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Amino-Functionalized MCM-41 Base-Catalyzed One-Pot Synthesis of 2-Amino-5,6-dihydropyrimidin-4(3*H*)-ones

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A simple and efficient one-pot method for the preparation of 2-amino-5,6-dihydropyrimidin-4(3*H*)-one from the cyclocondensation of Meldrum's acid, aldehydes and guanidinium carbonate using a catalytic amount of amino-functionalized MCM-41 is described. This efficient technique has the advantage of giving 2-NH_2-DHPM building blocks using a base catalyst in good to high yields, being completed in short reaction times and offering a simple product isolation procedure.

Keywords: 2-Amino-5,6-dihydropyrimidin-4(3*H*)-one, Guanidinium carbonate, 2-NH_2-DHPM building blocks, Base catalyst, Amino-functionalized MCM-41

INTRODUCTION

Dihydropyrimidines are well-known heterocycles that display a diverse range of biological activities such as antibacterial, antiviral, antitumor, anti-inflammatory and antihypertensive as well as calcium channel blockers, α-1a-antagonists, and neuropeptide Y (NPY) antagonists [1-3]. Consequently, the Biginelli reaction of carbonyl compounds, aldehydes and urea or thiourea has been reviewed and several improved procedures have recently been reported [3-9]. The use of a number of homogeneous catalysts, such as polyphosphate esters [10], $BF_3.OEt_2$ [11], $LaCl_3.H_2O$ [12] and $InBr_3$ [13], and heterogeneous solid catalysts, such as HY [14], Amberlyst-15 or Nafion-H [15], montmorillonite-KSF [16], MCM-41-R-SO_3H [17] and MCM-41 supported $FeCl_3$ [8] has been investigated in connection with this reaction.

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However, three-component condensation of carbonyl compounds, aldehydes and guanidine with base catalysts leading to DHPMs has been reported by only few researchers in few examples in the literature [18-23]. For example, Milcent *et al*. [21] synthesized ethyl 2-amino-4-aryl-1,4-dihydro-6-phenylpyrimidine-5-carboxylates from ethyl 3-aryl-2-benzoylpropenoates and guanidine in DMF in the presence of sodium hydrogen carbonate. The reaction required long times (8 to 48 h) and gave modest yields (40%). Eynde *et al*. [22] investigated the synthesis of the same compounds from condensation of ethyl benzoylacetate, aldehyde and guanidine in higher yields under similar conditions. On the other hand, Ostras *et al*. described the synthesis of 2-NH_2-DHPM building blocks under conventional and microwave conditions from three-component reaction of Meldrum's acid, aldehyde and guanidine in moderate yields [23].

It is of great practical importance to synthesize 2-NH_2-DHPM building blocks using easily separable and reusable

solid base catalysts. MCM-41 is a highly porous material with nano-size channel diameter (1.5-10 nm). It displays acidic silanol groups and high surface area [24-25]. In earlier studies by some of us on the chemical functionalization of MCM-41 catalyst with 3-aminopropyltriethoxysilane (APTES), we observed a change in the surface properties [26]. NH_2-functionalized MCM-41 was prepared by means of chemical bonding of aminosilane on the inner surface of the mesoporous material. Elemental analyses (CHN) indicated that the amine was immobilized on the surface and BET surface area measurement revealed that the MCM-41 surface area (1018 m^2 g^{-1}) decreased to 638 m^2 g^{-1} after grafting APTES molecules. Low-angle XRD pattern was slightly shifted to higher angles, which is in agreement with a decrease of the pore size [26]. This functionalization process converts the surface to be basic by incorporation of NH_2 terminal groups and thus offers an opportunity to be used as a base catalyst [27]. It should be noted that this water-tolerant [28-29] amino-functionalized MCM-41 catalyst attracts reagents to the surface quite easily, as compared to the hydrophilic surface of MCM-41 [30-31].

In continuation of our investigations on the synthesis of 1,4-dihydropyridines (DHPs) [32] and dihydropyrimidines (DHPMs) [33-35], herein we describe a simple and efficient method for the preparation of 2-amino-5,6-dihydropyrimidin-4(3*H*)-one building blocks using amino-functionalized MCM-41 catalyst. In this work, we introduce an efficient three-component reaction of Meldrum's acid, aldehydes and guanidinium carbonate that provides an easy access to 2-amino-5,6-dihydropyrimidin-4(3*H*)-one building blocks using MCM-41 catalyst functionalized with 3-aminopropyltriethoxy-silane (MCM-41-NH_2) [26]. This efficient method has the advantage of giving 2-NH_2-DHPM building blocks in good to high yields and being completed in short reaction times using a base catalyst (MCM-41-NH_2).

EXPERIMENTAL

Chemicals and Apparatus

Melting points were determined in evacuated capillaries with a Buchi B-545 apparatus. Mass spectra were obtained on a FISONS GC 8000/TRIO 1000 under 70 eV. ^{1}H NMR spectra were recorded on a Bruker 500, 400 or 80 MHz in DMSO-d_6 using tetramethylsilane as internal standard. Fourier transform infrared (FTIR) spectra were recorded from KBr disks using a Bruker Vector 22 FT-IR spectrometer. Elemental analyses were performed on a ThermoFinigan Flash EA 1112 series elemental analyzer.

KF/Al_2O_3 (40% by weight) was prepared as follows: KF (3 g) was dissolved in water (20 ml), mixed with neutral TLC grade chromatographic alumina (4.5 g) in water (20 ml), and then stirred for 30 min at room temperature. After evaporation of water under reduced pressure, the alumina was dried at 160 °C for 8 h.

MCM-41 was synthesized according to a previously reported procedure [37] and functionalized with 3-amino-propyltriethoxysilane (APTES) [38-39]. In a typical reaction, 1 g of MCM-41 was suspended in 50 ml of toluene and the mixture was stirred for an hour and then to this mixture 1.5 g of APTES was added and refluxed overnight. The white solid was removed from the solvent by filtration and was soxhleted by toluene and dried overnight under reduced pressure at 70 °C [26].

Typical Procedure for the Synthesis of 2-Amino-5,6-dihydropyrimidin-4(3*H*)-one 4a-k Derivatives

To a solution of Meldrum's acid **1** (1 mmol)**,** aldehyde **2a-k** (1 mmol) and guanidinium carbonate **3** (1 mmol) in DMF (0.5 ml) was added amino-functionalized MCM-41 (10 mol%). The reaction mixture was heated at 120 °C for appropriate time as indicated in Table 2. After completion of the reaction, the mixture was poured into crushed ice while stirring, filtered through a sintered funnel and washed by ethyl acetate-hexane (1:3).

For further purification, the product was recrystallized from ethanol. Spectroscopic data for the compounds **4a-k**:

2-Amino-6-phenyl-5,6-dihydropyrimidin-4(3*H*)-one (4a) [23]**.** m.p.: 257.3 °C; ^{1}H NMR (DMSO-d_6, 400 MHz): δ 2.27 (dd, J = 15.4, 8.3 Hz, 1H, CH_2), 2.49 (dd, J = 15.4, 6.0 Hz, 1H, CH_2), 4.61 (t, J = 7.5 Hz, 1H, CH), 6.54 (s, 2H, NH_2), 7.25-7.35 (m, 5H, 5CH arom.), 7.63 (s, 1H, NH); MS (70 eV): *m/z* (%) 189 (70) (M^+), 188 (45), 160 (25), 146 (37), 104 (100); IR (KBr): ν 3319, 3221, 3005, 2970, 2899, 2796, 1641, 1597, 1556, 1510, 1487, 1375 cm^{-1}, Anal. Calcd. for $C_{10}H_{11}N_3O$: C, 63.48; H, 5.86; N, 22.21%. Found: C, 62.43; H, 5.97; N, 22.88%.

2-Amino-6-(4-chlorophenyl)-5,6-dihydropyrimidin-4 (3*H*)-one (4b). m.p.: 280.3 °C; ^{1}H NMR (DMSO-d_6, 400

MHz): δ 2.24 (dd, *J* = 15.4, 8.0 Hz, 1H, CH_2), 2.49 (dd, *J* = 15.4, 6.0 Hz, 1H, CH_2), 4.63 (t, *J* = 6.0 Hz, 1H, CH), 6.55 (s, 2H, NH_2), 7.30 (d, *J* = 8.0 Hz, 2H, 2CH arom.), 7.40 (d, *J* = 8.0 Hz, 2H, 2CH arom.), 7.65 (s, 1H, NH); MS (70 eV): *m/z* (%) 223 (30) (M^+), 188 (20), 180 (20), 166 (5), 138 (40), 111(35); IR (KBr): ν 3356, 3008, 2837, 1639, 1591, 1506, 1398 cm^{-1}; Anal. Calcd. for $C_{10}H_{10}ClN_3O$: C, 53.70; H, 4.51; N, 18.79%. Found: C, 53.31; H, 4.58; N, 18.48%.

2-Amino-6-(4-bromophenyl)-5,6-dihydropyrimidin-4 (3*H*)-one (4c) [23]**.** m.p.: 285.2 °C; ^1H NMR (DMSO-d_6, 400 MHz): δ 2.24 (dd, *J* = 15.0, 7.6 Hz, 1H, CH_2), 2.47 (dd, *J* = 15.0, 6.0 Hz, 1H, CH_2), 4.61 (t, *J* = 6.0 Hz, 1H, CH), 6.51 (s, 2H, NH_2), 7.23 (d, *J* = 7.6 Hz, 2H, 2CH arom.), 7.53 (d, *J* = 7.6 Hz, 2H, 2CH arom.), 7.63 (s, 1H, NH); MS (70 eV): *m/z* (%) 269 (15) (M^++2), 267 (15) (M^+), 240 (8), 224 (10), 196 (4), 188 (25), 182 (20); IR (KBr): ν 3358, 3005, 1901, 1616, 1556, 1487, 1375 cm^{-1}; Anal. Calcd. for $C_{10}H_{10}BrN_3O$: C, 44.80; H, 3.76; N, 15.67%. Found: C, 44.49; H, 3.78; N, 15.13%.

2-Amino-6-(4-methylphenyl)-5,6-dihydropyrimidin-4 (3*H*)-one (4d). m.p.: 264.1 °C; ^1H NMR (DMSO-d_6, 80 MHz): δ 2.16 (s, 3H, CH_3), 2.36 (m, 2H, CH_2), 4.56 (t, *J* = 7.2 Hz, 1H, CH), 6.59 (s, 2H, NH_2), 7.15 (s, 4H, 4CH arom.), 7.60 (s, 1H, NH); MS (70 eV): *m/z* (%) 203 (70) (M^+), 188 (25), 174 (30), 160 (80), 118 (100); IR (KBr): ν 3354, 3022, 2860, 1639, 1589, 1406, 1375 cm^{-1}; Anal. Calcd. for $C_{11}H_{13}N_3O$: C, 65.01; H, 6.45; N, 20.68. Found: C, 64.13; H, 6.36; N, 20.29%.

2-Amino-6-(4-methoxyphenyl)-5,6-dihydropyrimidin-4 (3*H*)-one (4e) [23]. m.p.: 275.8 °C; ^1H NMR (DMSO-d_6, 500 MHz): δ 2.25 (dd, *J* = 15.0, 9.0 Hz, 1H, CH_2), 2.40 (dd, *J* = 15.5, 6.0 Hz, 1H, CH_2), 3.70 (s, 3H, OCH_3), 4.53 (t, *J* = 6.5 Hz, 1H, CH), 6.42 (s, 2H, NH_2), 6.88 (d, *J* = 8.2 Hz, 2H, 2CH arom.), 7.20 (d, *J* = 8.2 Hz, 2H, 2CH arom.), 7.49 (s, 1H, NH); MS (70 eV): *m/z* (%) 219 (75) (M^+), 218 (54), 204 (10), 190 (30), 176 (50), 134 (100); IR (KBr): ν 3356, 3032, 2829, 1637, 1560, 1377 cm^{-1}; Anal. Calcd. for $C_{11}H_{13}N_3O_2$: C, 60.26; H, 5.98; N, 19.17%. Found: C, 59.65; H, 6.05; N, 18.71%.

2-Amino-6-(3-methoxyphenyl)-5,6-dihydropyrimidin-4 (3*H*)-one (4f). m.p.: 262.0 °C; ^1H NMR (DMSO-d_6, 80 MHz): δ 2.07 (m, 1H, CH_2), 2.38 (m, 1H, CH_2), 3.71 (s, 3H, OCH_3), 4.58 (t, *J* = 6.5 Hz, 1H, CH), 6.42 (s, 2H, NH_2), 6.88 (m, 3H, 3CH arom.), 7.20 (m, 2H, 1CH arom., and 1NH); MS (70 eV): *m/z* (%) 219 (80) (M^+), 218 (54), 204 (10), 190 (30), 176 (50), 134 (100); IR (KBr): ν 3367, 3232, 3012, 2835, 1651, 1589, 1550, 1498 cm^{-1}; Anal. Calcd. for $C_{11}H_{13}N_3O_2$: C, 60.26; H, 5.98; N, 19.17%: Found: C, 59.71; H, 6.22; N, 19.25%.

2-Amino-6-(2-methoxyphenyl)-5,6-dihydropyrimidin-4(3*H*)-one (4g) [23]**.** m.p.: 251.5 °C; ^1H NMR (DMSO-d_6, 80 MHz): δ 2.34 (m, 1H, CH_2), 2.60 (m, 1H, CH_2), 3.79 (s, 3H, OCH_3), 4.85 (t, *J* = 6.5 Hz, 1H, CH), 6.52 (s, 2H, NH_2), 7.00-7.27 (m, 5H, 4CH arom. and 1NH); MS (70 eV): *m/z* (%) 219 (100) (M^+), 218 (100), 204 (85), 190 (35), 176 (90), 134 (75); IR (KBr): ν 3336, 3000, 2835, 1664, 1627, 1598, 1570 cm^{-1}; Anal. Calcd. for $C_{11}H_{13}N_3O_2$: C, 60.26; H, 5.98; N, 19.17%. Found: C, 59.28; H, 6.14; N, 19.24%.

2-Amino-6-(biphenyl-4-yl)-5,6-dihydropyrimidin-4 (3*H*)-one (4h). m.p.: 314.2 °C; ^1H NMR (DMSO-d_6, 80 MHz): δ 2.40 (m, 2H, CH_2), 4.68 (t, *J* = 8.0 Hz, 1H, CH), 6.48 (s, 2H, NH_2), 7.33-7.70 (m, 10H, 9CH arom., and 1NH); MS (70 eV): *m/z* (%) 265 (100) (M^+), 236 (35), 222 (27), 180 (75), 152 (65); IR (KBr): ν 3340, 3259, 3026, 1641, 1591, 1546, 1494, 1398 cm^{-1}; Anal. Calcd. for $C_{16}H_{15}N_3O$: C, 72.43; H, 5.70; N, 15.84%. Found: C, 71.39; H, 5.66; N, 15.80%.

2-Amino-6-(naphthalen-1-yl)-5,6-dihydropyrimidin-4 (3*H*)-one (4i). m.p.: 203.4 °C; ^1H NMR (DMSO-d_6, 80 MHz): δ 2.25 (m, 1H, CH_2), 2.60 (m, 1H, CH_2), 5.50 (t, *J* = 6.5 Hz, 1H, CH), 6.50 (s, 2H, NH_2), 7.42-8.20 (m, 8H, 7CH arom., and 1NH); MS (70 eV): *m/z* (%) 239 (20) (M^+), 210 (25), 196 (40), 181 (100), 178 (25), 126 (40); IR (KBr): ν 3410, 3325, 3061, 2968, 2910, 1653, 1602, 1571, 1496, 1357 cm^{-1}; Anal. Calcd. for $C_{14}H_{13}N_3O$: C, 70.28; H, 5.48; N, 17.56. Found: C, 71.29; H, 5.56; N, 17.01%.

2-Amino-6-(4-(dimethylamino)phenyl)-5,6-dihydro-pyrimidin-4(3*H*)-one (4j) [23]**.** m.p.: 266.2 °C; ^1H NMR (DMSO-d_6, 80 MHz): δ 2.40 (m, 2H, CH_2), 2.85 (s, 6H, $2CH_3$), 4.48 (t, *J* = 8.0 Hz, 1H, CH), 6.42 (s, 2H, NH_2), 6.67 (d, *J* = 8.8 Hz, 2H, 2CH arom.), 7.11 (d, *J* = 8.8 Hz, 2H, 2CH arom.), 7.47 (s, 1H, NH); MS (70 eV): *m/z* (%) 232 (30) (M^+), 203 (15), 189 (20), 147 (100), 131 (35); IR (KBr): ν 3425, 3034, 2968, 2885, 1654, 1620, 1560, 1521, 1402, 1369 cm^{-1}; Anal. Calcd. for $C_{12}H_{16}N_4O$: C, 62.05; H, 6.94; N, 24.12%. Found: C, 61.16; H, 6.91; N, 24.17%.

2-Amino-6-heptyl-5,6-dihydropyrimidin-4(3*H*)-one (4k). m.p.: 249.2 °C; ^1H NMR (DMSO-d_6, 500 MHz): δ 0.84 (t, *J* = 6.8, 3H, CH_3), 1.24 (m, 10H, $5CH_2$), 1.37 (m, 2H, CH_2),

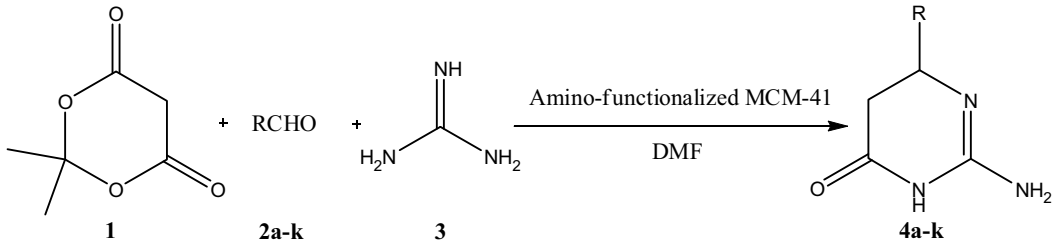
1.96 (dd, *J* = 15.4, 9.0 Hz, 1H, CH_2), 2.26 (dd, *J* = 15.4, 5.5 Hz, 1H, CH_2), 3.47 (m, 1H, CH), 6.22 (s, 2H, NH_2), 7.23 (s, H, NH), MS (70 eV): *m/z* (%) 211 (10) (M^+), 182 (5), 168 (10), 154 (5), 140 (5), 126 (20), 112 (100); IR (KBr): ν 3317, 3012, 2920, 2852, 1656, 1500, 1379, 1325, 1215, 1170 cm^{-1}; Anal. Calcd. for $C_{11}H_{21}N_3O$: C, 62.52; H, 10.02; N, 19.89%. Found: C, 61.76; H, 9.91; N, 19.96%.

RESULTS AND DISCUSSION

A three-component reaction of Meldrum's acid **1**, aldehydes **2a-k** and guanidinium carbonate **3** afforded the corresponding 2-amino-5,6-dihydropyrimidin-4(3*H*)-one building blocks (Scheme 1).

The use of Meldrum's acid for the synthesis of fused dihydropyridine-2-ones [36] and 2-NH_2-DHPM building blocks was previously described to take place in moderate yields [23]. For example, **4e** was obtained in 55 and 38% yield by conventional and microwave protocols, respectively [23].

At earlier stages of our work, we investigated the influence of solvent and temperature on the reaction yield of an equimolar quantity of 4-methoxybenzaldehyde **2e,** Meldrum's acid **1** and guanidinium carbonate **3**, as indicated in Table 1, by examining different catalysts. KF/Al_2O_3, K_2CO_3 and



1 **2a-k** **3** **4a-k**

Scheme 1. Three-component synthesis of 2-amino-5,6-dihydropyrimidin-4(3*H*)-one.

Table 1. Effects of the Catalyst Type, Solvent and Temperature on the Formation of 2-Amino-6-aryl-5,6-dihydropyrimidin-4(3*H*)-one

Entry	Catalyst	Solvent	Temperature (°C)	Product	Time (h)	Yield (%)[a]
1	KF/Al_2O_3	EtOH	78	**4e**	7	55
2	KF/Al_2O_3	DMF	120	**4e**	5	65
3	K_2CO_3	DMF	120	**4e**	3.5	65
4	MCM-41-NH_2	DMF	120	**4e**	1	72
5	MCM-41-NH_2	DMF	70	**4e**	1	50
6	MCM-41-NH_2	DMF	100	**4e**	1	57
7	MCM-41-NH_2[b]	DMF	120	**4e**	1	72
8	MCM-41-NH_2	DMF	120	**4c**	1	86
9	MCM-41-NH_2[b]	DMF	120	**4c**	1	86
10	MCM-41-NH_2[c]	DMF	120	**4c**	1	86
11	MCM-41-NH_2[d]	DMF	120	**4c**	1	85

[a]Isolated yield. Reaction conditions: Meldrum's acid (1 mmol), aldehyde (1 mmol), guanidinium carbonate (1 mmol), KF/Al_2O_3 40% by weight (1.2 g, 0.8 mmol) or K_2CO_3 (1 mmol) or MCM-41-NH_2 (10 mol%). [b]100 mol% of MCM-41-NH_2 was used. [c]Second run. [d]Third run.

MCM-41-NH_2 were used to explore the reaction scope. After the time indicated in Table 1, the crude products were poured into crushed ice while stirring, filtered through a sintered funnel and washed by ethylacetate-hexane (1:3). For further purification, the resulting products were recrystallized from ethanol. The results are summarized in Table 1.

2-Amino-6-(4-methoxyphenyl)-5,6-dihydropyrimidin-4 (3*H*)-one **4e** was obtained in 55% yield using KF/Al_2O_3 as a catalyst in refluxing ethanol for 7 h (entry 1, Table 1). When ethanol was replaced by DMF (0.5 ml) and the temperature was raised to 120 °C, the yield increased to 65% and the reaction time decreased to 5 h (entry 2, Table 1). The obtained results indicate that **4e** can be synthesized in good yields using KF/Al_2O_3 or K_2CO_3 as catalysts in DMF at 120 °C, but the reaction time was 5 and 3.5 h, respectively (entries 2 and 3, Table 1). A rise in yield (72%) and a drop in the reaction time (1 h) was observed when the amino-functionalized MCM-41 was used as catalyst in DMF at 120 °C (entry 4, Table 1). The results suggest that amino-functionalized MCM-41 in DMF can be used as an alternative catalyst. In order to find the optimum temperature for the formation of the product **4e**, the occurrence of the reaction of Meldrum's acid **1**, 4-methoxybenzaldehyde **2e** and guanidinium carbonate **3** was monitored at three different temperatures (70, 100 and 120 °C) (entries 4-6, Table 1). The results clearly indicate that the yields are better at 120 °C (entry 4, Table 1). Finding the suitable temperature and solvent, the influence of the catalyst concentration on the reaction yield was investigated. 2-NH_2-DHPM Building blocks **4c** and **4e** were obtained in 86 and 72% yield with a catalyst concentration of 100 and 10 mol%, respectively (entries 4, 7-9 Table 1). Increasing the catalyst concentration did not show any significant effect on the yield, indicating that 10 mol% of MCM-41-NH_2 is sufficient for this reaction. This catalyst also showed excellent reusability (entries 10 and 11, Table 1).

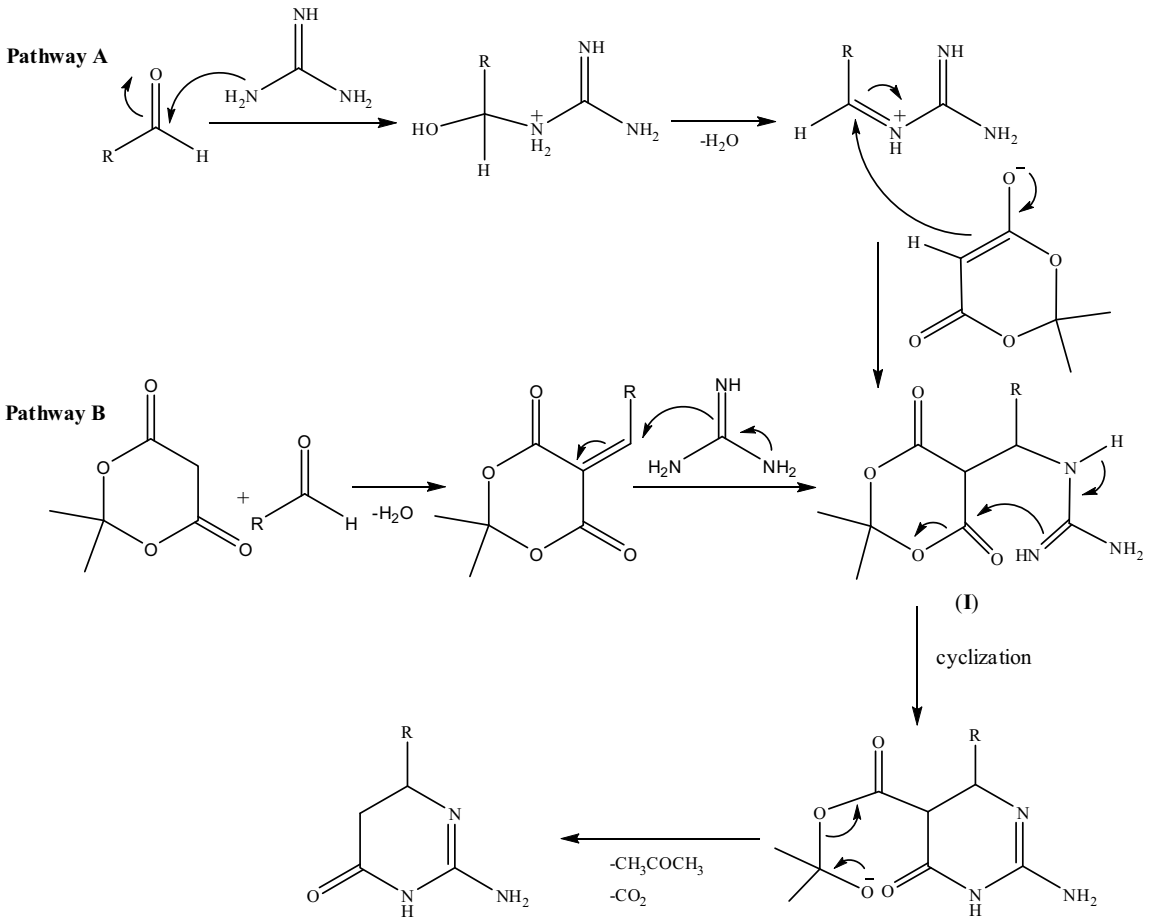
The optimal conditions were reached when the reaction of an equimolar solution of the Meldrum's acid **1**, aldehydes **2a-k** and guanidinium carbonate **3** in the presence of 10 mol% of the catalyst MCM-41-NH_2 took place at 120 °C in 0.5 ml of DMF as the solvent. Under these conditions, various aldehydes (**2a-k**) reacted smoothly to give the corresponding 2-amino-5,6-dihydropyrimidin-4(3*H*)-one derivatives (**4a-k**) after 1 h in 60-90% yield. The results in Table 2 show that the MCM-41-NH_2 base catalyst can be used in the synthesis of 2-NH_2-DHPM building blocks with various aldehydes in high yields and short reaction times.

Finally, a reasonable and appropriate mechanism is suggested for this reaction, as outlined in Scheme 2. The

Table 2. Three-component Synthesis of 2-NH_2-DHPM Building Blocks **4a-k** from Meldrum's Acid **1**, Aldehydes **2** and Guanidinium Carbonate **3** in the Presence of 10 mol% of Amino-functionalized MCM-41 Catalyst

Entry	R	Product	Time (h)	Yield (%)[a]	m.p. (°C) Found	Reported
1	C_6H_5	**4a**	1	72	257.3	257 [23]
2	*p*-Cl-C_6H_4	**4b**	1	88	280.3	-
3	*p*-Br-C_6H_4	**4c**	1	86	285.2	275-276 [23]
4	*p*-CH_3-C_6H_4	**4d**	1	80	264.1	-
5	*p*-OCH_3-C_6H_4	**4e**	1	72	275.8	278 [23]
6	*m*-OCH_3-C_6H_4	**4f**	1	60	262.0	-
7	*o*-OCH_3-C_6H_4	**4g**	1	74	251.5	206 [23]
8	*p*-C_6H_5-C_6H_4	**4h**	1	78	314.2	-
9	1-Naphthyl	**4i**	1	90	203.4	-
10	*p*-$(CH_3)_2N$-C_6H_4	**4j**	1	65	266.2	261-262 [23]
11	C_7H_{15}	**4k**	1	80	249.2	-

[a]Isolated yield.



Scheme 2. The proposed reaction mechanism for the synthesis of 2-NH_2-DHPM building blocks **4**

reaction may proceed through the arylideneguanidinium intermediate (formed in situ by the reaction of an aldehyde with guanidine), and the subsequent addition of the enolate to the arylideneguanidinium to generate the intermediate (**I**) (Pathway A, Scheme 2). However, as was previously reported [23], the reaction can proceed via pathway B to lead to the formation of arylidene Meldrum's acid, which then undergoes Michael addition of guanidine molecule to generate the intermediate (**I**) (Pathway B, Scheme 2). The cyclization of the intermediate (**I**) followed by its ring closure, elimination of acetone and decarboxylation of the intermediate acid affords the corresponding 2-NH_2-DHPM building blocks (Scheme 2).

In summary, we have developed a convenient and efficient alternative process for the synthesis of 2-amino-5,6-dihydropyrimidin-4(3*H*)-one derivatives through a three-component coupling of Meldrum's acid, aldehyde and guanidinium carbonate using amino-functionalized MCM-41 as a basic catalyst. Amino-functionalized MCM-41 is a highly promising environmentally-friendly base catalyst for the one-pot synthesis of 2-amino-5,6-dihydropyrimidin-4(3*H*)-one derivatives with high yields and short reaction times. The technique offers simplicity of operation and easy work-up procedure.

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