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FEAM as a Natural and Green Catalyst for Knoevenagel Condensation

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***Abstract*—A straightforward, gentle, and eco-friendly procedure has been established for the Knoevenagel condensation of aromatic aldehydes with malononitrile using FEAM (fruit extract of *Aegle marmelos*) at 100 ºC. The reaction provides the desired products in better yields with high purity. This green approach avoids the use of harmful acidic or basic catalysts, making the process both convenient to handle and environmentally safe.**

***Index Terms*—FEAM, Green chemistry, Knoevenagel condensation, natural catalyst.**

I. INTRODUCTION

The Knoevenagel condensation is regarded as a highly useful and dependable reaction for forming carbon– carbon bonds in organic synthesis. It was first reported by Emil Knoevenagel in 1894 [1]. In this reaction, aldehydes or ketones react with active methylene compounds in the presence of mild bases to generate α, βunsaturated products through a sequence involving enolate formation, nucleophilic addition, and subsequent removal of water [2,3]. Because the reaction is straightforward to perform and follows a well-understood mechanism, it has become an important method for constructing complex molecules used in pharmaceuticals, dyes, agrochemicals, fragrances, and advanced functional materials [4–6]. Despite its usefulness, conventional Knoevenagel procedures commonly rely on bases such as NaOH, KOH, or organic amines like piperidine, typically carried out in volatile organic solvents including ethanol, toluene, or benzene at higher temperatures for extended reaction periods [7,8]. These conditions create environmental and safety concerns, mainly due to solvent toxicity, increased chemical waste, and significant energy requirements [9]. In addition, the homogeneous catalysts employed in these systems are often difficult to separate and reuse, which raises operational costs and contributes to additional waste generation [10]. The use of harsh conditions can also restrict the applicability of the reaction, particularly when dealing with sensitive or sterically demanding substrates [11].

The concept of green chemistry, proposed by Paul Anastas and John C. Warner, promotes the design of chemical processes that minimize or eliminate hazardous substances and reduce waste while improving overall efficiency [12,13]. Applying these ideas to the Knoevenagel condensation has encouraged the development of alternative strategies. These include aqueous or solvent-free methods that employ water as an environmentally safer medium or avoid solvents entirely through grinding techniques or microwave assistance [14,15]. Another approach involves heterogeneous and recyclable catalysts such as metal oxides, clays, or calcined natural materials, which allow easier separation and repeated use [16,17]. Bio-derived catalysts from plant extracts, fruit ashes, or enzyme-based systems are also being explored as renewable and low-cost options [18,19]. Additionally, ionic liquids and deep eutectic solvents have been introduced as non-volatile and reusable substitutes for conventional solvents [20,21]. Natural materials are particularly attractive for sustainable catalysis. They are generally biodegradable, nontoxic, and readily available, and they often function effectively under mild reaction conditions. Many of these materials contain diverse functional groups that can promote catalytic activity without the need for additional reagents [22]. Converting plant-based resources or agricultural residues into catalysts also supports sustainable resource utilization and fits well with the principles of a circular economy [23].

The fruit of *Aegle marmelos* (Bael) is a nutritionally dense and medicinally valuable underutilized fruit containing diverse bioactive phytochemicals that mainly belong to alkaloids, coumarins, flavonoids, phenolic acids, terpenoids, carotenoids, and polysaccharides, which contribute to its extensive use in traditional medicine and modern therapeutics. Among the alkaloids reported from the fruit are aegeline, aegelenine, fragrine, dictamine, ethyl cinnamate, and aegelinosides A and B. The fruit also contains important coumarins including imperatorin, umbelliferone, scopoletin, marmesin, marmelosin, alloimperatorin, xanthotoxol, isoimperatorin, marmelide, marmin, psoralen, scoparone, and marmenol. Phenolic acids such as gallic acid, chlorogenic acid, p-coumaric acid, vanillic acid, 2,3-dihydroxy benzoic acid, and ellagic acid are present as major antioxidant constituents with total phenolic content of approximately 150–200 mg GAE/g extract. Flavonoids such as rutin and quercetin are the prominent flavonoid components contributing to free radical scavenging activity. Carotenoids including α-carotene, β-carotene, γ-carotene, and δ-carotene impart yellow color and provide significant antioxidant capacity. The fruit pulp is nutritionally rich with carbohydrates (31–42%), proteins (3.6–8.8%), vitamins (A, B_1, B_2, B_3, C), and minerals, particularly calcium (86.68%), along with Fe, Cu, Zn, and Mn in the range of 1.29–15.82% [24]. Unripe bael fruit is also **acidic** due to presence of organic acids, while the ash extract is alkaline, making it versatile for both acid and base-catalyzed reactions depending on preparation method [25].

In continuation of our ongoing efforts [26–37] to develop greener synthetic routes for value-added products, the present study reports, for the first time, the application of FEAM (fruit extract of *Aegle marmelos*) as a natural and environmentally benign catalyst for the Knoevenagel condensation carried out at 100 ºC without the use of additional solvents. This strategy is intended to integrate sustainability with practical simplicity and economic feasibility. The method demonstrates efficient catalytic activity under mild conditions, produces only minimal amounts of naturally degradable waste, shows good compatibility with a range of aldehydes and active methylene compounds, and allows straightforward work-up with potential for scale-up. By utilizing an easily available plant product, this work presents a new approach toward sustainable organic synthesis and supports the broader objectives of green chemistry and renewable catalytic systems.

II. EXPERIMENTAL SECTION

2.1 General Information

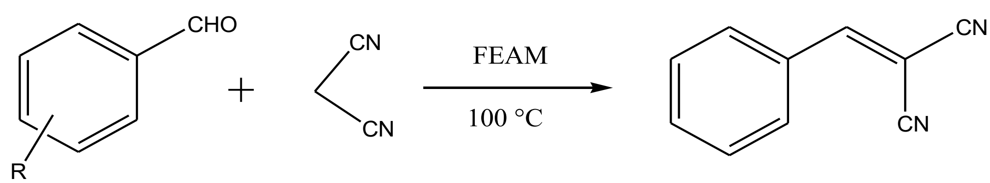
All chemicals were purchased from commercial sources and used without further purification. Melting points were recorded in open capillaries and are uncorrected. Reaction progress was monitored by thin-layer chromatography (TLC) using silica gel plates.

2.2 Preparation of FEAM (fruit extract of *Aegle marmelos*)

Fresh fruits of *Aegle marmelos* were thoroughly washed with clean water and then ground in a mechanical mixer–grinder using a small quantity of water to obtain a uniform mixture. The resulting slurry was subjected to simple filtration to separate the solid residues, yielding a fruit extract. This filtrate was used directly as both the catalyst and the reaction medium for the study. The pH of the freshly prepared FEAM was found to be 3.5, indicating that it is acidic in nature.

2.3 General Procedure for Knoevenagel Condensation

Aromatic aldehydes (10 mmol), malononitrile (10 mmol), and FEAM (5 gm) were placed in a round-bottom flask and the mixture was stirred at 100 °C. Once the reaction was complete, the reaction mixture was poured into ice-cold water to precipitate the product. The resulting solid was collected by filtration and thoroughly washed with water. The crude material obtained was then purified by recrystallization using absolute ethanol.



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III. RESULTS AND DISCUSSION

3.1 Optimization Studies

The reaction conditions were systematically optimized by studying the influence of amount of Ginger juice used and the reaction temperature. The results showed that the most favourable conditions were achieved using 5 gm FEAM at 100 ºC. further increase in quantity of FEAM and temperature do not affect the results obtained in the reaction. The reaction completed in 6 to 12 minutes depending upon the type of aldehyde reactant used in the reaction.

3.2 Scope of the work

The methodology was successfully applied to various aromatic aldehydes bearing different substituents. The results are summarized in Table 1.

Table 1. Knoevenagel Condensation of Aromatic Aldehydes with Malononitrile

Sr. No.	R	Product	M. P. (°C)	% Yield
1	H	3a	81-82	64
2	4-OCH_3	3b	111-112	82
3	4-Br	3c	165-166	87
4	4-Cl	3d	160-161	75
5	4-OH	3e	180-181	73
6	4$N(CH_3)_2$	3f	183-184	84
7	3,4 $(OH)_2$	3g	240-242	85

IV. CONCLUSIONS

A new and environmentally friendly approach for the Knoevenagel condensation has been established by employing FEAM as a natural catalyst. The reaction proceeds smoothly and affords better yields in the range of 64-87% along with good product purity. The method is simple to handle, cost-effective, and shows promise for large-scale or industrial use. Overall, this study highlights the usefulness of plant-derived product as sustainable substitute for traditional synthetic catalysts in organic synthesis.

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