos de reacción y rendimientos de buenos a excelentes.
**Palabras clave:** Óxido de molibdeno soportado en sílica, reacción multicomponente, condiciones libres de disolvente, 1,8-dioxodecahydroacridinas.

## Introduction

Multi component reactions (MCRs) make possible for the speedy synthesis of molecular libraries that have a high degree of structural diversity. Combinations of different starting materials can produce a variety of products with facility, which is of great value in the search for new drugs and chemical compounds [1].

Acridine and acridine-1, 8-dione derivatives are polyfunctionalized 1,4-dihydropyridine derivatives. They have a wide range of pharmacological properties such as antimalarial [2], anticancer [3], cytotoxic [4], antimicrobial [5], and widely prescribed as calcium β-blockers [6]. The discovery of acridines as antitumor agents has attracted the attention of organic chemists and led to intensive interest in the synthesis of several drugs based on acridine, therefore, we can conclude that anticancer is the most important pharmacologic property of these compounds [7]. Additionally, 1,8-dioxodecahydroacridines were created to act as laser dyes [8], and used as photo initiators [9]. Many procedures were explained for the synthesis of acridine derivatives from dimedone, aldehydes and different nitrogen sources in the presence of Amberlyst-15 [10], triethylbenzyl ammonium chloride (TEBAC) [11], ammonium acetate on basic alumina [12], *p*-dodecylbenzenesulfonic acid (DBSA) [13], and $[TBA]_2[W_6O_{19}]$ [14], and ionic liquids ([Hmim]TFA) [15]. However, most of these reported procedures have disadvantages, including low yields, prolonged reaction time, toxic organic solvents and harsh reaction conditions. Therefore, the development of simple, efficient, high-yielding, and environmentally friendly methods under mild conditions using new catalysts for the synthesis of 1,8-dioxodecahydroacridines is still necessary. Replacement of conventional toxic and pollutant Brønsted and Lewis acid catalysts with environmentally benign and reusable solid heterogeneous catalysts is an active area of current research. Using solid acid catalysts have some advantages such as ease of products separation, recycling of the catalyst and environmental acceptability as compared to a liquid acid catalyst [16]. There are many surfaces for supporting different acid catalysts, for example: $SiO_2$ [17], $Al_2O_3$ [18], $TiO_2$ [19], and hydroxyapatite [20], among them, silica has many advantages for supporting $MoO_3$ as an acidic catalyst. It is insoluble in common organic solvents, causes low corrosion, and shows environmental acceptability. Moreover, the products could be easily separated from the reaction mixture and the catalyst is recoverable without decreasing its activity [21, 22].

Due to our interest in the synthesis of heterocyclic compounds and in continuation of our previous works on the applications of reusable catalysts in organic reactions [23-30], herein we report a new and efficient method for the synthesis of 1,8-dioxodecahydroacridines using $MoO_3/SiO_2$ as a catalyst under solvent-free conditions (Scheme 1). To the best of our knowledge there are no examples on the use of $MoO_3/SiO_2$ as catalysts for the synthesis of 1, 8-dioxodecahydroacridines.




R= Ph, 4-MePh, 4-MeOPh

**Scheme 1.** Synthesis of 1,8-dioxodecahydroacridines in the presence of $MoO_3/SiO_2$.

## Results and discussion

### Catalyst characterization

*FT-IR analysis*

The FT-IR spectra of $SiO_2$, fresh and recovered $MoO_3/SiO_2$ (20 mol%), are presented in Fig. 1a-c, respectively. In the Fig. 1a, the intensive bands at around 1083, 811, and 470 $cm^{-1}$ are ascribed to Si-O vibration of $SiO_2$ and in the Fig. 1b, the additional bands observed at 996, 867 and 605 $cm^{-1}$ correspond to $MoO_3$. These bands confirm the successful supporting of the molybdenum oxide ($MoO_3$) on the silica [32]. Furthermore, the FT-IR spectrum of $MoO_3/SiO_2$ (Fig. 1c) shows that there is no substantial difference in the structure of recovered and fresh catalyst which was used the first time in the reaction. In addition, this evidence showed that $MoO_3$ was well supported on silica, and these interactions with silica are stable and during the reaction, no leaching occurs in the reaction medium.

*XRD analysis*

The X-Ray diffraction pattern of the $SiO_2$, fresh and used $MoO_3/SiO_2$ (20 mol%) is presented in Fig. 2a-c, respectively. The XRD patterns of 20% $MoO_3$ supported on silica (Fig. 2b) exhibit sharp peaks on the broad underlying peaks characteristic of the amorphous silica at $2\theta = 24°$. These intense peaks observed at $2\theta = 12.9$, 23.4, 25.8, and 27.4° are characteristic of the α-$MoO_3$ orthorhombic phase [33]. Furthermore, the XRD pattern of the used $MoO_3/SiO_2$ (Fig. 2c) showed that the structure of the catalyst remained almost the same after three times reuse. In addition, the weight of the recovered catalyst is the same as the amount of the fresh catalyst that was used the first time in the reaction.

*SEM analysis*

The scanning electron microscope (SEM) study provides an insight on the morphology of prepared catalyst. The SEM im-

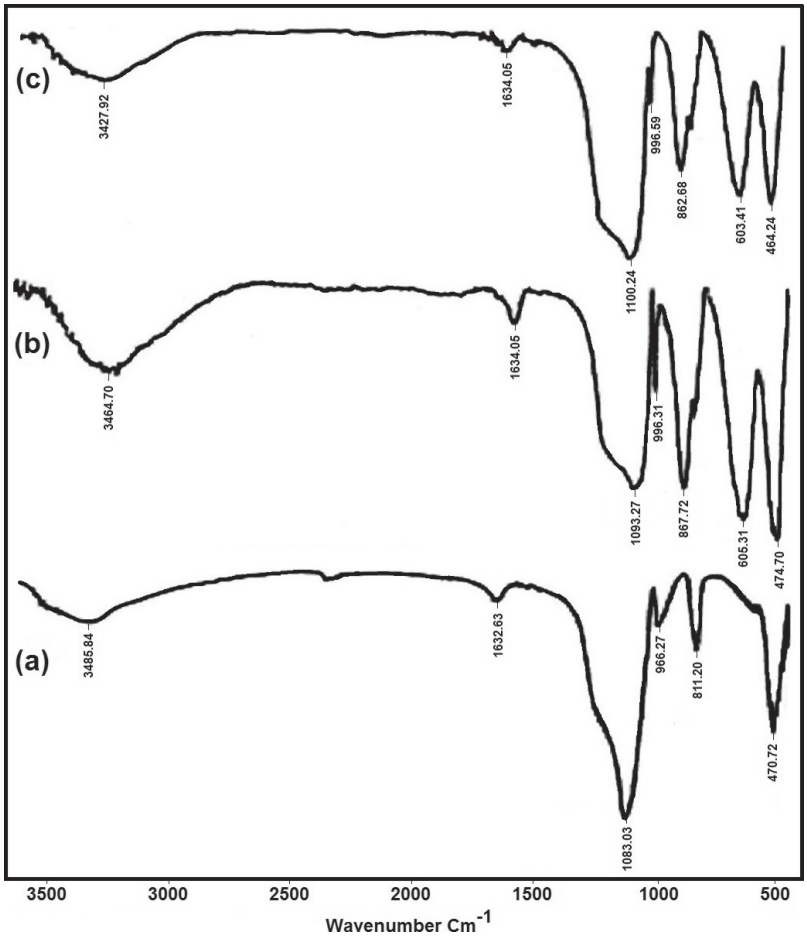



**Fig. 1.** FT-IR spectra of (**a**) $SiO_2$, (**b**) fresh $MoO_3/SiO_2$, and (**c**) recovered $MoO_3/SiO_2$.

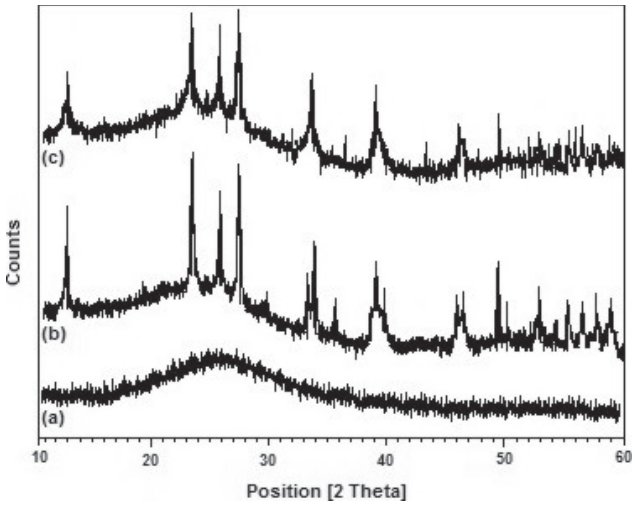



**Fig. 2.** X-ray diffraction pattern of a) $SiO_2$, b) Fresh $MoO_3/SiO_2$ (20 mol%) and c) Recovered $MoO_3/SiO_2$ (20 mol%) after reuse three times.

ages of fresh and recovered $MoO_3/SiO_2$ are shown in Fig. 3. These images demonstrate that the molybdenum oxide is well supported and distributed on silica. Comparison of these two images shows that $MoO_3$ was strongly adsorbed on the surface of $SiO_2$ and no leaching occurred during the reaction [34]. Moreover, the energy dispersive spectrum (EDS) (Fig. 4) reveals that the weight percentages of Mo, Si and O are 66.14%, 10.66% and 23.20%, respectively. The above results confirm the presence of molybdenum oxide in the catalyst structure.

*$MoO_3$-$SiO_2$ catalyzed synthesis of 1,8-dioxodecahydroacridines*

The one-pot synthesis of 1,8-dioxodecahydroacridines was achieved by the three-component condensation of aromatic aldehydes, dimedone, and ammonium acetate or aromatic amines in the presence of $MoO_3/SiO_2$ as a heterogeneous catalyst (Scheme 1). The $MoO_3/SiO_2$ was prepared according to the literature procedure [31]. To optimize the reaction conditions, the reaction of 4-chlorobenzaldehyde (1 mmol), dimedone (2

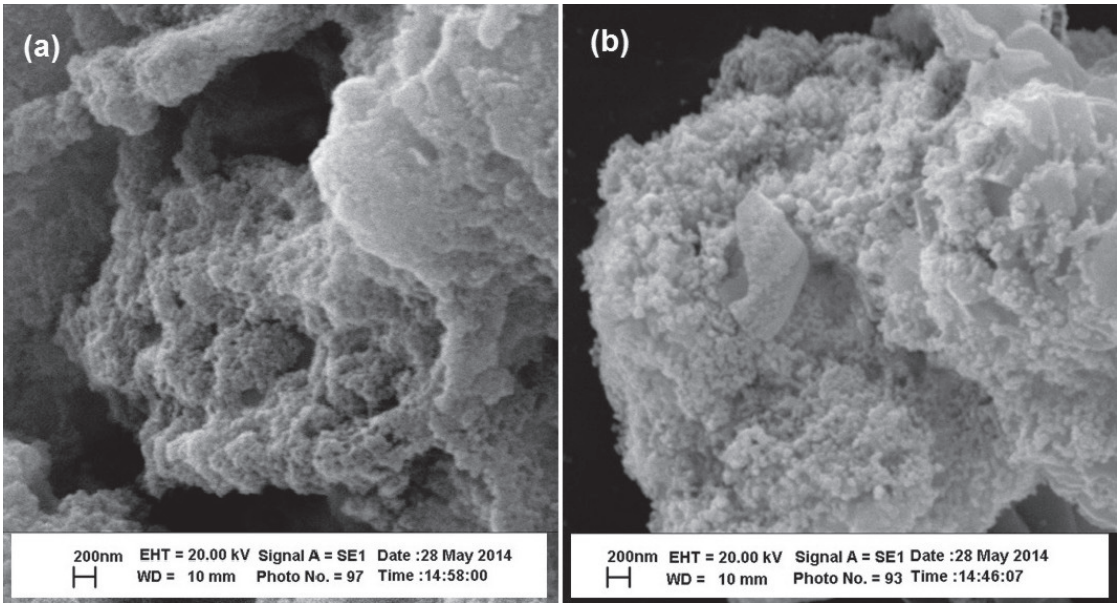



**Fig. 3.** SEM images of $MoO_3/SiO_2$.

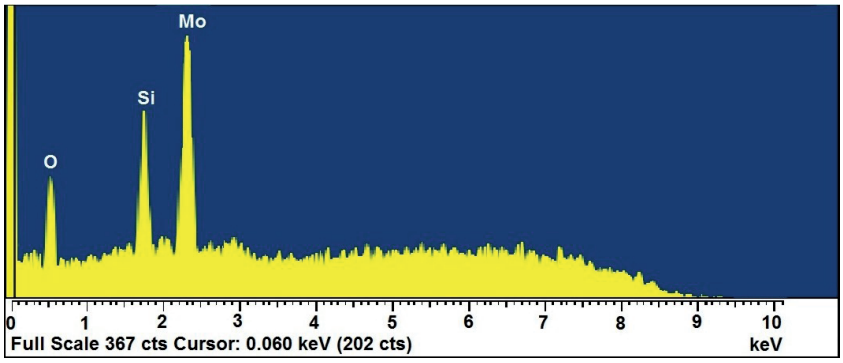



**Fig. 4.** EDS spectrum of $MoO_3/SiO_2$.

mmol) and ammonium acetate (1 mmol) in the presence of $MoO_3/SiO_2$ (20 mol%) was selected as a model.

The efficiency of the reaction is affected mainly by the amount of $MoO_3/SiO_2$ (Table 1). No product was obtained in the absence of the catalyst (entry 1), indicating that the catalyst is necessary for the reaction. Increasing the amount of the catalyst enhanced the yield of the products (entries 2, 3). The optimal amount of $MoO_3/SiO_2$ was 20 mol% (entry 4); increasing the amount of the catalyst beyond this value did not increase the yield noticeably (entries 5, 6). Furthermore, to optimize reaction conditions, the reaction was carried out in various solvents and under solvent-free conditions (entries 7-12). We observed that the reaction in polar solvents was done better than with non-polar solvents (entries 7-10), for example: the yield of the reaction in ethanol was 82% (entry 7) while, this amount for dichloromethane was trace (entry 10). However, in comparison of solvent and solvent-free conditions, the yields of the reaction under solvent-free conditions were greater, and the reaction times were shorter. Finally, the reaction was carried out at several temperatures under solvent-free conditions. Increasing the temperature did not improve the yield (entries 11, 12). The best result was obtained at 100 °C for 15 min; therefore, all reactions were carried out at 100 °C in the presence of 20 mol% $MoO_3/SiO_2$ under solvent-free conditions.

After optimization of the reaction conditions, to delineate this approach, particularly in regard to library construction, this methodology was evaluated with different aromatic aldehydes **1**, dimedone **2** and ammonium acetate or number of aromatic amines **3** (Table 2). A wide range of aromatic aldehydes bearing either electron-donating or electron-withdrawing substituents reacted successfully with dimedone and aromatic amines or ammonium acetate to give the corresponding 1,8-dioxodecahydroacridines products in high yields over short reaction times.

A probable mechanism for the condensation of an aromatic aldehyde, dimedone and ammonium acetate or aromatic amines in the presence of $MoO_3/SiO_2$ for the synthesis of 1,8-dioxodecahydroacridines is proposed as shown in (Scheme 2).

The recyclability of the catalyst in the reaction of 4-chlorobenzaldehyde (1 mmol), dimedone (2 mmol), and ammonium acetate (1 mmol) in the presence of $MoO_3/SiO_2$ (20 mol%) was also checked. After completion of the reaction, the reaction mixture was cooled to room temperature and hot ethanol was added. The solid residue was dissolved in hot ethanol and the mixture was stirred for 5 min. The catalyst was separated by simple filtration, dried at 100 °C under vacuum for 2 h, and reused for the similar reaction. As is shown in (Fig. 5), the catalyst could be reused at least three times without significant loss of activity.

## Conclusion

In summary, $MoO_3/SiO_2$ has been successfully applied as an efficient and green catalyst for the synthesis of 1,8-dioxodecahydroacridines at 100 °C and under solvent-free conditions. The catalyst could be recycled after a very simple work-up and reused at least for three runs without appreciable reduction of its catalytic activity. The mild reaction conditions, high yields, short reaction times, easy work-up, and absence of any volatile and hazardous organic solvents are some advantages of this protocol.

## Experimental

### Materials and techniques

All chemicals were available commercially and used without additional purification. The catalyst was synthesized according to the literature [31]. Melting points were recorded on an

**Table 1.** Effect of $MoO_3/SiO_2$ amount, solvent and temperature on the model reaction.[a]

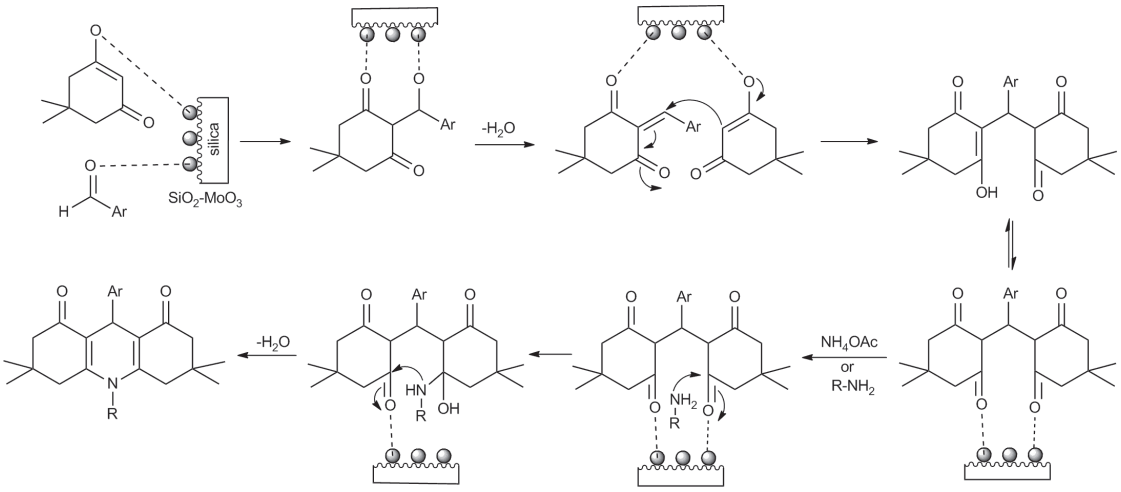
| Entry | Catalyst mol% | Time (min) | Conditions | Temperature (°C) | Yield (%)[b] |
|---|---|---|---|---|---|
| 1 | None | 200 | Solvent-free | 100 | None |
| 2 | 5 | 30 | Solvent-free | 100 | 41 |
| 3 | 10 | 15 | Solvent-free | 100 | 65 |
| 4 | 20 | 15 | Solvent-free | 100 | 93 |
| 5 | 30 | 15 | Solvent-free | 100 | 93 |
| 6 | 50 | 15 | Solvent-free | 100 | 91 |
| 7 | 20 | 150 | $C_2H_5OH$ | Reflux | 82 |
| 8 | 20 | 150 | $H_2O$ | Reflux | 74 |
| 9 | 20 | 150 | DMF | Reflux | 36 |
| 10 | 20 | 150 | $CH_2Cl_2$ | Reflux | Trace |
| 11 | 20 | 15 | Solvent-free | 110 | 92 |
| 12 | 20 | 15 | Solvent-free | 120 | 89 |

[a]4-chlorobenzaldehyde (1 mmol), dimedone (2 mmol), and ammonium acetate (1 mmol) at 100 °C.
[b]Isolated yields.

**Table 2.** Preparation of 1, 8-dioxodecahydroacridines using of $MoO_3/SiO_2$ (20 mol%) as a catalyst.[a]

| Entry | Ar | R | Products[b] | Time(min) | Yields(%)[c] | Mp °C | |
|---|---|---|---|---|---|---|---|
| | | | | | | Found | Reported |
| 1 | 4-$ClC_6H_5$ | $NH_4OAc$ | **4a** | 15 | 93 | 298-300 | 298-299 [14] |
| 2 | 4-$BrC_6H_4$ | $NH_4OAc$ | **4b** | 12 | 92 | 241-242 | 241-243 [35] |
| 3 | 2-$ClC_6H_4$ | $NH_4OAc$ | **4c** | 14 | 94 | 221-222 | 225-227 [14] |
| 4 | $C_6H_4$ | $NH_4OAc$ | **4d** | 10 | 88 | 192-194 | 192-195 [36] |
| 5 | 4-$MeC_6H_4$ | $NH_4OAc$ | **4e** | 20 | 90 | 270-272 | 279-281 [36] |
| 6 | 4-$MeOC_6H_4$ | $NH_4OAc$ | **4f** | 19 | 92 | 269-270 | 270-272 [36] |
| 7 | 3-$NO_2C_6H_4$ | $NH_4OAc$ | **4g** | 14 | 86 | 288-290 | 285-286 [35] |
| 8 | 3-$NO_2C_6H_4$ | $NH_4OAc$ | **4h** | 20 | 88 | 284-286 | 284-286 [36] |
| 9 | 4-$NO_2C_6H_4$ | $NH_4OAc$ | **4i** | 10 | 91 | 286-289 | 284-286 [14] |
| 10 | 4-$CNC_6H_4$ | $NH_4OAc$ | **4j** | 15 | 94 | 320-322 | 324-326 [35] |
| 11 | $C_6H_4$ | 4-$MeC_6H_4$ | **4k** | 20 | 88 | 255-257 | 260-262 [37] |
| 12 | $C_6H_4$ | 4-$MeOC_6H_4$ | **4l** | 18 | 89 | 212-215 | 215-217 [37] |
| 13 | 4-$ClC_6H_4$ | 4-$MeC_6H_4$ | **4m** | 20 | 91 | 270-272 | 269-271 [37] |
| 14 | 4-$MeC_6H_4$ | 4-$MeOC_6H_4$ | **4n** | 15 | 86 | 238-240 | 238-241 [37] |
| 15 | $C_6H_4$ | $C_6H_4$ | **4o** | 20 | 90 | 202-202 | 203-205 [37] |

[a]1 mmol aromatic aldehyde, 2 mmol dimedone, and 1 mmol aromatic amines or ammonium acetate at 100 °C under solvent-free conditions.
[b]All the products were characterized by IR spectral data and comparison of their melting points with those of authentic samples. Also, the structures of some products were confirmed by $^1H$ NMR spectral data.




**Scheme 2.** Plausible reaction mechanism for the formation of 1,8-dioxodecahydroacridines in the presence of $MoO_3/SiO_2$ catalyst.

electrothermal type 9100 melting point apparatus. The FT-IR spectra were obtained using a 4300 Shimadzu spectrophotometer and KBr disks. The $^1H$ NMR (500 MHz) spectra were recorded with a Bruker DRX500 spectrometer. Powder X-ray diffraction (XRD) of the catalyst was recorded with a Philips 1050 diffractometer using graphite monochromatized Cu-Kα radiation. Surface morphology and particle size were studied using a Hitachi S-4800 SEM-EDS instrument.

## Catalyst preparation

For preparation of molybdenum oxide supported on silica ($MoO_3/SiO_2$) with varying molar concentrations (5, 10, 20, 30, 50), the ammonium heptamolybdate and ethyl silicate-40 were used as molybdenum and silica sources, respectively. In a typical procedure, 20 mol% catalyst was synthesized by dissolving 14.11 g ammonium heptamolybdate in 40 mL water at

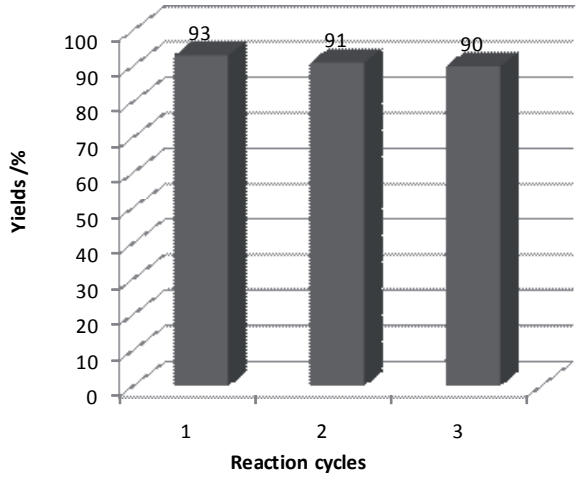



**Fig. 5.** Recycling experiment for $MoO_3/SiO_2$.

80 °C. This hot solution was added drop wise to the dry isopropyl alcohol solution of ethyl silicate-40 (48.0 g) with constant stirring. The obtained greenish gel was air dried and calcined at 500 °C in air in a muffle furnace for 12 h [31]. Similarly catalysts with 5, 10, 30, 50 mol% molybdenum oxide loadings were prepared.

### General procedure for synthesis of 1,8-dioxodecahydroacridines 4a-o

A mixture of aromatic aldehyde **1** (1 mmol), dimedone **2** (2 mmol), ammonium acetate **3** (1 mmol), and $MoO_3/SiO_2$ (20 mol%) was heated in the oil bath at 100 °C for 10-20 min. During the procedure, the reaction was monitored by TLC. Upon completion, the reaction mixture was cooled to room temperature and hot ethanol was added. The catalyst was insoluble in hot ethanol and it could be recycled by a simple filtration. The product was then collected from the filtrate after cooling to room temperature and recrystallized from ethanol to give compounds **4a-o** in high yields.

### Selected $^1$H NMR Data

*3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-chlorophenyl)-decahydroacridine.*
(**4a**): (500 MHz, $CDCl_3$) δ 1.00 (s, 6H, 2Me), 1.12 (s, 6H, 2Me), 2.19 (d, *J* = 16.3 Hz, 2H), 2.27 (d, *J* = 16.3 Hz, 2H), 2.29 (d, *J* = 16.7 Hz, 2H), 2.41 (d, *J* = 16.7 Hz, 2H), 5.07 (s, 1H, CH), 6.68 (brs, 1H, NH), 7.19 (d, *J* = 8.4 Hz, 2H, arom-H), 7.30 (d, *J* = 8.4 Hz, 2H, arom-H).

*3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-nitrophenyl)-decahydroacridine.*
(**4i**): (500 MHz, $CDCl_3$) δ 1.00 (s, 6H, 2Me), 1.14 (s, 6H, 2Me), 2.19 (d, *J* = 16.5 Hz, 2H), 2.28 (d, *J* = 16.5 Hz, 2H), 2.32 (d, *J* = 16.7 Hz, 2H), 2.46 (d, *J* = 16.7 Hz, 2H), 5.19 (s, 1H, CH), 6.12 (brs, 1H, NH), 7.54 (d, *J* = 8.7 Hz, 2H, arom-H), 8.10 (d, *J* = 8.7 Hz, 2H, arom-H).

*3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-chlorophenyl)-10-(4-methylphenyl) decahydroacridine.*
(**4m**): (500 MHz, $CDCl_3$) δ 0.83 (s, 6H, 2Me), 0.97 (s, 6H, 2Me), 1.86 (d, *J* = 17.4 Hz, 2H), 2.09 (d, *J* = 17.4 Hz, 2H), 2.15 (d, *J* = 16.5 Hz, 2H), 2.22 (d, *J* = 16.5 Hz, 2H), 2.52 (s, 3H, Me), 5.27 (s, 1H, CH), 7.11 (d, J = 8.3 Hz, 2H, arom-H), 7.24 (d, *J* = 8.3 Hz, 2H, arom-H), 7.37 (d, *J* = 8.7 Hz, 2H, arom-H), 7.39 (d, *J* = 8.7 Hz, 2H, arom-H).

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