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Ultrasound promotes one-pot synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives, catalyzed by ZnO nanoparticles

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Abstract Ultrasonic irradiation is being considered not only as a green approach but also as a powerful technique for the synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives. It can be carried out by using multicomponent reaction of cyclic enaminoketones, malononitrile, and aromatic aldehydes, in the presence of catalytic amounts of zinc oxide nanoparticles, in EtOH, at 80 °C. The preponderance of such a catalyst is due to its inexpensiveness, stability, and the potential of being easily obtained. Furthermore, high conversions, short reaction times, and cleaner reaction profiles are some of the advantages of this method.

Keywords Ultrasound irradiation · 1,4-Dihydropyridine and imidazo[1,2-*a*]quinoline derivatives · Multicomponent reaction · ZnO nanoparticles

Introduction

Ultrasound irradiation has been increasingly used in organic synthesis in the last three decades as an eco-friendly energy source. Compared to traditional methods, this technique is more convenient and easily controlled [1–3]. Moreover, enhanced reaction rates, formation of unadulterated products in high yields, easier

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manipulation, and waste minimization are some prominent features of this approach [4, 5]. More recently, ultrasonic irradiation has been used for the synthesis of azeto[2,1-*d*] [1, 5] benzothiazepines [6], 1,3-dipolar cycloaddition [7], and multicomponent reaction [8, 9] as a clean, practical, and usable protocol.

In this regard, multicomponent reactions (MCRs) are considered an invaluable strategy for the synthesis of many novel compounds [8–11]. The MCRs strategy depicted significant priorities over conventional linear-type synthesis, bringing about three or more simple and flexible molecules brought together to rapidly introduce structural complexity and diversity [12–15]. Because of the range of readily available starting materials and the simplicity of one-pot procedures, MCRs have been extensively used for the synthesis of heterocyclic compounds. Among these, the multicomponent synthesis of polyfunctionalized heterocyclic compounds has become more challenging in organic and medicinal chemistry [16, 17].

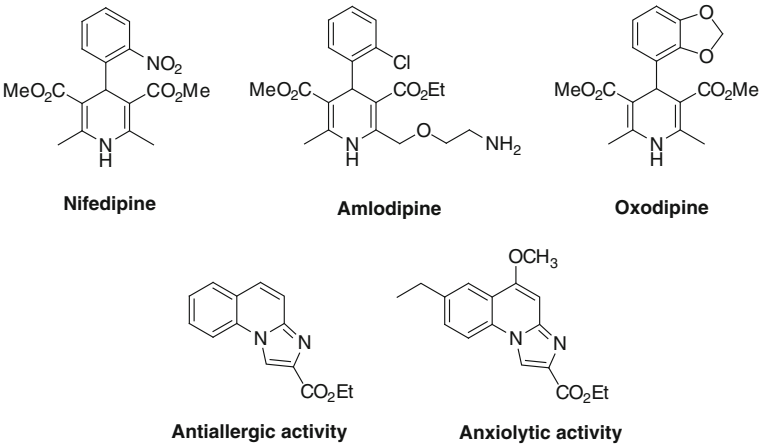
1,4-Dihydropyridines are an important class of compounds with a wide range of biological activities [18]. Due to being pharmacologically active and acting as an antitumor [19], calcium channel bloker [20], antitubercular [21], analgesic [22], antithrombotic [23], anti-inflammatory [24], and anticonvulsant agent [25], they are of many interests. Some of the marketed drug preparations containing 1,4-dihydropyridine moieties are Nifedipine, Amlodipine, Oxodipine, etc. (Scheme 1). In addition, imidazo[1,2-*a*]quinoline is a synthetically designed scaffold with a broad range of biological activities. Some of its derivatives have pharmacological properties such as antiallergic [26] and anxiolytic [27] activity (Scheme 1).

Nano-particles as heterogeneous catalysts have received considerable attention in recent years owing to their interesting structures and outstanding catalytic activities [28–31]. These nano-metal oxides have also shown high activity, strong oxidizing power, recyclability, and long-term stability [32, 33]. Nano-crystalline metal oxides such as nano-zinc oxide exhibit versatile applications as active adsorbents for gases or destruction of hazardous chemicals [34, 35], and also as catalysts in various organic transformations [8, 9, 36–39]. Hence, we decided to synthesize 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives, by using ultrasound irradiation at EtOH and in the presence of catalytic amounts of zinc oxide nano-particles.

Experimental

General

Melting points were measured on a Electrothermal-9100 apparatus and are uncorrected. IR spectra were recorded on a Brucker FT-IR Tensor 27 infrared spectrophotometer. ^{1}H-NMR and ^{13}C-NMR spectra were recorded on a Avance III 400 Bruker spectrometer at 400 and 100 MHz, respectively. Elemental analyses were performed using a Heracus CHN-O-Rapid analyzer. The cyclic enaminoketones **1a–d** [43] and **1e**, **f** [40] were prepared from the according to procedures described in the literature.



Scheme 1 Some of the marketed drugs containing 1,4-dihydropyridine moieties and imidazo[1,2-*a*]quinoline derivatives with biological activity

Preparation of ZnO nanoparticles

Zinc acetate dihydrate (2.19 g, 10 mmol) and $CO(NH_2)_2$ (12 g, 200 mmol) were dissolved in deionized water (200 ml) at room temperature to form a transparent solution, which was then refluxed for 12 h. It was cooled in an ice bath to stop the reaction. The precipitate was centrifuged and washed with deionized water and absolute EtOH, dried at 80 °C for 8 h, and then calcinated in a furnace in the presence of air at 400 °C for 2 h [44].

General procedure for the preparation of 1,4-dihydropyridine (**4a–l**) and imidazo[1,2-*a*]quinoline (**4m–r**) derivatives

A mixture of cyclic enaminoketones **1** (2 mmol), malononitrile **2** (2 mmol), aldehyde **3** (2 mmol), and ZnO nanoparticles (10 mol %) in ethanol (10 ml) was irradiated at 80 °C for the times reported in Table 3 (the progress of the reaction being monitored by TLC and was used hexane/ethyl acetate as an eluent). After completion of the reaction, the catalyst was separated from the reaction mixture by centrifugation. Then, the reaction mixture was poured into ice-cold water; the crude product was filtered, dried, and recrystallized from ethanol.

*2-amino-4-(4-methoxyphenyl)-7,7-dimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4b**)*

Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,392, 3,328, 3,232 (NH_2, NH), 2,192 (CN), 1,654 (C=O), 1,596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 8.82 (s, 1H, NH), 6.94 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 6.71 (d, 2H, $^3J_{HH}$ = 4 Hz, CH–Ar), 5.65 (s, 2H, NH_2), 4.16 (s, 1H, CH), 3.61 (s, 3H, OCH_3), 2.32 (d, $^2J_{HH}$ = 8 Hz, CH), 2.20 (d, $^2J_{HH}$=8 Hz, CH), 2.08 (d, $^2J_{HH}$=8 Hz, CH), 1.89 (d, $^2J_{HH}$=8 Hz, CH), 0.92 (s, 3H,

CH_3), 0.81 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 192.71 (C=O), 156.08, 153.27, 152.59, 137.96, 136.48, 132.71, 131.48, 113.35 (CN), 61.68 (C_3), 55.63 (OCH_3), 48.25 (CH_2), 37.36 (CH_2), 36.58 (CH), 32.69 (CMe_2), 29.34 (CH_3), 27.24 (CH_3). Anal. calcd. for $C_{19}H_{21}N_3O_2$: C, 70.57; H, 6.55; N, 12.99 %. Found: C, 70.43; H, 6.48; N, 12.82 %.

*2-amino-4-(4-bromophenyl)-7,7-dimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4c**)*

Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,392, 3,328, 3,232 (NH_2, NH), 2,192 (CN), 1,654 (C=O), 1,596 (C=C). ^1H NMR (400 MHz, DMSO-d_6) δ: 8.93 (s, 1H, NH), 7.45 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 7.08 (d, 2H,$^3J_{HH}$=4 Hz, CH–Ar), 5.82 (s, 2H, NH_2), 4.30 (s, 1H, CH), 2.42 (d,$^2J_{HH}$=8 Hz, CH), 2.31 (d, $^2J_{HH}$=8 Hz, CH), 2.18 (d,$^2J_{HH}$=8 Hz, CH), 1.98 (d,$^2J_{HH}$=8 Hz, CH), 1.02 (s, 3H, CH_3), 0.90 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 191.89 (C=O), 155.88, 154.12, 136.70, 134.34, 132.19, 131.79, 128.86, 113.41 (CN), 60.61 (C_3), 49.59 (CH_2), 40.89 (CH_2), 39.91 (CH), 33.53 (CMe_2), 30.22 (CH_3), 28.82 (CH_3). Anal. calcd. for $C_{18}H_{18}BrN_3O$: C, 58.08; H, 4.87; N, 11.29 %. Found: C, 57.91; H, 4.75; N, 11.05 %.

*2-amino-7,7-dimethyl-5-oxo-1-phenyl-4-(p-tolyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4e**)*

Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,472, 3,344 (NH_2), 2,176 (CN), 1,651 (C=O), 1,590 (C=C). ^1H NMR (400 MHz, DMSO-d_6) δ: 7.63–7.12 (m, 9H, CH–Ar), 5.31 (s, 2H, NH_2), 4.42 (s, 1H, CH), 2.21 (d, $^2J_{HH}$=4 Hz, CH), 2.17 (d, $^2J_{HH}$=4 Hz, CH), 2.00 (d,$^2J_{HH}$=8 Hz, CH), 1.69 (d,$^2J_{HH}$=8 Hz, CH), 1.03 (s, 3H, CH_3), 0.88 (s, 3H, CH_3), 0.73 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 193.76 (C=O), 156.23, 153.14, 135.02, 134.31, 132.25, 131.66, 130.79, 130.18, 127.77, 126.26, 120.94, 114.82 (CN), 62.70 (C_3), 49.80 (CH_2), 42.10 (CH_2), 38.42 (CH), 34.51 (CMe_2), 32.02 (CH_3), 29.63 (CH_3), 21.11 (CH_3). Anal. calcd. for $C_{25}H_{25}N_3O$: C, 78.30; H, 6.57; N, 10.96 %. Found: C, 78.08; H, 6.45; N, 10.83 %.

*2-amino-4-(4-bromophenyl)-7,7-dimethyl-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4f**)*

White crystals; IR (KBr, ν_{max},cm^{-1}): 3,456, 3,328 (NH_2), 2,160 (CN), 1,648 (C=O), 1,587 (C=C). ^1H NMR (400 MHz, DMSO-d_6) δ: 7.63–7.23 (m, 9H, CH–Ar), 5.71 (s, 2H, NH_2), 4.46 (s, 1H, CH), 2.21 (d, $^2J_{HH}$=4 Hz, CH), 2.17 (d, $^2J_{HH}$=4 Hz, CH), 2.01 (d, $^2J_{HH}$=8 Hz, CH), 1.70 (d,$^2J_{HH}$=8 Hz, CH), 0.88 (s, 3H, CH_3), 0.73 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 192.25 (C=O), 157.14, 154.66, 135.29, 133.27, 132.92, 131.62, 131.43, 128.36, 126.03, 122.88, 120.95, 114.95 (CN), 61.69 (C_3), 49.75 (CH_2), 43.22 (CH_2), 38.58 (CH), 35.90 (CMe_2), 31.69 (CH_3), 29.50 (CH_3). Anal. calcd. for $C_{24}H_{22}BrN_3O$: C, 64.29; H, 4.95; N, 9.37 %. Found: C, 64.07; H, 4.78; N, 9.19 %.

*2-amino-5-oxo-4-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4g**)*

Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,408, 3,328, 3,232 (NH_2, NH), 2,176 (CN), 1,638 (C=O), 1,596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 8.96 (s, 1H, NH), 7.27–7.12 (m, 5H, CH–Ar), 5.75 (s, 2H, NH_2), 4.34 (s, 1H, CH), 2.28–1.76 (m, 6H, $3CH_2$). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 192.94 (C=O), 156.59, 153.98, 137.94, 133.74, 132.82, 131.70, 130.29, 113.40 (CN), 62.12 (C_3), 49.10 (CH_2), 42.89 (CH_2), 38.58 (CH), 25.40 (CH_2). Anal. calcd. for $C_{16}H_{15}N_3O$: C, 72.43; H, 5.70; N, 15.84 %. Found: C, 72.29; H, 5.52; N, 15.67 %.

*2-amino-5-oxo-4-(p-tolyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4h**)*

Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,424, 3,328, 3,232 (NH_2, NH), 2,176 (CN), 1,651 (C=O), 1,596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 8.93 (s, 1H, NH), 7.04 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 7.01 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 5.73 (s, 2H, NH_2), 4.30 (s, 1H, CH), 2.23 (s, 3H, CH_3), 2.20–1.72 (m, 6H, $3CH_2$). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 192.58 (C=O), 155.97, 153.22, 138.00, 133.13, 132.73, 131.85, 129.64, 113.43 (CN), 62.29 (C_3), 49.28 (CH_2), 40.90 (CH_2), 38.36 (CH), 26.12 (CH_2), 21.57 (CH_3). Anal. calcd. for $C_{17}H_{17}N_3O$: C, 73.10; H, 6.13; N, 15.04 %. Found: C, 72.89; H, 5.98; N, 14.88 %.

*2-amino-4-(4-chlorophenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4i**)*

Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,392, 3,328, 3,232 (NH_2, NH), 2,176 (CN), 1,657 (C=O), 1,600 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 9.00 (s, 1H, NH), 7.31 (d, 2H, $^3J_{HH}$= 4 Hz, CH–Ar), 7.14 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 5.81 (s, 2H, NH_2), 4.35 (s, 1H, CH), 2.28–1.73 (m, 6H, $3CH_2$). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 193.05 (C=O), 156.11, 154.01, 135.56, 133.77, 132.79, 131.57, 127.61, 114.94 (CN), 63.14 (C_3), 48.92 (CH_2), 41.40 (CH_2), 37.29 (CH), 25.28 (CH_2). Anal. calcd. for $C_{16}H_{14}ClN_3O$: C, 64.11; H, 4.71; N, 11.83 %. Found: C, 63.87; H, 4.59; N, 11.69 %.

*2-amino-5-oxo-1,4-diphenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4j**)*

Pink crystals; IR (KBr, ν_{max},cm^{-1}): 3,472, 3,296 (NH_2), 2,176 (CN), 1,648 (C=O), 1,590 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 7.52–7.09 (m, 10H, CH–Ar), 5.26 (s, 2H, NH_2), 4.41 (s, 1H, CH), 2.21–1.49 (m, 6H, $3CH_2$).). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 192.75 (C=O), 155.18, 153.49, 142.28, 140.33, 135.26, 131.61, 131.23, 131.08, 130.50, 125.88, 124.68, 114.06 (CN), 62.89 (C_3), 49.12 (CH_2), 41.75 (CH_2), 38.34 (CH), 26.60 (CH_2). Anal. calcd. for $C_{22}H_{19}N_3O$: C, 77.40; H, 5.61; N, 12.31 %. Found: C, 77.18; H, 5.47; N, 12.13 %.

*2-amino-5-oxo-1-phenyl-4-(p-tolyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4k**)*

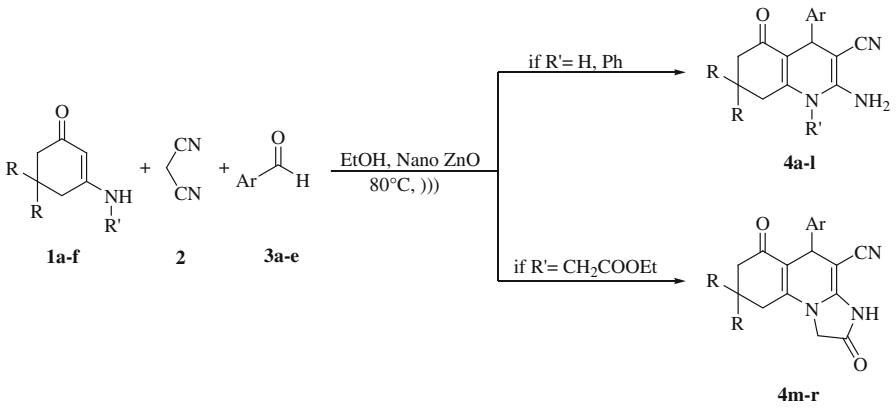
Yellow crystals; IR (KBr, ν_{max},cm^{-1}): 3,472, 3,328 (NH_2), 2,192 (CN), 1,638 (C=O), 1,590 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 7.61–7.11 (m, 9H, CH–Ar), 5.32 (s, 2H, NH_2), 4.46 (s, 1H, CH), 2.27 (s, 3H, CH_3), 2.22–1.56 (m, 6H, 3CH_2). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 192.86 (C=O), 156.82, 154.27, 136.07, 135.51, 134.52, 134.14, 133.54, 131.63, 131.32, 128.85, 125.96, 115.44 (CN), 63.23 (C_3), 48.69 (CH_2), 41.24 (CH_2), 37.69 (CH), 25.88 (CH_2), 21.46 (CH_3). Anal. calcd. for $C_{23}H_{21}N_3O$: C, 77.22; H, 5.96; N, 11.82 %. Found: C, 77.01; H, 5.78; N, 11.69 %.

*2-amino-4-(4-chlorophenyl)-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4l**)*

Brown crystals; IR (KBr, ν_{max},cm^{-1}): 3,408, 3,328 (NH_2), 2,176 (CN), 1,657 (C=O), 1,600 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 7.61–7.41 (m, 5H, CH–Ar), 7.39 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 7.30 (d, 2H, $^3J_{HH}$=4 Hz, CH–Ar), 5.41 (s, 2H, NH_2), 4.51 (s, 1H, CH), 2.29–1.56 (m, 6H, 3CH_2). ^{13}C NMR (100 MHz, DMSO-d_6) δ: 193.11 (C=O), 155.12, 153.68, 140.94, 135.16, 132.03, 130.87, 130.83, 129.94, 128.51, 127.47, 126.97, 115.35 (CN), 63.69 (C_3), 49.28 (CH_2), 41.52 (CH_2), 38.14 (CH), 25.30 (CH_2). Anal. calcd. for $C_{22}H_{18}ClN_3O$: C, 70.30; H, 4.83; N, 11.18 %. Found: C, 70.04; H, 4.68; N, 10.99 % .

Results and discussion

There are several methods known for the synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives; these compounds were conventionally prepared using multicomponent reaction of cyclic enaminoketones, malononitrile, and aromatic aldehydes [40–42]. Although, many reported methods are effective enough, the use of expensive or poisonous catalysts, low yields, tedious work-up



Scheme 2 Synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives

processes, long reaction times, and hazardous conditions make them less favorable. In addition, the catalysts are not recyclable and are destroyed during the work-up procedure. Therefore, the introduction of a mild, easy, efficient, and environmentally benign method to synthesize 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives is still needed.

In order to synthesis 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives, we have investigated the multicomponent reaction of cyclic enaminoketones, malononitrile, and aromatic aldehydes in EtOH, in the presence of catalytic amounts of zinc oxide nanoparticles by using ultrasound irradiation (Scheme 2).

To optimize the reaction conditions, the reaction between 3-amino-5,5-dimethylcyclohex-2-enone **1a**, malononitrile **2**, and benzaldehyde **3a** was chosen as a model reaction. When this multicomponent reaction was carried out in the presence of ZnO nanoparticles (10 mol %), in ethanol, under ultrasound irradiation and at 80 °C, the 2-amino-7,7-dimethyl-5-oxo-4-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile **4a** was obtained in 92 % yield within 30 min. This reaction was also carried out in different solvents such as toluene and acetonitrile, and the best results in terms of reaction time and yield of the desired product **4a**, which was obtained when the reaction was conducted in ethanol (Table 1, entries 1–3). Decreasing the catalyst loading from 10 to 4 mol % significantly lowered the yield of the reaction (Table 1, entries 4–6). The best catalyst loading was found at 10 mol %, which gave an excellent yield of **4a** after only 30 min. It is worth mentioning that the reaction temperature was optimized to 80 °C in ethanol (Table 1, entries 7–9).

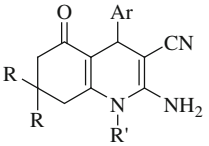
Table 1 Optimization of the model reaction between 3-amino-5,5-dimethylcyclohex-2-enone (**1a**), malononitrile **2** and benzaldehyde (**3a**)

Entry	Solvent	Catalyst (mol %)	Temperature (°C)	Time (min)	Yield (%)
1	EtOH	10	80	30	92
2	CH_3CN	10	80	45	85
3	Toluene	10	80	65	78
4	EtOH	8	80	33	90
5	EtOH	6	80	38	87
6	EtOH	4	80	45	83
7	EtOH	10	70	35	89
8	EtOH	10	60	38	87
9	EtOH	10	r.t.	50	80

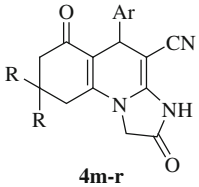
Table 2 Reusability of nano-ZnO on the three-component reaction for the synthesis of (**4a**)

Entry	Time (min)	Yield (%)
1	50	67
2	38	88
3	40	85
4	35	89

Table 3 Multicomponent reaction of cyclic enaminoketones (**1a–f**), malononitrile (**2**), and aromatic aldehydes (**3a–e**)



4a-l



4m-r

Compd. no.	R	R′	Ar	Time (min)	Yield (%)	M.P. observed (°C)	M.P. reported (°C)
4a	Me	H	C_6H_5	30	92	262–264	265–267 [40]
4b	Me	H	4–CH_3O–C_6H_4	35	90	279–281	
4c	Me	H	4-Br-C_6H_4	27	93	210 (dec.)	
4d	Me	C_6H_5	C_6H_5	32	91	244–245	246–248 [40]
4e	Me	C_6H_5	4–CH_3–C_6H_4	35	90	243–245	
4f	Me	C_6H_5	4–Br–C_6H_4	28	92	269–271	
4g	H	H	C_6H_5	32	92	254 (dec.)	
4h	H	H	4–CH_3–C_6H_4	37	90	269–271	
4i	H	H	4–Cl–C_6H_4	30	93	296–298	
4j	H	C_6H_5	C_6H_5	33	90	110 (dec.)	
4k	H	C_6H_5	4–CH_3–C_6H_4	38	88	235 (dec.)	
4l	H	C_6H_5	4–Cl–C_6H_4	30	92	228–230	
4m	Me	CH_2–COOEt	C_6H_5	45	90	>300	>300 [41]
4n	Me	CH_2–COOEt	4–CH_3–C_6H_4	48	88	>300	>300 [41]
4o	Me	CH_2–COOEt	4–Cl–C_6H_4	42	92	>300	>300 [41]
4p	H	CH_2–COOEt	C_6H_5	48	89	270–272	274–275 [41]
4q	H	CH_2–COOEt	4–CH_3–C_6H_4	55	88	283–285	287–2289 [41]
4r	H	CH_2–COOEt	4–Cl–C_6H_4	45	90	289–290	290–291 [41]

We also attempted to reuse the catalysts by a variety of methods (Table 2). Direct reuse of the catalysts (Table 2, entry 1) led to a greater than 25 % decrease in activity, while washing of the catalysts with dichloromethane, ethyl acetate, and chloroform prior to reuse also resulted in lower conversions (Table 2, entries 2–4). This phenomenon probably arose because the reactant and product were not completely desorbed from nano-ZnO and, therefore, the active sites were blocked.

According to our collected data, we decided to apply this method for synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives, using a multicomponent reaction of cyclic enaminoketones, malononitrile, and aromatic aldehydes under ultrasound irradiation and in the presence of Zno nanoparticles (Table 3).

Conclusions

In summary, we have reported an efficient procedure for the multicomponent reaction of cyclic enaminoketones, malononitrile, and aromatic aldehydes, which

leads to the synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives. This reaction was carried out in the presence of ZnO nanoparticles (10 mol %), in ethanol, by using ultrasound irradiation at 80 °C. The procedure offers several advantages including high yields, operational simplicity, and clean reaction conditions in comparison with existing methods, which makes it a useful practical process for the synthesis of these compounds.

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