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Synthesis and Characterization of Bifunctional Basic Mesoporous Organosilica Catalyst as an Efficient and Ecofriendly Nanocomposite in Biginelli Condensation Reaction

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Abstract

An organic–inorganic hybrid nanocomposite was prepared by immobilizing *β*-cyclodextrin (*β*-CD) and amino groups onto mesoporous MCM-41 via surfactant-templated sol-gel procedure and post-modification method. The heterogeneous hybrid nanocomposite, MCM-41-*β*-CD/NH_2, was characterized by SEM, TEM, XRD, TGA, BET and FT-IR. The potential application of this covalently linked basic catalyst was also investigated as an efficient, heterogeneous and recyclable stationary micro-vessel and basic heterogeneous catalyst in Biginelli multicomponent condensation reaction. High yield, high efficiency and reusability are among the advantages of this environmentally friendly method. The nano catalyst can be easily separated from the reaction mixture and reused after washing for several runs without less in activity.

Keyword: MCM-41-*β*-CD/NH_2, heterogeneous catalyst, *β*-cyclodextrin (*β*-CD), Biginelli reaction.

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Introduction

The headmost name given to a series of mesoporous material is the Mobil Composition of Matter (MCM) [1]. Among this class of materials, MCM-41 has attracted a great deal of attention due to its interesting advantages such as tunable pore size, controlled size and morphology, and dual-functional surface (external and internal) [2]. MCM-41 consists of hexagonal channels with the surface area around (~1000 m^2/g) and has high thermal stability [3-5]. Thus, this nano material has the potential applications in many fields, such as decomposition and absorption, photocatalyst, sensors, nano electronics, encapsulation of enzymes and medicinal properties [6-9]. Furthermore, the organic–inorganic hybrid nanocomposite of MCM-41 was used for investigation in the three-component synthesis of 3,4-dihydropyrimidin-2-(1*H*)-one derivatives from aromatic aldehydes, ethyl acetoacetate and urea. [10]

Heterocyclic frameworks have been found in various biologically active natural products, agrochemicals and pharmacological relevance molecules [11]. Among these heterocyclic compounds containing nitrogen atom such as dihydropyrimidinones are of special interest as medically potent lead molecules and a key intermediate for the synthesis of various biologically active compounds [12]. Preparation of these non-planar compounds [13-15] is now recognized as a powerful heterocyclic synthesis with many essential applications and it has been the subject of several reviews [16-19]. Recently, Farhadi et al. reported the synthesis of some 4-Aryl-1, 3, 4, 6 ,7, 8-hexahydroquinazolin-2,5 (1*H*,6*H*)-diones (HHQs) derivatives using K_3AlF_6 and its nano form. [20, 21]. βCD as a one of the phase-transfer catalyst is known as remarkable natural maccrocycle host, having a hydrophpbic cavity which forms inclusion complexes with a large variety of guest molecules [22-24]. However, this report describes a one-pot multicomponent process for the synthesis of various 4-Aryl-1, 3, 4, 6, 7, 8-hexahydroquinazolin-2,5 (1*H*,6*H*)-diones (HHQs) derivatives using new nano dual organo-modification MCM-41 as the catalyst.

Experimental

General

All chemical materials were purchased from Aldrich and Merck Chemical companies. Tetraethyl orthosilicate, (TEOS (98%, Aldrich)) was selected as a source of silica and cetyltrimethyl-ammonium bromide, (CTAB (98%, Aldrich)) was used as the structure directing agent. Deionized water was obtained from a system of two ionic interchange columns, cole–Parmer instruments. Melting points were determined on an electrothermal SI550 apparatus. FT-IR spectra were recorded from KBr discs on a Perkinelemer BX_ll. ^{1}H NMR and ^{13}C NMR spectra were recorded using a Bruker Avance 500 MHz instrument in DMSO-*d6*. Mass spectra were obtained on Platform II

spectrometer from Micromass. EI mode at 70 eV. UV/vis spectra (in EtOH) were taken with a CINTRAL 101 spectrophotometer. The support surface area and pore size distribution were measured by the nitrogen adsorption–desorption method (ASAP 2000, Micromeritics). Transmission electron microscope (TEM) images were obtained using Zeiss – EM10C –80 kV instrument.

Synthesis of aminopropyl and β-cyclodextrin grafted mesoporous MCM-41

Initially, 0.5g CTAB was added to 96 mL of deionized H_2O and stirred for2h. After the solution turned clear, 34 mL of ethanol was added to the solution. Then 10 mL of aqueous ammonia solution was added to the mixture and allowed to be mixed for 5 min. Next, 2.0 mL of TEOS was poured into the solution immediately under stirring for 3h at room temperature. The solid product was recovered by filtration and dried at room temperature overnight. CTAB was removed from the mesoporous MCM-41 by calcinating the sample at 540 ˚C for 9h. Next, the obtained mesoporous MCM-41 (1g) was dispersed in dry DMF (30 mL) by sonication. Then solution of hexamethylene disocyanate (HMDI) (3 mL) in 5 mL of dry DMF was added dropwise to the mixture. After mechanically agitation for 3h, the suspended substance was separated with filtration for removing the unreacted HDMI. In the next step, the product was dissolved in 15 mL of DMF solvent, and then 15 mL of DMF solvent containing 2 mmol *β*-CD was added dropwise to the solution. To synthesize β-cyclodextrin and grafted mesoporous MCM-41, the reaction mixture was stirred at 70 °C for 3 h. MCM-41-*β*-CD was washed with water and acetone several times and dried in vacuum for 24 h. Finally, to generate the MCM-41-*β*-CD/NH_2 compound, 2 g of triethoxypropyl silyl amine was added to the suspension of MCM-41-*β*-CD in 80 mL of toluene under the reflux condition for 24h. The mesoporous MCM-41-*β*-CD/NH_2 was then filtrated and washed with water and acetone several times and dried under the vacuum condition.

General procedure for the synthesis of 4-Aryl-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (HHQs) derivatives catalyzed by MCM-41-β-CD/NH_2

A mixture of the aldehyde (10 mmol), 1,3-cyclohexadione (10 mmol), urea (12 mmol) in acetonitrile (10 mL) and MCM-41-*β*-CD.NH_2 (0.15 g) was stirred under reflux condition for the appropriate time (30-90 min). The progress of the reaction was followed by TLC using n-hexane/ethyl acetate (2:1) as eluents until the total disappearance of the 1,3-cyclohexadione. Afterward, the catalyst was filtered out and the crude product was washed with water and recrystallized in ethanol. All these products were characterized by UV/Vis, FT-IR, ^{1}H-NMR, ^{13}C-NMR and MS Spectra.

Spectroscopic data

4-phenyl-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**a**)

White powder, m.p. 228-230 °C; 86% yield; -FT-IR (KBr): 3380.25, 2920.93, 1725.05, 1710.01, 1610.17 cm^{-1}-UV/Vis (EtOH): λ_{max} (logε) = 265.66 nm (5.50).

4-(4-methylphenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**b**)

White powder, m.p. 192-193 °C; 37% yield; -FT-IR (KBr): 3336.85, 2941.02, 1722.08, 1602.37 cm^{-1}. -^{1}H NMR (500 MHz, DMSO-d_6): δ= 1.90 (m, *J*=7Hz, 2H, H-8), 2.01 (m, 2H, H-7), 2.19 (m, 2H, H-6), 2.36 (m, 3H, CH_3), 2.94 (d, *J*=10.7 Hz, 1H, H-4), 3.90 (d , *J*=9.6 Hz, 1H, NH), 6.83 (s,1H, NH), 6.94 (m, 2H, Ar-H), 7.08 ppm (m, 2H, Ar-H). -^{13}C NMR (125 MHz, DMSO-d_6): δ= 20.20, 21.02, 29.05, 32.38, 33.50, 35.41, 37.24, 60.54, 100.45, 101.41, 116.39, 128.08, 128.35, 128.72, 134.02, 134.41, 141.68, 142.60, 195.83, 205.25 ppm. -MS (EI, 70 eV): *m/z* (%): 255.1 ($M^{+\cdot}$, $C_{15}H_{15}N_2O_2$), 253.2 ($M^{+\cdot}$ – 2H), 240.1 ($M^{+\cdot}$ – $C_{15}H_{14}NO_2$), 227.2($M^{+\cdot}$ – $C_{15}H_{15}O_2$), 164.1 ($M^{+\cdot}$ – C_7H_7), 148.1 ($M^{+\cdot}$ – C_7H_7-CH-NH-CO-NH), 131.1 ($M^{+\cdot}$ – C_7H_7-CH-CH=CH_2), 119.1 ($M^{+\cdot}$ – C_7H_7-CH-NH), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2). -UV/Vis (EtOH): λ_{max} ($\log_\varepsilon$) = 257.98 nm (5.49).

4-(3-methylphenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**c**)

Pale yellow powder, m.p. 211-213 °C; 44% yield; -FT-IR (KBr): 3359.99, 2975.01, 1720.79, 1609.14 cm^{-1}. -^{1}H NMR(500MHz, DMSO-d_6): δ=1.89 (m, 2H, H-8), 2.10 (m, 2H, H-7), 2.16 (m, 2H, H-6), 2.38 (m, 3H, CH_3), 2.99 (d, *J*=10.65 Hz, 1H, H-4), 3.90 (d, *J*=9.6 Hz, 1H, NH), 6.78 (s, 1H, NH), 6.84 (m, 1H , Ar-H), 6.91 (m, 1H, Ar-H), 7.00 (m, 1H, Ar-H), 7.04 (m, 1H, Ar-H). -^{13}C NMR (125 MHz, DMSO-d_6): δ=20.40, 21.63, 28.95, 29.09, 32.68, 33.44, 35.39, 37.24, 100.44, 101.39, 111.57, 116.38, 125.43, 126.02, 127.58, 128.79, 129.43, 136.45, 144.74, 145.63, 167.72, 169.47, 189.82, 195.86, 196.27, 205.22, 206.67 ppm. -MS (EI, 70 eV): *m/z* (%): 255.2 ($M^{+\cdot}$; $C_{15}H_{15}N_2O_2$), 253.2 ($M^{+\cdot}$ – 2H.), 240.1 ($M^{+\cdot}$ – $C_{15}H_{14}NO_2$), 227.2 ($M^{+\cdot}$ – $C_{15}H_{15}O_2$), 164.1 ($M^{+\cdot}$ – C_7H_7), 148.1 ($M^{+\cdot}$ – C_7H_7-CH-NH-CO-NH), 131.1 ($M^{+\cdot}$ – C_7H_7-CH-CH=CH_2), 119.1 ($M^{+\cdot}$ – C_7H_7-CH-NH), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2). -UV/Vis (EtOH): λ_{max} ($\log_\varepsilon$)=268.22 nm (5.50).

4-(2-methylphenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**d**)

Pale yellow powder, m.p. 219-220 °C; 42% yield; -FT-IR (KBr): 3314.86, 2936.99, 1712.01, 1617.22 cm^{-1}. -^{1}H NMR(500 MHz, DMSO-d_6): δ=1.84(m, 2H, H-8), 2.08 (m, 2H, H-7), 2.20 (m, 2H, H-6), 2.36 (m, 3H, CH_3), 3.15 (d, *J*=10.9 Hz, 1H, H-4), 4.01 (d, *J*=10.65 Hz, 1H, NH), 6.90 (s, 1H, NH), 6.93 (m, 1H, Ar-H), 6.97 (m, 1H, Ar-H), 7.02 (m, 2H, Ar-H). -^{13}C NMR (125 MHz, DMSO-d_6): δ=20.69, 21.35, 28.95, 29.48, 35.98, 37.82, 61.81, 101.07, 101.89, 112.77, 118.22, 125.72, 126.18, 130.13, 135,49, 139.36, 144.54, 167.67, 169.97, 196.42, 206.32 ppm. -MS (EI, 70 eV): *m/z* (%): 255.1 ($M^{+\cdot}$; $C_{15}H_{15}N_2O_2$), 253.2 ($M^{+\cdot}$ - 2H), 240.1 ($M^{+\cdot}$ – $C_{15}H_{14}NO_2$), 227.2 ($M^{+\cdot}$ – $C_{15}H_{15}O_2$), 164.1 ($M^{+\cdot}$ – C_7H_7), 148.1($M^{+\cdot}$ – C_7H_7-CH-NH-CO-NH), 131.1 ($M^{+\cdot}$ – C_7H_7-CH-CH=CH_2), 119.1 ($M^{+\cdot}$ – C_7H_7-CH-NH), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2). -UV/Vis (EtOH): λ_{max} ($\log_\varepsilon$)=258.40 nm (5.49).

4-(4-methoxyphenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**e**)

Pale yellow powder, m.p. 200-202 °C; 60% yield; -FT-IR (KBr): 3389.23, 2959.93, 1722.04, 1601.58, 1375.17 cm^{-1}. -^{1}H NMR (500 MHz, DMSO-d_6): δ=1.86 (m, 2H, H-8), 2.15 (m, H-7), 2.39 (m, 2H, H-6), 2.96 (d, *J*=10.7 Hz, 1H, H-4), 3.68 (s, 3H, OCH_3), 3.88 (d, *J*=10.75 Hz, 1H, NH), 6.75 (m, 2H, Ar-H), 6.84 (s, 1H, NH), 7.10 (m, 2H, Ar-H). -^{13}C NMR (125 MHz, DMSO-d_6): δ=20.99, 29.07, 31.97, 32.18, 35.41, 37.16, 55.38, 60.58, 100.50, 101.44, 113.71, 116.25, 129.70, 137.51, 157.37, 167.61, 195.87, 205.37 ppm. -MS (EI, 70 eV): *m/z* (%): 271.1 (M^{+}, $C_{15}H_{15}N_2O_3$), 269.2 (M^{+} –2H), 256.1($M^{+\cdot}$ – $C_{15}H_{14}NO_3$), 255.1 ($M^{+\cdot}$ – CH_3), 243.1 ($M^{+\cdot}$ – $C_{15}H_{15}O_3$), 164.1 ($M^{+\cdot}$ – C_7H_7O-CH-NH-CO-NH), 147.1 ($M^{+\cdot}$ – C_7H_7O-CH-CH=CH_2), 135.1 ($M^{+\cdot}$ – C_7H_7O-CH-NH), 107.1 ($M^{+\cdot}$ – C_7H_7O), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2). -UV/Vis (EtOH): λ_{max} ($\log_\varepsilon$)=265.66 nm (5.50).

4-(3-methoxyphenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**f**)

Pale yellow powder, m.p. 201-203 °C; 38% yield; -FT-IR (KBr): 3374.80, 2941.18, 1719.71, 1614.34, 1374.19 cm^{-1}. -^{1}H NMR (500 MHz, DMSO-d_6): δ=1.89 (m, 2H, H-8), 2.16 (m, 2H, H-7) 2.39 (m, 2H, H-6), 2.98 (d, *J*=10.7 Hz, 1H, H-4), 3.69 (s, 3H, OCH_3), 3.91 (d, *J*=10.6 Hz, 1H, NH), 6.85 (s, 1H, NH), 6.62 (m, 1H, Ar-H), 6.73 (s, 1H, Ar-H), 6.79 (d, *J*=6.55 Hz, 1H, Ar-H), 7.07 (t, *J*=6.85 Hz, 1H, Ar-H). -^{13}CNMR (125 MHz, DMSO-d_6): δ=20.27, 21.09, 28.98, 29.12, 32.67, 33.47, 35.42, 36.15, 37.27, 55.22, 59.66, 60.38, 100.48, 101.24, 110.36, 111.54, 114.47, 115.04, 116.00, 120.72, 121.33, 128.64, 146.48, 147.31, 159.11, 167.90, 169.59, 189.83, 195.92, 196.31, 205.29, 206.66 ppm. -MS (EI, 70 eV): *m/z* (%): 271.1(M^{+}, $C_{15}H_{15}N_2O_3$), 269.2 (M^{+} –2H), 256.1

($M^{+\cdot}$ – $C_{15}H_{14}NO_3$), 255.1 ($M^{+\cdot}$ – CH_3), 243.2 ($M^{+\cdot}$ – $C_{15}H_{15}O_3$), 164.1 ($M^{+\cdot}$ – C_7H_7O-CH-NH-CO-NH), 147.1 ($M^{+\cdot}$ – C_7H_7O-CH-CH=CH_2), 135.1 ($M^{+\cdot}$ – C_7H_7O-CH-NH), 107.1 ($M^{+\cdot}$ – C_7H_7O), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2). -UV/Vis (EtOH): λ_{max} ($\log_\varepsilon$)=275.12 nm(5.49).

4-(2-methoxyphenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**g**)

Pale yellow powder, m.p. 209-211 °C; 35% yield; -FT-IR (KBr): 3260.57, 2955.07, 1710.75, 1611.88, 1383.05 cm^{-1}.-^{1}H NMR (500 MHz, DMSO-d_6): δ=1.87 (m, 2H, H-8), 2.36 (m, 2H, H-7), 2.38 (m, 2H, H-9), 3.72 (s, 3H, OCH_3), 2.90 (s, 1H, H-4), 4.55 (s, 1H, NH), 6.81 (s, 1H, NH), 7.05 (t, *J*=7.8 Hz, 1H, Ar-H), 6.74 (t, *J*=8.3 Hz, 1H, Ar-H), 6.88 (m, 2H, Ar-H).

-^{13}C NMR (125 MHz, DMSO-d_6): δ=20.50, 20.98, 28.99, 37.26, 55.61, 101.60, 110.41, 111.47, 119.83, 126.55, 129.20, 131.69, 156.54, 169.76, 196.07, 206.42 ppm.-MS (EI, 70 eV): *m/z* (%): 271.1 (M^+; $C_{15}H_{15}N_2O_3$), 269.2 (M^+ –2H), 256.1 ($M^{+\cdot}$ – $C_{15}H_{14}NO_3$), 256.1 ($M^{+\cdot}$ – CH_3), 243.2 ($M^{+\cdot}$ – $C_{15}H_{15}O_3$), 164.1 ($M^{+\cdot}$ – C_7H_7O-CH-NH-CO-NH), 147.1 ($M^{+\cdot}$ – C_7H_7O-CH-CH=CH_2), 135.1 ($M^{+\cdot}$ – C_7H_7O-CH-NH), 107.1 ($M^{+\cdot}$ – C_7H_7O), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2)**.**-UV/Vis (EtOH): λ_{max} (logε)=267.79 nm (5.50).

4-(4-Chlorophenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**h**)

Pale yellow powder, m.p. 232-234 °C; 88% yield; -FT-IR (KBr): 3321.06, 2938.79, 1716.15, 1614.90, 773.18 cm^{-1}.^{1}H NMR (500 MHz, DMSO-d_6): δ=1.86 (m, 2H, H-8), 2.04 (m, 2H, H-7), 2.29 (m, 2H, H-9), 3.06 (s, 1H, H-4), 4.64 (s, 1H, NH), 6.93 (s, 1H, NH), 7.12 (d, *J*=7.85 Hz, 2H, Ar-H), 7.30 (d, *J*=7.55 Hz, 2H, Ar-H).-^{13}C NMR (125 MHz, DMSO-d_6): δ=20.53, 21.01, 28.93, 37.16, 101.55, 110.92, 126.23, 127.34, 128.93, 131.53, 132.50, 141.28, 170.13, 196.22, 205.65 ppm.-MS (EI, 70 eV): *m/z* (%): 277.1 ($M^{+\cdot}$;$C_{14}H_{13}N_2ClO_2$), 274.1 (M^+ - 2H), 262.1 ($M^{+\cdot}$ – $C_{14}H_{12}NClO_2$), 249.1 ($M^{+\cdot}$ – $C_{14}H_{13}ClO_2$), 247.1 ($M^{+\cdot}$ – $C_{14}H_{11}ClO_2$), 182.1 ($M^{+\cdot}$ – C_6H_4 Cl -CH-NH-CO-NH), 151.1 ($M^{+\cdot}$ – C_6H_4 Cl -CH-CH=CH_2), 139.1 ($M^{+\cdot}$ – C_6H_4 Cl-CH-NH), 111 ($M^{+\cdot}$ – C_6H_4 Cl), 71.1($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=262.24 nm (5.49).

4-(3-Chlorophenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**i**)

Pale yellow powder, m.p. 219-220 °C; 71% yield; -FT-IR (KBr): 3074.81, 2984.07, 1718.42, 1603.53, 782.22 cm^{-1}.-^{1}H NMR (500 MHz, DMSO-d_6): δ=1.89 (m, 2H, H-8), 2.18 (m, 2H, H-7),

2.41 (m, 2H, H-9), 3.04 (d, *J*=10.85 Hz, 1H, H-4), 3.94 (d, *J*=10.8Hz, 1H, NH), 6.94 (s, 1H, NH), 7.21 (d, *J*=8.45 Hz 1H, Ar-H), 7.18 (m, *J*=7.65 Hz, 1H, Ar-H), 7.16 (m, *J*=7.45 Hz, 1H, Ar-H), 7.14 (m, 1H, Ar-H).-^{13}C NMR (125 MHz, DMSO-d_6): δ=20.43, 20.87, 29.07, 32.95, 35.23, 37.02, 59.76, 100.48, 101.46, 125.71, 127.53, 128.18, 128.90, 129.50, 129.67, 132.33, 132.40, 147.62, 148.20, 168.47, 195.95, 205.40 ppm.-MS (EI, 70 eV): *m/z* (%): 277.1 ($M^{+\cdot}$; $C_{14}H_{13}N_2ClO_2$), 274.1 ($M^{+\cdot}$ – 2H), 262.1 ($M^{+\cdot}$ – $C_{14}H_{12}NClO_2$), 249.1 ($M^{+\cdot}$ – $C_{14}H_{13}ClO_2$), 247.1 ($M^{+\cdot}$ – $C_{14}H_{11}ClO_2$), 182.1 ($M^{+\cdot}$ – C_6H_4 Cl -CH-NH-CO-NH), 151.1 ($M^{+\cdot}$ – C_6H_4 Cl -CH-CH=CH_2), 139.1 ($M^{+\cdot}$ – C_6H_4 Cl-CH-NH), 111.1 ($M^{+\cdot}$ – C_6H_4 Cl), 71.1($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=262.24 nm (5.49).

4-(2-Chlorophenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**j**)

Pale yellow powder, m.p. 222-224 °C; 80% yield; -FT-IR (KBr): 3084.82, 2944.99, 1719.08, 1603.92, 796.06cm^{-1}.-^{1}H NMR (500 MHz, DMSO-d_6): δ=1.89 (m, 2H, H-8), 2.17 (m, 2H, H-7), 2.39 (m, 2H, H-9), 3.01 (d, *J*=10.85 Hz, 1H, H-4), 3.88 (d, *J*=9 Hz, 1H, NH), 6.87 (s, 1H, NH), 7.21 (m, 1H, Ar-H), 7.19 (m, 2H, Ar-H), 7.07 (d, *J*=8.4 Hz, 1H, Ar-H).-^{13}C NMR (125 MHz, DMSO-d_6): δ=20.14, 20.87, 29.04, 31.93, 32.55, 35.29, 37.04, 59.92, 100.45, 101.44, 127.71, 129.75, 130.09, 130.15, 130.77, 143.91, 144.63, 168.25, 169.87, 195.91, 196.37, 205.33 ppm.-MS (EI, 70 eV): *m/z* (%): 277.1 ($M^{+\cdot}$; $C_{14}H_{13}N_2ClO_2$), 274.1 ($M^{+\cdot}$ –2H), 262.1 ($M^{+\cdot}$ – $C_{14}H_{12}NClO_2$), 249.1 ($M^{+\cdot}$ – $C_{14}H_{13}ClO_2$), 247.1 ($M^{+\cdot}$ – $C_{14}H_{11}ClO_2$), 182.1 ($M^{+\cdot}$ – C_6H_4 Cl -CH-NH-CO-NH), 151.1 ($M^{+\cdot}$ – C_6H_4 Cl -CH-CH=CH_2), 139.1 ($M^{+\cdot}$ – C_6H_4 Cl-CH-NH), 111.1 ($M^{+\cdot}$ – C_6H_4 Cl), 71.1 ($M^{+\cdot}$ – NH-CH=CH-COH), 70.1 ($M^{+\cdot}$ – CH_3-CO-CH=CH_2), 57.1 ($M^{+\cdot}$ – NH-CO-NH), 51.1 ($M^{+\cdot}$ – CH_2=CH-COH), 42.1 ($M^{+\cdot}$ – CH_2-CH_2-CH_2)**.**-UV/Vis (EtOH): λ_{max} (logε)=258.83 nm (5.49)**.**

4-(4-Boromophenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,6H)-diones (2**k**)

Pale yellow powder, m.p. 216-219 °C; 64% yield; -FT-IR (KBr): 3174.20, 2945.97, 1720.89, 1603.05 cm^{-1}.-^{1}H NMR (500 MHz, DMSO-d_6): δ=1.89 (m, 2H, H-8), 2.15 (m, 2H, H-7), 2.40 (m, 2H, H-9), 3.00 (d, *J*=10.87 Hz, 1H, H-4), 3.88 (d, *J*=10.85 Hz, 1H, NH), 6.91 (s, 1H, NH), 7.16 (d, *J*=8.43 Hz, 2H, Ar-H), 7.30 (d, *J*=8.43 Hz, 2H, Ar-H).-^{13}C NMR (125 MHz, DMSO-d_6): δ= 20.65, 21.06, 28.95, 32.43, 36.71, 37.21, 56.71, 101.63, 111.22, 127.74, 131.90, 132.24, 142.73, 170.16, 196.24, 205.46 ppm.-MS (EI, 70 eV): *m/z* (%): 321 ($M^{+\cdot}$; $C_{14}H_{13}N_2BrO_2$), 318 ($M^{+\cdot}$ –2H), 306 ($M^{+\cdot}$- $C_{14}H_{12}NBrO_2$), 293 ($M^{+\cdot}$- $C_{14}H_{13}BrO_2$), 241 ($M^{+\cdot}$- $C_{14}H_{13}N_2O_2$), 213.1 ($M^{+\cdot}$- C_6H_4 Br -CH-NH-CO-NH), 197 ($M^{+\cdot}$- C_6H_4 Br-CH-CH=CH_2), 185 ($M^{+\cdot}$- C_6H_4 Br-CH-NH), 157 ($M^{+\cdot}$- C_6H_4 Br), 71.1 ($M^{+\cdot}$-

NH-CH=CH-COH), 70.1 ($M^{+\cdot}$- CH_3-CO-CH=CH_2), 57.8 ($M^{+\cdot}$- NH-CO-NH), 51.1 ($M^{+\cdot}$- CH_2=CH-COH), 42.1 ($M^{+\cdot}$- CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=255.42 nm (5.48).

4-(3-Boromo phenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,2H)-diones (2**l**)

White powder, m.p. 215-217 °C; 79% yield; -FT-IR (KBr): 3100.14, 2939.21, 1718.91, 1598.64 cm^{-1}.^{1}H NMR (500 MHz, DMSO-d_6): δ=1.83 (m, 2H ,H-8), 2.13 (m, 2H, H-7), 2.36 (m, 2H, H-9), 3.03 (d, *J*=10.85 Hz, 1H, H-4), 3.88 (d, *J*=9.6 Hz, 1H, NH), 6.93 (s, 1H, NH), 7.31 (s, 1H, Ar-H), 7.22 (d, *J*=7.6 Hz, 1H, Ar-H), 7.18 (d, *J*=7.9 Hz, 1H, Ar-H), 7.11 (t, *J*=7.7 Hz, 1H, Ar-H).-MS (EI, 70 eV): *m/z* (%): 321.1 ($M^{+\cdot}$; $C_{14}H_{13}N_2BrO_2$), 318.1 ($M^{+\cdot}$ –2H), 306.1 ($M^{+\cdot}$- $C_{14}H_{12}NBrO_2$), 293.1 ($M^{+\cdot}$- $C_{14}H_{13}BrO_2$), 241.1 ($M^{+\cdot}$- $C_{14}H_{13}N_2O_2$), 213.1 ($M^{+\cdot}$- C_6H_4 Br -CH-NH-CO-NH), 197.1 ($M^{+\cdot}$- C_6H_4 Br-CH-CH=CH_2), 185 ($M^{+\cdot}$- C_6H_4 Br-CH-NH), 157 ($M^{+\cdot}$- C_6H_4 Br), 71.1 ($M^{+\cdot}$- NH-CH=CH-COH), 70.1 ($M^{+\cdot}$- CH_3-CO-CH=CH_2), 57.8 ($M^{+\cdot}$- NH-CO-NH), 51.1 ($M^{+\cdot}$- CH_2=CH-COH), 42.1 ($M^{+\cdot}$- CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=260.54 nm (5.49).

4-(2-Boromophenyl) -1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,2H)-diones (2**m**)

Pale yellow powder, m.p. 219-220 °C; 85% yield; FT-IR (KBr): 3328.76, 2935.95, 1713.36, 1615.06 cm^{-1}.-^{1}H NMR (500 MHz, DMSO-d_6): δ=1.86 (m, 2H, H-8), 2.16 (m, 2H, H-7), 2.37 (m, 2H, H-9), 3.06 (s, 1H, H-4), 4.51 (s, 1H, NH), 7.03 (s, 1H, NH), 7.45 (d, *J*=7.65 Hz, 1H, Ar-H), 7.12 (d, *J*=3.85 Hz, 2H, Ar-H), 7.04 (d, *J*=4.2 Hz, 2H, Ar-H).^{13}C NMR (125 MHz, DMSO-d_6): δ=20.65, 21.06, 28.95, 32.43, 36.71, 37.21, 56.71, 101.63, 111.22, 127.74, 131.90, 132.24, 142.73, 170.16, 196.24, 205.46 ppm.-MS (EI, 70 eV): *m/z* (%): 321.1 ($M^{+\cdot}$; $C_{14}H_{13}N_2BrO_2$), 318.1 ($M^{+\cdot}$ –2H), 306.1 ($M^{+\cdot}$- $C_{14}H_{12}NBrO_2$), 293.1 ($M^{+\cdot}$- $C_{14}H_{13}BrO_2$), 241.1 ($M^{+\cdot}$- $C_{14}H_{13}N_2O_2$), 213.2 ($M^{+\cdot}$- C_6H_4 Br -CH-NH-CO-NH), 197.1 ($M^{+\cdot}$- C_6H_4 Br-CH-CH=CH_2), 185.1 ($M^{+\cdot}$- C_6H_4 Br-CH-NH), 157.1 ($M^{+\cdot}$- C_6H_4 Br), 71.1 ($M^{+\cdot}$- NH-CH=CH-COH), 70.1 ($M^{+\cdot}$- CH_3-CO-CH=CH_2), 57.8 ($M^{+\cdot}$- NH-CO-NH), 51.1 ($M^{+\cdot}$- CH_2=CH-COH), 42.1 ($M^{+\cdot}$- CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=260.54 nm (5.49).

4-(4-Nitrophenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,2H)-diones (2**n**)

White powder, m.p. 225-226 °C; 78% yield; -FT-IR (KBr): 3123.71, 2950.15, 1719.29, 1600.86, 1515.07, 1343.04cm^{-1}.^{1}H NMR (500 MHz, DMSO-d_6): δ=1.90 (m, 2H, H-8), 2.19 (m, 2H, H-7), 2.39 (m, 2H, H-9), 3.08 (d, *J*=10.95 Hz, 1H, H-4), 4.01 (d, *J*=9.95 Hz, 1H, NH), 7.10 (s, 1H, NH), 7.49 (d, *J*=8.55 Hz 2H, Ar-H), 8.02 (d, *J*=8.55 Hz, 2H, Ar-H).-^{13}C NMR (125 MHz, DMSO-d_6): δ=20.43, 21.03, 28.88, 29.07, 32.33, 33.51, 35.23, 36.38, 37.04, 59.39, 100.45, 101.50, 110.54, 115.15, 122.92, 123.00, 129.59, 130.28, 145.78, 153.76, 154.17, 168.89, 170.32, 195.99, 196.51,

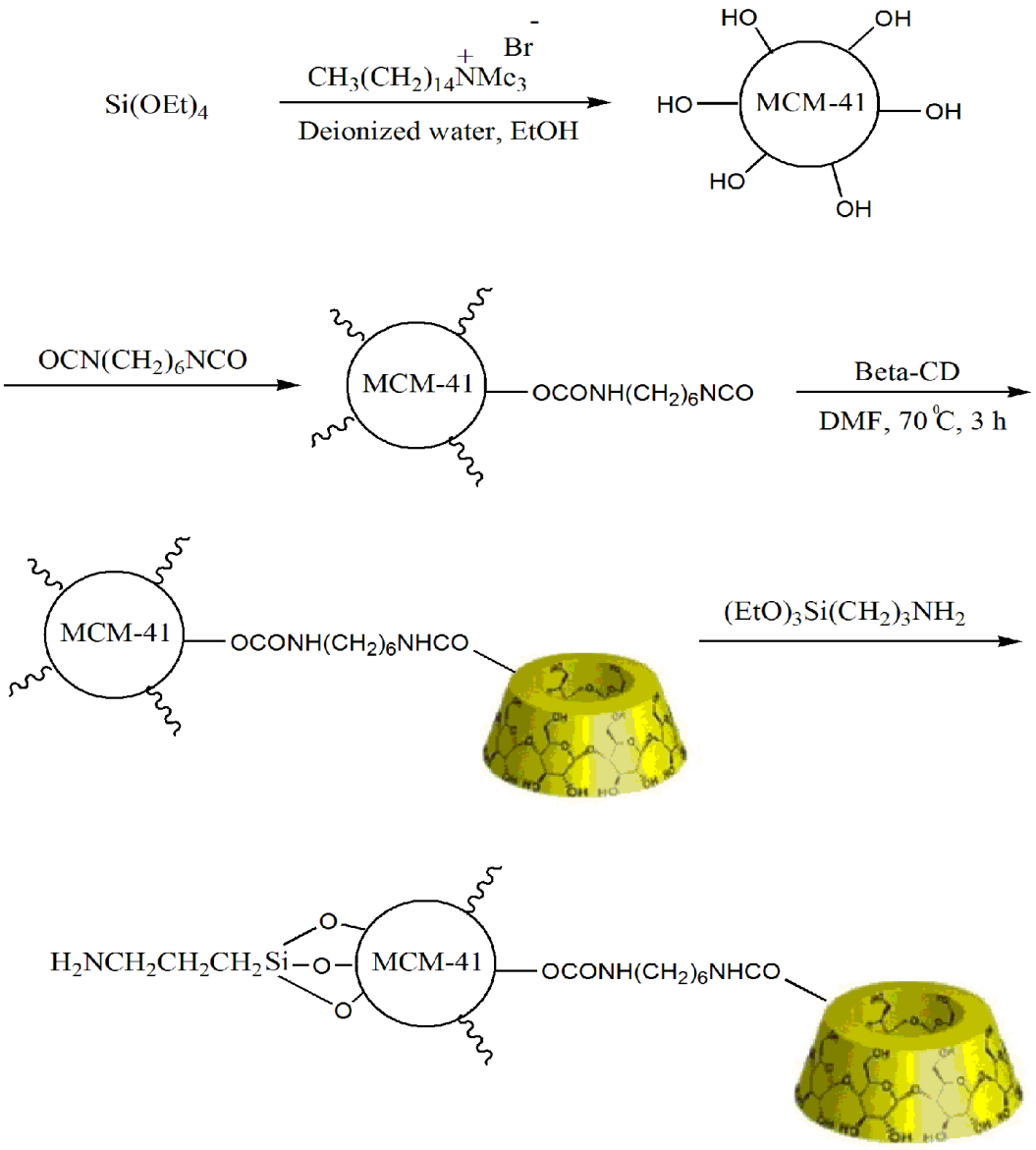
205.88 ppm.-MS (EI, 70 eV): *m/z* (%): 287.1 ($M^{+\cdot}$; $C_{14}H_{13}N_3O_4$), 285.1 ($M^{+\cdot}$ –2H), 272.1 ($M^{+\cdot}$-$C_{14}H_{12}N_2O_4$), 259.1 ($M^{+\cdot}$-$C_{14}H_{13}NO_4$), 258.1 ($M^{+\cdot}$- $C_{14}H_{13}N_2O_3$), 241.1 ($M^{+\cdot}$-$C_{14}H_{13}N_2O_2$), 193.1 ($M^{+\cdot}$-C_6H_4 NO_2 -CH-NH-CO-NH), 165.1 ($M^{+\cdot}$-C_6H_4 NO_2), 162.1 ($M^{+\cdot}$-C_6H_4 NO_2-CH-CH=CH_2), 150.1 ($M^{+\cdot}$-C_6H_4 NO_2-CH-NH), 71.1 ($M^{+\cdot}$-NH-CH=CH-COH), 70.1 ($M^{+\cdot}$-CH_3-CO-CH=CH_2), 57.8 ($M^{+\cdot}$-NH-CO-NH), 51.1 ($M^{+\cdot}$-CH_2=CH-COH), 42.1 ($M^{+\cdot}$-CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=262.67 nm (5.50)**.**

4-(3-Nitrophenyl)-1,3,4,6,7,8-hexahydroquinazolin-2,5(1H,2H)-diones (2**o**)

Pale yellow powder, m.p. 216-218 °C; 71% yield; -FT-IR (KBr): 3119.91, 2953.97, 1719.17, 1601.62, 1524.68, 1351.53 cm^{-1}.^{1}H NMR (500 MHz, DMSO-d_6): δ=1.96 (m, 2H, H-8), 2.12 (m, 2H, H-7), 2.38 (m, 2H, H-9), 3.17 (d, *J*=10.95 Hz, 1H, H-4), 4.01 (d, *J*=10.05 Hz, 1H, NH), 7.04 (s, 1H, NH), 8.01 (s, 1H, Ar-H), 7.95 (d, *J*=9.4 Hz, 1H, Ar-H), 7.48 (m, 1H, Ar-H), 7.67(m, 1H, Ar-H).-^{13}C NMR (125 MHz, DMSO-d_6): δ=20.28, 26.95, 29.09, 31.87, 33.29, 35.16, 36.76, 59.29, 100.56, 114.93, 115.15, 120.95, 123.23, 129.23, 130.08, 147.65, 165.94, 169.04, 196.04, 205.57 ppm.-MS (EI, 70 eV): *m/z* (%): 287.1 ($M^{+\cdot}$;$C_{14}H_{13}N_3O_4$), 285.2 ($M^{+\cdot}$ –2H), 272.1 ($M^{+\cdot}$- $C_{14}H_{12}N_2O_4$), 259.1 ($M^{+\cdot}$-$C_{14}H_{13}NO_4$), 258.1 ($M^{+\cdot}$- $C_{14}H_{13}N_2O_3$), 241.1 ($M^{+\cdot}$- $C_{14}H_{13}N_2O_2$), 193.1 ($M^{+\cdot}$- C_6H_4 NO_2 -CH-NH-CO-NH), 165.1 ($M^{+\cdot}$- C_6H_4 NO_2), 162.1 ($M^{+\cdot}$- C_6H_4 NO_2-CH-CH=CH_2), 150.1 ($M^{+\cdot}$- C_6H_4 NO_2-CH-NH), 71.1 ($M^{+\cdot}$- NH-CH=CH-COH), 70.1 ($M^{+\cdot}$- CH_3-CO-CH=CH_2), 57.8 ($M^{+\cdot}$- NH-CO-NH), 51.1 ($M^{+\cdot}$- CH_2=CH-COH), 42.1 ($M^{+\cdot}$- CH_2-CH_2-CH_2).-UV/Vis (EtOH): λ_{max} (logε)=263.52 nm (5.50).

Results and discussion

The systematic steps of aminopropyl and *β*-cyclodextrin grafted mesoporous MCM-41, MCM-41-*β*-CD.NH_2 synthesis is shown in Scheme 1.



Scheme 1. Synthetic procedure of MCM-41-*β*-CD/NH_2.

The structure of MCM-41-*β*-CD/NH_2 was confirmed by FT-IR spectra. The typical Si–O–Si bands around 1228, 1063, 794 and 462 cm^{-1} associated with the formation of a condensed silica network are present in the spectra**.** The strong peak around 1630 cm^{-1} is related to the bending vibration of adsorbed H_2O. The peaks at 2800-3400 cm^{-1} region are attributed to amino groups, which are covered by O–H vibration located in silica surface and physically adsorbed water. The bands in the range of 2800–3000 cm^{-1} corresponded to the stretching vibration of the C–H bonds of the methylene groups, which indicates successful grafting of organic groups to MCM-41. By using a Philips XL30 scanning electron microscope, SEM determined the morphology and particle size distribution of MCM-41-β-CD/NH2. The nanocomposite has spherical shape with nano dimension of about 300 nm (Figure 1).

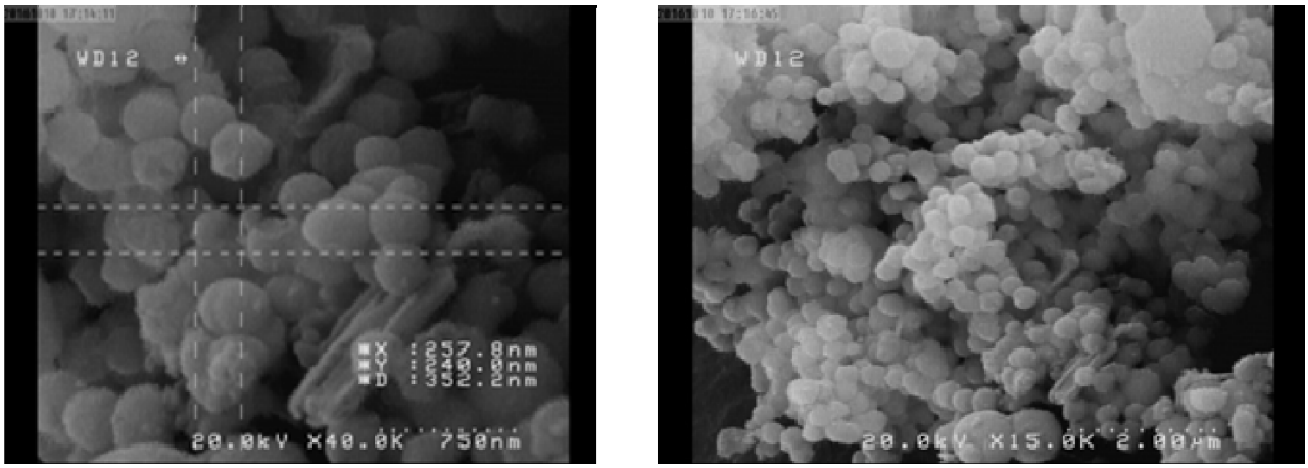


Figure 1. The SEM Images of MCM-41-β-CD/NH_2.

Transmission electron microscopy (TEM) revealed that MCM-41-β-CD/NH_2 has an average particle size about 300 nm (Figure 2).

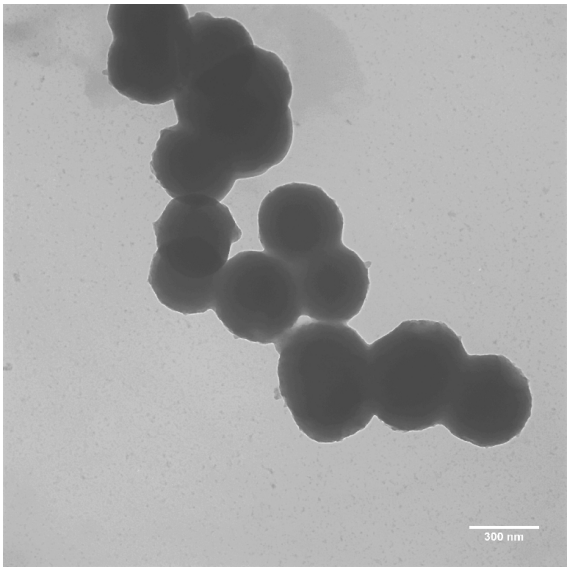


Figure 2. The TEM of MCM-41-β-CD/NH_2.

X-ray diffraction (XRD) pattern of the MCM-41-β-CD/NH_2 powder is shown in Figure 3. The broad peak around 2° in the XRD pattern is attributed to amorphous silica.

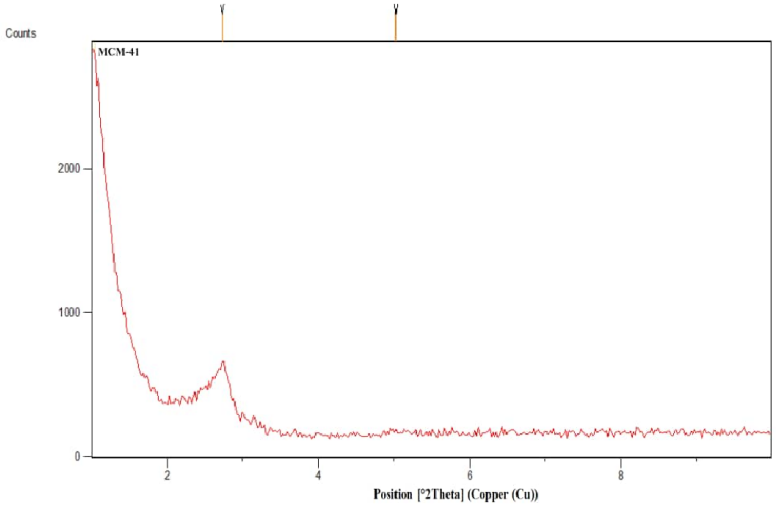


Figure 3. The XRD Analysis of MCM-41-β-CD/NH_2.

The loss of organic components attached to the MCM-41 can be quantified with the weight loss in thermogravimetric analysis (TGA-DTG). Hence, the presence of organic parts, β-CD, and amino propyl in the MCM-41 mesoporous network were confirmed through TGA-DTG. Figure 4 shows two distinct weight loss steps in the combined TGA-DTG curves. The curves show that the first weight loss occurs before 200 ^{o}C, which can be attributed completely to the loss of adsorbed water molecules (11%). The secondary weight losses at about 250 ^{o}C shows the decomposition of organic substances in MCM-crown composites (24%). The decomposition of organic substance is complete at 680 ^{o}C to form the constituent inorganic oxides. TGA of the samples demonstrated high thermal stability, with decomposition starting at around 250 ^{o}C under a nitrogen atmosphere.

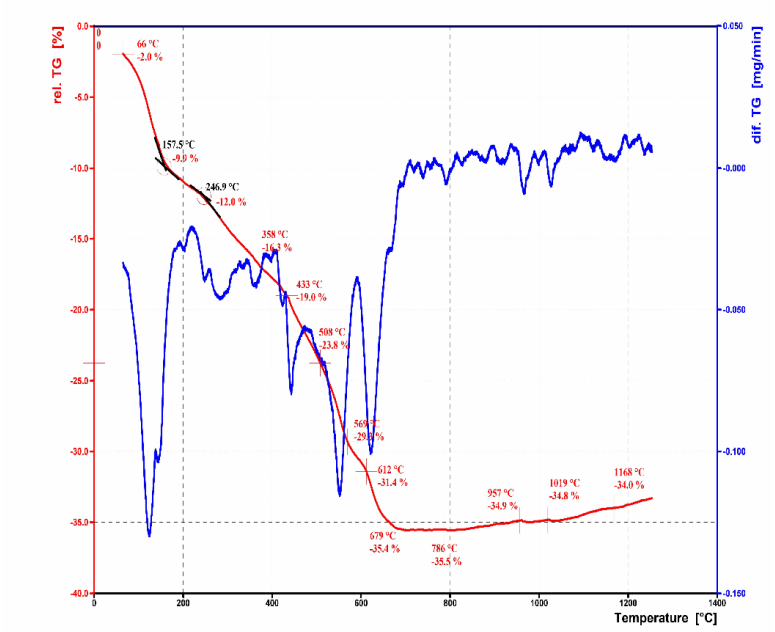


Figure 4. The TGA-DTG of MCM-41-β-CD/NH_2.

The specific surface area and the pore size distribution were also calculated by Brunauer-Emmett-Teller (BET) method. The pore size distribution was calculated using desorption branches of nitrogen isotherms. The total surface of catalyst is 39.9 m^2/g and the BET surface is 6.981 m^2 (Figure 5).

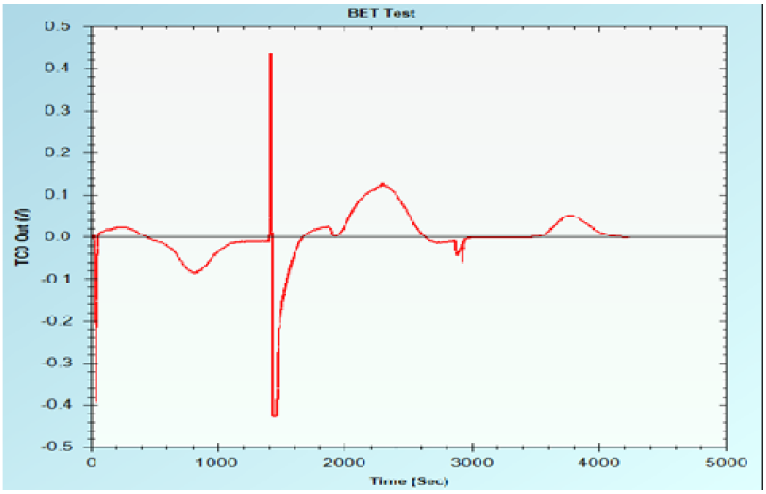
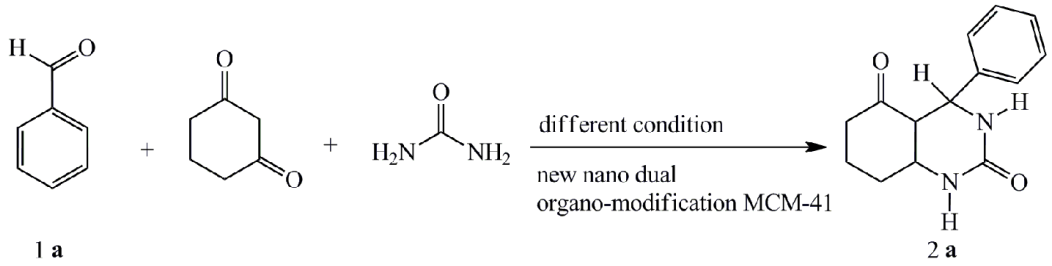


Figure 5. The BET curve of MCM-41-*β*-CD/NH_2.

The basic capacity of the hybrid nanocomposite was measured based on the previously reported procedures [25]. The potential application of the covalently linked basic nanocomposite as a stationary micro-vessel basic heterogeneous catalyst was to the Biginelli multicomponent condensation reaction. At first, one pot multicomponents condensation of 1,3-cyclohexadione, benzaldehyde and urea were investigated in the presence of nanocomposite. TLC followed the progress of the reaction until the total disappearance of the benzaldehyde (Scheme 2). The results of reaction optimaization are reported in the Table 1.



1 a **2 a**

Scheme 2. MCM-41-*β*-CD/NH_2 catalyzed synthesis of the 4-phenyl-1,3,4,6,7,8-hexahydroquinazolin-2,5(1*H*,6*H*)-diones (2a).

Table 1. Optimization of reaction conditions in the synthesis of of 4-phenyl-1,3,4,6,7,8-hexahydroquinazolin-2,5 (1*H*,6*H*)-diones product (2a) under different conditions.

Comp	Catalyst (g)	T (˚C)	Solvent	Yield (%) [a]	Time (min) [b]
2a	Without	r.t.	Solvent free	30	198
2a	Without	100	Solvent free	35	180
2a	0.1	reflux	H_2O (10 mL)	66	144
2a	0.1	reflux	EtOH (10 mL)	45	132
2a	0.1	reflux	CH_3CN (10 mL)	78	120
2a	0.15	reflux	CH_3CN (10 mL)	86	45
2a	0.05	reflux	CH_3CN (10 mL)	63	144
2a	0.2	reflux	CH_3CN (10 mL)	80	78

a. Isolated Yield; b. Times are given after maximum progress of reaction.

The best condition was achieved using mixture of as aldehyde (10 mmol), 1,3-cyclohexadione (10 mmol), urea (12 mmol) and nano MCM-41-*β*-CD/NH_2 (0.15 g) in acetonitrile (10 mL) under reflux condition. The results for the application of aromatic aldehaydes with electron donating and withdrawing groups are shown in Table 2.

Table 2. New nano MCM-41-*β*-CD/NH_2 catalyzed synthesis of some 4-aryl-3,4,6,7,8 hexahydroquinazolin-2,5(1*H*,6*H*)-diones 2**a-o**.

Comp.	Ar	Time (min)	Yield%	m.p (˚C)
2**a**	C_6H_5	45	86	228-230
2**b**	4-CH_3-C_6H_4	90	37	192-193
2**c**	3-CH_3-C_6H_4	50	44	211-213
2**d**	2-CH_3-C_6H_4	50	42	219-220
2**e**	4-CH_3O-C_6H_4	60	60	200-202
2**f**	3-CH_3O-C_6H_4	70	38	201-203
2**g**	2-CH_3O-C_6H_4	45	35	209-211
2**h**	4-Cl-C_6H_4	30	88	232-234
2**i**	3-Cl-C_6H_4	50	71	219-220
2**j**	2-Cl-C_6H_4	30	80	222-224
2**k**	4-Br-C_6H_4	80	64	216-219
2**l**	3-Br-C_6H_4	35	79	215-217
2**m**	2-Br-C_6H_4	30	85	219-220
2**n**	4-NO_2-C_6H_4	40	78	225-226
2**o**	3-NO_2-C_6H_4	50	71	216-218

The Comparison of the application of MCM-41-β-CD/NH_2 as a catalyst for the preparation of hexahydroquinazolins with recently reported catalysts (nano K_3AlF_6 and K_3AlF_6) in this reaction were reported in Table 3.

Table 3. Comparison of MCM-41-*β*-CD/NH_2 with nano K_3AlF_6 and K_3AlF_6.

Comp.	Ar	MCM-41-*β*-CD/NH_2		nano *K3AlF6*		*K3AlF6*	
		Time (min)	Yield %	Time (min)	Yield %	Time (min)	Yield %
2a	C_6H_5	45	86	90	95	120	80
2b	4-CH_3-C_6H_4	90	37	50	90	150	82
2c	3-CH_3-C_6H_4	50	44	80	85	180	85
2d	2-CH_3-C_6H_4	50	42	50	90	120	83
2e	4-CH_3O-C_6H_4	60	60	90	85	180	85
2f	3-CH_3O-C_6H_4	70	38	90	85	150	75
2g	2-CH_3O-C_6H_4	45	35	50	95	150	75
2h	4-Cl-C_6H_4	30	88	60	95	120	82
2i	3-Cl-C_6H_4	50	71	50	85	129	80
2j	2-Cl-C_6H_4	30	80	55	80	120	85
2k	4-Br-C_6H_4	80	64	85	85	120	77
2l	3-Br-C_6H_4	35	79	50	90	150	87
2m	2-Br-C_6H_4	30	85	65	90	150	84
2n	4-NO_2-C_6H_4	40	78	65	80	120	85
2o	3-NO_2-C_6H_4	50	71	85	80	150	80

As shown in Table 3, this catalyst is better than the others in high yield and time. The catalyst recyclability was confirmed in the 4-phenyl-3,4,6,7,8 hexahydroquinazolin-2,5(1*H*,6*H*)-diones **2a** and the results are shown in Figure 6.

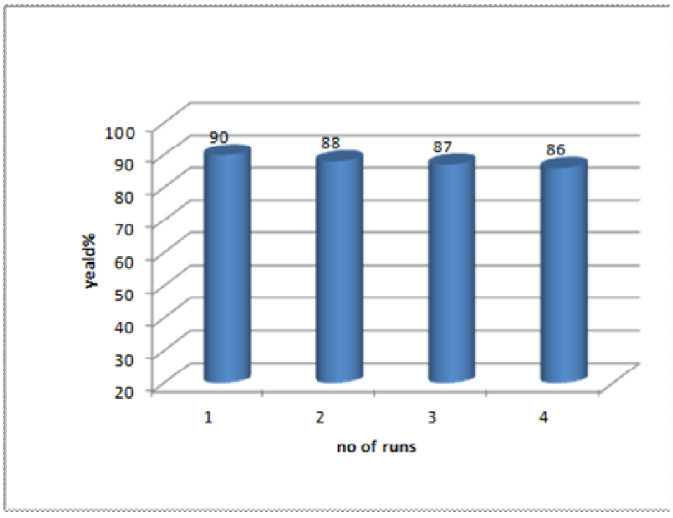


Figure 6. Recyclability of MCM-41-β-CD/NH_2.

Conclusion

In the present study, a mesoporous MCM-41 having *β*-CD and amino basic units with pore channels were synthesized via a surfactant-templated sol–gel methodology and a post modification process. The catalytic activity of the basic nanocomposite has been successfully applied to the one-pot three-components reaction 1,3-cyclohexadione, aromatic aldehyde and urea in CH_3CN as the solvents. This catalytic system certainly contributes to better environmental and green technology for the facile preparation of the 4-Aryl-1,3,4,6,7,8-hexahydroquinazolin-2,5(1*H*,6*H*)-diones derivatives. The current methodology has the advantages of operational simplicity, short reaction times, good yields and the desired products which can be separated directly from the reaction mixture with high purity.

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