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One-pot, three-component, selective synthesis of the polyfunctionalized 4*H*-pyran and 4*H*-benzo[*b*]pyran derivatives in the presence of a highly efficient molecular sieve-supported zinc catalyst

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Abstract: A series of pentasubstituted 4*H*-pyrans and tetrahydrobenzo[*b*]pyrans are synthesized with excellent yields via a one-pot condensation of aromatic aldehydes, malononitrile, and a dicarbonyl compound, ethyl acetoacetate, acetyl-acetone or dimedone, in the presence of 4 Å molecular sieve modified with zinc(II) as heterogeneous catalyst, in ethanol. The process offers numerous advantages, such as better yield, short reaction time, and mild reaction conditions. The catalyst's preparation is simple and it could be reused while still maintaining its activity.

Keywords: heterogeneous catalysis; molecular sieve; multicomponent reactions; polyfunctionalized 4*H*-pyrans and 4*H*-benzo[*b*]pyrans; zinc.

1 Introduction

The synthesis of the polyfunctionalized 4*H*-pyrans and 4*H*-benzo[*b*]pyrans is a popular research field as these are constituents of several natural products [1, 2]. These compounds possess a wide range of biological and pharmaceutical properties [3–6], and these heterocycles are utilized as antibacterial, anticancer, anti-coagulant, anti-anaphylactic and antispasmolytic agents, among others [7–11]. Moreover, 4*H*-pyran derivatives are considered as potential calcium channel antagonists [12] because their structure is similar to that of the biologically active 1,4-dihydropyridines. Apart from their own biological activities, the 4*H*-pyrans can serve as useful intermediates for the synthesis of various heterocyclic compounds, such as the pyrano[2,3-b]pyridine derivatives [13], polyazanaphthalenes [13], pyrano[2,3-c]pyrazoles [13], pyrano[2,3-d]pyrimidines [13, 14], and pyridin-2-ones [15, 16], which are also potentially biologically active.

Initially, the 2-amino-4*H*-pyran derivatives were synthetized through the cyclization reaction of the arylidenmalononitriles and β-dicarbonyl compounds in the presence of triethylamine [17] or piperidine [13]. One can still find examples for this approach nowadays. For example, Wang and co-workers [18] gained pyran derivatives with good yields using arylmethylidene-malononitriles and acetoacetic esters in aqueous media catalyzed by triethylbenzylammonium chloride (TEBAC). The conventional methods for the synthesis of the 4*H*-benzo[*b*]pyrans involve the condensation of aromatic aldehydes, malononitrile, and dimedone in refluxing acetic acid [19] or the condensation of dimedone with α-cyano-cinnamonitriles in the presence of piperidine in ethanol [20].

A more convenient and atom-efficient approach for the preparation of the 4*H*-pyran and 4*H*-benzo[*b*]pyran derivatives is the one-pot, three-component reaction of aldehydes, malononitrile, and a compound with an active methylene group. The multicomponent reactions (MCRs) are valuable tools in the hands of organic chemists, as they have numerous advantages over classical reaction strategies: they provide complex molecules in a single step and within shorter reaction times as well as yield less side products. The MCRs are also advantageous because they are more practical and environment-friendly compared with other options. Several methods have been developed for the one-pot, three-component, homogeneous catalytic synthesis of the 4*H*-pyrans, such as piperidine [21], DBU in water [22], L-proline [23], NaOH in water [24], LiBr in water [25], tetramethylammonium hydroxide [26], and sodium acetate combined with microwave irradiation [27]. The use of ionic liquids alone [28–30] or in combination with microwave irradiation [31] as well as deep eutectic solvents [32] have also been reported for this conversion.

Heterogeneous catalytic methods offer several advantages compared to homogeneous ones as the catalyst can be easily recovered from the reaction mixture by simple filtration and can often be reused, thus making the process both economically and environmentally more advantageous. Recently, several heterogeneous catalysts have

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been elaborated for the synthesis, such as the Cu(II) oxymetasilicate [33], the MgO-SnO_2 solid superbase [34], the silica-bonded N-propylpiperazine sodium n-propionate (SBPPSP) [35], the silica nanoparticles [36], $SnCl_2$/nano-SiO_2 [37], nano-crystalline ZnO [38], Amberlyst A21 [39], and the strong base Mg/La mixed oxide [40]. After all, some of these methods have disadvantages, such as long and complex preparation of the catalyst [34, 35, 37], use of toxic or harmful reagents [35, 37], and lower yields [34]. Hence, the elaboration of novel routes for the synthesis of the 4*H*-pyran and 4*H*-benzo[*b*]pyran derivatives may be important.

The aim of our research group is to develop new heterogeneous catalytic methods for the preparation of different organic compounds using supported metal catalysts. In this work, palladium [41–44], nickel [45], copper [46–49], titanium [50], or lanthanum [51] on different supports (4 Å molecular sieve, Mg:La 3:1 mixed oxide) were used with good yields in organic syntheses. Here, we present a simple method for the three-component synthesis of the polyfunctionalized 4*H*-pyrans and 4*H*-benzo[*b*]pyrans in the presence of a 4 Å molecular sieve (4A)-supported, slightly basic zinc catalyst (Zn^{2+}/4A).

2 Materials and methods

The morphology of the catalyst samples was investigated by a JEOL 6380LVa type scanning electron microscope (JEOL, Tokyo, Japan). Elemental mapping was accomplished using the energy-dispersive X-ray detector of the equipment. The samples were fixed by conductive double-sided carbon adhesive tape. Accelerating voltage and working distance were set at 15–30 kV and 10–12 mm, respectively.

The nitrogen adsorption/desorption isotherms were measured at –196°C with a computer-controlled Nova 200e (Quantachrome) instrument. The apparent surface area (S_{BET}) was calculated using the Brunauer-Emmett-Teller (BET) model. The total pore volume (V_t) was derived from the amount of vapor adsorbed at $p/p_0 \rightarrow 1$, assuming that the pores were already filled with liquid adsorbate. The micropore volume (W_0) was derived from the Dubinin-Radushkevich (DR) plot. Prior to the adsorption measurement, the samples were evacuated at 110°C for 24 h. Next, ^{1}H and ^{13}C NMR spectra were generated on a BRUKER Avance-500 instrument in $CDCl_3$ or DMSO-d_6. TMS served as the internal standard. The melting points (uncorrected) were determined on a Gallenkamp apparatus. All compounds and solvents were purchased from Merck Hungary Ltd.

2.1 Preparation of the catalyst

The preparation of the catalyst was effectuated as described [50, 51]; 1 mmol of anhydrous $ZnCl_2$ was dissolved in 100 ml of deionized water and stirred with 1 g 4 Å molecular sieve (4A) at room temperature for 24 h. The solid was filtered, washed with deionized water and with acetone, and then dried in an oven at 150°C for 1 h.

2.2 Determination of the pH of the catalyst

The catalyst (1 g) was stirred in 30 ml deionized water under continuous measuring of the pH. The values were accepted after reaching a constant value at least during 10 min (see also [50, 51]).

2.3 Typical reaction conditions

The general procedure for the one-pot synthesis of 2-amino-4*H*-pyrans and 4*H*-benzo[*b*]pyrans: Aldehyde (1 mmol), malononitrile (1.2 mmol), a β-dicarbonyl compound (ethyl acetoacetate, acetylacetone or dimedone) (1 mmol) and Zn^{2+}/4A (0.1 g) were stirred in a 10-ml flask in refluxing ethanol (3 ml) for 4 h. Then the solid was filtered and washed with acetone, after which the filtrate was evaporated. The residue was suspended in diethyl ether, and the precipitated solid was filtered and subjected to ^{1}H NMR spectroscopy. All products yielded satisfactory ^{1}H NMR data. The spectral data of the known compounds were identical with those reported in the literature.

2.4 Representative physical and spectroscopic data of the products

6-Amino-4-(4-chlorophenyl)-5-cyano-2-methyl-4*H*-pyran-3-carboxylic acid ethyl ester (4e): Off-white solid. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 1.11 (t, *J* = 7 Hz, 3H), 2.37 (s, 3H), 4.03–4.05 (q, *J* = 5 Hz, 2H), 4.43 (s, 1H), 4.50 (s, 2H), 7.14 (d, *J* = 8 Hz, 2H), 7.27 (d, *J* = 8 Hz, 2H).

6-Amino-5-cyano-4-(2-fluorophenyl)-2-methyl-4*H*-pyran-3-carboxylic acid ethyl ester (4f): Off-white solid. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 1.09 (t, *J* = 7 Hz, 3H), 2.38 (s, 3H), 4.01–4.04 (m, 2H), 4.51 (s, 2H), 4.75 (s, 1H), 6.98–7.02 (m, 1H), 7.06–7.09 (m, 1H), 7.14–7.20 (m, 2H). ^{13}C NMR (500 MHz, $CDCl_3$): δ = 13.79, 18.51, 32.76 (d), 32.80 (d), 59.60, 60.61, 106.43, 115.50 (d), 119.23, 124.24 (d), 128.70 (d), 129.81, 129.86, 130.90 (d), 158.20 (d), 165.73. Anal. Calcd. for $C_{16}H_{15}FN_2O_3$: C 63.51, H 4.96, N 9.26%, found: C 63.55, H 4.91, N 9.32%.

5-Acetyl-2-amino-3-cyano-6-methyl-4-phenyl-4*H*-pyran (4l): Yellow solid. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 2.05 (s, 3H), 2.30 (s, 3H), 4.43 (s, 1H), 4.87 (s, 2H), 7.19–7.25 (m, 3H), 7.31–7.34 (m, 2H). ^{13}C NMR (500 MHz, $CDCl_3$): δ = 18.70, 29.31, 30.89, 35.66, 59.13, 114.21, 119.07, 127.84, 128.64, 129.82, 130.00, 132.41, 140.76, 155.75, 158.26, 198.45. Anal. Calcd. for $C_{15}H_{13}ClN_2O_2$: C 62.34, H 4.50, N 9.69%, found: C 62.34, H 4.57, N 9.64%.

5-Acetyl-2-amino-3-cyano-6-methyl-4-(4-nitrophenyl)-4*H*-pyran (4p): Yellow solid. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 2.11 (s, 3H), 2.35 (s, 3H), 4.58 (s, 1H), 5.37 (s, 2H), 7.39 (d, *J* = 8.5 Hz, 2H), 8.18 (d, *J* = 8.5 Hz, 2H). ^{13}C NMR (500 MHz, $CDCl_3$): δ = 19.12, 30.17, 39.38, 59.36, 114.62, 118.89, 124.25, 128.31, 147.10, 150.70, 156.36, 158.20, 197.51. Anal. Calcd. for $C_{15}H_{13}N_3O_4$: C 60.20, H 4.35, N 14.04%, found: C 60.27, H 4.33, N 14.01%.

2-Amino-7,7-dimethyl-4-(4-chloro-phenyl)-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (5d): White solid. ^{1}H NMR (500 MHz, DMSO-d_6): δ = 0.95 (s, 3H), 1.03 (s, 3H), 2.10 (d, *J* = 9.9 Hz,

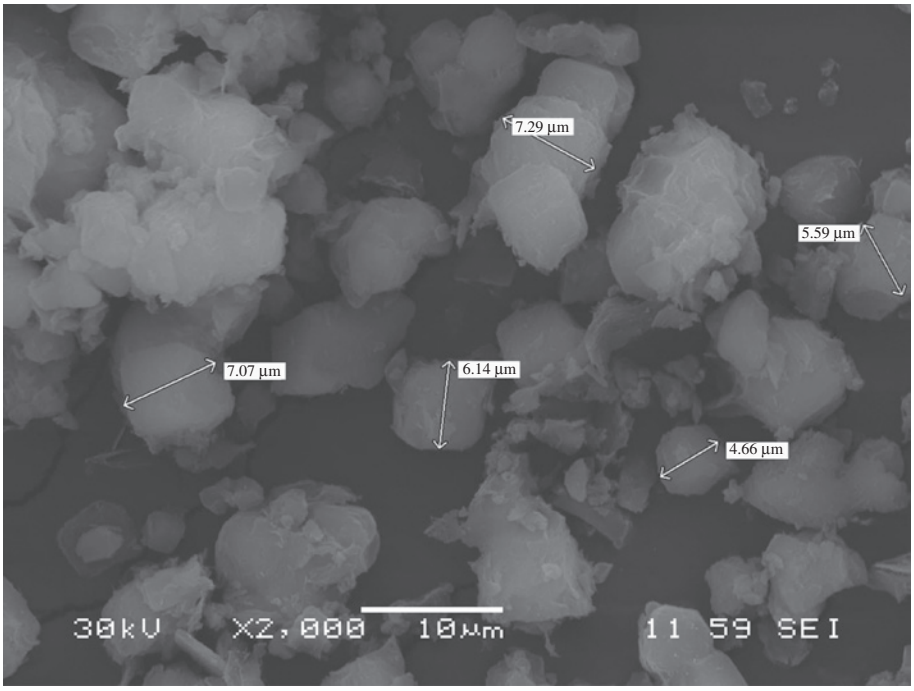


Figure 1: SEM image of the catalyst.

1H), 2.25 (d, J = 9.9 Hz, 1H), 2.50 (s, 2H), 4.19 (s, 1H), 7.04 (s, 2H), 7.17 (d, J = 5.1 Hz, 2H), 7.34 (d, J = 5.1 Hz, 2H).

2-Amino-7,7-dimethyl-4-(3-nitrophenyl)-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (5g): White solid. ^{1}H NMR (500 MHz, DMSO-d_6): δ = 0.96 (s, 3H), 1.05 (s, 3H), 2.12 (d, J = 9.6 Hz, 1H), 2.27 (d, J = 9.6 Hz, 2H), 2.55 (s, 2H), 4.42 (s, 1H), 7.16 (s, 2H), 7.62–7.68 (m, 2H), 7.98 (s, 1H), 8.08 (d, J = 4.8 Hz, 1H) (see Supplementary Material).

3 Results and discussion

The scanning electron microscopy (SEM) investigation of the Zn^{2+}/4A catalyst showed the peculiar cuboctahedron shape of the molecular sieve support as can be seen on Figures 1 and 2. The average size of the well-defined particles ranged from 6 to 7 µm. EDS determination showed 38.89 w/w % zinc on the surface (Figure 3). The zinc content determined by ICP-OES was 5.98 w/w %, indicating that the zinc particles were distributed on the surface of the support. The even distribution of zinc can be verified by the SEM image (Figure 2 lower). The nitrogen adsorption measurements showed the significant diminution of the surface of the support. The specific surface of 4A decreased from 800 to 332 m^2/g. The total pore volume of the molecular sieve varied from 0.3 to 0.201 cm^3/g, and the micropore volume was 0.266 cm^3/g.

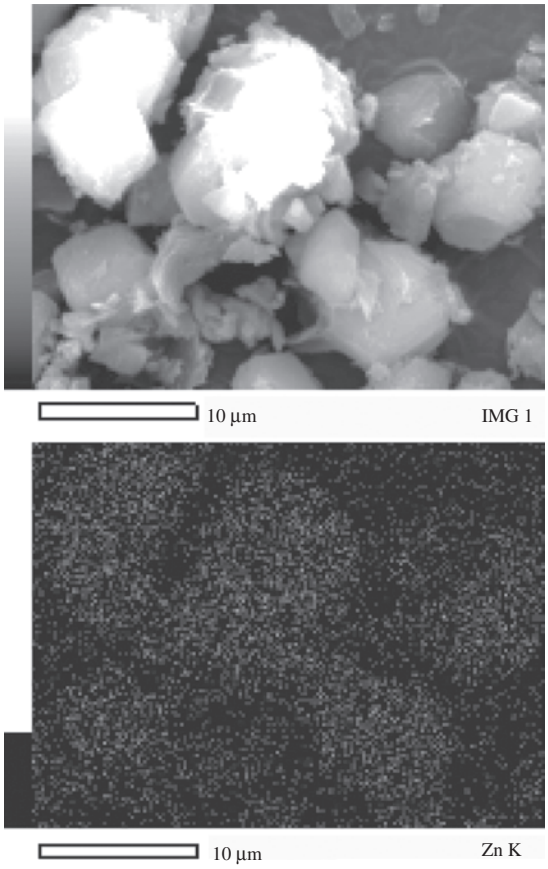


Figure 2: SEM image of the catalyst (upper), distribution of the zinc on the particle (lower).

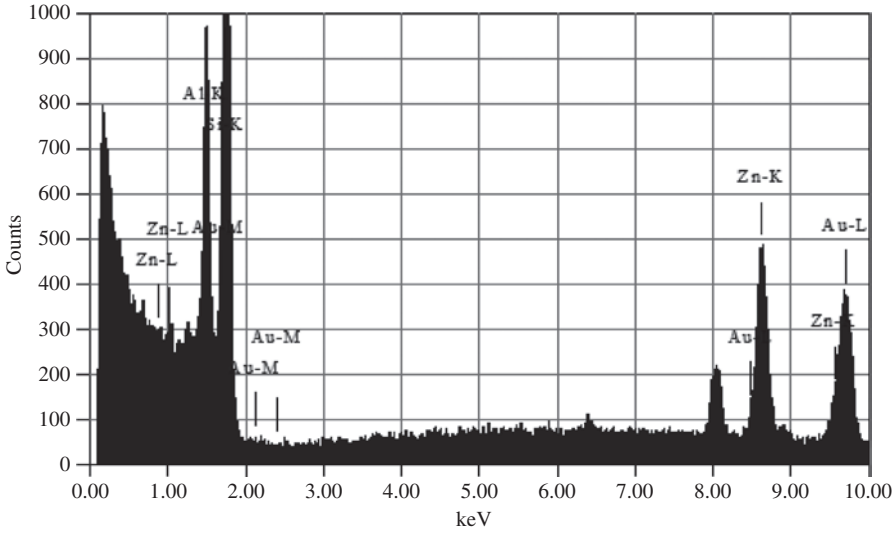
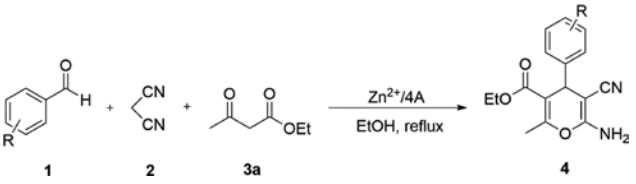


Figure 3: The standard quantitative analysis of the catalyst (magnification: 4000×).

The catalyst was slightly basic in nature, with a pH value of 9.71 (see Section 2.2).

Continuing our work aiming to develop new heterogeneous catalysts and testing them in different organic reactions, we tested the applicability of our Zn^{2+}/4A catalyst in the MCR of aldehydes, malononitrile, and ethyl acetoacetate (Table 1). In our previous experiments (e.g. [50, 51]) generally 0.1 g catalyst/1 mmol substrate was the most efficient ratio in the case of the 4A-supported catalysts, thus we used the same amount. In the literature, ethanol has been mainly used in this cyclization; accordingly, we used ethanol in the current study as well. In toluene and acetonitrile, complex reaction mixtures were obtained. In ethanol, the desired 4*H*-pyran derivatives were formed with excellent yield. The reagents were applied in a 1:1:1.2 (ester:aldehyde:nitrile) molar ratio, and 0.1 g Zn^{2+}/4A catalyst was used in 3 ml ethanol. The products obtained were subjected to ^{1}H NMR spectroscopy.

Table 1: The reactions of different aromatic aldehydes, malononitrile, and ethyl acetoacetate.[a]



Entry	R	Product	Yield (%)[b]	Mp (°C)
1	H	**4a**	95	166–168 (177 [52])
2	3-Br	**4b**	99	155–156 (156–158 [21])
3	4-Br	**4c**	96	161–162 (172–173 [24])
4	2-Cl	**4d**	94	178–180 (179–181 [27])
5	4-Cl	**4e**	94	169–170 (171–172 [27])
6	2-F	**4f**	93	158–160 (–)
7	3-MeO	**4g**	95	122–124 (–)
8	3-NO_2	**4h**	97	182–183 (182–184 [33])
9	4-NO_2	**4i**	97	171–172 (175–176 [27])

[a]Reaction conditions: 1 mmol aldehyde, 1.2 mmol malononitrile, 1 mmol ethyl acetoacetate, 0.1 g catalyst, 3 ml ethanol, reflux temperature, 4 h.
[b]Isolated yields.

One can find examples in the literature for the use of zinc containing catalysts in the synthesis of these compounds [16, 38]. For example, Zhang and co-workers [16] declared that the selective synthesis of 2-pyridinones and 2-amino-4*H*-pyrans were always a challenge because of the bad chemoselectivity of the cyclocondensation reaction of the carbonyls and methylene compounds that usually gave the mixture of the two products (Figure 4). They elaborated a Zn-SSA catalyzed method for the synthesis of the 2-pyridinones through the cleavage and recyclization of the corresponding pyran derivatives. In our case only the appropriate pyran derivatives were obtained. No trace of the 2-pyridinone side products could be observed in the ^{1}H NMR spectra.

We also examined the reactivity of the heteroaromatic aldehydes, but in the reaction of both the furan-2-carbaldehyde and thiophene-2-carbaldehyde, the yield of the desired product was below 50% and several side products were also formed. Replacing ethyl acetoacetate by acetylacetone, the expected products were also obtained with excellent yields, as shown in Table 2. In some cases, based on a TLC-examination, the reaction mixture contained small amounts of side products originating from an aldol-type reaction of acetylacetone, but

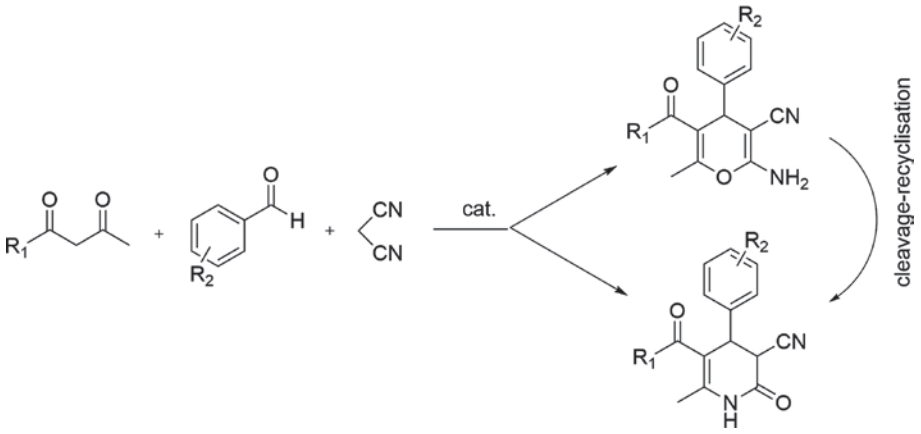
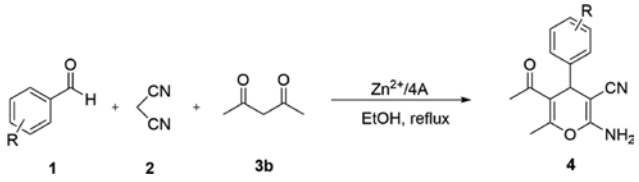


Figure 4: Reaction route described by Zhang et al. [16].

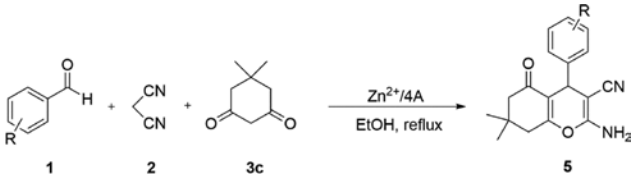
Table 2: The reactions of different aromatic aldehydes, malononitrile, and acetylacetone.[a]



Entry	R	Product	Yield (%)[b]	Mp (°C)
1	H	**4j**	80	122–124 (158–160 [28])
2	4-Br	**4k**	92	62–64 (–)
3	2-Cl	**4l**	90	131–132 (–)
4	4-Cl	**4m**	87	118–120 (154 [38])
5	2-F	**4n**	98	128–130 (–)
6	3-MeO	**4o**	98	144–145 (–)
7	4-NO_2	**4p**	98	152–154 (–)

[a]Reaction conditions: 1 mmol aldehyde, 1.2 mmol malononitrile, 1 mmol acetylacetone, 0.1 g catalyst, 3 ml ethanol, reflux temperature, 4 h.
[b]Isolated yields.

Table 3: The reactions of different aromatic aldehydes, malononitrile and dimedone.[a]



Entry	R	Product	Yield (%)[b]	Mp (°C)
1	H	**5a**	98	221–222 (224–225 [53])
2	3-Br	**5b**	91	218–220 (224–226 [54])
3	4-Br	**5c**	85	206–208 (207–209 [26])
4	4-Cl	**5d**	98	204–205 (203–206 [55])
5	2-F	**5e**	98	221–222 (232–233 [56])
6	3-MeO	**5f**	92	193–194 (188–189 [57])
7	3-NO_2	**5g**	94	210–212 (210–212 [58])
8	4-NO_2	**5h**	91	138–140 (151–152 [56])

[a]Reaction conditions: 1 mmol aldehyde, 1.2 mmol malononitrile, 1 mmol dimedone, 0.1 g catalyst, 3 ml ethanol, reflux temperature, 4 h.
[b]Isolated yields.

these could easily be eliminated during the workup of the reaction mixture. In the reaction of aldehydes, malononitrile, and dimedone, the appropriate tetrahydro-benzo[*b*]pyran derivatives were also obtained with excellent yield (Table 3).

No significant substituent effect was observed in the reactions; the aromatic aldehydes containing both electron-withdrawing groups (e.g. halide, nitro group) and the electron-donating groups (e.g. alkoxy group) gave nearly the same results in short reaction times in every case examined.

The workup of the reaction mixtures was very simple, in which the catalyst was filtered out and washed with both ethanol and acetone. The residue obtained after the evaporation of the filtrate was diluted with diethyl ether, after which the precipitated solid was filtered.

We also studied the reusability of the zinc catalyst in the reaction of 3-nitrobenzaldehyde (**1h**), malononitrile (**2**), and ethyl acetoacetate (**3a**) in ethanol. After a 4-h reflux, the reaction mixture was worked up as described above, the catalyst was heated at ~150°C for 1 h. It was reused in two more runs without considerable decrease in its activity. The isolated yields for the three successive runs were 97% each time, clearly demonstrating the practical recyclability of this catalyst.

The potential leaching of the zinc from the surface of the support was also investigated. After the filtration of the catalyst from the reaction mixture, the filtrate

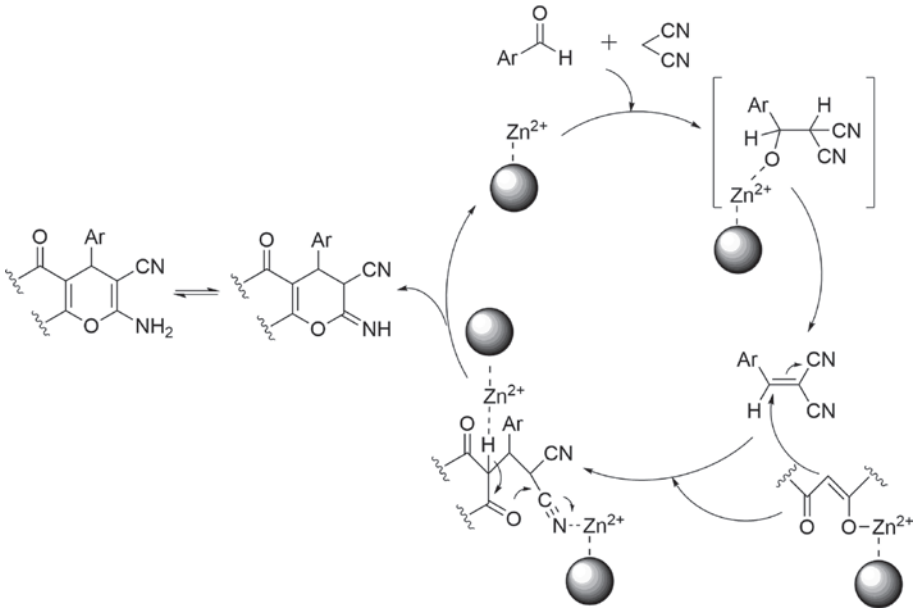


Figure 5: Proposed mechanism of the reaction.

was examined by XRF. No appreciable amount of zinc could be detected, verifying both the real heterogeneous catalytic reaction and the reusability results of the catalyst.

We propose a plausible mechanism for the formation of the 4*H*-pyran derivatives involving three consequent steps, a Knoevenagel condensation, then a Michael addition, and an intramolecular ring closure in the presence of a 4A molecular sieve-supported zinc catalyst (Figure 5). We assume a mechanism that is similar to the described one by Bhattacharyya et al. [38]. The role of the zinc might be to facilitate the reaction steps through the coordination of the heteroatoms in the transition states.

4 Conclusions

The slightly basic zinc on 4 Å molecular sieve catalyst can be used successfully for the one-pot, three component, selective synthesis of the polysubstituted 4*H*-pyrans and tetrahydro-benzo[*b*]pyrans. The preparation of the catalyst is simple and it can be reused without the loss of activity.

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Conflict of interest statement: The authors declare that they have no conflict of interest regarding this article.

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