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Zinc Oxide Nanoparticles Promoted Highly Efficient and Benign Synthesis of 3,4-Dihydropyrimidine-2(1*H*)-one/thione Derivatives

VINAYAK R. BAGUL

Department of Chemistry, Mahatma Gandhi Vidyamandir's Arts, Science and Commerce College, Surgana (Affiliated to S. P. P. University, Pune), Nasik-422211, India.

Abstract

Using the synthetic potential of recyclable zinc oxide (ZnO) nanoparticles (NPs), a proficient, elegant, and rapid one-pot synthesis of a variety of 3,4-dihydropyrimidine-2 (1*H*)-one/thione derivatives from the 1,3-dicarbonyl compound, urea/thiourea, and various aromatic aldehydes have been unveiled in the present research. The ZnONPs were synthesized by the co-precipitation method. The powder X-ray diffraction method was employed for the determination of the crystallite size of the synthesized ZnONPs. The hexagonal phase was obtained in the XRD pattern of the synthesized ZnO NPs with an average crystallite size of 25 nm.The current synthetic strategy offers excellent yields, a short reaction time, favorable reaction conditions, easy transformation, non-chromatographic product purification, and catalyst recyclability. Furthermore, the catalyst could be retrieved and reused without losing any of its catalytic activity. As a result, this elegant protocol is an adequate method for dihydropyrimidinone/thione synthesis.

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Introduction

Pietro Biginelli reported the Biginelli reaction, a three-component reaction, in 1893. P. Biginelli reported the acid-catalyzed synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones (DHPMs) through a multicomponent reaction (MCR) of an aromatic aldehyde, ethyl acetoacetate, and urea. The Biginelli adduct is formed in the classic version of this reaction, which involves an acid-catalyzed three-component reaction between benzaldehyde, ethylacetoacetate, and urea in ethanol at reflux. DHMPs and related compounds demonstrate potential in the treatment of cancer,[1-3] calcium channel inhibition,[4-5] antioxidant,[6,7] antimicrobial,[8-10] and anti-inflammatory function.[11] The noteworthy medicinal profile of the DHPMs has prompted the synthesis of new Biginelli reaction methodologies.

CONTACT Vinayak R. Bagul bagulvinayak99@gmail.com Department of Chemistry, Mahatma Gandhi Vidyamandir's Arts, Science and Commerce College, Surgana (Affiliated to S. P. P. University, Pune), Nasik-422211, India.

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In the past few years, researchers have focused on green concepts in order toto establish environmentally sustainable synthetic methodologies for synthesising a wide range of organic compounds.[12-21] Lewis acid catalysis,[22] polymer-supported,[23] ionic liquids,[24] microwave-assisted synthesis,[25] solvent-free techniques,[26, 27] and other similar methods have all been published in the last few years. However, most reported dihydropyrimidinone synthesis strategies to have drawbacks such as intolerant reaction conditions, long reaction times, unsatisfactory yields, higher reaction temperatures, and the use of expensive catalysts that are not recognized as environmentally friendly.

The metal oxide NPs are thought to be more reactive as a catalyst because they have a larger surface area that is more readily accessible to substrate molecules, resulting in better catalytic action.[28, 29] Nanocatalysts have exceptional properties due to their high surface area to volume ratio, which makes them superior to mass materials in terms of catalysis. To achieve organic reactions in a short amount of time with higher yields, a variety of nano-catalysis techniques have been used.[30-32] The ZnO NPs were reported as anexcellent catalysts for gas sensing,[33, 34] multicomponent reactions,[35-37] and dye degradation[38, 39] applications.Following that, here an attempt is made to develop a better catalysts for the synthesis of dihydropyrimidinones that was both easy to use and rendered high yields. On this basis, the present study reports ZnONPs as an effective catalyst for the synthesis of dihydropyrimidinones.

Experimental

Materials

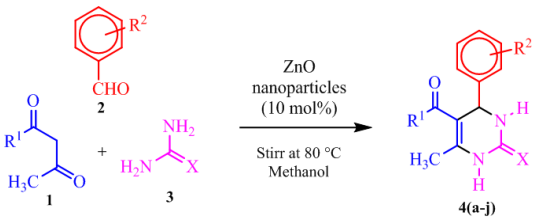
$Zn(NO_3)_2•6H_2O$ (purity: >98%), NaOH (purity: >98%), aromatic aldehydes (purity: >97%), ethyl acetoacetate(purity: >97%), acetylacetone (purity: >97%), urea(purity: >98%), and thiourea (purity: >97%), were purchased from Sigma Aldrich, SDFCL and Avra synthesis and were used without any further purification. Melting points were determined in open capillaries and are uncorrected. ^{1}H NMR and ^{13}C NMR spectra were recorded with a Bruker using $CDCl_3$ solvent. The powder X-Ray Diffraction (XRD) technique was used to investigate the average crystallite size of the prepared nanomaterials on a Bruker D8 Advance X-Ray Diffraction instrument.

Synthesis and characterization of ZnONPs

The co-precipitation method was used to produce the ZnO NPs. 1M of $Zn(NO_3)_2•6H_2O$ was dissolved in distilled water and the solution was kept under constant stirring using a magnetic stirrer for 30 minutes. Following full dissolution, a 2M sodium hydroxide solution was added while stirring constantly. The reaction was allowed to continue for two hours. The white solution was produced at the end of the reaction and was allowed to settle for 12 hours.The precipitate was then washed with distilled water several times before being dried in an oven. In a muffle furnace, the obtained product was kept at 400°C for 3 hours to obtain ZnONPs.

General Procedure for Preparation of 3,4-dihydropyrimidine-2(1H)-ones/thiones

A mixture of aromatic aldehyde (0.01mol), urea/ thiourea (0.01mol), and ethyl acetoacetate/ acetylacetone (0.01mol) were taken in a flat bottom flask. 15 mL methyl alcohol was added to the solvent, and the resulting mixture was stirred until a clear solution was obtained. Then ZnONPs (10 mol %) was added. This mixture was heated on a magnetic stirrer at 80°C with continuous stirring and then it was allowed to cool at room temperature after completion of the reaction (monitored by TLC). Afterward, ethyl acetate was introduced and the extract was processed to acquire crude products free of ZnO NPs. The ethyl acetate extract was dried over sodium sulphate before being evaporated with a rotary evaporator. Spectroscopic data such as ^{1}HNMR and ^{13}C NMR spectral data were used to validate the structure of all synthesized products.



Scheme 1 ZnONPs catalyzed dihydropyrimidine-2(1*H*)-one/thione synthesis

Physicochemical and Spectral Data of the Selected Compounds

6-methyl-2-oxo-4-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate

Yield: 96 %, Colour: white solid, m.p. : 232°C; ^{1}HNMR (400MHz, DMSO-d6) δ (ppm): 1.10 (t, *J* = 7.1 Hz, 3H), 2.27 (s, 3H), 4.00 (q, *J* = 7.1 Hz, 2H), 5.14 (s, 1H), 7.27-7.35 (m, 5H), 7.75 (s, 1H), 9.18 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d6); δ (ppm) 14.54, 18.24, 54.43, 59.65, 99.73, 126.71, 127.73, 128.86, 145.34, 148.83, 152.60, 165.81.

Ethyl-4-(4-isopropylphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate

Yield: 90 %, Colour: white solid, m.p. : 176°C; ^{1}HNMR (400MHz, DMSO-d6); δ (ppm):1.19 (t, *J* = 7.1 Hz, 3H) 1.23 (d, *J* = 7.2 Hz, 6H), 2.33 (s, 3H), 2.85 (sept, *J* = 7.2 Hz, 1H), 4.07 (q, *J* = 7.1 Hz, 2H), 5.37 (d, 1H), 5.91 (s, 1H), 7.17(d, *J* = 8.2 Hz, 2H), 7.25 (d, *J* = 8.2 Hz, 2H), 8.55 (s, 1H);^{13}C NMR (100 MHz, DMSO-d6) δ (ppm):14.54, 18.24, 54.43, 59.65, 99.73, 126.71, 127.73, 128.86, 145.34, 148.83, 152.60, 165.81.

5-acetyl-4-(4-chlorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one

Yield: 92 %, Colour: white solid, m.p. : 220°C; ^{1}HNMR (400MHz, DMSO-d6); δ (ppm): 2.11 (s, 3H), 2.30 (s, 3H), 5.25 (d, *J* = 3.8Hz, 1H), 7.40 (d, *J* = 8.4 Hz, 2H), 7.20 (d, *J* = 8.4 Hz, 2H), 9.78 (s, 1H), 10.30 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d6) δ (ppm): 18.85, 31.07, 53.53, 110.85, 128.94, 129.15, 132.78,142.33, 145.49, 174.72, 195.19.

Ethyl-4-(4-hydroxy-3-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate

Yield: 89 %, Colour: white solid, m.p. : 232°C; ^{1}HNMR (400MHz, DMSO-d6) δ (ppm): 1.09 (t, *J* = 7.2 Hz, 3H). 2.20 (s, 3H), 3.96 (q, *J* = 7.2 Hz, 2H), 6.75 (d, *J* = 2.1 Hz, 1H), 5.03 (d, *J* = 3.4 Hz, 1H), 6.67 (d, *J* = 8.1 Hz, 1H), 6.57 (dd, *J* = 8.1, 2.1 Hz, 1H), 8.89 (s, 1H), 9.09 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d6) δ (ppm):14.68, 18.26, 54.06, 56.05, 59.65, 100.04, 111.35, 115.78, 118.78, 136.43, 146.29, 147.75, 148.44, 152.76, 165.97

Result and Discussion

Powder XRD is an analytical method which is used to determine the phase of a crystallite substance and can also be used to determine unit cell dimensions. The hexagonal phase is visible in the XRD pattern of the synthesized ZnONPs (Figure 1). The synthesized ZnONPs showed sharper and stronger diffraction peaks, according to the XRD spectrum. The diffraction peaks at 31.65°, 34.32°, 36.14°, 47.43°, 56.46°, 62.74°, 66.18°, 67.79°, and 68.91° are correlated with the (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes, respectively, and are mentioned in Table 1. The (hkl) values are in good agreement with the ZnO standard cards (JCPDS file No: 79-2205). Debye Scherrer's formula is used to calculate the average crystallite size of the sample (D): $D = 0.9\lambda/\beta\cos\theta$, where, λ is the wavelength of the X-ray radiation, θ is the diffraction angle and β is the full width half maximum (FWHM) intensity. From the line broadening of the X-ray diffraction peak, the crystallite size of the NPs was determined using the formula above. The average crystallite size is 25 nm.

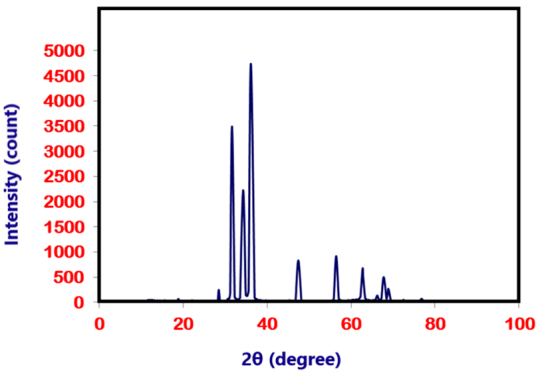


Fig.1: XRD pattern plot of synthesized ZnO NPs

Table 1: The observed and standard 2θ values of XRD data of synthesized ZnONPs

Observed 2θ	h kl
31.65	100
34.32	002
36.14	101
47.43	102
56.46	110
62.74	103
66.18	200
67.79	112
68.91	201

All of the experiments were carried out with an optimized catalyst concentration (Table 2). In a comparative study of the typical reaction of benzaldehyde, ethyl acetoacetate, and urea at 80°C, the best result was obtained using a catalyst concentration of 10 mol percent. There was no apparent loss of catalytic activity after recycling and reusing the catalyst for three runs in a phase. Surprisingly, I found that different substitution patterns of benzaldehydes produced similar amounts of 3,4-dihydropyrimidine-2(1*H*)-ones/thiones (Table 3). The disclosed method stands out as compared to previous synthetic approaches. (Table 4).

Conclusion

The catalytic activity of ZnO NPs in the Biginelli reaction for the one-pot synthesis of 3,4-dihydropyrimidine-2(1*H*)-one/thione derivatives was examined, and it was found that these ZnO NPs furnished dihydropyrimidinones with remarkable efficiency. A short reaction time, the use of a cost-effective, non-toxic, green catalyst, and clean reaction transformation with excellent yields are among the protocol's key highlights.

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Conflict of Interest

The author declares that he do not have any conflict of interest.

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