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Original Research

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Surfactant TBAB as a Catalyst for the Synthesis of 3, 4-Dihydropyrimidine Derivatives

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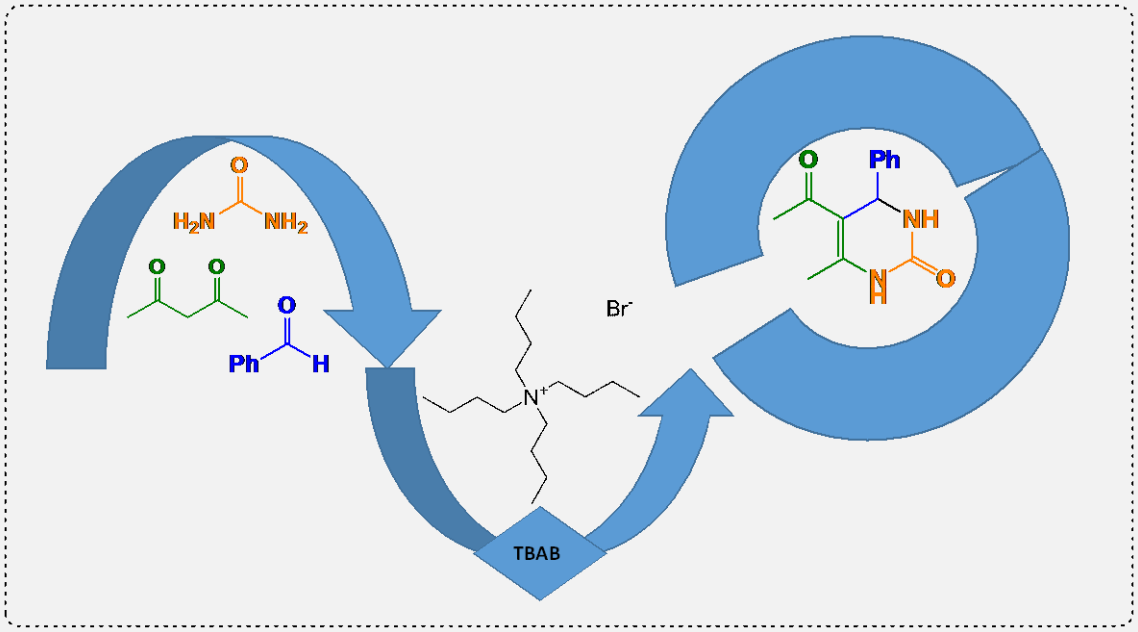
ABSTRACT: 3, 4-Dihydropyrimidine compounds are widely used in the pharmaceutical industry due to their biological, antibacterial, antiviral, anti-HIV and anti-tumor properties and other medicinal properties, and therefore, considering the practical importance of these compounds, providing new synthetic methods and synthesis New derivatives of them are necessary. In this research, the synthesis of new derivatives of these compounds using tetrabutylammonium bromide (TBAB) surfactant as a green and environmentally friendly catalyst is reported. From the condensation of β-dicarbonyl with urea or thiourea and aromatic aldehydes in the presence of catalyst TBAB at 80°C in solvent-free conditions, new dihydropyrimidine derivatives were substituted. The structure of the synthesized compounds was confirmed using spectroscopic data. The features of this method include high efficiency, mild conditions, single container, not need for solvent, and compatibility of the catalyst with the environment.



KEYWORDS: 3, 4-Dihydropyrimidine, tetrabutylammonium bromide, β-dicarbonyl, antibacterial, Solvent free.

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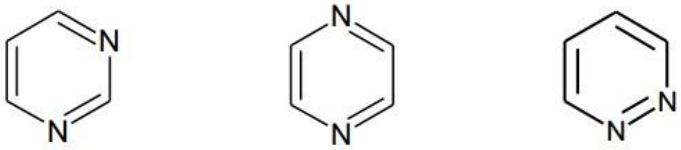
Introduction

Multicomponent reactions were discovered in 1850 by Strecker. The most important criteria for the efficiency and feasibility of a process is to minimize the number of synthesis steps and reaction purification steps [1]. Carrying out multi-component reactions is considered as a promising field, fundamental and a great success in chemistry [2]. In the multicomponent reaction technique, useful and valuable complex structures are synthesized in a very fast, efficient and effective way and with little time, without the need to separate the intermediates [3]. Therefore, by reducing the steps of synthesis compared to traditional methods, it will have a faster reaction and more efficiency [4]. This method allows us to avoid separating the limits of the means, changing the conditions or adding any additional reactivity [5]. Minimize the amount of waste and wastage of products and save energy, time and raw material (cost) to get the maximum benefit of synthetic products [6]. The attention and interest of organic chemists to multicomponent reactions has led to the development and progress of multicomponent reactions in the direction of greater reactivity and optimization [7]. For this reason, partial reactions are considered to be superior techniques in green chemistry; And it has been increasingly important in organic chemistry and medicinal chemistry, and has turned into one of the most effective and economical tools for simultaneous synthesis of compounds [8]. These reactions have been widely used for the simultaneous synthesis of many compounds under mild conditions in one pot due to their high speed and efficiency [9].

In 1893, an Italian chemist named Pietro Biginelli performed the cyclization reaction of an aldehyde with a -β-ketostere and urea in a one-pot reaction under reflux conditions in ethanol in an acidic environment, which led to the synthesis of dihydropyrimidine became [10]. Although the Biginelli reaction had a low yield at first and was limited to few derivatives, the biological properties of the products of this reaction made scientists look for ways to optimize the conditions, use new catalysts, and use various solvents to synthesize new derivatives [11]. Diazines are a group of compounds derived from benzene in which two carbon atoms in a six-membered ring have been replaced by two nitrogen atoms. All these compounds are aromatic. According to the position of nitrogen atoms, diazines include pyrimidine (1), pyrazine (2) and pyridazine (3) [12].



1 2 3

In pyrimidine, nitrogen atoms are located in positions 1 and 3. These three diazines are colorless compounds, stable and soluble in water [13]. The origin of the word pyrimidine dates back to 1884, when Pinerquind chose this name for pyrimidine due to its similarity to pyridine and amidine [14]. The electronic structure of pyrimidine is similar to benzene, in that it has 6 electrons and follows Hoeckel's law (4n+2), and because it is flat, it is known as an aromatic compound [15]. The first pyrimidine derivative was obtained by Brugnatelli in 1818, this compound, called alloxan, was formed from the oxo-reduction reaction of uric acid with nitric acid. In 1878, barbituric acid was synthesized from the condensation of

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malonic acid with urea [16]. This compound has antidiabetic properties in animals. Pyrimidines are more important than other compounds of diazines, because most pyrimidine derivatives play a key role in biological processes [17]. For this reason, there are many synthetic methods for pyrimidine nuclei. One of the most common methods of pyrimidine synthesis is the condensation of a three-carbon unit with species having an N-C-N bond [18]. This general method is very versatile because there are many different molecules that perform cyclization. For example, the three-carbon unit may be a β-dialdehyde, β-ketoester, β-diketone, malonic ester, β-ketonitrile, or other such compounds. Also, the nitrogen-containing unit may be urea, thiourea, guanidine or amidine [19]. Due to the high electronegativity of two nitrogen atoms, unsubstituted diazines are resistant to electrophilic substitution reactions [20]. The electronic structure of pyrimidine is similar to 3,1-dinitrobenzene, because this compound has two nitrogens in its structure, which, like the nitro groups in benzene, causes the electron deficiency of this compound, and its electron deficiency is felt mostly on carbons 2, 4, and 6 [21]. Therefore, this ring performs the nucleophilic substitution reaction easily because the negative charge resulting from the nucleophilic reaction is placed on the electronegative nitrogen atoms [22]. To perform electrophilic substitution reactions, it is necessary to have strong electron-donating groups such as hydroxy and amino groups on the pyrimidine ring [23]. The electrophilic substitution reaction takes place at carbon 5 because this position has the least electron deficiency [24]. To perform a successful nitration, the presence of at least two electron-donating groups is necessary [25]. The position of these groups is important in nitration [26]. Positions 2, 4 and 6 of pyrimidine are electron poor due to the strong electrophilic effect of nitrogen atoms, so they are suitable for carrying out nucleophilic substitution reactions [27]. The presence of proper leaving groups is usually necessary in these situations [28]. Substituents such as chlorine, methoxy, methylthio or methylsulfonyl are used [29]. The use of chlorine compared to other leaving groups facilitates the reaction conditions and increases the yield of the reaction [30]. The presence of electron withdrawing groups in position 5 increases the electron poverty of the ring and increases the rate of nucleophilic substitution reactions [31].

The review of scientific literature shows that the most common method of synthesis of 3,4-dihydropyrimidines is the reaction of acetylacetone with aldehyde urea or thiourea. This method is exclusively applicable for the synthesis of 3,4-dihydropyrimidines with substitutions in positions 2, 4, 5, and 6. Many methods have been reported for the synthesis of these compounds, but due to the widespread use of these compounds, scientists are looking for more efficient, practical and less expensive methods for the synthesis of these compounds.A review of the scientific literature shows that it was used in the Beguinelli reaction for the synthesis of 3,4-dihydropyrimidine; But until now, surfactant has not been reported as a catalyst in the synthesis of these compounds. In this project, new derivatives of substituted 3,4-dihydropyrimidine were synthesized in the presence of catalyst TBAB in solvent-free conditions at 80°C with high yield.

■ Results and Discussion

The studies conducted show that the Beguinelli reaction was used for the synthesis of 3,4-dihydropyrimidine, in which TBAB surfactant was reported as a catalyst in the synthesis of these compounds. In this project, new substituted 3,4-dihydropyrimidine derivatives were synthesized in the

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presence of TBAB catalyst under solvent-free conditions at 80°C with high yield. The IR spectrum of 3,4-dihydropyrimidine compound is shown in **Figure 1**. In the IR spectrum of this compound, which was taken in the form of a KBr tablet, the absorption related to the NH_2 group of the original material has been removed, and the absorption peak of the NH group at 3235 and 3120 cm^{-1} and the absorption peak of C=O groups at 1725 cm^{-1} are related to the ketone group and peak C=O at 650 cm^{-1} related to carbamide group confirms the reaction.

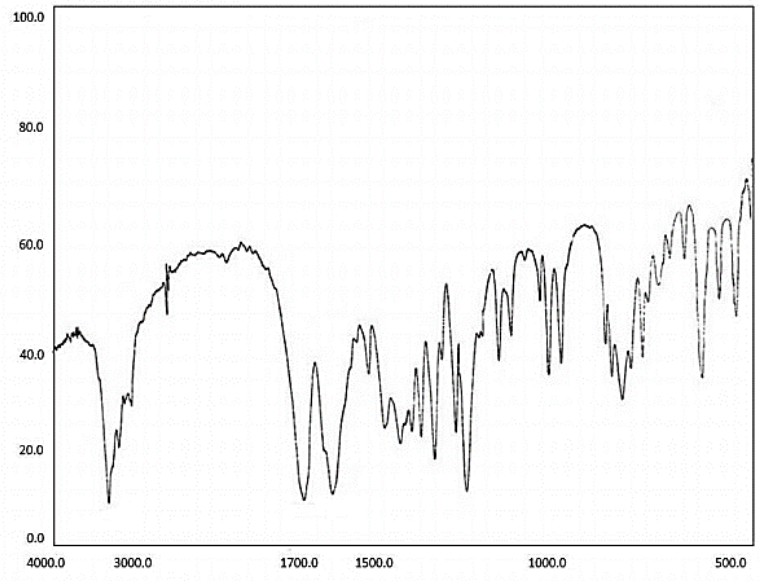
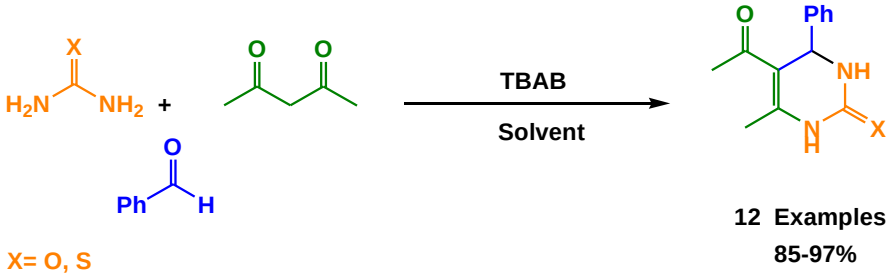


Figure 1. FT-IR spectra for 3,4-dihydropyrimidine compound.

To optimize the reaction conditions, the reaction of benzaldehyde, urea and acetylacetone was used as the base reaction. In this reaction, the effects of solvent and catalyst amounts and the effect of microwaves on the reaction efficiency were investigated. The results of the reactions are shown in **Table 1**. According to the obtained results, this reaction was investigated in different polar and non-polar solvents, and the results showed that the reaction yield is low in non-polar solvents, and the highest reaction yield is related to the use of 15 mg of catalyst TBAB in solvent-free conditions at 80 °C and for 30 minutes. These conditions were used for the synthesis of 3,4-dihydropyrimidine derivatives substituted in positions 2, 4, 5, and 6.

Table 1. Optimizing reaction conditions for 3,4-dihydropyrimidines synthesis [a]



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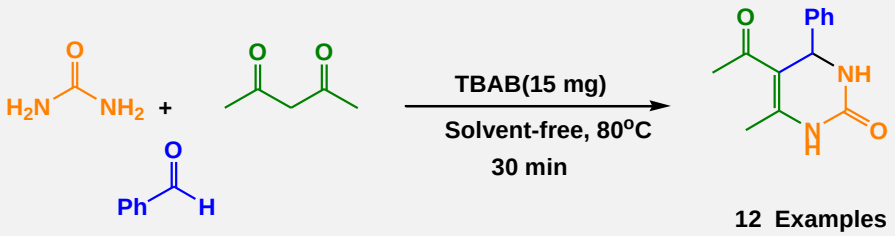
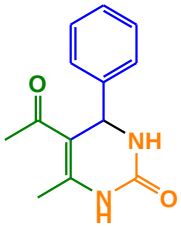
Entry	Catalyst (mg%)	Solvent	Temp (°C)	Yield (%) [a]
1	--	Neat	80	No
2	5	Neat	80	35
3	10	Neat	80	81
4	15	Neat	80	97
5	20	Neat	80	96
6	15	DMF	80	61
7	15	H_2O	80	71
8	15	THF	80	42
9	15	CH_3CN	80	62
10	15	$CHCl_3$	80	39
11	15	MeOH	80	81
12	15	DMSO	100	58
13	15	Neat	40	49
14	15	Neat	rt	37

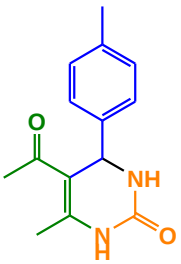
The reaction of various benzaldehydes with thio/urea and acetylacetone was carried out in solvent-free conditions with 15 mg of TBAB and a temperature of 80°C, and new derivatives of substituted 3,4-dihydropyrimidines were synthesized in different situations with high yield. The results are shown in **Table 2**. According to the results of this table, electron-killing substitutions had a higher benefit than electron-donating substitutions.

Table 2. Reaction yield of substituted 3,4-dihydropyrimidine derivatives [a]

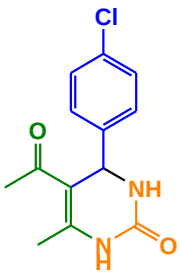




4a (97%)



4b (89%)



4c (92%)

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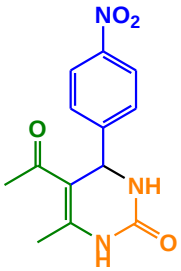
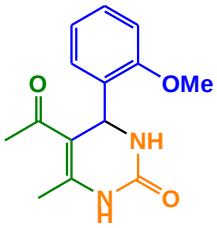
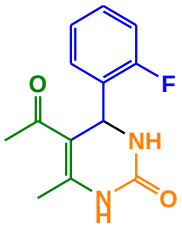


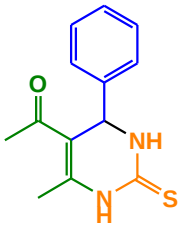
4d (94%)



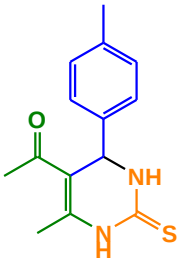
4e (87%)



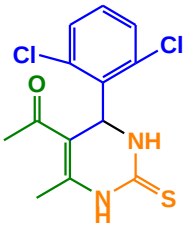
4f (90%)



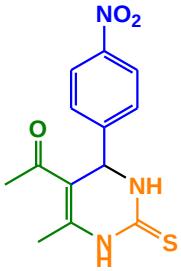
4g (95%)



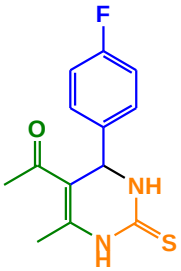
4h (88%)



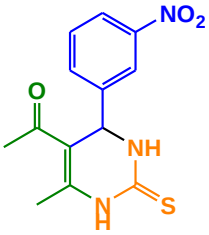
4i (89%)



4j (93%)



4k (90%)



4l (85%)

The proposed mechanism of this reaction, which is prepared in **scheme 1**, is a two-step mechanism. It seems that TBAB acts as an ionic liquid and causes a homogenous environment of the reaction mixture. Tetrabutylammonium bromide (TBAB) activates the carbonyl group of the aldehyde for nucleophilic attack.

A: Formation of N-acylimine: This step is known as the rate-determining step.

B: Increasing nucleophilicity, cyclization and water removal.

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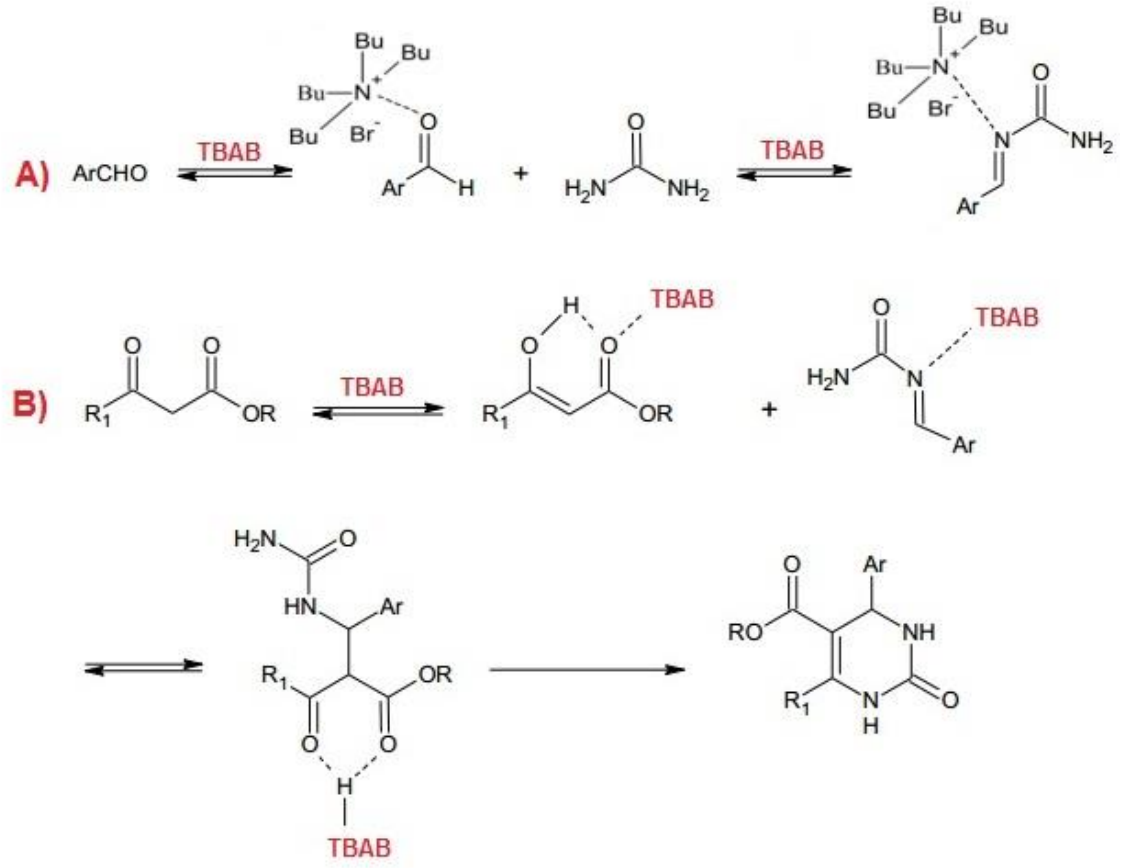

Scheme 1. Plausible mechanism.

■ Conclusion

In this research work, we succeeded in synthesizing various derivatives of 3,4-dihydropyrimidines from the reaction of benzaldehyde, urea and acetylacetone using TBAB catalyst. One of the features of this method is the one-pot reaction, in which there is no need to separate intermediates and carry out the reaction in subsequent steps that lead to a reduction in the yield of the reaction. The reaction is carried out in solvent-free conditions, and organic solvents, which are toxic and harmful to the environment, have not been used. Using catalyst TBAB, which is an ionic liquid, non-toxic and environmentally friendly. Products were synthesized with high yield in a short period of time.

■ Experimental

Hydrogen nuclear magnetic resonance 1HNMR with 400 MHz field was performed by Bruker's instrumental analysis. Tetramethylsilane (TMS) has also been used as an internal standard. Infrared (IR) spectra were recorded using a Shimadzu 470 IR Spectrometer. The spectra of solid compounds were taken in the form of KBr tablets. The melting points of the compounds have been measured using the Bamstead/Electrothermal device. All raw materials were purchased from Acros, Floka and Merck.

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Preparation of derivatives of 3,4-dihydropyrimidines substituted in different positions

The reaction mixture of benzaldehyde derivatives (1 mmol), urea (1.5 mmol), acetylacetone (1 mmol) and TBAB (15 mg) was stirred under solvent-free conditions at 80 °C until completion (the end of the reaction was determined by TLC became). After the reaction, the reaction mixture was cooled, then the resulting solid was washed with cold water and crystallized in ethanol.

Supporting Information

5-acetyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (4a):

1H NMR (400 MHz, DMSO) δ 10.25 (s, 1H), 9.51 (s, 1H), 7.24-7.12 (m, 5H), 5.13 (d, J=3.8 Hz, 1H), 2.22 (s, 3H), 1.05 (s, 3H).13C NMR (100 MHz, DMSO) δ 175.0, 165.2, 145.2, 129.6, 128.4, 127.1, 101.5, 54.6, 17.6, 14.3.

1-(6-methyl-4-(3-nitrophenyl)-2-thioxo-1,2,3,4-tetrahydropyrimidin-5-yl)ethanone (4l):

1H NMR (400 MHz, DMSO) δ 10.44 (s, 1H), 9.66 (s, 1H), 8.12-8.05 (m, 2H),7.72-7.65 (m, 3H), 5.28 (d, J=4.1 Hz, 1H), 2.26 (s, 3H), 1.06 (s, 3H).13C NMR (100 MHz, DMSO) δ 175.1, 165.5, 148.4, 146.6, 146.1, 133.6, 131.0, 123.3, 121.7, 100.5, 54.1, 17.8, 14.5.

References

[1] S. S. Panda, P. Khanna, L. Khanna, Biginelli Reaction: A Green Perspective, Curr. Org. Chem. 16 (2012) 507–520. https://doi.org/10.2174/138527212799499859.

[2] R.O.M.A. De Souza, E.T. da Penha, H.M.S. Milagre, S.J. Garden, P.M. Esteves, M.N. Eberlin, O.A.C. Antunes, The Three-Component Biginelli Reaction: A Combined Experimental and Theoretical Mechanistic Investigation, Chem. – A Eur. J. 15 (2009) 9799–9804. https://doi.org/10.1002/chem.200900470.

[3] Y. Nishimura, H. Kikuchi, T. Kubo, R. Arai, Y. Toguchi, B. Yuan, K. Sunaga, H. Cho, Synthesis of 4,4-Disubstituted 3,4-Dihydropyrimidin-2(1H)-ones and -thiones, the Corresponding Products of Biginelli Reaction Using Ketone, and Their Antiproliferative Effect on HL-60 Cells, Chem. Pharm. Bull. 70 (2022) c21-00794. https://doi.org/10.1248/cpb.c21-00794.

[4] G.C. Tron, A. Minassi, G. Appendino, Pietro Biginelli: The Man Behind the Reaction, European J. Org. Chem. 2011 (2011) 5541–5550. https://doi.org/10.1002/ejoc.201100661.

[5] L. Gong, X. Chen, X. Xu, Asymmetric Organocatalytic Biginelli Reactions: A New Approach To Quickly Access Optically Active 3,4-Dihydropyrimidin-2-(1 H)-ones, Chem. – A Eur. J. 13 (2007) 8920–8926. https://doi.org/10.1002/chem.200700840.

[6] D. Ding, C. Zhao, Primary Amine Catalyzed Biginelli Reaction for the Enantioselective Synthesis of 3,4-Dihydropyrimidin-2(1 H)-ones, European J. Org. Chem. 2010 (2010) 3802–3005. https://doi.org/10.1002/ejoc.201000448.

[7] N. Jagadishbabu, K. Shivashankar, Biginelli Reaction of Vicinal Diols: A New Route for One-Pot Synthesis of 3,4-Dihydropyrimidin-2(1H)-One Derivatives, Lett. Org. Chem. 14 (2017). https://doi.org/10.2174/1570178614666170310123511.

DOI: 10.22034/jsc.2024.457661.1074
J. Synth. Chem. 2024, 3, 49-60

[8] F. Sánchez-Sancho, M. Escolano, D. Gaviña, A.G. Csáky, M. Sánchez-Roselló, S. Díaz-Oltra, C. del Pozo, Synthesis of 3,4-Dihydropyrimidin(thio)one Containing Scaffold: Biginelli-like Reactions, Pharmaceuticals. 15 (2022) 948. https://doi.org/10.3390/ph15080948.

[9] A. Verma, G. Chaudhary, D. Verma, A. Prabhakar, K. Behera, Ammonium tetrafluoroborate: Novel Unprecedented Catalyst for the Synthesis of 3,4-dihydropyrimidin-2(1 H)-ones through Biginelli Reaction, ChemistrySelect. 9 (2024). https://doi.org/10.1002/slct.202304179.

[10] R. Tayebee, M. Ghadamgahi, Solvent free one-pot multi-component synthesis of 3,4-dihydropyrimidin-2(1 H)-ones catalyzed by mesoporous NH 4 H 2 PO 4 /MCM-41 as an environmentally friendly, cheap, and effective catalyst, Arab. J. Chem. 10 (2017) S757–S764. https://doi.org/10.1016/j.arabjc.2012.12.001.

[11] J.-P. Wan, Y. Liu, Synthesis of Dihydropyrimidinones and Thiones by Multicomponent Reactions: Strategies Beyond the Classical Biginelli Reaction, Synthesis (Stuttg). 2010 (2010) 3943–3953. https://doi.org/10.1055/s-0030-1258290.

[12] R. Esmaeili, L. Kafi-Ahmadi, S. Khademinia, A highly efficient one-pot multicomponent synthesis of 3,4-dihydropyrimidin-2-(1H)-ones/thiones catalyzed by strontium pyroarsenate nano-plates, J. Mol. Struct. 1216 (2020) 128124. https://doi.org/10.1016/j.molstruc.2020.128124.

[13] F. Vitório, T.M. Pereira, R.N. Castro, G.P. Guedes, C.S. Graebin, A.E. Kümmerle, Synthesis and mechanism of novel fluorescent coumarin–dihydropyrimidinone dyads obtained by the Biginelli multicomponent reaction, New J. Chem. 39 (2015) 2323–2332. https://doi.org/10.1039/C4NJ02155J.

[14] O.M. Singh, N.S. Devi, Application of β-Oxodithioesters in Domino and Multicomponent Reactions: Facile Route to Dihydropyrimidines and Coumarins, J. Org. Chem. 74 (2009) 3141–3144. https://doi.org/10.1021/jo802585b.

[15] D. Meng, Y. Xie, Q. Peng, J. Wang, NHC-Catalyzed Enantioselective [3 + 3] Annulation to Construct 5,6-Dihydropyrimidin-4-ones, Org. Lett. 22 (2020) 7635–7639. https://doi.org/10.1021/acs.orglett.0c02832.

[16] M. Puripat, R. Ramozzi, M. Hatanaka, W. Parasuk, V. Parasuk, K. Morokuma, The Biginelli Reaction Is a Urea-Catalyzed Organocatalytic Multicomponent Reaction, J. Org. Chem. 80 (2015) 6959–6967. https://doi.org/10.1021/acs.joc.5b00407.

[17] L.-R. Wen, W.-K. Yuan, M. Li, Dual Roles of β-Oxodithioesters in the Copper-Catalyzed Synthesis of Benzo[e]pyrazolo[1,5- c][1,3]thiazine Derivatives, J. Org. Chem. 80 (2015) 4942–4949. https://doi.org/10.1021/acs.joc.5b00288.

[18] G.C. Nandi, S. Samai, M.S. Singh, Biginelli and Hantzsch-Type Reactions Leading to Highly Functionalized Dihydropyrimidinone, Thiocoumarin, and Pyridopyrimidinone Frameworks via Ring Annulation with β-Oxodithioesters, J. Org. Chem. 75 (2010) 7785–7795. https://doi.org/10.1021/jo101572c.

[19] Z.-L. Shen, X.-P. Xu, S.-J. Ji, Brønsted Base-Catalyzed One-Pot Three-Component Biginelli-Type Reaction: An Efficient Synthesis of 4,5,6-Triaryl-3,4-dihydropyrimidin-2(1 H)-one and Mechanistic Study, J. Org. Chem. 75 (2010) 1162–1167. https://doi.org/10.1021/jo902394y.

[20] Y.-C. Guo, X.-D. Song, W. Deng, W. Rao, H. Xu, Z.-L. Shen, An efficient synthesis of 4,5-diaryl-3,4-dihydropyrimidin-2(1 H)-one via a cesium carbonate-promoted direct condensation of 1-aryl-2-propanone with 1,1′-(arylmethylene)diurea, RSC Adv. 10 (2020) 30062–30068.

DOI: 10.22034/jsc.2024.457661.1074
J. Synth. Chem. 2024, 3, 49-60

https://doi.org/10.1039/D0RA05480A.

[21] T.M. Shaikh, S. Nagarajan, E. Kandasamy, One Pot Multicomponent Biginelli Reaction Employing Ionic Liquids as an Organocatalyst, Curr. Organocatalysis. 7 (2020) 96–107. https://doi.org/10.2174/2213337206666191001214521.

[22] S.E. Sadati Sorkhi, M.M. Hashemi, A. Ezabadi, Introduction of a novel dicationic Brönsted acidic ionic liquid based on pyrazine and its application in the synthesis of xanthenediones and 3, 4-dihydropyrimidin-2(1H)-ones under solvent-free conditions, Res. Chem. Intermed. 46 (2020) 2229–2246. https://doi.org/10.1007/s11164-020-04089-9.

[23] Z. Liu, R. Zhang, Y. Liu, Y. Guo, Q. Meng, The Effects of Different Catalysts, Substituted Aromatic Aldehydes on One-Pot Three-Component Biginelli Reaction, Curr. Org. Synth. 16 (2019) 181–186. https://doi.org/10.2174/1570179416666181122100405.

[24] G. Zhang, D. Tian, W. Shi, Efficient Catalytic Synthesis of 3,4-Dihydropyrimidin-2-ones/thiones via Little Acidic Ionic Liquid Combined with Rapid Heating Ways, J. Heterocycl. Chem. 55 (2018) 2522–2531. https://doi.org/10.1002/jhet.3292.

[25] E. Marqués-López, R.P. Herrera, Biginelli Multicomponent Reactions, in: Multicomponent React., Wiley, 2015: pp. 306–330. https://doi.org/10.1002/9781118863992.ch9.

[26] G. Bosica, F. Cachia, R. De Nittis, N. Mariotti, Efficient One-Pot Synthesis of 3,4-Dihydropyrimidin-2(1H)-ones via a Three-Component Biginelli Reaction, Molecules. 26 (2021) 3753. https://doi.org/10.3390/molecules26123753.

[27] C.O. Kappe, The Generation of Dihydropyrimidine Libraries Utilizing Biginelli Multicomponent Chemistry, QSAR Comb. Sci. 22 (2003) 630–645. https://doi.org/10.1002/qsar.200320001.

[28] A.C. Boukis, B. Monney, M.A.R. Meier, Synthesis of structurally diverse 3,4-dihydropyrimidin-2(1 H)-ones via sequential Biginelli and Passerini reactions, Beilstein J. Org. Chem. 13 (2017) 54–62. https://doi.org/10.3762/bjoc.13.7.

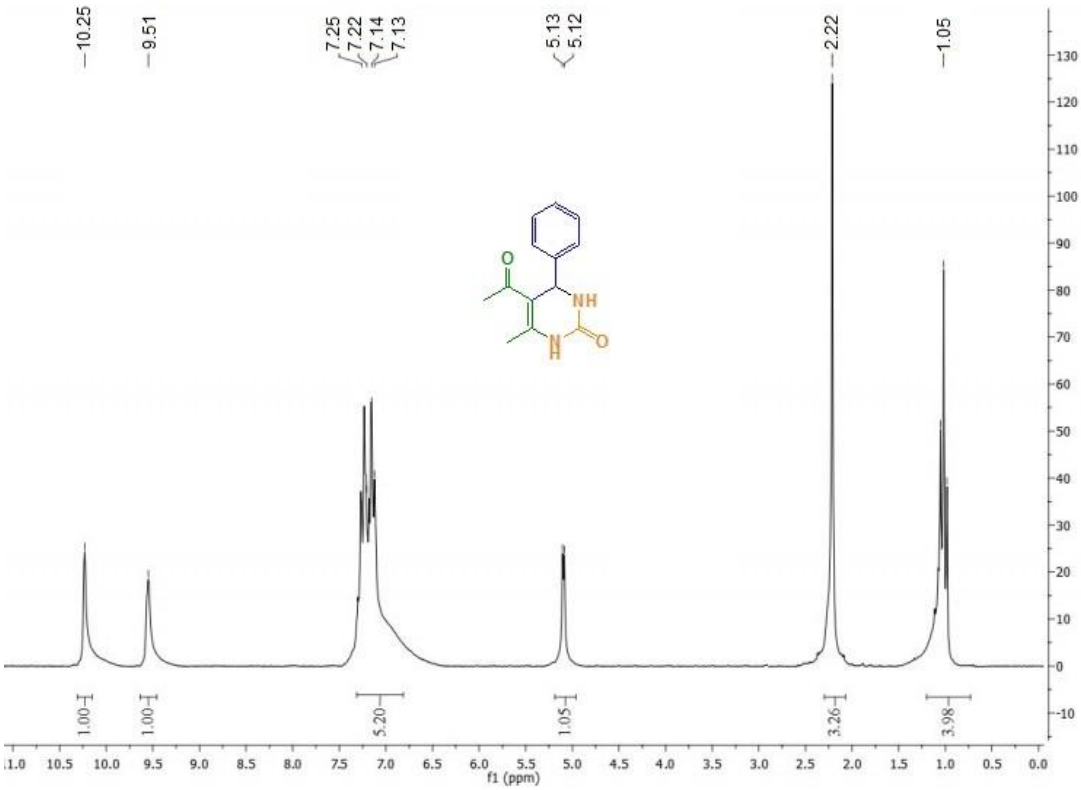
[29] M. Gopalakrishnan, P. Sureshkumar, J. Thanusu, V. Kanagarajan, M. Ezhilarasi, An Expedient One-Pot Synthesis of 3,4-Dihydropyrimidin-2(1H)-Ones and -Thiones Catalyzed by ZIRCONIA Nanopowder. An Improved Protocol for the Biginelli Cyclocondensation Compounds in ‘Dry Media’, Lett. Org. Chem. 5 (2008) 142–147. https://doi.org/10.2174/157017808783743911.

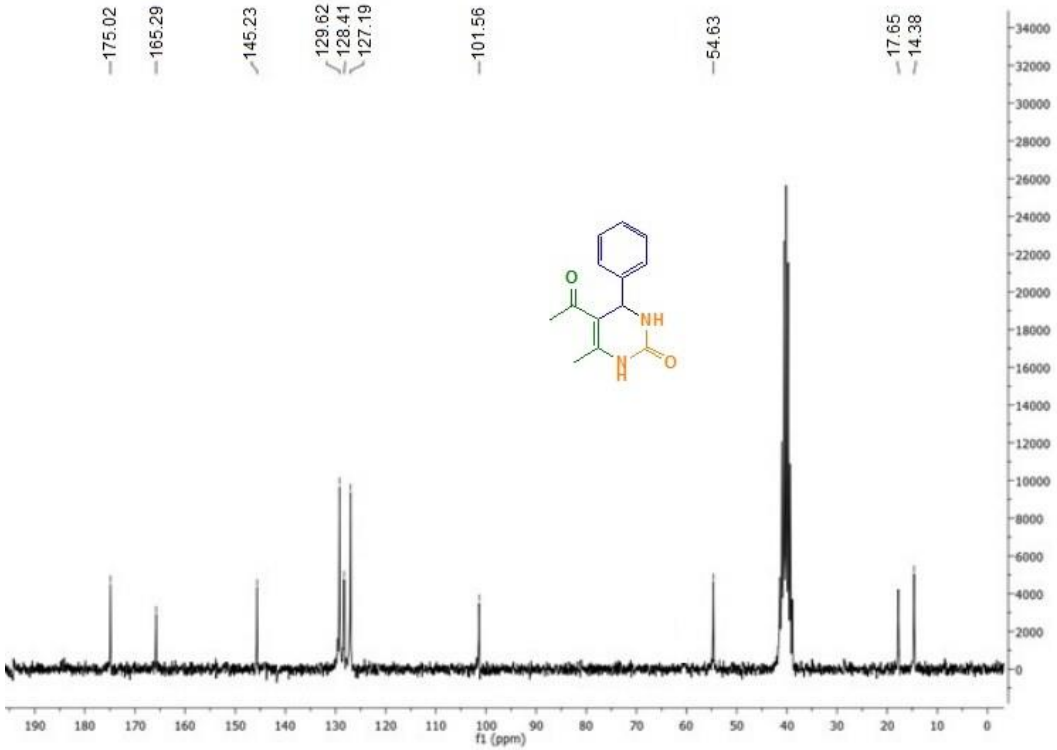
[30] S. Andleeb, I.-D. Imtiaz-ud-Din, M.K. Rauf, S.S. Azam, A. Badshah, H. Sadaf, A. Raheel, M.N. Tahir, S. Raza, A one-pot multicomponent facile synthesis of dihydropyrimidin-2(1H)-thione derivatives using triphenylgermane as a catalyst and its binding pattern validation, RSC Adv. 6 (2016) 79651–79661. https://doi.org/10.1039/C6RA19162B.

[31] C.K. Khatri, D.S. Rekunge, G.U. Chaturbhuj, Sulfated polyborate: a new and eco-friendly catalyst for one-pot multi-component synthesis of 3,4-dihydropyrimidin-2(1H)-ones/thiones via Biginelli reaction, New J. Chem. 40 (2016) 10412–10417. https://doi.org/10.1039/C6NJ03120J.

Copies of NMR spectra of synthesized compounds:

DOI: 10.22034/jsc.2024.457661.1074
J. Synth. Chem. 2024, 3, 49-60





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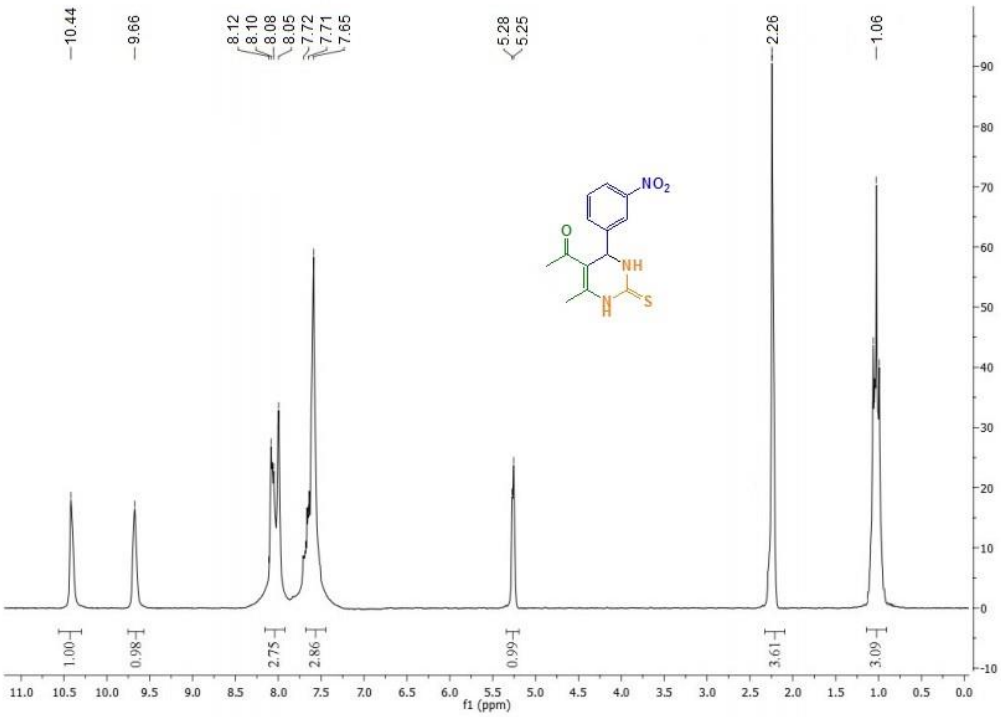

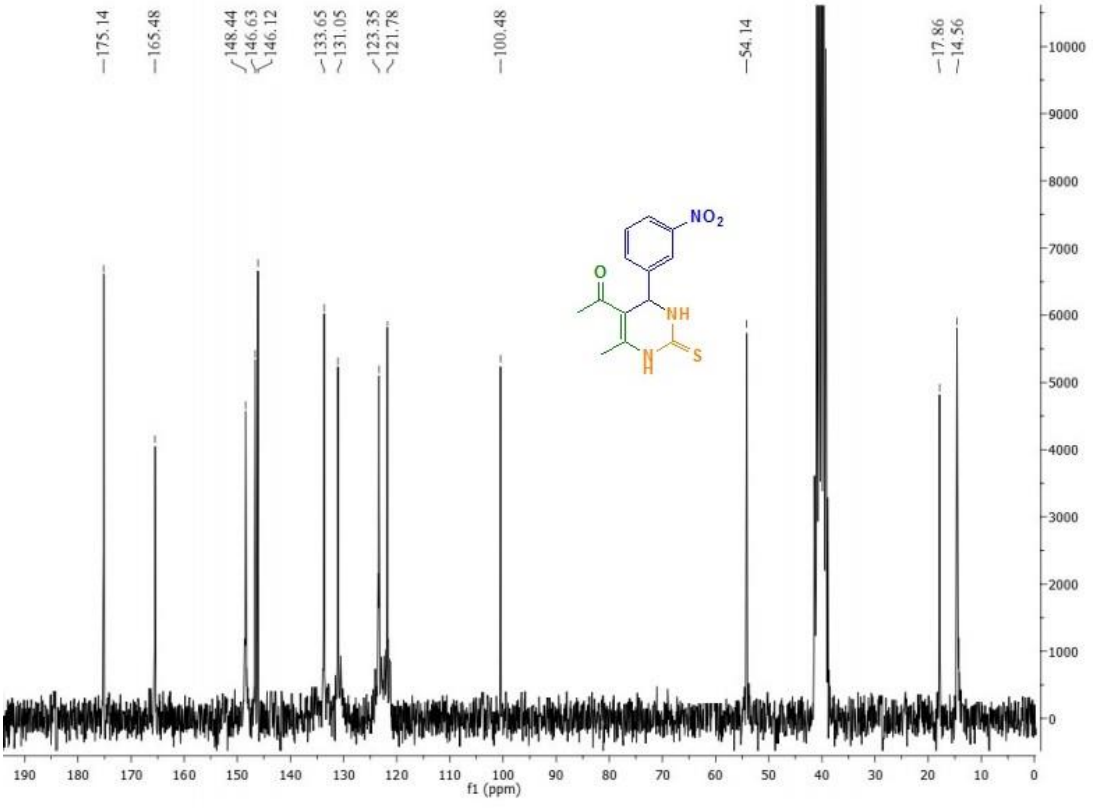

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