Fruit Extract Catalyzed Synthesis of Heterocycles: A Mini-Review

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Abstract: This paper reports the recently published work on using the extract of fruits as natural catalysts in the formation of heterocycles. Twelve green principles are described in this review through the recent synthesis of heterocyclic compounds. These catalysts have many benefits, such as non-hazardous, clean reaction profile, easy handle, low cost, easily available, etc. The Fruit extract catalyst is one of the best options for the recyclization of organic heterocycles in the future.  

Keywords: fruit juice; heterocyclic compounds; green catalyst.  
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1. Introduction  
Presently, Multi-component organic syntheses have received considerable attention in pharmaceutical and medicinal chemistry due to the significant benefits associated with this route [1]. In addition, green and efficient reactions are challenging tasks in heterocyclic synthesis [2]. Concomitant with these needs has been the requirement for improved catalysts.  

Recently, fruit extracts, vegetable extracts, and industrial and even agricultural wastages are being explored for their catalytic activities [3].  

Figure 1. Synthesis of Heterocycles from Fruit extract(Juice) as a catalyst.
In particular, various juices and vegetables are broadly attractive for green chemistry applications due to their environmentally friendly, non-hazardous, easily available, and low cost [4]. Many organic syntheses have been carried out using the homogeneous catalytic activity of fruit and vegetable juices [5]. These have proven to be useful for preparing such organic heterocyclic compounds [6]. The present overview covers the uses of fruit juices as green catalysts to organic heterocyclic synthesis.

2. Lemon Juice

Khan and co-workers [7] reported the synthesis of poly-substituted pyrrol-2-one, furn-2-one, and tetrahydropyrpyri-dines (1) from easily available biodegradable lemon juice catalyst. The synthesized derivatives were achieved from dialkyl acetylenedicarboxylates, different amines, and aldehydes in a short reaction time using 0.25 ml lemon juice at 110°C under without solvent conditions.

\[ \text{Lemon Juice} \]

Lokeshwari and Kariyappa [8] reported the synthesis of conjugated pyrazoles (2) from chalcone and phenylhydrazine hydrochloride with tetrabutylammonium bromide phase transfer catalyst in freshly extracted lemon juice as a catalyst medium by an accessible and eco-friendly approach.

\[ \text{Scheme 2. Synthesis of conjugated pyrazole derivatives.} \]

Metwally and co-workers [9] reported the synthesis of thiopyran[2,3-d][1,3]thiazoles (3) derivatives through Diels Alder Reaction using lemon juice as a catalyst.

Bhosale and co-workers [10] reported a three-component one-pot synthesis of 2-amino-4H-chromens (4) by simply mixing Malononitrile, aromatic aldehyde α-napthol, and lemon juice catalyst of natural origin and avoiding using hazardous organic solvents. They conclude main advantages of this method are its green method character, short reaction time, simple workup procedure, and gives good yield.
Scheme 3. Synthesis of new thiopyran[2,3-d][1,3]thiazoles.

Scheme 4. Synthesis of 2-amino-4H-chromens.

Kawle and Kamble [11] reported a one-pot four-component synthesis of 2, 5, 6-triaryl pyrazines, 1, 2, 4, 5-tetraaryl imidazoles and 2, 2, 4, 5-tetraaryl imidazoles (5) from benzoin, aryl amine, ammonium acetate, and aryl ketone or aryl aldehydes using lemon juice (citrus lemon) as an eco-friendly catalyst gives good yield under mild conditions.

Scheme 5. Synthesis of 2, 5, 6-triaryl pyrazines, 1, 2, 4, 5-tetraaryl imidazole’s and 2, 2, 4, 5-tetraaryl imidazole’s.

Niralwad and Ghorade [12] reported the eco-friendly synthesis of 2-arylbenzothiazoles (6) from 2-aminothiophenols and aromatic aldehydes using catalyzed lemon juice under microwave irradiation. They conclude that lemon juice was the green catalyst to synthesis of arylbenzthiazole in excellent yield and easy reaction workup procedure.

Scheme 6. Synthesis of 2-arylbenzothiazoles.

El-Saghie and co-workers [13] reported a synthesis of 2-amino-5(2-hydroxyphenyl)-1, 2, 4-triazole carboxylic acids (7) by using 2-hydroxybenzaldehyde or 2-hydroxy...
acetophenone, substituted amino acids, and thiosemicarbazide in solvent water in the presence of lemon juice.

\[
\begin{align*}
\text{Scheme 7. Synthesis of 2-amino-5(2-hydroxyphenyl)-1, 2, 4-triazole carboxylic acid.}
\end{align*}
\]

Saha and co-workers [14] reported a one-pot three-component synthesis of biologically important tricyclic fused imidazoles using lemon juice as a catalyst. They conclude that all the reactions provided good to excellent yield.

\[
\begin{align*}
\text{Scheme 8. Synthesis of tricyclic fused imidazole.}
\end{align*}
\]

Wadhawa and co-workers [15] reported lemon juice catalyzed one-pot-multicomponent reaction, synthesis of benzimidazoles derivatives (9) from phenylenediamine and different aldehydes in the absence of solvent. All reactions were smoothly conducted, produced excellent yield and easy workup.

\[
\begin{align*}
\text{Scheme 9. Synthesis of substituted benzimidazoles.}
\end{align*}
\]

Pal [16] reported the synthesis of bis-, tris- and tetraindoles (10) from aldehydes and indoles by using citrus lemon juice as a natural catalyst with good yield in aqueous ethanol at pH 3 at 60°C under ultrasound irradiation.

\[
\begin{align*}
\text{Scheme 10. Synthesis of bis-, tris- and tetraindoles derivatives.}
\end{align*}
\]

Vekariya and co-workers [17] reported, one-pot three-component reaction of b-oxoesters with hydroxylamine hydrochloride and various aromatic aldehydes afforded 3,4-
disubstituted isoxazole-5(4H)-ones (11) and 6-amino-1,4-dihydropyran[2,3-c]-pyrazole-5-carbonitrile compounds under hydroalcoholic medium, 90°C temperature by using citrus lemon juice as a natural and renewable catalyst in excellent yields.

Scheme 11. Synthesis of 6-amino-1, 4-dihydropyran [2, 3-c] - pyrazole-5-carbonitrides catalyzed by lemon juice as a green catalyst.

Jelena and co-workers [18] reported experimentally simple, innovative, efficient, and one-pot-multicomponent biocatalyzed synthesis of 3, 4-dihydro-2(1H)-quinoxalinones and 3, 4-dihydro-1, 4-benzoxazin-2-ones (13) by tetracyclization of ethyl 2-hydroxy-4-alkyl(aryl)-4-oxo-2-butenoate or their corresponding salts with o-phenylenediamine or o-aminophenol in lemon juice as a solvent gives excellent yields.

Scheme 13. The general outline of the synthesis of 3, 4-dihydro-2(1H)-quinoxalinones and 3, 4-dihydro-1, 4-benzoxazin-2-ones.

Ayoubi and co-workers [19] reported, synthesis of 4-(benzylidene) aminopyrimidine, (benzo[d]thiazol-2-ylimino)methylphenol, n-benzylidenezine, and n-benzylideneaniline with lemon juice as a natural catalyst and solvent instead of a chemical catalyst. The synthesized product is characterized by melting point, IR, 1H NMR, and 13C NMR spectra. They also conclude that the use of natural catalysts without organic solvent are eco-friendly, affordable, very safe, and easy reaction conditions.

Scheme 14. Synthesis of Schiff base.
Bakht [20] reported lemon juice catalyzed synthesis of Schiff’s base (15) from o-amino benzoic acid and 4-hydroxy-3-methoxy benzaldehyde. All prepared Schiff’s bases were characterized by various spectroscopic techniques and confirmed their particle size, crystallinity, and morphology.

![Scheme 15. Synthesis of classical Schiff’s base.](image)

Das [21] reported lemon juice catalyzed synthesis of fused tricyclic imidazole (16); it is biodegradable, non-hazardous, easily available, alternative for metal-based catalysts. All twelve principles had been covered by this catalyst.

![Scheme 16. Three-component syntheses of fused tricyclic imidazole.](image)

Ishak [22] reported efficient and greener approach had been developed for the lemon juice catalyzed synthesis of 6-arylamino-5-cyano-2,3-dihydro-1,3-thiazin-4(1H)-ones (17). It was prepared via the treatment of 3-arylamino-2-cyano-3-mercptoacrylamides with several aldehydes. They also conclude that the reaction proceeds in a short period with excellent yields.

![Scheme 17. Synthesis of 6-arylamino-5-cyano-2,3-dihydro-1,3-thiazin-4(1H)-ones.](image)

Pal [23] reported condensation of indole with various aldehydes using lemon extract catalyst at 80°C under water solvent and room temperature to afford bis- and tris(3-indolyl)methanes (18).
Scheme 18. Synthesis of bis- and tris(3-indolyl)methane.

Esam [24] reported, synthesis of 6-arylamino-5-cyano-2,3-dihydro-1,3-thiazin-4(1H)-ones (19) carried out by the condensation of 3-arylamino-2-cyano-3- mercapto-acrylamides with various aldehydes under microwave irradiation using lemon extract catalyst. The advantages of the reported method are as follows: available starting materials, short reaction time (min), simple workup, environment friendly, and appreciable yields of reactions.

Scheme 19. Synthesis of 6-arylamino-5-cyano-2, 3-dihydro-1, 3- thiazin-4(1H)-ones.

Sheikh and co-workers [25] reported synthesizing N-substituted pyrroles (20) through Paal-Knorr reaction catalyzed by naturally available, inexpensive, non-hazardous lemon juice. During synthesis, aromatic amines were condensed with 1,4-dione to obtain the pyrrole in the presence of lemon juice.

Scheme 20. Synthesis of N-substituted pyrrole.

Sachdeva and co-workers [26] reported a one-pot multi-component condensation synthesis of Schiff base (21) from substituted 1H-indole-2, 3-diones, and various amino acids thiosemicarbazone is found to be natural acid-catalyzed lemon juice using water as a green solvent in excellent yields.

Scheme 21. Synthesis of Schiff base.
Milovanovi and co-workers [27] reported a one-pot synthesis of 1-benzoyl-1H-pyrazole (22) from acetylacetone with a variety of substituted benzoyl hydrazides in a lemon juice as an eco-friendly medium in a short reaction time in a high yield.

\[
\text{Ar} \text{CONH}_2 + \text{Ar} \text{CON} = \text{O} \xrightarrow{\text{lemon juice, r.t.}} \text{Ar} \text{CON} - \text{N} - \text{N}
\]

**Scheme 22.** Synthesis of 1-benzoyl-1H-pyrazole.

3. **Citric Fruit Juice**

Napoleon and Khan [28] reported a simple and eco-friendly synthesis of 9-aryl/heteroaryl-1,8-dioxo-octahydroxanthenes (23) in the aqueous and alcoholic medium using citrus fruit extract and commercially available citric acid is reported. It involves green catalysts as dibasic, citric acid and includes various benefits such as excellent product yield, non-hazardous solvent, easily available, reusable. This described technique is very easy for operation, environmentally friendly.

\[
\text{CHO} + \text{R} = \text{O} \xrightarrow{\text{citric fruit juice}} \text{R} \text{O} \text{O}
\]

**Scheme 23.** Synthesis of 9-aryl/heteroaryl 1, 8-dioxo-octahydroxanthenes.

Marvi and Fekri [29] reported the one-pot two components synthesis of various β substituted enaminones (24) via the reaction of β dicarbonyl compounds with various primary amines using naturally catalyzed citrus juice.

\[
\text{R}_1 \text{CON} = \text{O} + \text{R} \text{(Ar)} \text{-NH}_2 \xrightarrow{\text{Citrus juice, silica gel grinding, solvent free}} \text{R} \text{(Ar)HN} \text{CON}
\]

**Scheme 24.** Citrus juice catalyzed grinding synthesis of enaminones under solvent-free conditions on silica.

4. **Tamarind Juice**

Pal [30] reported the synthesis of bis and tris(indolyl)methane, and tetraindolyl compounds (25) by electrophilic substitution reaction of indole with aldehydes using microwave irradiations under tamarind fruit juice as a solvent.

\[
\text{R} = \text{H, Me} \quad \text{R}_1 = \text{Aryl, Heteroaryl Alkyl and 3-Indolyl}
\]

**Scheme 25.** Synthesis of bis-, tris(indolyl)methanes, and tetraindolyl compounds.
5. Verjuice

Safari and co-workers [31] reported 5-arylmethylenepyrimidine-2,4,6-trione (26), pyrano[2,3-d] pyrimidinones (27), and pyrimido[4,5-d]pyrimidinone (28) derivatives via Knoevenagel condensation reaction between barbituric or thiobarbituric acid and aldehydes by using verjuice as a catalyst.

\[
\text{ArCHO} + \text{O} = \text{N} - \text{N} + \text{H} - \text{CN} \xrightarrow{\text{Ver juice, } 60^\circ C} \text{Ar} - \text{N} - \text{N} - \text{O} - \text{Ar}
\]

**Scheme 26.** Synthesis of 5-aryl-methylene-pyrimidine-2, 4, 6-trione derivatives in verjuice.

\[
\text{ArCHO} + \text{O} = \text{N} - \text{N} + \text{CN} \xrightarrow{\text{Verjuice 10 ml, reflux}} \text{Ar} - \text{N} - \text{N} - \text{O} - \text{Ar}
\]

**Scheme 27.** Synthesis of pyrano[2, 3-d] pyrimidinone derivatives in verjuice.

\[
\text{ArCHO} + \text{O} = \text{N} - \text{N} + \text{H}_2\text{N} - \text{NH}_2 \xrightarrow{\text{Verjuice reflux}} \text{Ar} - \text{N} - \text{N} - \text{O} - \text{Ar}
\]

**Scheme 28.** Synthesis of pyrimido[4, 5-d] pyrimidinone derivatives in verjuice.

6. Banana Peels

Bagul and co-workers [32] reported the synthesis of 3-carboxycoumarin by one-pot multi-component Knoevenagel condensation and intramolecular cyclization of various 2-hydroxybenzaldehyde with Meldrum's acid, using aqueous extract of banana. The product of yield was produced between 76 to 94% within 490 min at room temperature by filtration method.

\[
\text{WEB} \text{ mol 5%} \xrightarrow{\text{r.t.}} \text{WEB} = \text{water extract of banana}
\]

**Scheme 29.** Synthesis of 3-carboxycoumarin.

7. Orange Juice and Shell Ash

Sheikh [33] reported the synthesis of quinoxaline (30) from the condensation of 1, 2-dicarboxylic acid with 1, 2-diamine had been carried out in the presence of orange juice.
Khatavi and co-workers [34] described, synthesis of 5-aryl-1,2,4-triazolidine-3-thiones and 1,2,4-triazospiro-3-thiones (31) from aromatic aldehyde, cyclic ketone, and thiosemicarbazide in the presence of water extract of orange fruit shell ash (WEOFSA) as an agro-waste catalyst. They concluded that the developed method added advantages like not requiring an external base, inexpensive, environment-friendly, moderate to excellent yield isolation in a straightforward method.

Badiger and Kamanna [35] reported a novel Knoevenagel reaction for the condensation of aromatic/heteroaromatic aldehydes with Malononitrile to give α, β–unsaturated benzylidene derivatives (32) by using water extract of orange fruit peel ash (WEOFPA).

Gulati and co-workers [36] reported a novel benzimidazole derivatives (33) synthesis from the condensation of substituted aldehydes and o-phenylenediamine using Cocos nucifera L., Citrus limetta, and Citrus sinensis L. fruit juice under the solvent-free condition at room temperature. The synthesized compounds in vitro herbicidal activity against Raphanus sativus L. (Radish) seeds and antibacterial evaluation was carried out using Erwinia carovora and Xanthomonas citri stains using the inhibition zone method. Also, antifungal activity was also determined against Rhizoctonia solani and Colletotrichum gloeosporioides stains using poisoned food techniques.
9. **Pineapple Juice.**

Patil and co-workers [37] reported a synthesis of dihydropyrimidinone derivatives (34) accomplished via a multi-component cyclocondensation between aryl aldehyde, diketone/ketoester, and urea by using pineapple extract catalyst. The reaction was done without solvent.

\[
\text{R-CHO} + \text{R}_1\text{C} = \text{O} + \text{H}_2\text{N-CONH}_2 \xrightarrow{\text{Pineapple juice}} \text{R}_1\text{C} = \text{N} - \text{CO} \text{(34)}
\]

**Scheme 34.** Synthesis of dihydropyrimidinones.

Patil and co-workers [38] reported a simple, eco-friendly, and economical method for Knoevenagel condensation (35) of substituted and heteroaromatic aldehydes with Malononitrile, catalyzed by pineapple juice at room temperature in the absence of any other catalyst.

\[
\text{R-CHO} + \text{CN-CN} \xrightarrow{\text{Pineapple juice}} \text{R-CN-CN} \text{(35)}
\]

**Scheme 35.** The general reaction of Knoevenagel condensation.

Gulati and co-workers [39] reported a synthesis of substituted coumarins (36) from various phenols and methyl acetoacetate in the presence of the extract of *Citrus limon, Vitis vinifera L.*, and banana peels had been carried out. Prepared compounds have been screened for *in vitro* herbicidal activity against Raphanus sativus L. (Radish seeds), fungicidal activity against *Rhizoctonia solani*, and *Colletotrichum gloeosporioides* by poisoned food techniques. The antibacterial evaluation was carried out using *Erwinia cartovora* and *Xanthomonas citri* using the inhibition zone method. The activity data found some derivatives most active against Raphanus sativus L. (root) and Raphanus sativus L. (shoot), *R. solani* fungus, and *Xanthomonas citri* bacterium the highest concentration.

\[\text{Method: A)} \ \text{Citrus limon L. juice,} \\
\text{B)} \ \text{Vitis vinifera L. juice} \\
\text{C)} \ \text{banana peels}
\]

**Scheme 36.** Synthesis of coumarin derivatives.

10. **Mango Fruit.**

Hiremath and Kamanna [40] reported WEMPA (Mango Fruit peels) catalyzed synthesis of 1H-Pyrazolo[1,2-b]phthalazine-5,10-dione derivatives (37) from the condensation of aldehydes, Malononitrile, and phthalhydrazide under microwave irradiations in ethanol.
solvent. Mango peels are a wastage material; therefore, it is easily available and free of cost, non-hazardous, etc.

Scheme 37. Synthesis of 1H-Pyrazolo[1,2-b]phthalazine-5,10-dione derivatives.

11. Dragon Fruit

Saifan and Al-horaibi [41] reported Dragon fruit peel extract catalyzed the synthesis of dihydropyridines (38) from the condensation of substituted aromatic aldehydes, ethyl acetoacetate, and ammonium acetate without solvent. Dragon fruit is commercially available, low cost, easily handled, non-hazardous, etc.

Scheme 38. Synthesis of dihydropyridines.

Overviewed of recently published research articles, the pH of fruit juices is weakly acidic. Therefore, those juice could play the role of green catalyst in organic heterocyclic reactions.

12. Conclusions

We conclude that the recently published work on using the extract of fruits as natural catalysts in the formation of heterocycles. All twelve green principles are described in this review through the recent synthesis of heterocyclic compounds. These catalysts have many benefits, such as non-hazardous, clean reaction profile, easy handle, low cost, easily available, etc. We concluded that fruit extract catalyst is one of the best options for recyclization of organic heterocycles in the future.

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Conflicts of Interest

The authors declare no conflict of interest.

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