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Modified Biginelli condensation by molybdate sulfuric acid as a powerful and reusable catalyst

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ABSTRACT

Molybdate sulfuric acid (MSA) has been prepared and used as catalyst for the Biginelli synthesis of some quinazolinones/thiones under solvent-free conditions. The catalyst loading is low and it shows recyclability. This method has advantages such as avoidance of the organic solvents, high yield of pure products, short reaction times and operational simplicity.

***Keywords**: Biginelli, Molybdate sulfuric acid, Quinazolinones/thiones, Solvent-free.*

1. Introduction

Among various acids, solid acids are well known and they are widely used in both industry and academic researches as efficient catalysts for chemical reactions instead of traditional acids [1,2]. More recently, pressure from environmentalists has led to a search for more environmentally friendly forms of catalysis [3,4]. In the last two decades, multicomponent reactions (MCRs) have also drawn special attention owing to the advent of high-throughput screening techniques that enabled rapid identification of potential new medicines among large collections of organic compounds [5,6]. Moreover, the chemistry of quinazoline system has received an increasing interest because of its biological significance. They are a class of drugs which function as hypnotic/sedatives. For example, the Afloqualone, Cloroqualone, and Diproqualone have been also used in the treatment of cancer [7].

The most general method for the preparation of octahydroquinazolinones/thiones involves the one-pot Biginelli reaction of cyclic 1,3-dione, aromatic aldehydes and urea/thiourea in the presence of a Lewis or mineral acid. Although, synthesis of octahydro-quinazolinones/thiones via the Biginelli reaction using several reagents have been previously reported, most of the present procedures have disadvantages such as long reaction time, use of strongly acidic condition or organic solvents, unsatisfactory product yield, and producing side products [8-13].

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2. Experimental

2.1. General

The chemicals were purchased from Merck, Fluka and Aldrich chemical companies. The reactions were monitored by TLC (silica-gel 60 F_{254}, hexane: AcOEt). IR spectra were recorded on a FT-IR Shimadzu-470 spectrometer and the ^{1}H NMR spectra was obtained on a Bruker-Instrument DPX-400 Avance 2 model.

*2.2. General procedure for the preparation of MSA (**1**)*

Firstly 25 mL of dry n-hexane was taken in a 100 mL round bottom flask, equipped with ice bath and overhead stirrer, and 4.118 g (20 mmol) of anhydrous sodium molybdate was added to the flask, then 0.266 mL (40 mmol) of chlorosulfonic acid was added dropwise to the flask for 30 min. This solution was stirring for 1.5 h. Afterwards the reaction mixture was gradually poured into 25 mL of chilled distilled water with agitation. The bluish solid which separated out was filtered. Then catalyst was washed with distilled water for five times till the filtrate showed negative test for chloride ion, and was dried at 120 °C for 5 h. The catalyst was obtained in 90% yield (5.8 g) as a bluish solid, which decomposed at 354 °C.

2.3. General procedure for the synthesis of octahydroquinazolinones/thiones

A mixture of urea/thiourea (1.2 mmol), aldehyde (1 mmol), cyclic 1,3-dione (1 mmol), and MSA (0.32 g, 0.1 mmol) was stirred and heated at 100 °C in a preheated oil bath for an appropriate time. After completion of the reaction as indicated by TLC, the reaction mixture was cooled to room temperature and dichloromethane (10 mL) was added. Then the resulting mixture was stirred for 3 min. The catalyst was separated by filtration. The solvent was removed by distillation, then washed with cold water and recrystallized from methanol to afford the pure product **5.** The recovered catalyst from the model reaction was regenerated by washing with EtOAc and drying at 120 °C for 1 h.

Selected spectral data

*Compound **5b***:

IR (KBr): $\bar{\nu}$ = 3285 (s), 3200 (s), 1640 (s), 1605 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 0.91 (3H, s, CH_3), 1.04 (3H, s, CH_3), 1.97-2.01 (1H, d, J = 16 Hz, CH_2), 2.17-2.19 (1H, d, J = 8 Hz, CH_2), 2.30-2.34 (1H, d, J = 16 Hz, CH_2), 2.47-2.51 (1H, d, J = 16 Hz, CH_2), 3.66 (3H, s, OCH_3), 4.76 (1H, s, CH), 7.07-6.86 (m, 4H, Arom), 7.76 (1H, s, NH), 9.27 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 27.34 (CH_3), 29.16 (CH_3), 31.56 (C), 32.75 (CH_2), 50.52 (CH_2), 51.84 (CH), 55.29 (OCH_3), 107.84 (C), 113.39 (CH), 128.99 (CH), 135.15 (C), 149.52 (C), 153.61 (C), 157.55 (CO), 194.86 (CO) ppm. Anal. Calcd. for $C_{17}H_{20}N_2O_3$: C, 67.98; H, 6.71; N, 9.33 %. Found: C, 68.228; H, 6.41; N, 9.25 %.

*Compound **5c***:

IR (KBr): $\bar{\nu}$ = 3443 (s), 3192 (s), 1681 (s), 1665 (s), 1624 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 0.74 (3H, s, CH_3), 0.81 (3H, s, CH_3), 1.75-1.93 (2H, m,CH_2), 2.09-2.24 (2H, m,CH_2), 5.34 (1H, s, CH), 7.05-7.18 (4H, m, Arom), 7.50 (1H, s, NH), 9.33 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 28.64 (CH_3), 30.33 (C), 33.85 (CH_2), 51.38 (CH), 107.41 (C), 128.99 (CH), 130.55 (CH), 131.01 (CH), 133.46 (CH), 142.79 (CH), 154.66 (C), 161.84 (CO), 194.24 (CO) ppm. Anal. Calcd. for $C_{16}H_{17}ClN_2O_2$: C, 63.05; H, 5.62; N, 9.19 %. Found: C, 63.26; H, 5.37; N, 9.12 %.

*Compound **5h***:

IR (KBr) ν_{max} / cm^{-1}: 3285 (s), 3190 (s), 1646 (s), 1605 (s); ^{1}H NMR (DMSO-d_6, 400 MHz) δ / ppm : 0.79 (3H, s, CH_3), 0.879 (3H, s, CH_3), 1.93-1.97 (2H, d, J = 16 Hz, CH_2), 2.00-2.04 (2H, d, J = 16 Hz, CH_2), 2.26 (3H, s, CH_3), 4.50 (1H, s, CH), 6.80-6.98 (4H, m, Arom), 7.06 (2H, br, NH) ppm; Anal. Calcd. for $C_{17}H_{20}N_2O_2$: C, 71.81; H, 7.09; N, 9.85 %. Found: C, 72.03; H, 6.90; N, 9.72 %.

*Compound **5j***:

IR (KBr): $\bar{\nu}$ = 3228 (s), 2962 (s), 1649 (s), 1620 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 1.890-1.833 (2H, m,CH_2) ,1.94 -2.00 (2H, m,CH_2) ، 2.306 -2.347 (2H, m, CH_2), 4.594 (1H, s, CH), 7.096-7.241 (5H, m, Arom),7.516 (1H, s, NH), 9.347 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 21.00 (CH_2), 27.77 (CH_2), 36.75 (CH_2), 50.94 (CH), 107.69 (C), 113.39 (CH), 129.03 (CH), 136.07 (C), 149.51 (C), 154.81 (C), 158.74 (CO), 191.82 (CO) ppm. Anal. Calcd. for $C_{14}H_{14}N_2O_2$: C, 69.41; H, 5.82; N, 11.56 %. Found: C, 69.22; H, 5.92; N, 11.41 %.

*Compound **5n***:

IR (KBr): $\bar{\nu}$ = 3302 (s), 3190 (s), 1688 (s), 1667 (s), 1636 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 1.35-1.61 (2H, m, CH_2), 1.74-1.91(2H, m, CH_2), 1.96-2.18 (2H, m, CH_2), 3.50 (3H, s, OCH_3), 4.39 (1H, s, CH), 6.56-6.65 (4H, m, Arom), 6.96 (1H, s, NH), 8.11 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz) δ / ppm : 20.99 (CH_2), 29.00 (CH_2), 37.27 (CH2), 55.02(CH), 101.62 (C), 110.42 (CH), 111.48 (CH), 119.84 (CH), 126.56 (CH), 129.25 (CH), 131.70 (CH), 156.55 (C), 169.11 (CO), 196.02 (CO) ppm. Anal. Calcd. for $C_{15}H_{16}N_2O_3$: C, 66.16; H, 5.92; N, 10.29 %. Found: C, 66.40; H, 5.752; N, 10.11 %.

*Compound **5q***:

IR(KBr): $\bar{\nu}$ = 3437 (s), 3305 (s), 1664 (s), 1613 (s), 1589 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 0.679 (3H, s, CH_3), 0.803 (3H, s, CH_3), 1.82-2.01 (2H, m, CH_2), 2.06-2.22 (2H, m, CH_2), 4.94 (1H, s, CH), 7.06-7.16 (4H, m, Arom), 7.61 (1H, s, NH), 9.35 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 27.28 (CH_3), 29.12 (C), 32.78 (CH_2), 50.21 (CH_2), 52.08 (CH), 107.25 (C),125.32 (CH), 126.25 (CH), 127.35 (CH), 130.48 (CH), 133.26 (CH), 145.90 (CH), 147.42 (C), 158.11 (CO), 193.03 (CO) ppm. Anal. Calcd. for $C_{16}H_{17}ClN_2O_2$: C, 63.05; H, 5.62; N, 9.19 %. Found: C, 63.22; H, 5.55; N, 9.08 %.

*Compound **5r***:

IR (KBr): $\bar{\nu}$ = 3278 (s), 3162 (s), 1642 (s), 1572 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 0.903 (3H, s, CH_3), 1.041 (3H, s, CH_3), 2.053-2.093 (2H, m, CH_2), 2.212 (2H, s,CH_2), 2.247 (3H , s, CH_3), 5.137 (1H, s, CH), 7.091-7.154 (m, 4H, Arom), 9.657 (1H, s, NH), 10.559 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 20.33 (CH_3), 27.34 (CH_3), 29.60 (CH_3), 32.75 (C), 37.84 (CH2), 50.75 (CH2), 51.51 (CH), 105.84 (C), 123.29 (CH), 129.00 (CH), 137.37 (C), 140.00 (C), 149.47 (C), 174.61 (CS) , 195.06 (CO)

ppm. Anal. Calcd. for $C_{17}H_{20}N_2OS$: C, 67.97; H, 6.71; N, 9.32; S, 10.67 %. Found: C, 68.18; H, 6.50; N, 9.21; S, 10.45 %.

*Compound **5t***:

IR (KBr): $\bar{\nu}$ = 3262 (s), 3165 (s), 1666 (s), 1641 (s), 1584 (s) cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 0.909 (3H, s, CH_3), 1.033 (3H, s, CH_3), 2.086-2.219 (2H, m, CH_2), 2.399-2.431 (2H, m, CH_2), 3.729 (3H, s, OCH_3), 5.122 (1H, s, CH), 6.888-6.910 (2H, m, Arom), 7.146-7.124 (2H, m, Arom), 9.640 (1H, s, NH), 10.546 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 27.25 (CH3), 29.29 (CH_3), 32.73 (C), 38.93 (CH_2), 50.31 (CH_2), 52.09 (CH), 55.56 (OCH_3), 108.75 (C), 114.27 (CH), 128.09 (CH), 136.06 (C), 148.92 (C), 159.09 (C), 174.82 (CS)، 194.11 (CO) ppm. Anal. Calcd. for $C_{17}H_{20}N_2O_2S$: C, 64.53; H, 6.37; N, 8.85; S, 10.13 %. Found: C, 64.61; H, 6.39; N, 8.72; S, 9.90 %.

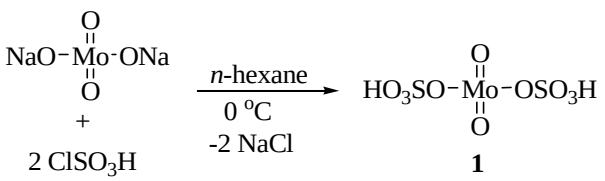
*Compound **5u***:

IR (KBr): $\bar{\nu}$ = 3262 (s), 3173 (s), 1698 (s), 1620 (s), 1567 (s) cm^{-1}. ^{1}H NMR (DMSO-d_6, 400 MHz): δ= 0.897 (3H, s, CH_3),1.035 (3H, s, CH_3), 2.059-2.229 (2H, m, CH_2), 2.364-2.484 (2H, m, CH_2), 5.193 (1H, s, CH), 7.223-7.280 (m, 3H, Arom), 7.328-7.365 (2H, m, Arom), 9.696 (1H, s, NH), 10.594 (1H, s, NH) ppm. ^{13}CNMR (DMSO-d_6, 100 MHz): δ= 27.24 (CH_3), 29.28 (C), 32.73 (CH_2), 50.31 (CH_2), 52.69 (CH), 108.61 (C), 126.89 (CH), 128.02 (CH), 128.96 (CH), 143.83 (CH), 149.16 (C), 175.08 (CS), 194.12 (CO) ppm. Anal. Calcd. for $C_{16}H_{18}N_2OS$: C, 67.10; H, 6.33; N, 9.78; S, 11.20 %. Found: C, 67.22; H, 6.40; N, 9.65; S, 11.35 %.

3. Results and Discussion

Molybdate sulfuric acid (MSA) was synthesized from the reaction of sodium molybdate and chlorosulfonic acid (Scheme 1). The reaction is simple and proceeds efficiently to produce the desired solid acid. MSA was characterized by X-ray fluorescence (XRF), X-ray diffraction (XRD), and FT-IR spectroscopy [14].

Fig. 1 shows the XRD patterns of molybdate sulfuric acid (MSA). It was reported that high degree mixing of Mo–S in chlorosulfonic acid often led to the absence of XRD pattern for anhydrous sodium molybdate. The broad peaks around 23°, 29° and 34° (2θ) (θ is the Bragg′s angle) from the smaller inset could be attributed to linking of Mo to the chlorosulfonic acid.



Scheme 1. Synthesis of molybdate sulfuric acid (MSA).

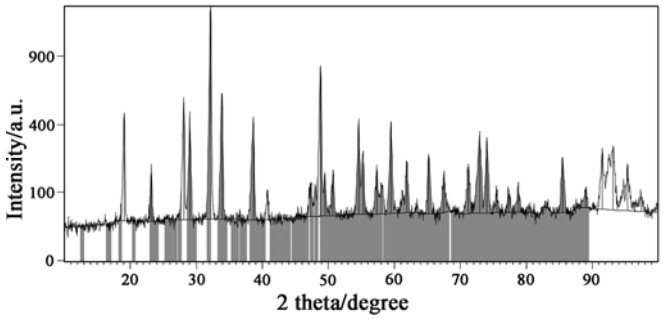


Fig. 1. The powder X-ray diffraction pattern of the molybdate sulfuric acid (MSA **1**).

The XRF data of molybdosulfuric acid (MSA **1**) indicates the presence of MoO_3 and SO_3 in this catalyst (Table 1).

The FT-IR spectra of anhydrous sodium molybdate and molybdate sulfuric acid (MSA **1**) are shown in Fig. 2. The spectrum of molybdate sulfuric acid (MSA **1**) shows the characteristic bonds of anhydrous sodium molybdate and chlorosulfonic acid. The adsorbtion in 3459, 2110, 1635, 1129, 909, 771, 637, 616 and 451 cm^{-1} in the catalyst spectrum reveal both bonds in anhydrous sodium molybdate and –OSO_3H group. Hence titration of catalyst whit NaOH (0.1 N) was done. Firstly, 1 mmol of catalyst dissolved in 100 mL of water.

Table 1. XRF data of MSA **1**.

Entry	Compound	Concentration (%W/W)
1	SO_3	49.52
2	Na_2O	1.15
3	MoO_3	39.02
4	Cl	0.150
5	K_2O	0.064
6	Nb_2O_5	0.019
7	Fe_2O_3	0.012
8	CuO	0.010
9	LOI[a]	10.03
10	Total	99.98

[a] Loss on ignition.

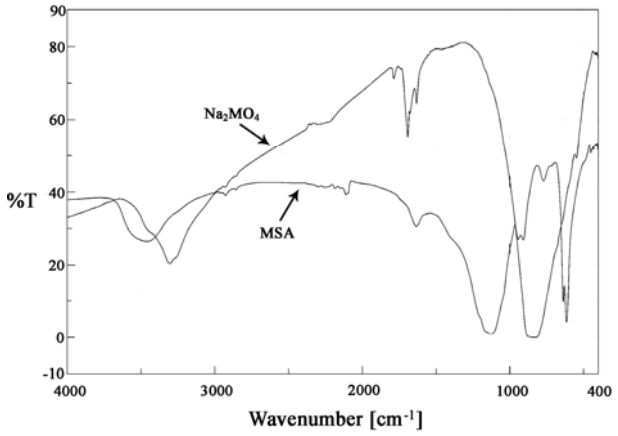


Fig. 2. FT-IR spectra of MSA **1** and sodium molybdate.

Therefore, in the presence of phenolphthalein as an indicator with NaOH (0.1 N) titrated. In equivalent point, was seen that for 1 mmol of catalyst, 2 mmol of NaOH utilized. The result of catalyst titration is shown two acidic valences.

In order to investigate the acid capacity of MSA, a solution of it (0.0805 g) in distilled water (100 mL) was titrated with standard solution of NaOH (0.1 N) in the presence of phenolphthalein as indicator. At the endpoint of titration 5 mL of titrant was consumed. The capacity of MSA was determined according the following equation as 2. (m/MW) × n = N_2V_2, (0.0805/322) × n = 0.1 × 0.005, thus n = 2. Therefore, MSA can be considered as a solid heterogeneous alternative to sulfuric acid.

In connection with our previous programs on synthesis of organic compounds [15-18], herein, we wish to report a simple and convenient synthesis of octahydroquinazolinones/thiones **5** through the reaction of aromatic aldehyde **2**, urea/thiourea **3**, and cyclic 1,3-dione **4**, in the presence of catalytic amount of MSA (**1**) under solvent-free conditions (Scheme 2). The yield of products was good to excellent without the formation of octahydoxanthenes **6**, which is the major product of the procedure reported by the literature [19].

A solvent-free or solid state reaction obviously reduce pollution, and bring down handling costs due to simplification of experimental procedure, work up technique and saving in labour. However, interest in the environmental control of chemical processes has increased remarkably during three decades ago as a response to public concern about the use of hazardous chemicals. Therefore, to improve the effectiveness of this method in preventing chemical waste, it is important to investigate optimal reaction conditions. To find the simple and suitable conditions for the preparation of **5** using MSA (**1**), the treatment of benzaldehyde, dimedone, and urea was chosen as a model reaction. At first, we found that in the absence of **1**, the reaction did not proceed even at a high temperature after long reaction time (Fig. 3).

After examining the various amounts of **1** and a wide range of temperatures (Fig. 4), we found that this reaction can be efficiently carried out by adding 10 mol% of the catalyst at 100 °C under solvent-free conditions in a short time span of 60 min. The use of excessive amounts of the catalyst does not increase the product yield or reaction rate.

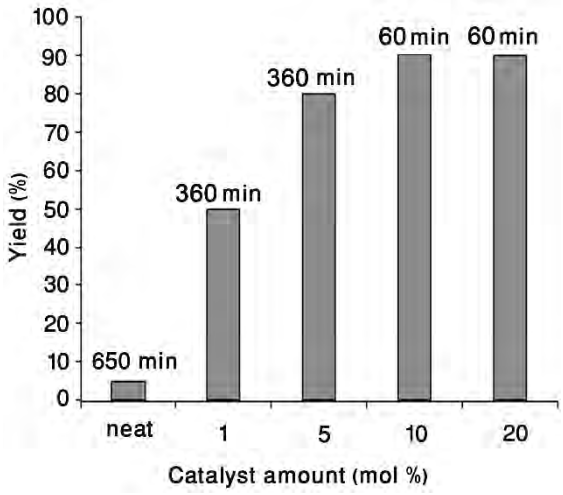
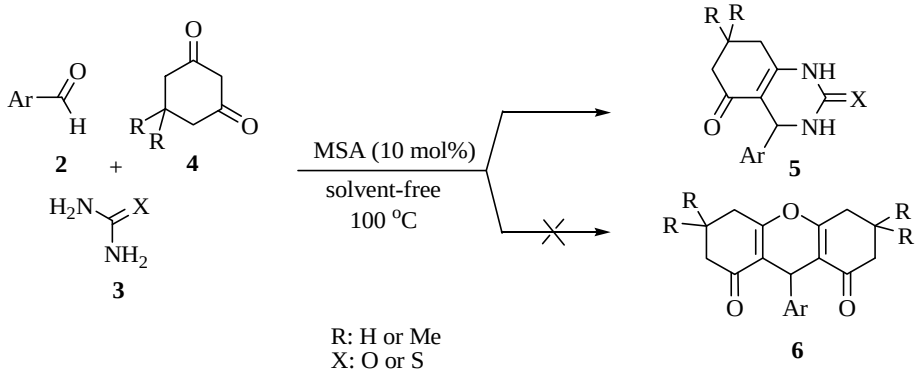


Fig. 3. Optimization of catalyst amount in synthesis of **5a** at 100 °C under solvent-free conditions.



Scheme 2. Three-component condensation of arylaldehydes, cyclic 1,3-dione, and urea/thiourea using MSA (**1**).

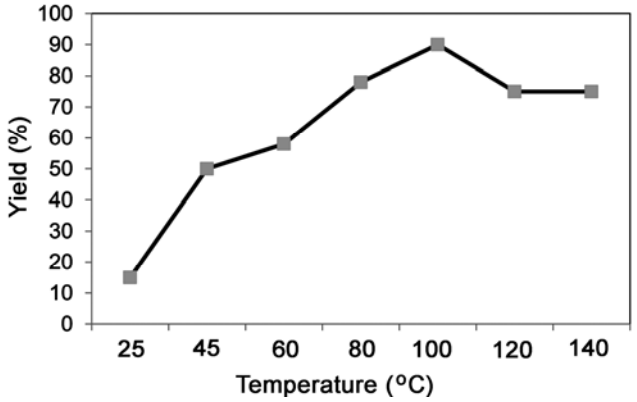


Fig. 4. Optimization of temperature in synthesis of **5a**. Reaction time: 60 min.

The scope of this MCR was examined using a variety of starting materials including various benzaldehydes, two types of cyclic 1,3-diones, and urea/thiourea. By employing different substituted benzaldehydes bearing either electron-withdrawing or electron-donating groups, it was found that the reactions are successful and the reactions proceeded well to afford the corresponding products **5** in good to excellent yields. The obtained results have been summarized in Table 2.

All products were characterized by comparison of their spectral (FT-IR and NMR) and physical data (melting point) with those reported in the literature and it was found that they are in agreement.

It should be mentioned that our efforts on the synthesis of quinazolinones/thiones employing aliphatic aldehydes were unsuccessful. The problem with alkyl aldehydes is likely to be because they can undergo enolization.

In view of eco-friendly procedure, the recovery and reuse of this catalyst is quite preferable. MSA (**1**) was easily separated from the reaction mixture by filtering, followed by drying. The catalyst was reused three times for synthesis of **5a** without significant loose of activity (Fig. 5).

Table 2. Synthesis of **5** via the Biginelli method using MSA (**1**) at 100 °C under solvent-free conditions.

Entry	R	Ar	X	Time (min)	Yield (%)[a]	m.p. (°C)	Ref.
5a	Me	C_6H_5	O	60	90	288-290	[13]
5b	Me	4-MeO-C_6H_4	O	70	85	272-274	[20]
5c	Me	2-Cl- C_6H_4	O	75	80	271-273	[13]
5d	Me	4-Br- C_6H_4	O	45	80	324-326	[21]
5f	Me	4-F- C_6H_4	O	50	78	300-302	[13]
5g	Me	2-MeO- C_6H_4	O	30	80	197-199	[13]
5h	Me	4-Me-C_6H_4	O	35	90	300-302	[21]
5i	Me	3-O_2N- C_6H_4	O	25	88	297-299	[22]
5j	H	C_6H_5	O	45	92	275-277	[23]
5k	H	4-Cl-3-O_2N- C_6H_3	O	35	82	209-211	[24]
5l	H	4-Br- C_6H_4	O	40	80	275-277	[20]
5m	H	4-Cl- C_6H_4	O	40	88	281-282	[23]
5n	H	2-MeO- C_6H_4	O	45	78	197-199	[25]
5o	Me	2,4-Cl_2- C_6H_3	O	45	90	263-265	[22]
5p	Me	3-Br-C_6H_4	O	40	90	265-267	[24]
5q	Me	3-Cl-C_6H_4	O	55	80	290-292	[24]
5r	Me	4-Me-C_6H_4	S	60	80	280-282	[22]
5s	Me	4-Br-C_6H_4	S	75	77	290-292	[24]
5t	Me	4-MeO-C_6H_4	S	70	80	268-270	[25]
5u	Me	C_6H_5	S	70	83	280-282	[25]

[a]Isolated yields.

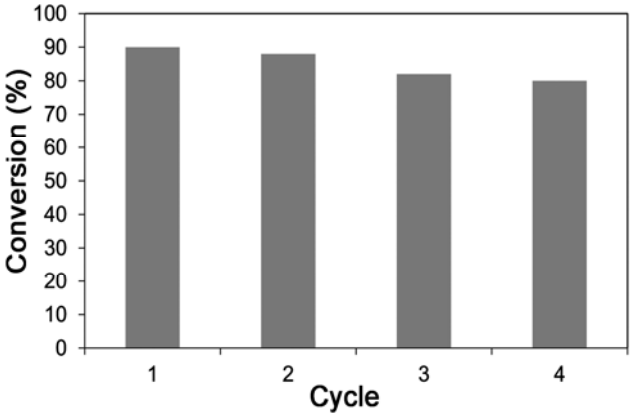


Fig. 5. Recyclability of MSA in synthesis of **5a** under optimized conditions. Reaction time: 60 min.

4. Conclusions

To conclude, this simple catalytic system is remarkably tolerant to a variety of functional groups on the arylaldehyde and offers significant advantages such as, low catalyst loading, high yields, avoidance of the organic solvents, and the use of safe and recyclable catalyst, short reaction times and operational simplicity. Therefore, in employing a small amount of safe, inexpensive, and powerful catalyst under solvent-free conditions, this protocol is economic and eco-friendly.

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