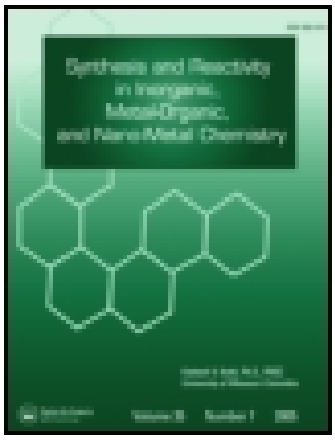

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Nanosized MgO as a Heterogeneous Base Catalysts, Catalyses Multicomponent Reaction of Cyclic Enaminoketones, Malononitrile and Aromatic Aldehydes

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Abstract

Nanosized MgO is an efficient catalyst for the synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives from multicomponent reaction of cyclic enaminoketones, malononitrile and aromatic aldehydes in EtOH at reflux condition. This method provides several advantages such as mild reaction and high yield with short reaction time.

Keywords: Nanosized MgO; multicomponent reaction; cyclic enaminoketones; malononitrile; aromatic aldehydes;

1. Introduction

In recent decades, multicomponent reactions (MCRs) have remarkably developed as a prevailing method in preparation of many diverse natural and pharmaceutical products and organic molecules. They are of stupendous attention to chemists due to their high yield, short reaction time, being easily handled and their competency in synthesis of miscellaneous structures. [1] Nowadays, MCRs are being used in order to produce heterocyclic compounds with various scaffolds and in combinatorial libraries for the synthesis of compounds owning biological activities. [2] As previously documented, among these biologically active compounds 1,4-dihydropyridines and imidazo[1,2-*a*]quinolines are particularly well known for their therapeutic applications. [3] Dihydropyridines were reported for their antitumor, [4] calcium channel blokers, [5] antitubercular, [6] analgesic, [7] antithrombotic, [8] anti-inflammatory, [9] anticonvulsant [10] and other pharmacological activities. Some of the marketed drug preparation containing 1,4-dihydropyridine moieties are Nifedipine, Amlodipine, Oxodipine, *etc*. Moreover, some of imidazo[1,2-*a*]quinoline derivatives have pharmacological properties such as antiallergic [11] and anxiolytic [12] activity (Scheme **1**).

The field of nanocatalysis is a rapidly growing field which involves the use of nanoparticles as catalysts for a variety of homogeneous and heterogeneous catalysis applications since nanoparticles have a large surface to volume ratio compared to bulk materials. Nanocatalysts are esteemed as materials of enormous surface areas and with new investigation, developments may offer expanding catalytic abilities. Heterogeneous catalysis characterizes one of the oldest commercial practices of nanoscience, nanoparticles of metals and other compounds have been

extensively used for important chemical reactions. [13] Nanosized magnesium oxide (MgO) is known as heterogeneous catalysts and requires no special handling. Nanosized magnesium oxide (MgO), obtained using a novel but simple procedure, was methodically investigated as a heterogeneous base catalyst for reactions taking place in the liquid phase, specifically the Michael addition and the Knoevenagel condensation. [14]

Considering the fact that nanosized magnesium oxide (MgO), has been hugely used in many chemical reactions, we investigated its use in multicomponent reactions as catalyst and then scrutinized the three-component reaction of cyclic enaminoketones, malononitrile and aromatic aldehydes in the presence of nanosized magnesium oxide (MgO), as the catalyst for the synthesis of both 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives.

2. Experimental

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** [15] and **1e-f** [16] were prepared from the according to procedures described in the literature.

2.1. Preparation of nanosized MgO

The MgO nanoparticles were synthesized by precipitation of the magnesium hydroxide gels in aqueous solution using $Mg(NO_3)_2$ as salt and liquid ammonia as the precipitating agent. Initially, the pH of 200 mL of distilled water was adjusted to 10.5 by addition of liquid ammonia. To this solution, 0.1 M magnesium nitrate solution (0.0148 g/mL) was added drop wise with continuous

stirring. The rate of addition of the salt solution was kept at 20 mL/h. During the addition, the pH of the mixture decreased due to hydrolysis of the salt. The pH was maintained at 10.5 by controlled addition of liquid ammonia solution. After completion of the precipitation procedure, the mixture was stirred at room temperature for 12 h, filtered, repeatedly washed with distilled water, dried at 120 °C, and calcined at 500 °C for 2 h. [17]

2.2. 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 **1a-f** (2 mmol), malononitrile **2** (2 mmol), aromatic aldehydes **3a-e** (2 mmol), and nanosized MgO (55 mol%) in ethanol (10 mL) at reflux for the time 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. The reaction mixture was poured into ice-cold water; the crude product was filtered, dried, and recrystallized from ethanol.

*2-amino-7,7-dimethyl-5-oxo-1-phenyl-4-(p-tolyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile **(4b)**:* Yellow crystals; IR (KBr, ν_{max}, cm^{-1}): 3472, 3344 (NH_2), 2176 (CN), 1651 (C=O), 1590 (C=C). ^1H NMR (400 MHz, DMSO-d_6) $\check{u}_H$: 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) $\check{u}_C$: 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 (C3), 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 (**4c**):* White crystals; IR (KBr, ν_{max}, cm^{-1}): 3456, 3328 (NH_2), 2160 (CN), 1648 (C=O), 1587 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 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_C: 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 (C3), 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-4-(4-methoxyphenyl)-7,7-dimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4e**):* Yellow crystals; IR (KBr, ν_{max}, cm^{-1}): 3392, 3328, 3232 (NH_2, NH), 2192 (CN), 1654 (C=O), 1596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 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_C: 192.71 (C=O), 156.08, 153.27, 152.59, 137.96, 136.48, 132.71, 131.48, 113.35 (CN), 61.68 (C3), 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 (**4f**):* Yellow crystals; IR (KBr, ν_{max}, cm^{-1}): 3392, 3328, 3232 (NH_2, NH), 2192 (CN), 1654 (C=O), 1596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.93 (s, 1H, NH), 7.45 (d, 2H, $^3J_{HH}$=4

Hz, CH-Ar), 7.08 (d, 2H, $^{3}J_{HH}$=4 Hz, CH-Ar), 5.82 (s, 2H, NH_2), 4.30 (s, 1H, CH), 2.42 (d, $^{2}J_{HH}$=8 Hz, CH), 2.31 (d, $^{2}J_{HH}$=8 Hz, CH), 2.18 (d, $^{2}J_{HH}$=8 Hz, CH), 1.98 (d, $^{2}J_{HH}$=8 Hz, CH), 1.02 (s, 3H, CH_3), 0.90 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6) ŭ$_C$: 191.89 (C=O), 155.88, 154.12, 136.70, 134.34, 132.19, 131.79, 128.86, 113.41 (CN), 60.61 (C3), 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-5-oxo-1,4-diphenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4g**):* Pink crystals; IR (KBr, ν_{max}, cm^{-1}): 3472, 3296 (NH_2), 2176 (CN), 1648 (C=O), 1590 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) ŭ$_H$: 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) ŭ$_C$: 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 (C3), 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 (**4h**):* Yellow crystals; IR (KBr, ν_{max}, cm^{-1}): 3472, 3328 (NH_2), 2192 (CN), 1638 (C=O), 1590 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) ŭ$_H$: 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) ŭ$_C$: 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 (C3), 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 (**4i**):* Brown crystals; IR (KBr, ν_{max}, cm^{-1}): 3408, 3328 (NH_2), 2176 (CN), 1657 (C=O), 1600 (C=C).

^{1}H NMR (400 MHz, DMSO-d_6) ŭ$_H$: 7.61-7.41 (m, 5H, CH-Ar), 7.39 (d, 2H, $^{3}J_{HH}$=4 Hz, CH-Ar), 7.30 (d, 2H, $^{3}J_{HH}$=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) ŭ$_C$: 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 (C3), 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 %.

*2-amino-5-oxo-4-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (**4j**):* Yellow crystals; IR (KBr, ν_{max}, cm^{-1}): 3408, 3328, 3232 (NH_2, NH), 2176 (CN), 1638 (C=O), 1596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) ŭ$_H$: 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) ŭ$_C$: 192.94 (C=O), 156.59, 153.98, 137.94, 133.74, 132.82, 131.70, 130.29, 113.40 (CN), 62.12 (C3), 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 (**4k**):* Yellow crystals; IR (KBr, ν_{max}, cm^{-1}): 3424, 3328, 3232 (NH_2, NH), 2176 (CN), 1651 (C=O), 1596 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) ŭ$_H$: 8.93 (s, 1H, NH), 7.04 (d, 2H, $^{3}J_{HH}$=4 Hz, CH-Ar), 7.01 (d, 2H, $^{3}J_{HH}$=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) ŭ$_C$: 192.58 (C=O), 155.97, 153.22, 138.00, 133.13, 132.73, 131.85, 129.64, 113.43 (CN), 62.29 (C3), 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 (**4l**):* Yellow

crystals; IR (KBr, ν_{max}, cm^{-1}): 3392, 3328, 3232 (NH_2, NH), 2176 (CN), 1657 (C=O), 1600 (C=C). ^{1}H NMR (400 MHz, DMSO-d_6) ŭ$_H$: 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) ŭ$_C$: 193.05 (C=O), 156.11, 154.01, 135.56, 133.77, 132.79, 131.57, 127.61, 114.94 (CN), 63.14 (C3), 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 %.

3. Results and discussion

There are several methods 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. [18] Although, many reported methods are effective enough, use of expensive or poisonous catalysts, low yields, tedious work-up processes, long reaction times and hazardous conditions make it less favorable. 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.

Our contribution is intended to describe the application of a new catalyst in their synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives. Our investigations show that multicomponent reaction of cyclic enaminoketones, malononitrile and aromatic aldehydes in the presence of a catalytic amount of nanosized MgO under reflux in EtOH to afford the corresponding 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives in good to excellent yields and short reaction time (Scheme **2**).

Moreover, in order to optimize the conditions of reactions, several polar and non-polar solvents

were used in the reaction of 3-anilino-5,5-dimethylcyclohex-2-en-1-one **1a**, malononitrile **2** and benzaldehyde **3a** as the model for reactions to study the solvent effects for preparing compound **4a**. In each case, the substrates were mixed together with 55 mol% nanosized MgO agitated with 10 mL solvent at reflux. The results are shown in Table **1**. It is noteworthy to mention that the polar solvents such as EtOH afford better yields than non polar solvents. The polar solvent may stabilize the activated intermediates and this increase reaction rate. In addition homogenization of the solid mixture through dissolution in the polar solvents improved the yields and decreased reaction time. In the absence of solvent the target product is afforded with yields <20% at the same time.

In addition, the amount of used catalyst was optimized. The best results were obtained when the reactions were carried out in the presence of 55 mol% nanosized MgO. The results are shown in Table **2**.

Finally based on the findings, multicomponent reaction of cyclic enaminoketones, malononitrile and aromatic aldehydes in the presence of nanosized MgO were examined so as to synthesize 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives (Table **3**). The reaction of various aromatic aldehydes containing electron-withdrawing groups (**4c**, **4f**, **4i**, **4l**, **4o**, **4r**) and electron-donating groups (**4b**, **4e**, **4h**, **4k**, **4n**, **4q**) were examined. They all gave the products in goods yields (Table **3**). The aldehydes containing electron-withdrawing groups gave the products in shorter time as compared to the aldehydes containing electron-donating groups. The presence of electron-withdrawing substituents on aromatic rings markedly increases the electrophilicity of the corresponding aromatic aldehydes and 2-arylidenemalononitrile derivatives (**I**) in the

Knoevenagel condensation and Michael addition reaction and this increase the reactivity and then reaction rate.

In these reactions, in first step Knoevenagel condensation takes place to form the 2-arylidenemalononitrile derivatives (**I**). The active methine of cyclic enaminoketones reacts with the electrophilic C=C double bond of 2-arylidenemalononitrile giving the intermediate **II**. The latter is then cyclized by nucleophilic attack of the NHR' group on the cyano (CN) moiety, giving intermediate **III**. Finally, the expected product **4a-l** (1,4-dihydropyridine derivatives) is afforded by tautomerization. Here, the catalytic activity of MgO was established for Knoevenagel condensation and Michael addition reactions (Scheme **3**).

Wherein R'=CH_2COOEt, upon intermolecular cyclization and elimination of EtOH gave rise to imidazo[1,2-*a*]quinoline derivatives (**4m-r**) (Scheme **4**).

Conclusion

In summary nanosized MgO could be used as a catalyst for synthesis of 1,4-dihydropyridine and imidazo[1,2-*a*]quinoline derivatives which are often encountered in biologically active compounds. This method was simple and efficient and the reaction products were isolated by easy work-up procedure and do not need any further purification steps.

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Table 1: Solvent effects on the multicomponent reaction of 3-anilino-5,5-dimethylcyclohex-2-en-1-one **1a**, malononitrile **2** and benzaldehyde **3a**

Entry	Solvent	Time (min)	Yield (%)
1	Toluene	75	73
2	THF	42	82
3	EtOAc	40	85
4	CH_3CN	35	87
5	Ethanol	20	91

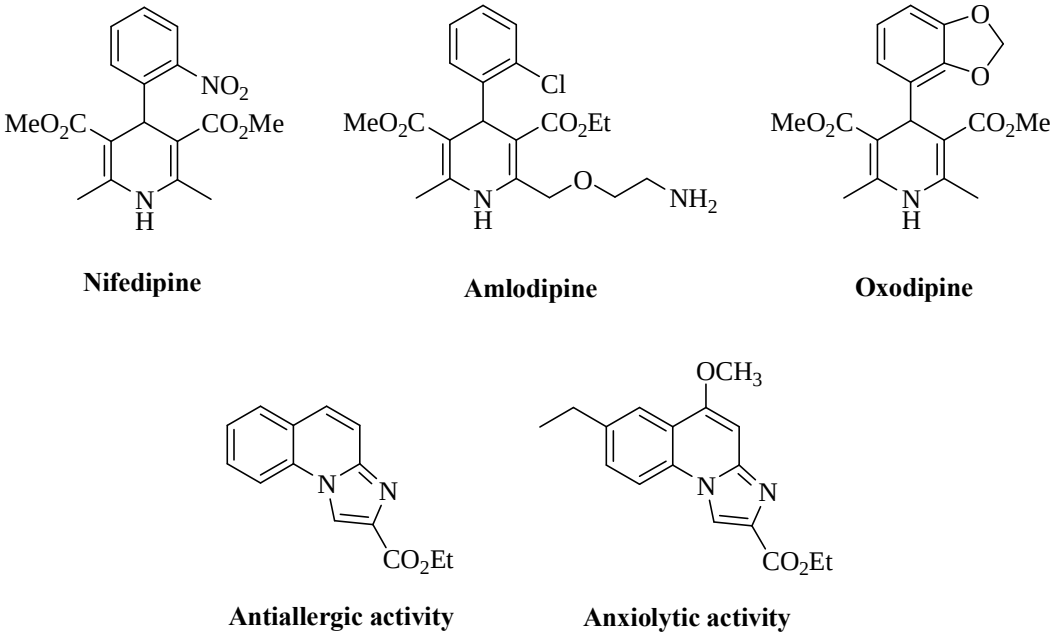
Table 2: Optimized the quantity of catalysts on the multicomponent reaction of 3-anilino-5,5-dimethylcyclohex-2-en-1-one **1a**, malononitrile **2** and benzaldehyde **3a**

Entry	Mol% catalyst	Time (min)	Yield (%)
1	25	38	80
2	35	33	84
3	45	27	87
4	55	20	91
5	65	20	91

Table 3: Multicomponent reaction of cyclic enaminoketones **1a-f**, malononitrile **2** and aromatic aldehydes **3a-e**

M. P. reported (°C)	M. P. observed (°C)	Yield (%)	Time (min)	Ar	R'	R	Compd. No.
246-248 [18a]	244-245	91	20	C_6H_5	C_6H_5	Me	4a
	243-245	89	24	4-CH_3-C_6H_4	C_6H_5	Me	4b
	269-271	92	17	4-Br-C_6H_4	C_6H_5	Me	4c
265-267 [18a]	262-264	90	21	C_6H_5	H	Me	4d
	279-281	88	27	4-CH_3O-C_6H_4	H	Me	4e
	210 (dec.)	91	18	4-Br-C_6H_4	H	Me	4f
	110 (dec.)	89	23	C_6H_5	C_6H_5	H	4g
	235 (dec.)	87	25	4-CH_3-C_6H_4	C_6H_5	H	4h
	228-230	92	20	4-Cl-C_6H_4	C_6H_5	H	4i
	254 (dec.)	88	24	C_6H_5	H	H	4j
	269-271	87	29	4-CH_3-C_6H_4	H	H	4k
	296-298	91	22	4-Cl-C_6H_4	H	H	4l
>300 [18b]	>300	90	36	C_6H_5	CH_2-COOEt	Me	4m

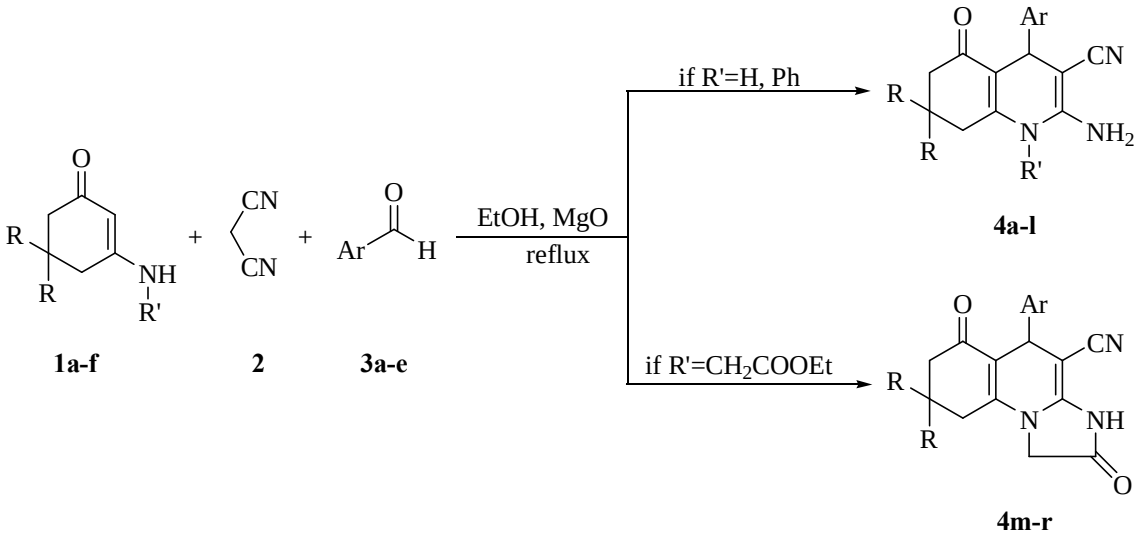
>300 [18b]	>300	89	40	4-CH_3-C_6H_4	CH_2-COOEt	Me	4n
>300 [18b]	>300	92	33	4-Cl-C_6H_4	CH_2-COOEt	Me	4o
274-275 [18b]	270-272	89	40	C_6H_5	CH_2-COOEt	H	4p
287-289 [18b]	283-285	87	43	4-CH_3-C_6H_4	CH_2-COOEt	H	4q
290-291 [18b]	289-290	90	35	4-Cl-C_6H_4	CH_2-COOEt	H	4r



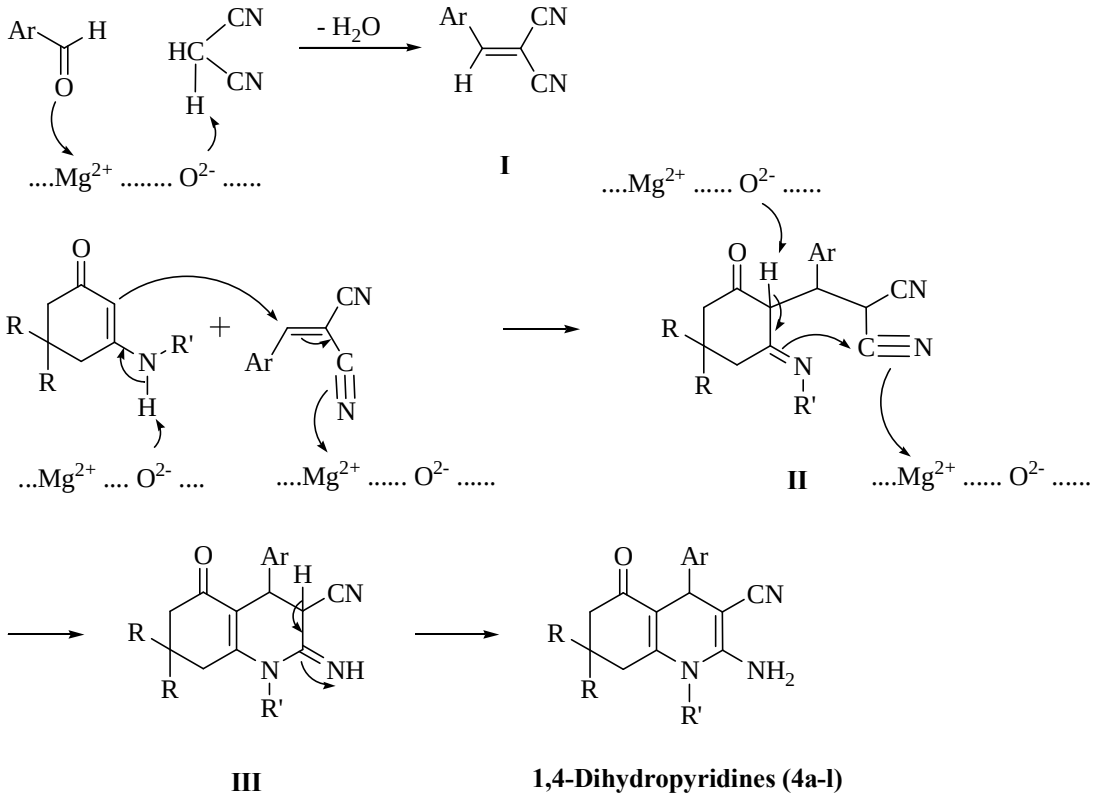
Nifedipine **Amlodipine** **Oxodipine**

Antiallergic activity **Anxiolytic activity**

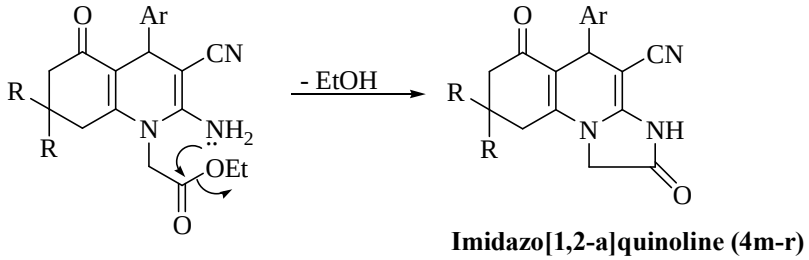
Scheme 1: Some of the marketed drug preparation containing 1,4-dihydropyridine moieties and some of imidazo[1,2-*a*]quinoline derivatives with pharmacological properties



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



Scheme 3: The suggested mechanism for the synthesis of 1,4-dihydropyridine derivatives (**4a-l**)



Imidazo[1,2-a]quinoline (4m-r)

Scheme 4: The suggested mechanism for the synthesis of imidazo[1,2-*a*]quinoline derivatives (**4m-r**)