



# Effectual and Rapid Synthesis of Hantzsch Derivatives in Solvent-Free Conditions Catalyzed by a Mesoporous Basic Silica-Based Nanomaterial

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## Abstract

In this investigation, a very fast and effective protocols for the synthesis polyhydroquinoline and polyhydroacridine derivatives have been developed using nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] as a mesoporous basic catalyst via the Hantzsch synthesis under solvent free conditions. In comparison to other catalysts employed for the synthesis of these derivatives, this catalyst benefits from a series of advantages such as being prepared using affordable and readily available raw materials, durability and high-performance, recoverability and reusability (7 times without considerable loss of activity), environmental friendliness and non-metallic nature. Other notable features of these protocols include: excellent yields, high atom economy, easy workup and moderate conditions.

**Keywords** Nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] · Mesoporous basic catalyst · Polyhydroquinolines · Polyhydroacridines · Solvent-free MCRs

## 1 Introduction

In the new era, an applied field that has generated much attention of many chemists and pharmacists is the

chemistry of nanoscale materials [1–3]. One of the significant branches of nanomaterials is nanocatalysts, that increasing of their use is due to their superior physical and chemical characteristics in comparison to bulk materials

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[1–3]. Nanocatalysts are the bridge connecting the attractive advantages of homogeneous and heterogeneous catalysts because, like homogeneous catalysts, they have a high reaction speed (by reason of the high surface-to-volume ratio), and also, like heterogeneous catalysts, they can be easily separated from the reaction mixture (due to insolubility in the reaction medium) [3–7]. These benefits, beside characteristics such as low density and toxicity, high permeability and mechanical stability, easy functionalization and biocompatibility, have created it additional motivation to employ these materials as catalysts [3–7].

In terms of combinatorial chemistry, the use of multi-component reactions (MCRs) is very important in order to produce industrial, pharmaceutical and chemical compounds [8, 9]. In MCRs, three or more starting materials react together in one pot to form a product in which all or most of the raw materials are involved [9, 10]. This feature is extremely important in terms of atom economy [10]. Ecologically-friendly approach through reduction on energy intake, the number of synthetic steps, generation of waste, reaction time and removing of separation of unstable intermediates are including the benefits of MCRs [9–11]. In recent decades, with expanding environmental challenges, the redesign of MCRs with green and environmentally friendly approaches have become convenient tools, particularly in the scopes of organic synthesis, drug discovery and materials science [12, 13]. In these areas, an effective step in achieving the goals of green chemistry is to perform MCRs in solvent-free conditions [12, 13]. Solvent-free environment, in addition to the reduction in reactor size and the elimination of environmental hazards of organic solvents, makes MCRs more efficient (especially in terms of reaction time and efficiency) [12–15]. Therefore, it is natural that solvent-free MCRs have covered large areas of chemical companies today, and the demand for them as cleaner chemical techniques is increasing [12–15].

Heterocyclic compounds with a nitrogen nucleus have been identified across a broad range of pharmacological and bio-active compounds [16, 17]. Among this category of heterocycles, derivatives of polyhydroquinoline and polyhydroacridine are valuable compounds due to their

diverse biological activities [17]. Current research shows that these compounds can act as neuroprotectors and chemo sensitizer platelet in cancer control [17, 18]. In addition, these derivatives have the general characteristics of miscellaneous bioactive compounds such as anti-tumour, antiathero-sclerotic, geroprotective, vasodilator, hep-atoprotective activity and bronchodilator [17]. With regards to broad application of polyhydroquinoline and polyhydroacridine derivatives, various protocols had been informed for the production of these derivatives such as usual heating [19], ultrasound and microwave irradiation [20–23] and visible-light irradiation procedure [24]. Furthermore, various catalysts, e.g. Gallium oxide nano-material [25], urease [26], Fe<sub>3</sub>O<sub>4</sub>@Schiff-base-Cu [27], Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@PTS-THEIC-(CH<sub>2</sub>)<sub>3</sub>OB(OH)<sub>2</sub> [28], [βCD/Im](OTs)<sub>2</sub> [29], Zn-MOF microspheres [30], Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-PEG/NH<sub>2</sub> [31], SBA-15@AMPD-Co [32], β-CD-mono-SO<sub>3</sub>H [33], Cell-Pr-NHSO<sub>3</sub>H [34], MCM-41-SO<sub>3</sub>H [35] were also used for such transformation.

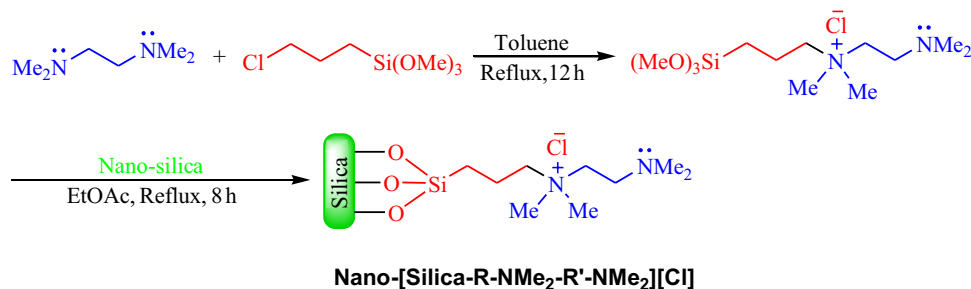
Even though most of the mentioned reports provide obvious benefits, the employ of large amounts of catalysts [23], costly metal precursors [25, 27], high temperatures [31, 35], low yields [24, 30, 35], tedious work-up procedures and using of toxic organic solvents restrict the use of these protocols [22, 30, 33]. Therefore, there is considerable scope for finding alternative and potential experimental methods in this field, particularly that are in great demand and having benefits such as non-toxic, lower cost and environmentally friendly.

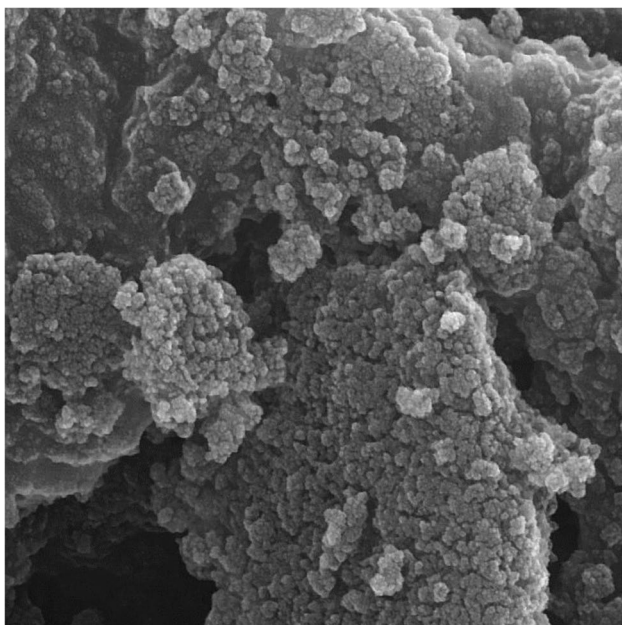
According to the mentioned challenges and in order to solve them, herein, we describe application of a mesoporous basic nanocatalyst (nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]) as a highly efficient catalyst for the rapid production of polyhydroquinolines and polyhydroacridines through the Hantzsch synthesis in the absence of solvent.

## 2 Results and Discussion

Nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] was synthesized based on Scheme 1 [36]. The images of FE-SEM, HR-TEM and EDS analyzes of the catalyst are demonstrated in Figs. 1, 2 and 3, respectively.

**Scheme 1** The preparation of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

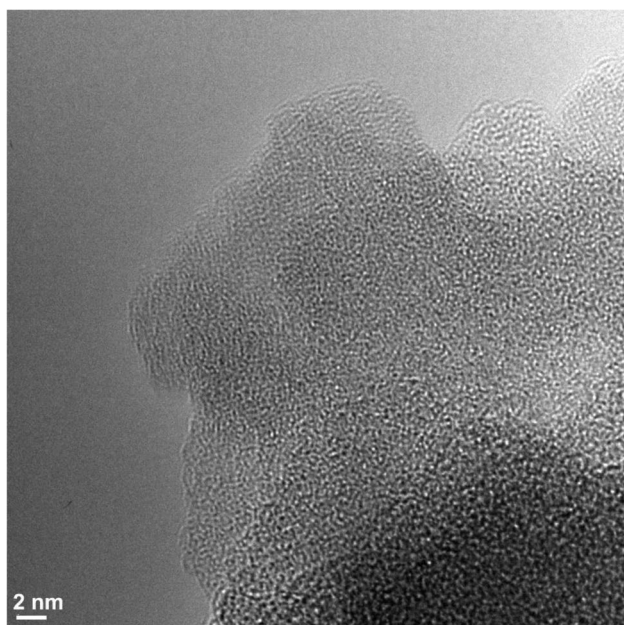




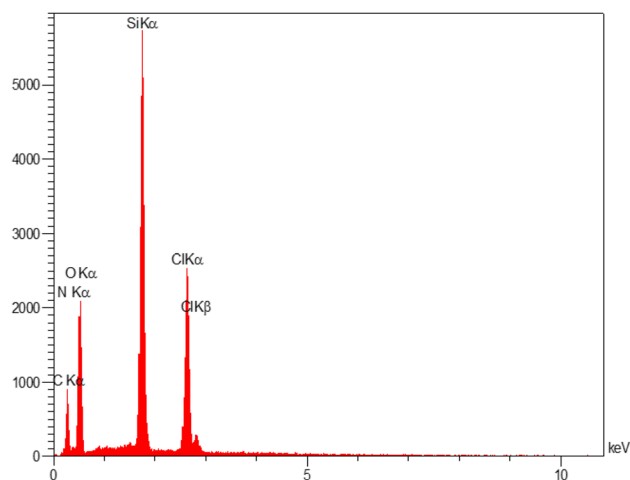
**Fig. 1** The FE-SEM image of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

## 2.1 The Synthesis of Polyhydroquinolines by nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

To determine the most ideal conditions for the synthesis of polyhydroquinoline derivatives using nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl], initially, the reaction of 4-chlorobenzaldehyde (1 mmol) with dimedone



**Fig. 2** The HR-TEM image of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

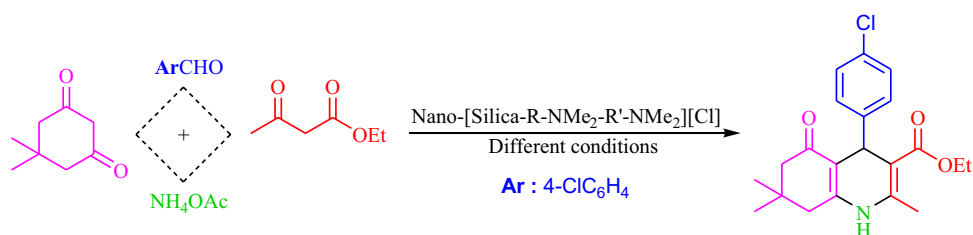


**Fig. 3** The EDS spectrum of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

(1 mmol), NH<sub>4</sub>OAc (1.4 mmol) and EtOAc (1 mmol) were considered as a model reaction (Scheme 2), then the effect of temperature and catalyst amount on the model reaction in solvent-free conditions was investigated; the corresponding results are briefed in Table 1. As the data in this Table demonstrate, the best results were acquired once 10 mg of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] was employed at 60 °C (Table 1, entry 4). In another study, the progress of the model reaction was checked in the presence of the starting materials for the prepare of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] as well as bases such as DBU and K<sub>2</sub>CO<sub>3</sub> (Table 5, entries 7–10). The results of Table 5 clearly show that these catalysts were not effectual for the synthesis of polyhydroquinoline, and our idea for the synthesis of these derivatives with nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] catalyst is reasonable.

In continue, the production of various derivatives of polyhydroquinolines was carried out using various arylaldehydes, dimedone, NH<sub>4</sub>OAc and EtOAc under the optimized conditions, for the purpose of recognize generality and efficiency of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]; the relevant results are demonstrated in Table 2. The data from this Table showed that all substituents on the arylaldehydes comprising bromo, chloro, methyl, methoxy, and nitro on para, meta and ortho positions, provided the corresponding products with excellent yields in short times.

Based on previous reports [25], a reasonable mechanism for the synthesis of polyhydroquinolines using nano[silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] is proposed in Scheme 3. Initially, the  $\alpha$ -proton of dimedone is abstracts with the help of the basic group

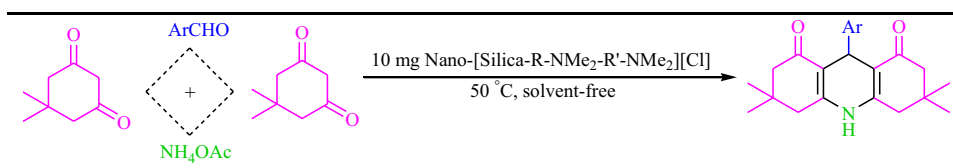
**Scheme 2** The model reaction**Table 1** Optimization of temperature and amount of catalyst in the synthesis of polyhydroquinolines

Entry	Catalyst	Temp (°C)	Catalyst amount (mg)	Time (min)	Yield <sup>a</sup> (%)
1	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	70	5	10	82
2	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	70	10	10	98
3	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	70	20	10	98
4	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	60	10	10	98
5	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	50	10	10	70
6	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	120	-	80	42
7	1,2-diamine	60	10	60	50
8	Nano-silica	60	10	60	45
9	K <sub>2</sub> CO <sub>3</sub> (as a base)	60	10	60	trace
10	DBU (as a base)	60	10	60	45

<sup>a</sup>Yield of isolated product

of the catalyst (NMe<sub>2</sub>) and produces carbanion (**I**); then, Knoevenagel condensation between aldehyde and carbanion (**I**) occurs, which leads to the formation of intermediate **II**. In the other hand, the reaction of ammonia (generated from NH<sub>4</sub>OAc) with EtOAc

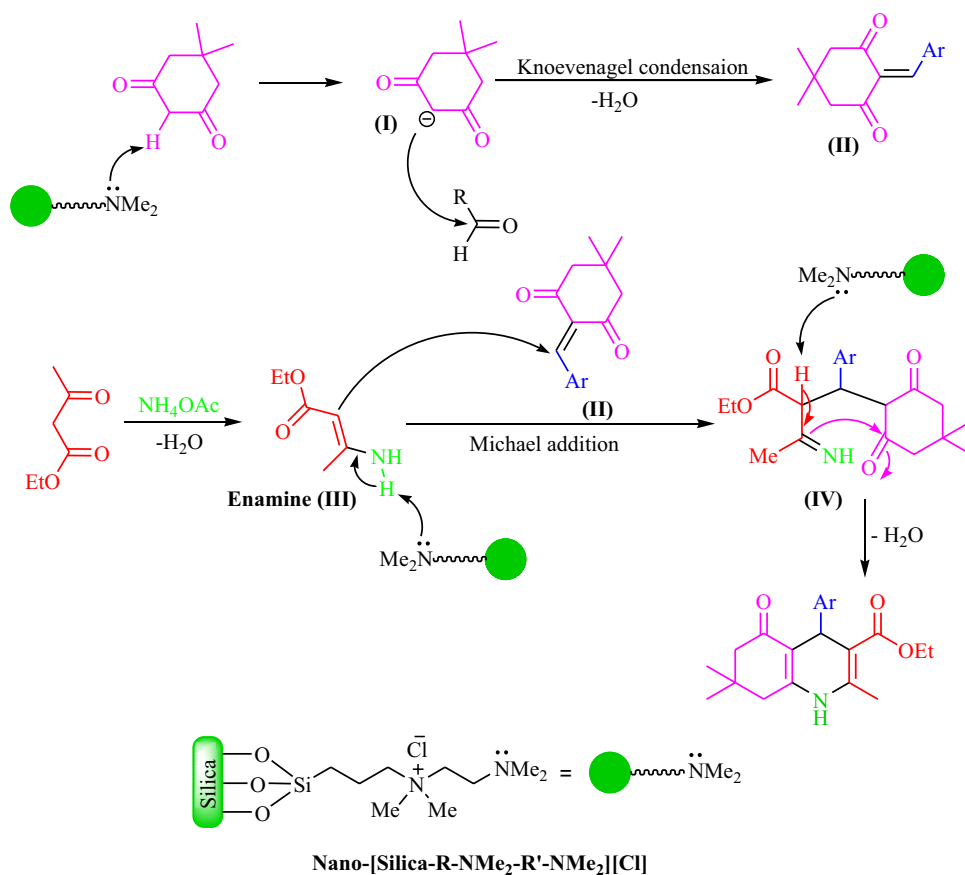
gives intermediate **III** after removal of H<sub>2</sub>O molecule. Michael-type addition of **II** with **III** affords intermediate **IV**. Eventually, the nanocatalyst assists cyclization of **IV**, and then elimination of water molecule to provide the product.

**Table 2** The synthesis of polyhydroquinolines using nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

Compd. no	Ar	Time (min)	Yield <sup>a</sup> (%)	M.p. (°C)	
				Found	Reported
<b>1a</b>	C <sub>6</sub> H <sub>5</sub>	9	98	222–224	223–225 [25]
<b>2a</b>	4-BrC <sub>6</sub> H <sub>4</sub>	14	98	248–250	248–250 [25]
<b>3a</b>	2-ClC <sub>6</sub> H <sub>4</sub>	10	94	178–180	179–181 [25]
<b>4a</b>	4-ClC <sub>6</sub> H <sub>4</sub>	10	98	238–240	239–241 [25]
<b>5a</b>	4-MeC <sub>6</sub> H <sub>4</sub>	13	93	259–261	260–262 [25]
<b>6a</b>	4-MeOC <sub>6</sub> H <sub>4</sub>	15	91	248–250	247–250 [30]
<b>7a</b>	2-O <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	6	98	201–203	200–202 [25]
<b>8a</b>	3-O <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	7	96	179–181	180–182 [25]
<b>9a</b>	4-O <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	5	95	235–237	236–238 [25]

<sup>a</sup>Isolated yield

**Scheme 3** The suggested mechanism for the synthesis of polyhydroquinolines



## 2.2 The Production of Polyhydroacridines with nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

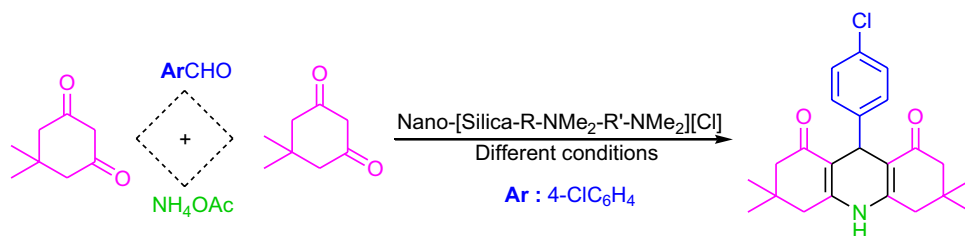
Afterwards the success in use of nano[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] as catalyst for the production of polyhydroquinolines, we determined to evaluate its catalytic performance to achieve polyhydroacridine derivatives in solvent-free conditions. In order to find the best conditions to achieve this goal, we chose the reaction shown in Scheme 4 as a model reaction and then we explored the effect of temperature and catalyst amount on this reaction (Table 3, entries 1–10). The optimum state were achieved utilizing 10 mg of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] at 50 °C in the absence of solvent (Table 3, entry 4).

Afterwards optimisation, we investigated different electron-acceptor and electron-donor benzaldehydes in Hantzsch synthesis catalyzed by nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] for production of polyhydroacridines for designation the performance and generality of the catalytic system (Table 4). It is obvious from Table 4, both electron-accepting and electron-donating benzaldehydes produced the polyhydroacridine derivatives with high yields in short times.

## 2.3 Recyclability and Durability of the Catalyst

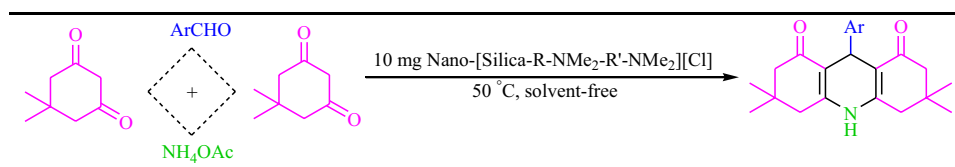
Industrially, the recoverability and reusability heterogeneous catalysts is an important feature. Therefore,

**Scheme 4** The model reaction



**Table 3** Optimisation of temperature and amount of catalyst in the synthesis of polyhydroacridines

Entry	Catalyst	Temp (°C)	Catalyst amount (mg)	Time (min)	Yield <sup>a</sup> (%)
1	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	60	5	8	84
2	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	60	10	8	98
3	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	60	20	8	98
4	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	50	10	8	98
5	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	40	10	8	74
6	Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl]	120	-	100	48
7	1,2-diamine	50	10	60	45
8	Nano-silica	50	10	60	50
9	K <sub>2</sub> CO <sub>3</sub> (as a base)	50	10	60	trace
10	DBU (as a base)	50	10	60	40

<sup>a</sup>Yield of isolated product**Table 4** The synthesis of polyhydroacridines by nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

Compd. no	Ar	Time (min)	Yield <sup>a</sup> (%)	M.p. (°C)	
				Found	Reported
<b>1b</b>	C <sub>6</sub> H <sub>5</sub>	8	98	243-245	244-246 [28]
<b>2b</b>	4-BrC <sub>6</sub> H <sub>4</sub>	10	98	330-332	330-332 [35]
<b>3b</b>	2-ClC <sub>6</sub> H <sub>4</sub>	9	93	308-310	309-310 [35]
<b>4b</b>	4-ClC <sub>6</sub> H <sub>4</sub>	8	98	310-312	311-313 [28]
<b>5b</b>	4-MeC <sub>6</sub> H <sub>4</sub>	8	94	215-217	216-218 [31]
<b>6b</b>	4-MeOC <sub>6</sub> H <sub>4</sub>	10	93	245-247	246-248 [31]
<b>7b</b>	2-O <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	5	98	281-283	280-282 [28]
<b>8b</b>	3-O <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	5	98	267-269	268-270 [28]
<b>9b</b>	4-O <sub>2</sub> NC <sub>6</sub> H <sub>4</sub>	5	98	197-200	197-200 [28]

<sup>a</sup>Isolated yield

recovery capability and reproducibility of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] were explored for the producing of compound **1a** and **1b** from the derivative series polyhydroquinolines and polyhydroacridines, respectively. The results showed that in both synthesis protocols, the recycling process was repeated for seven times without significant change in activity of the catalyst (Figs. 4 and 5). The catalyst recycling procedure is explained in the experimental section.

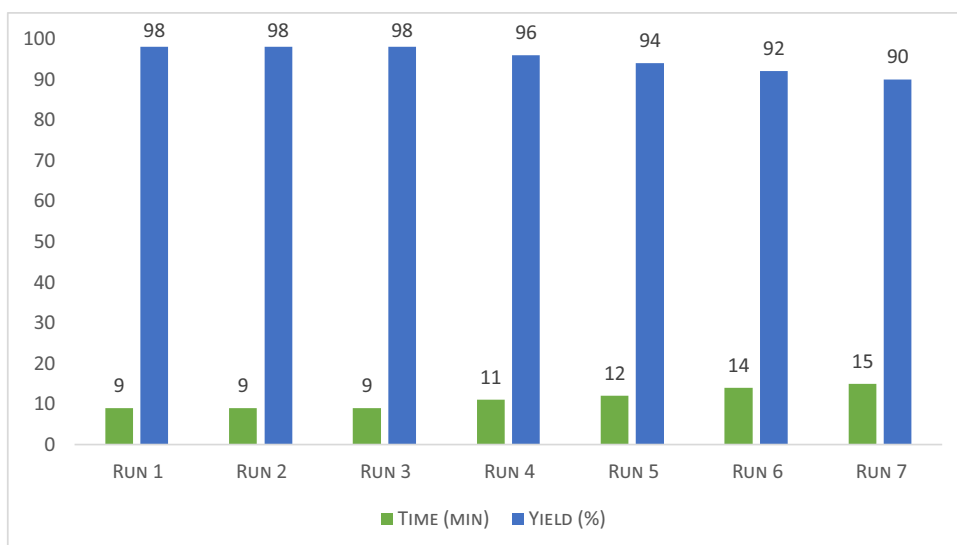
In another study, in order to check the durability and efficiency of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl], we synthesized all the derivatives in Tables 2 and 4 under optimal conditions with a sample of our catalyst that was stored for

two years in a closed container at room temperature. Obtaining results identical to the data in these tables indicate the durability and high efficiency of the catalyst despite long-term storage under simple conditions.

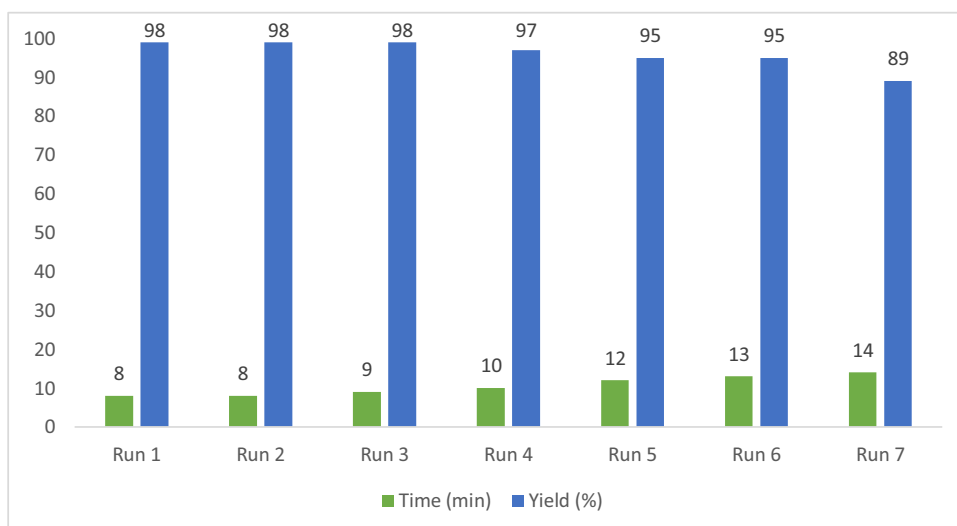
## 2.4 Comparison

For the purpose of illustrate the superiority of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] to other reported catalysts available in previous literature for the synthesis of polyhydroquinolines (A) and polyhydroacridines (B), we have presented the reaction outcomes for the production of compound 1a and 1b in Table 5. This Table

**Fig. 4** Recyclability of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] in the synthesis of compound (1a)



**Fig. 5** Recyclability of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] in the synthesis of compound (1b)



shows that nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] was better than the other catalysts in terms of the reaction temperature, yield, time and no solvent use; furthermore, in our procedures, visible-light, microwave and ultrasound irradiations (which require special devices) have not used.

### 3 Experimental Section

#### 3.1 Materials and Methods

High-purity starting materials and solvents were bought from Fluka or Merck Chemical Companies. The known products were detected by comparison their melting points/spectral data with those recorded in the earlier articles. The

melting points were recorded on a Büchi B-545 apparatus in open capillary tubes. Monitoring progress of the reactions was accomplished by TLC using silica gel SIL G/UV 254 plates. <sup>1</sup>H NMR (500 MHz) and <sup>13</sup>C NMR (125 MHz) were recorded on Bruker Avance DPX, FT-NMR spectrometers (δ in ppm). The particle morphology was examined by FE-SEM (MIRA3TESCAN-XMU) and HR-TEM (FEI Titan 80–300 kV). Energy dispersive X-ray spectroscopy (EDS) was taken on a SAMX-EDS apparatus.

#### 3.2 General Protocol for the Synthesis of Polyhydroquinolines using nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

A mixture of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] (0.01 g), dimedone (0.140 g, 1 mmol), ethyl

**Table 5** Comparison of the reaction results of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] with other catalysts available for the production of polyhydroquinolines and polyhydroacridines

Catalyst	Conditions	Type of reaction	Time (min)	Yield (%)	Ref
Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl] (0.01 g)	Solvent-free, 60 °C	A	9	98	This work
Nano-[Silica-R-NMe <sub>2</sub> -R'-NMe <sub>2</sub> ][Cl] (0.01 g)	Solvent-free, 50 °C	B	8	98	This work
catalyst-free	Ultrasonic irradiation	B	20	90	[21]
Ni-Nanoparticles (10 mol%)	Microwave irradiation	A	1	95	[22]
Silica (2 g)	Microwave irradiation	B	5	90	[23]
catalyst-free *	visible-light irradiation	A	155–160	80–91	[24]
Nano-Ga <sub>2</sub> O <sub>3</sub> (0.06 g)	EtOH, Reflux	A	60	90	[25]
urease (0.01 g)	H <sub>2</sub> O, 65 °C	B	210	87	[26]
Fe <sub>3</sub> O <sub>4</sub> @Schiff-base-Cu (0.25 mol%)	H <sub>2</sub> O, Reflux	A	20	99	[27]
Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -R-OB(OH) <sub>2</sub> (0.01 g)	EtOH, Reflux	B	100	85	[28]
Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -R-OB(OH) <sub>2</sub> (0.01 g)	EtOH, Reflux	A	45	96	[28]
[βCD/Im](OTs) <sub>2</sub> (1 mmol)	Solvent-free, 90 °C	B	10	95	[29]
Zn-MOF microspheres (0.008 g)	PEG-400, 80 °C	A	150	83	[30]
Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -PEG/NH <sub>2</sub> (0.005 g)	Solvent-free, 120 °C	B	10	91	[31]
SBA-15@AMPD-Co (0.008 g)	Solvent-free, 100 °C	A	65	96	[32]
β-CD-mono-SO <sub>3</sub> H (0.03 g)	H <sub>2</sub> O, 50 °C	B	120	91	[33]
Cell-Pr-NHSO <sub>3</sub> H (0.05 g)	EtOH, reflux	A	50	90	[34]
MCM-41-SO <sub>3</sub> H* (0.005 g)	Solvent-free, 110 °C	B	10–110	60–98	[35]

\* In these articles, the desired product (1a or 1b) was not synthesized, therefore, we have reported times and yields as ranges

acetoacetate (0.130 g, 1 mmol), ammonium acetate (0.108 g, 1.4 mmol) and aromatic aldehydes (1 mmol) was stirred by a glass agitator at 60 °C. After that the reaction was completed (as observed by TLC), the reaction mixture was diluted with 10 mL of hot EtOH. Then, with the help of centrifugation and decantation, the catalyst insoluble in this solvent was separated from the reaction mixture soluble in it. The separated catalyst was washed by EtOH (3 × 3 mL), dried (under vacuum at 80 °C), and used for the next run. Finally, the solvent acquired from the centrifugation and decanting was evaporated, and the resulting precipitate was recrystallized from EtOH (95%) to give the pure polyhydroquinoline.

### 3.3 General Process for the Synthesis of Polyhydroacridines using nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]

Dimedone (0.280 g, 2 mmol), ammonium acetate (0.108 g, 1.4 mmol), aromatic aldehydes (1 mmol) and nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] (0.01 g) were mixed, and stirred strongly with a glass rod at 50 °C. After ensuring the completion of the reaction (confirmed by TLC) and separating the catalyst from the reaction

mixture (like the above method for the preparation polyhydroquinolines), the pure product was obtained by recrystallization of the resulting precipitate from ethanol (95%).

## 4 Conclusions

Concisely, we have employed the efficient, recoverable and mesoporous basic catalyst (nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl]) for the rapid synthesis of Hantzsch derivatives (polyhydroquinolines and polyhydroacridines) under solvent-free conditions. The recovery and reusability of the catalyst for seven runs did not show a significant decrease in product yields, in addition, the catalyst had a very high durability while maintaining the catalytic performance. The utilization of nano-[Silica-R-NMe<sub>2</sub>-R'-NMe<sub>2</sub>][Cl] permitted for the reactions to have excellent yields of the biologically efficient products in short times by easy purification process in mild, solvent-free and green conditions.

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**Authors' Contributions** Abeer Abdullah Al Anazi: synthesis of all polyhydroquinoline derivatives (1a–9a).

Rukhsana Satar: synthesis of some polyhydroacridine derivatives (1b–5b) and helped to edit the manuscript.

Hijran Sanaan Jabbar: synthesis of some polyhydroacridine derivatives (6b–9b) and helped to edit the manuscript.

I.B. Sapaev: optimizing the synthesis conditions of polyhydroquinoline derivatives.

Farag M. A. Altalbawy: writing and reviewing the manuscript.

Ameer A. Alameri: optimizing the synthesis conditions of polyhydroacridine derivatives.

Rasha Fadhel Obaid: identifying the structure of the catalyst by means of FE-SEM, HR-TEM and EDS analyzes.

Andrés Alexis Ramírez-Coronel: identifying the structure of some polyhydroquinoline derivatives by  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR (4a and 6a) and analyzing their resulting data.

RAED H. C. ALFILH: identifying the structure of some polyhydroacridine derivatives by  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR (1b and 6b) and analyzing their resulting data.

Yasser Fakri Mustafa: Assisting in writing and editing the manuscript.

Majid Heidarpour: synthesized the catalyst and helped to edit the manuscript.

**Data Availability** We have confirmed.

## Declarations

**Ethics Approval and Consent to Participate** Author's state that the manuscript is not be submitted simultaneously to another journal. The submitted work is original and has not been published elsewhere in any form or language, and the authors have no conflict of interest regarding this manuscript. The authors agree to participate in submitting our manuscript to this journal, and agree to the publication of our research data in this journal.

**Consent for Publication** We have confirmed.

**Competing interests** We declare that we have no conflict of interest.

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