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HIGHLY EFFICIENT ONE-POT SYNTHESIS OF PYRANO[2,3-*d*]PYRIMIDINES: ANOTHER APPLICATION OF A KEPLERATE TYPE GIANT NANOPOROUS ISOPOLYOXOMOLYBDATE AS A REUSABLE CATALYST

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Abstract: A novel and efficient improved procedure for the synthesis of pyrano[2,3-*d*]pyrimidines based on the one-pot three-component cyclocondensation of 1,3-dimethylbarbituric acid, aromatic aldehydes, and malononitrile is developed using $(NH_4)_{42}[Mo^{VI}_{72}Mo^{V}_{60}O_{372}(CH_3COO)_{30}(H_2O)_{72}]$, a Keplerate-type giant-ball nanoporous isopolyoxomolybdate denoted as ($\{Mo_{132}\}$), as catalyst under solvent-free conditions. This protocol demonstrates several notable advantages, including operational simplicity, high yields, short reaction times, and environmentally friendly conditions. Furthermore, the catalyst could be recovered conveniently and reused efficiently such that a considerable catalytic activity still could be achieved after fifth run.

Keywords: Giant nanoporous isopolyoxomolybdate; Keplerate; $\{Mo_{132}\}$; Pyrano[2,3-*d*]pyrimidines; Solvent-free conditions.

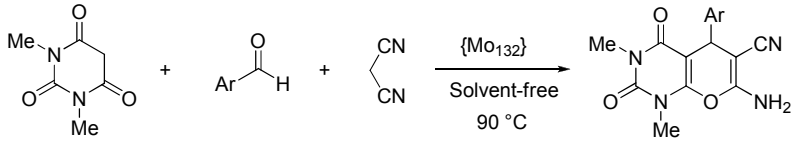
Introduction

Pyrano[2,3-*d*]pyrimidines, having two active pharmacophores pyran and pyrimidine, have received considerable attention over the past years due to their wide range of the diverse pharmacological properties such as antitumor, cardiotonic, hepatoprotective, antihypertensive and antibronchitic activity[i-v]. Also, a number of these compounds have been considered as antimicrobial[vi], antifungal[vii], anti-biofilm[vii], anti-diabetic[viii] as well as α-amylase and α-glucosidase inhibitors[viii]. These compounds are generally synthesized *via* the one-pot three-component reaction of barbituric acid or 1,3-dimethylbarbituric acid, an aldehyde, and malononitrile or ethyl cyanoacetate in the presence of a catalyst such as $ZnFe_2O_4$ nanoparticles[ix], KF[x], $H_3PW_{12}O_{40}$[xi],1,8-diazabicyclo[5.4.0]undec-7-ene (DBU)[xii], Mn/ZrO_2[xiii], MgO[xiv], and urea-based ionic liquid stabilized on silica-coated Fe_3O_4 magnetic nanoparticles[xv]. Synthesis of these compounds using microwave irradiation[xvi] and electrocatalytic procedure in the presence of sodium bromide as electrolyte[xvii] have been also reported. Although these methods are relatively satisfactory, many of them employ

considerable amounts of organic solvents, which are not environmentally friendly. Some of them also suffer from drawbacks such as harsh reaction conditions, long reaction times, unsatisfactory yields, tedious work-up, and environmental pollution. Therefore, a need still exists for developing more efficient procedures using new reusable catalysts with high activity, which allow the high yield synthesis of pyrano[2,3-*d*]pyrimidines in short reaction times under simple experimental set up and eco-friendly conditions.

Polyoxometalates (POMs) are a subset of metal oxide clusters of early transition metals in their high oxidation state, e.g., W, Mo, V, Nb, Ta, and so on, bridged by oxygen atoms[xviii]. They are known to have a variety of sizes and structures, and have been studied due to their attractive electronic and molecular properties such as extremely versatile redox potentials, acidities, polarities, and solubilities, which give rise to a variety of applications in catalysis, biomedicine, magnetism, and nano and materials science[xix-xxii]. A number of very large POM anions with a variety of applications in fundamental and applied science[xxiii,xxiv] have been synthesized and structurally characterized[xxv-xxviii]. Müller and co-workers, for the first time, reported the famous remarkable giant nanosized porous Keplerate-type POM, $(NH_4)_{42}[Mo^{VI}_{72}Mo^{V}_{60}O_{372}(CH_3COO)_{30}(H_2O)_{72}]$, which denoted as $\{Mo_{132}\}$[xxix]. This ball shaped POM has been characterized using the transmission electron microscopy (TEM) image by Polarz et al[xxx]. The TEM picture clearly shows a periodic structure with an average size of approximately 3 nm in diameter. This experimentally obtained diameter fits nicely with the theoretical value for the inner diameter of this ball-shaped POM that was calculated to be 2.9 nm[xxix, xxxi].

In this view, recently, a series of organic transformations has been reported by our group using $\{Mo_{132}\}$ as catalyst. This new reusable catalyst performed well and showed a high level of catalytic activity in the synthesis of 1,2,4,5-tetrasubstituted imidazoles[xxxii], 1,8-dioxo-octahydroxanthenes and 1,8-dioxodecahydroacridines[xxxiii], biscoumarins[xxxiv], 1,4-dihydropyridines[xxxv], polyhydroquinolines[xxxvi], and dihydropyrano[3,2-*c*]chromenes[xxxvii]. Due to our interest in the synthesis of heterocyclic compounds[xxxviii-xlii], and as part of our research on the development of environmentally friendly methods for the synthesis of organic compounds using reusable catalysts[xliii-xlix], we report here another successful application of $\{Mo_{132}\}$ as highly efficient and reusable catalyst for the synthesis of pyrano[2,3-*d*]pyrimidines by one-pot three-component reaction of 1,3-dimethylbarbituric acid, aromatic aldehydes, and malononitrile under solvent-free conditions (Scheme 1).



Scheme 1. Synthesis of pyrano[2,3-*d*]pyrimidines catalyzed by $\{Mo_{132}\}$

Experimental

All chemicals were available commercially and used without additional purification. Melting points were recorded on a Stuart SMP3 melting point apparatus. The IR spectra were obtained using a Tensor 27 Bruker spectrophotometer as KBr disks. The ^{1}H NMR spectra were recorded with a Bruker 300 FT spectrometer.

Synthesis of the Keplerate $\{Mo_{132}\}$. $N_2H_4.H_2SO_4$ (0.8 g, 6.1 mmol) was added to a solution of $(NH_4)_6Mo_7O_{24}.4H_2O$ (5.6 g, 4.5 mmol) and CH_3COONH_4 (12.5 g, 162.2 mmol) in H_2O (250 ml). The solution was then stirred for 10 min (color change to bluegreen) and 50% CH_3COOH (83 ml) was subsequently added. The reaction solution, now green, was stored in an open 500-ml Erlenmeyer flask at 20 °C without further stirring (slow color change to dark

brown). After 4 d the precipitated red-brown crystals were filtered off, washed with absolute ethanol and diethyl ether, respectively, and finally dried in air[xxix].

General procedure for the synthesis of pyrano[2,3-*d*]pyrimidines 4a-j catalyzed by {Mo_{132}}. A mixture of 1,3-dimethylbarbituric acid **1** (1 mmol), an aromatic aldehyde **2a-j** (1 mmol), malononitrile **3** (1 mmol), and {Mo_{132}} (0.1 g) was heated in the oil bath at 90 ºC for 15-40 min. During the procedure, the reaction was monitored by TLC. Upon completion of the transformation, the reaction mixture was cooled to room temperature and hot ethanol was added. This resulted in the precipitation of the catalyst, which was collected by filtration. The catalyst was washed with a small portion of hot ethanol. The combined filtrate was concentrated by half and allowed to stand at room temperature. The precipitated solid was collected by filtration, and recrystallized from ethanol to give compounds **4a-j** in high yields. All the products were characterized according to comparison of their melting points with those of authentic samples. The structures of some products were also confirmed by their IR and ^{1}H NMR spectral data.

7-Amino-1,3-dimethyl-2,4-dioxo-5-phenyl-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4a**): FT-IR (KBr disc): υ 3384, 3310, 3195, 3031, 2954, 2885, 2198, 1703, 1646, 1496, 1390, 1233, 1193, 1041, 506 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 3.09 (s, 3H, NCH_3), 3.37 (s, 3H, NCH_3), 4.33 (s, 1H, CH), 7.18-7.32 (m, 5H, arom-H), 7.34 (s br., 2H, NH_2).

7-Amino-5-(2-chlorophenyl)-1,3-dimethyl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4b**): FT-IR (KBr disc): υ 3395, 3314, 3193, 2959, 2195, 1710, 1488, 1390, 1231, 1190, 1039, 751, 501 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 3.11 (s, 3H, NCH_3), 3.42 (s, 3H, NCH_3), 4.92 (s, 1H, CH), 7.22-7.33 (m, 2H, arom-H), 7.34-7.45 (m, 4H, arom-H and NH_2).

7-Amino-5-(3-chlorophenyl)-1,3-dimethyl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4c**): FT-IR (KBr disc): υ 3395, 3310, 3197, 2962, 2197, 1689, 1640, 1494, 1387, 1231, 1186, 1036, 749, 498 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 3.10 (s, 3H, NCH_3), 3.36 (s, 3H, NCH_3), 4.38 (s, 1H, CH), 7.22-7.36 (m, 4H, arom-H), 7.41 (s br., 2H, NH_2).

7-Amino-5-(4-chlorophenyl)-1,3-dimethyl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4d**): FT-IR (KBr disc): υ 3428, 3301, 2191, 1704, 1686, 1644, 1489, 1384, 1232, 1188, 1034, 772 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 3.09 (s, 3H, NCH_3), 3.51 (s, 3H, NCH_3), 4.36 (s, 1H, CH), 7.29 (d, 2H, J = 8.7 Hz, arom-H), 7.36 (d, 2H, J = 8.7 Hz, arom-H), 7.39 (s br., 2H, NH_2).

7-Amino-1,3-dimethyl-5-(2-nitrophenyl)-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4f**): FT-IR (KBr disc): υ 3380, 3308, 3186, 2960, 2197, 1689, 1525, 1392, 1310, 1234, 1193, 1041, 969, 786, 570, 500 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 3.03 (s, 3H, NCH_3), 3.36 (s, 3H, NCH_3), 5.15 (s, 1H, CH), 7.46 (td, 1H, J = 8.2, 1.5 Hz, arom-H), 7.50-7.57 (m, 3H, arom-H and NH_2), 7.66 (td, 1H, J = 7.5, 1.2 Hz, arom-H), 7.86 (dd, 1H, J = 8.1, 1.5 Hz, arom-H).

7-Amino-1,3-dimethyl-5-(4-nitrophenyl)-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4g**): FT-IR (KBr disc): υ 3390, 3306, 3195, 3075, 2958, 2875, 2204, 1634, 1514, 1381, 1230, 1184, 1034, 867, 822, 716, 571 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 3.10 (s, 3H, NCH_3), 3.40 (s, 3H, NCH_3), 4.56 (s, 1H, CH), 7.52 (s br., 2H, NH_2), 7.60 (d, 2H, J = 8.7 Hz, arom-H), 8.19 (d, 2H, J = 8.7 Hz, arom-H),.

7-Amino-5-(3-hydroxyphenyl)-1,3-dimethyl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (**4h**): FT-IR (KBr disc): υ 3417, 3302, 3189, 2949, 2833, 2193, 1691, 1508, 1386, 1227, 1184, 1036, 970, 847, 754, 568 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-

d_6): δ 3.11 (s, 3H, NCH_3), 3.36 (s, 3H, NCH_3), 4.23 (s, 1H, CH), 6.58-6.70 (m, 3H, arom-H), 7.08 (t, 1H, J = 7.5 Hz, arom-H), 7.33 (s br., 2H, NH_2), 9.30 (br., 1H, OH).

Results and discussion

We commenced our experiments by examining the model reaction of 1,3-dimethylbarbituric acid **1** (1 mmol), 4-chlorobenzaldehyde **2d** (1 mmol), and malononitrile **3** (1 mmol), for the synthesis of compound **4d** in different sets of reaction conditions. The results are summarized in Table 1. First, an uncatalyzed reaction was tested under solvent-free condition but no significant yield was obtained (entry 1). Pleasingly, we discovered that the reaction was efficiently catalyzed by $\{Mo_{132}\}$ under solvent-free conditions at a relatively elevated temperature, providing a high yield of the product **4d**. The best result was conducted at 90 ºC in the presence of 0.1 g of $\{Mo_{132}\}$ (entry 12). Next, the reaction was carried out in different solvents including H_2O, MeOH, EtOH, EtOH/H_2O, and CH_3CN. As shown, the yields of the reaction under solvent-free conditions were greater and the reaction times were generally shorter than the conventional methods.

Table 1
Optimization of reaction conditions for synthesis of compound **4d** catalyzed by $\{Mo_{132}\}$[a]

Entry	Catalyst (g)	Solvent	T (°C)	Time (min)	Isolated Yield (%)
1	-----	-----	90	90	Trace
2	0.04	-----	70	120	48
3	0.04	-----	90	90	64
4	0.04	-----	110	80	63
5	0.06	-----	70	60	75
6	0.06	-----	90	50	79
7	0.06	-----	110	50	81
8	0.08	-----	70	45	80
9	0.08	-----	90	30	84
10	0.08	-----	110	30	84
11	0.10	-----	70	30	89
12	0. 10	-----	90	20	95
13	0. 10	-----	110	20	93
14	0. 12	-----	70	25	87
15	0. 12	-----	90	25	92
16	0. 12	-----	110	30	92
17	0.10	H_2O	Reflux	75	50
18	0.10	MeOH	Reflux	105	58
19	0.10	EtOH	Reflux	120	61
20	0.10	EtOH/H_2O	Reflux	130	60
21	0.10	CH_3CN	Reflux	120	55

[a]Reaction conditions: 1,3-dimethylbarbituric acid **1** (1 mmol), 4-chlorobenzaldehyde **2d** (1 mmol), and malononitrile (1 mmol).

In order to evaluate the general character of this model reaction, we included a range of other aromatic aldehydes in the reaction of 1,3-dimethylbarbituric acid and malononitrile under the optimized reaction conditions. The results are summarized in Table 2. As shown, in all cases, the expected products were obtained in high yields over short reaction times. Under the same conditions however, no reaction occurred when aliphatic aldehydes were used.

In view of the green chemistry, the recycling performance of $\{Mo_{132}\}$ was also investigated in the model reaction. After completion of the reaction, the catalyst was recovered according to the procedure described in the Experimental section. The separated catalyst was washed with hot ethanol and then dried at 50 ºC under vacuum for 1 h before being reused in a similar reaction. We found that the catalyst could be used at least five times without significant reduction in its activity (Fig. 1).

Table 2
Synthesis of pyrano[2,3-*d*]pyrimidines **4a-j** using $\{Mo_{132}\}$[a]

Entry	R	Product	Time (min)	Isolated Yield (%)	m.p. (ºC)	
					Found	Reported
1	C_6H_5	**4a**	35	89	219-221	218-220 [ix]
2	2-ClC_6H_4	**4b**	40	90	235-236	236-238 [x]
3	3-ClC_6H_4	**4c**	35	92	251-253	247-248 [ix]
4	4-ClC_6H_4	**4d**	20	95	234-235	239-241 [x]
5	4-FC_6H_4	**4e**	25	91	236-238	238-240 [xv]
6	2-$O_2NC_6H_4$	**4f**	35	88	203-205	202-204 [xi]
7	4-$O_2NC_6H_4$	**4g**	15	93	215-216	217-219 [ix]
8	3-HOC_6H_4	**4h**	40	90	201-203	196-198 [xiii]
9	4-MeC_6H_4	**4i**	30	90	226-228	229-230 [xv]
10	4-$MeOC_6H_4$	**4j**	35	91	218-219	222-224 [xi]

[a]Reaction conditions: 1,3-dimethylbarbituric acid **1** (1 mmol), an aromatic aldehyde **2a-j** (1 mmol), malononitrile **3** (1 mmol), $\{Mo_{132}\}$ (0.10 g), 90 ºC, solvent-free.

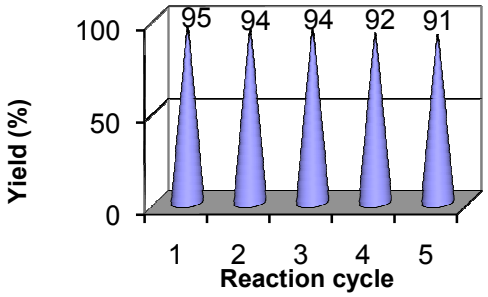
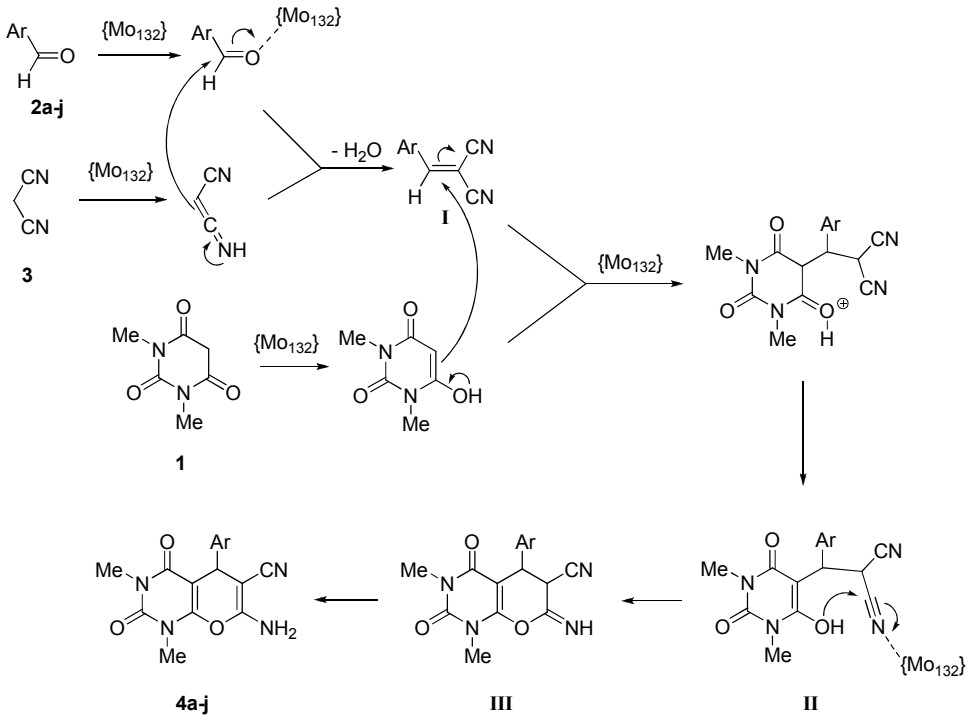


Fig. 1. Effect of recycling on the catalytic performance of $\{Mo_{132}\}$ in the synthesis of **4d**.

A mechanistic rationalization for this reaction is provided in Scheme 2. The $\{Mo_{132}\}$ catalyst has several accessible Mo sites and NH_4 groups, which could act as Lewis acid and Brönsted acid centers, respectively, and therefore promote the reaction. The catalyst would play a significant role in increasing the electrophilic character of the electrophiles in the reaction. As shown, the dicyano olefin **I** is readily formed in situ by Knoevenagel condensation of aldehydes **2a-j** and **3**. 1,3-Dimethylbarbituric acid **1**, in enolic form, easily reacts with olefin **I** followed by cyclization to produce intermediates **II** and **III**, respectively. Finally, the products **4a-j** are obtained from the latter intermediate after tautomerization. Under these conditions, however, attempts to isolate the proposed intermediates failed even after careful monitoring of the reactions.

Conclusion

In conclusion, we have developed a convenient one-pot three component cyclocondensation reaction between 1,3-dimethylbarbituric acid, aromatic aldehydes, and malononitrile for the preparation of pyrano[2,3-*d*]pyrimidines using $\{Mo_{132}\}$, a Keplerate-type giant-ball nanoporous isopolyoxomolybdate, as catalyst. Some attractive features of this protocol are high yields, short reaction times, solvent-free conditions, easy work-up, high catalytic activity and recyclability and reusability of the catalyst. The catalyst can be used at least five times without substantial reduction in its catalytic activity.



Scheme 2. Plausible mechanism for the formation of pyrano[2,3-*d*]pyrimidines in the presence of {Mo_{132}} as catalyst

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