

Using a Green Method, the Biginelli Reaction is used to Create 1, 2, 3, and 4-tetrahydropyrimidine

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Abstract: Using recycled aluminum foil as an environmentally friendly acid catalyst, this article discusses a one-pot, multicomponent synthesis of 3,4-dihydropyrimidinone derivatives from aldehydes, beta esters, and solvent-free urea. Shorter reaction times, higher yields, different operational dangers, and no environmental contamination are all advantages of this method with the Biginelli reaction's typical circumstances; it is a simple, solvent-free, environmentally friendly process. No particular reaction environment is required.

Keywords: Sonication, 3, 4-dihydropyrimidinone, and red mud one-pot multicomponent synthesis.

I. INTRODUCTION

Sonication works well for a variety of chemical processes. Sonication has replaced the conventional approach as a more effective and advantageous way to induce chemical reactions. [1-4]. Sonication has advanced into a more advanced and useful technology in the field of organic chemistry using an unconventional energy source [6-10]. Accelerations are necessary for a wide variety of chemical reactions in these systems in the absence of solvents. Coupled sonication without solvents, conditions for reactions without solvents, and an environmentally acceptable approach [11]. Due to their biological characteristics in drug development, dihydropyrimidines have become important target molecules for drugs like anticarcinogenic, antimetotics, calcium channel blockers, antivirals, antihypertensives, and alpha-antagonists [12-15]. According to the Biginelli method, ethyl acetoacetate, urea, and maybe another ester aromatic aldehyde undergo a multicomponent reaction with dihydropyrimidines (DHPM) under very acidic circumstances [16]. The reactions are often carried out in the presence of a potent protic acid catalyst, other compounds, and ethanol or tetrahydrofuran [17-20]. Throughout this time, the aldehydes in the original multicomponent scheme, the catalyst, the reaction conditions, and the carbonyl functionality of the acyclic ketone all underwent modifications. Several articles [21-26] have described the synthesis of dihydropyrimidines modified by the Biginelli reaction using 1,3-cyclic ketones rather than acyclic ketones, such as dimedone, indane-1,3-dione, or other heterocyclic ketones. We used leftover aluminum foil from previous projects as the project's catalyst because it is inexpensive and widely accessible in the neighborhood.[26].

II. MATERIAL AND METHODS

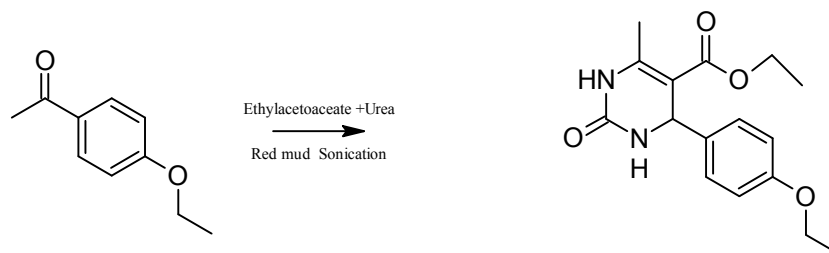
Experimental:

The paraffin tube method was used to estimate the uncorrected melting points. Without using KBr pellets, IR spectra were obtained on a Shimadzu spectrophotometer. Using TMS as an internal standard, ¹H-NMR spectra were obtained using a Bruker AMX-400 spectrometer. Visualization was performed in an ultraviolet chamber or with iodine.

General Procedure

Sonicate ethyl acetoacetate (10 mmol), urea (10 mmol), aldehyde (10 mmol), discarded aluminum foil (0.4 g), and 12 M HCl (1 drop) in a 25 mL Borosil beaker. Using thin-layer chromatography, the reaction was observed. After the reaction was complete, the reaction mixture was chilled and the precipitated solid was filtered from ethyl acetate or another solvent and recrystallized. The produced 1,2,3,4-tetrahydropyrimidine was compared to the determined literal melting point of the Biginelli product. To prepare the entire series of chemicals, the preceding method was utilized.

Reaction Scheme.



Observation Table

Sr. No	Aldehyde	Time in Min	Yield	Physical Constant in 0C
1	P-Nitrobenzaldehyde	50	78	198
2	P-chloro Benzaldehyde	55	80	218
3	P-methyl Benzaldehyde	58	82	217
4	P-Methoxy Benzaldehyde	50	84	196
5	furfur aldehyde	55	90	192
6	Benzaldehyde	50	78	190
7	cinnaldehyde	50	80	228
8	M-methyl Benzaldehyde	58	82	242
9	M-chloro Benzaldehyde	50	84	217

III. RESULT AND DISCUSSION

The feasibility of using leftover Red mud as a catalyst in the Biginelli reaction (scheme 1) employing other building blocks is investigated by modifying aromatic aldehydes. The reaction was carried out using the catalyst in the range of 0.1 to 0.4 g. 82 percent of the desired output was completed after the necessary amount of time. Second, the amount of catalyst was changed in the reaction conditions to maximize the product yield. The product yield unexpectedly increased to between 85 and 90% with the addition of 0.4 grammes of catalyst. The same amount of catalyst was added to both reactions, but no additional advancement was seen. This study's major focus is on the synthesis of more contemporary tetrahydropyrimidine derivatives using the Biginelli reaction and sonication. Several techniques, including melting point analysis, thin-layer chromatography, infrared analysis, and nuclear magnetic resonance (NMR) analysis, were used to look at the structures of the produced compounds.

Several aromatic and heteroaromatic aldehydes were treated with urea and 1,3-dicarbonyl compounds in an ecologically safe manner to demonstrate the generalizability of the current method. Compounds bearing both electron-withdrawing and electron-donating substituents on the aromatic ring effectively interacted under the prescribed reaction conditions to create the corresponding 1, 2, 3, 4-tetrahydropyrimidine with very high yields and environmental friendliness. The inventive feature of the proposed approach was effectively applied to the synthesis of several heterocyclic aldehydes, resulting in good to outstanding yields of the crucial 1, 2, 3, and 4-tetrahydropyrimidine. The initial stage of N-acyl imine synthesis is the rate-limiting step of the process.

The intermediate forms a complex with waste Red mud to create N, which serves as the electrophile for the nucleophilic addition of the ketoester enol. Throughout the procedure, adducts were produced. Condensation with urea-NH₂ during the synthesis results in a cyclized product having an ester bond.

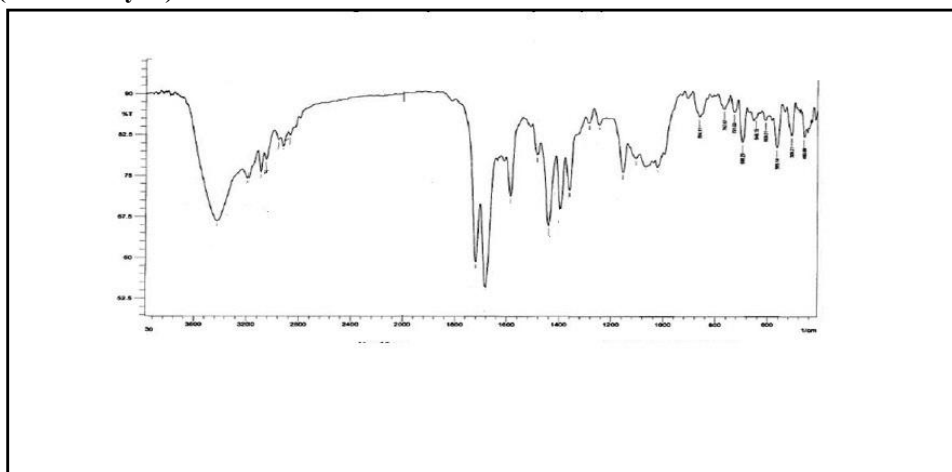
III. CONCLUSION

As a result, we have created a brand-new, effective procedure for the modified Biginelli reaction using ultra sonication, aromatic or heterocyclic aldehyde 1, 3-diketones, aldehydes, and urea in 60-70 minutes. When combined with sonication irradiation, the reaction takes place without the use of any solvents, and the work-up is rapid and environmentally safe. Our procedure offers a quick and efficient way to create a number of ring-fused pyrimidine derivatives.

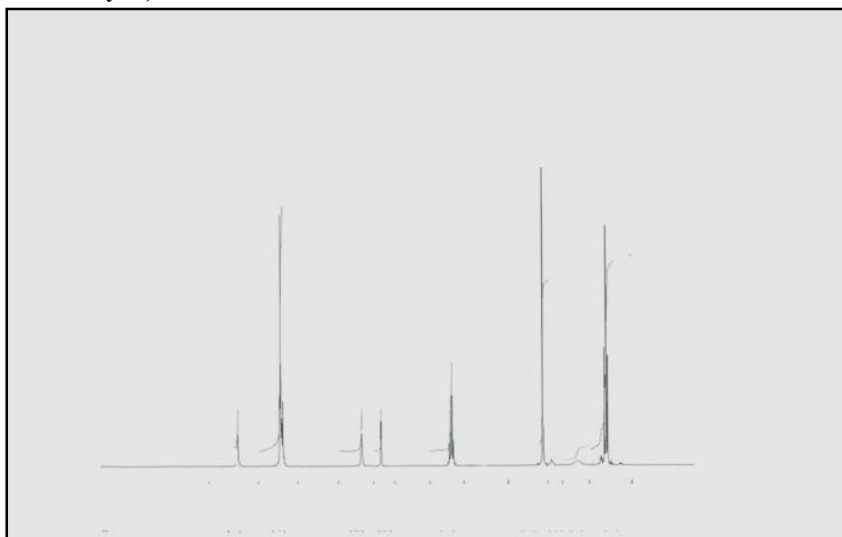
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IR Spectrum (Benzaldehyde)



NMR Spectrum (Benzaldehyde)



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