

GREEN AND ENVIRONMENTALLY BENIGN ORGANIC SYNTHESIS BY USING FRUIT JUICE AS BIOCATALYST: A REVIEW

ABSTRACT: Facile and green synthetic approaches are important issues in organic synthesis. Green chemistry has become a motivational and inspirational tool for organic chemists to develop mild and benign pathways for the synthesis of biologically active compounds. The naturally available fruit juice as a biocatalyst in synthesis fulfills almost all the terms and conditions of green chemistry and attracted the interest of researchers. The best thing is that most of fruits are easily available, cheap and can be easily extracted. The purpose of this review is to look out present aspects of fruit juice in organic transformations.

KEYWORDS: Fruit juice, Eco-friendly, Biocatalyst, Organic Synthesis

1. INTRODUCTION

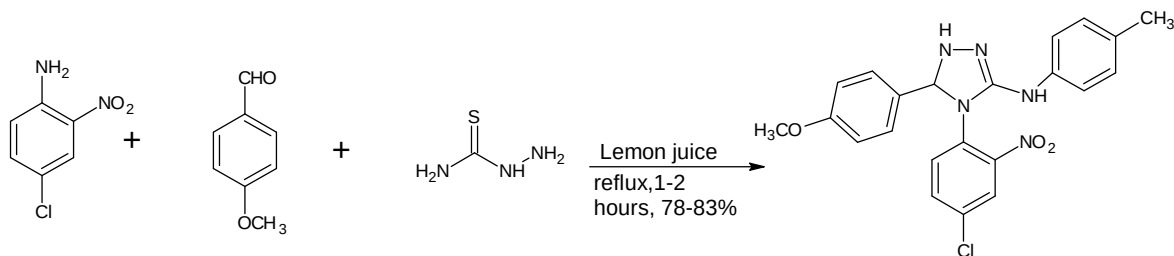
With the start of twenty-first century, the public is much more aware of the hazardous substances used and produced during the chemical reactions, and hence the ‘*concept of green and sustainable chemistry*’ has been evolved. The main aim of this concept is to develop the smooth and nonpolluting pathways and to find creative ways to reduce the use of toxic reagents, solvents, harsh reaction conditions and expensive catalysts [1-3]. The experimental trials of new catalyst in an environmentally benign manner have become much more important in recent years [4]. The conventional synthesis of chemicals produces large amount of toxic wastes and by-products [5]. These threats indicate that it must be important to develop methods which satisfy green principles [6-7]. Among various green chemistry aspects, selection of catalysts for mild running of chemical reaction with optimum yield is most important part of the reaction procedure. Fruit juice plays an important role as a biocatalyst in many of the chemical reactions and this biocatalyst follows all the parameters of green chemistry. The multi-component reactions are the most important tools in organic transformations and pharmaceuticals. The concept of “Green Chemistry” has been widely accepted to complete the basic scientific challenges of protecting human health and environment [8]. In order to complete these needs, chemical reactions are proceeds in solvent-free [9-10], water as a solvent [11], ionic liquids [12], bio-based chemicals [13] and supercritical fluids [14] as green solvents. The increasing interest in fruit juice because these

are available at low cost and non-toxic agents that can carry out an organic transformation in an environmentally benign manner. Fruit juice acts as homogenous catalyst for various organic reactions in easy and smooth way. The main motive of the present review is to represent the environmentally healthy use of fruit juice in organic transformations.

2. FRUIT JUICE AS A BIOCATALYST IN ORGANIC SYNTHESIS



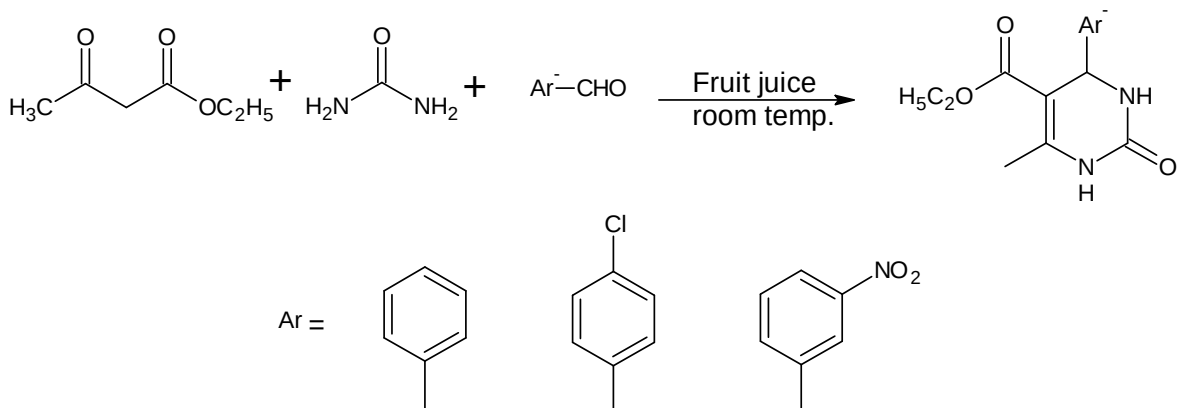
2.1 Sachdeva *et al.*, 2013 reported the multi-component synthesis of substituted -2H-1,2,3-triazoles derivatives using lemon juice in ethanol by the reaction of 4-chloro-2-nitro aniline and 4-methoxy aldehyde with thiosemicarbazide in maximum yield. They found that lemon juice plays a role of biocatalyst which provides non hazardous and mild conditions which are basic principles of green chemistry (**Scheme-1**).



R= OH, OCH₃, CH₃, Cl

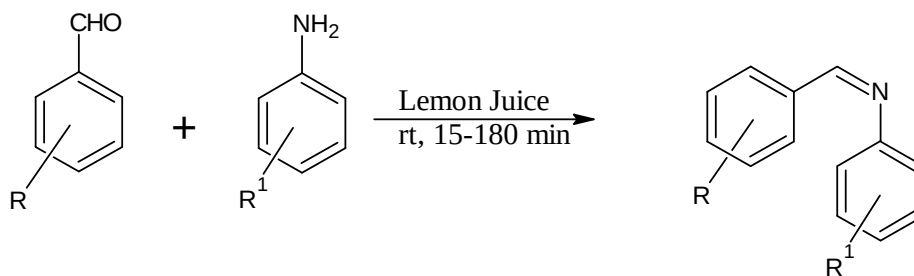
Scheme - 1

2.2 Dihydropyrimidinone **has** been synthesized by Patil *et al* (2011). These derivatives synthesized by reacting aldehydes, 1,3-dicarbonyl compounds with urea at room temperature under solvent free conditions. This is a one-pot multi-component system reaction where lemon juice or pineapple acts as a green catalyst (**Scheme - 2**).



Scheme – 2

2.3 Patil *et al* (2012) reported the synthesis of Schiff Base by using lemon juice (*Citrus limonium*) as an effective and mild acid catalyst for condensation reaction. This synthesis shows the formation of selective imine by the reaction of aryl aldehyde and aromatic primary amine. This method provides a cost effective idea and benefits from the elimination of production of acidic waste (**Scheme - 3**).

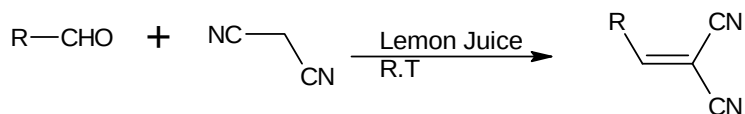


R= H, OH, OMe, NO₂, NMe₂

R¹= H, Me, OMe, Br, NO₂

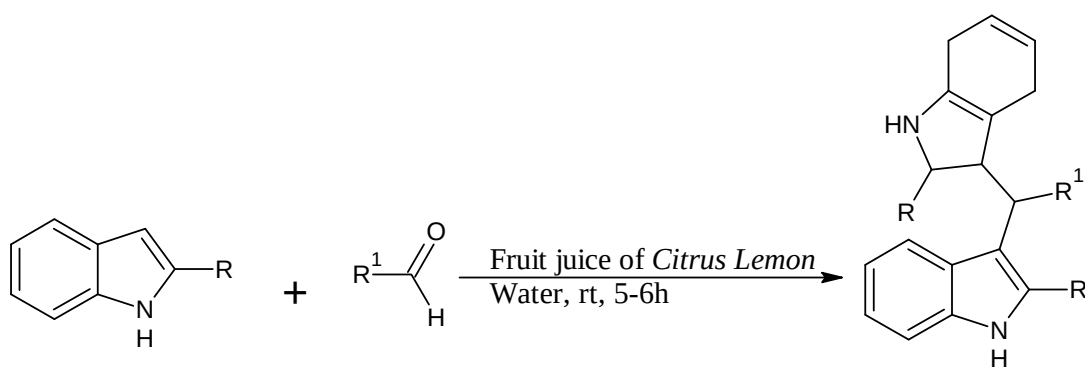
Scheme - 3

2.4 A green procedure for Knoevenagel condensation was reported by Deshmukh *et al* (2012). They showed that lemon juice (*Citrus limonium*) act as environmentally benign acid catalyst for the reaction between aldehydes and malanonitriles. The mixture was stirred at room temperature for 30-120 minutes. This is new procedure by lemon juice qualifying it is a green method (**Scheme - 4**).



Scheme- 4

2.5 Pal *et al* (2013) reported the biocondensation of indoles and aldehydes for the synthesis of bis-, and tris (indoyl) methanes. This reaction proceeds indoles and aldehydes were treated in lemon juice (*Citrus limonium*) mixture at pH 3 in stirring at room temperature for 5-6 hour. This method is suitable in term of short reaction time, reaction procedure is very simple and attained better yield of product (**Scheme - 5**).

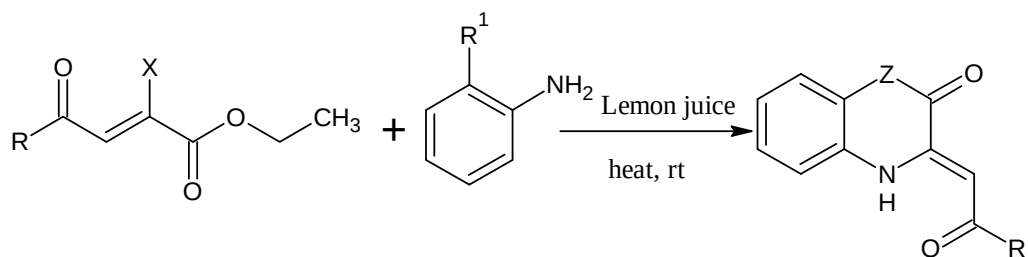


R= H, Me

R¹= aryl, heteroaryl, alkyl

Scheme - 5

2.6 Petronijevic *et al* (2017) reported ecofriendly and clean one-pot synthesis of 3,4-dihydro-2(1H)-quinoxalinones and 3,4-dihydro-1,4-benzoxazine-2-ones. Lemon juice (*Citrus limonium*) acts as an alternative to toxic solvents and catalysts. This reaction proceeds by substituted ester keto ester and substituted amines with lemon juice and reflux for 24h. After simple work up the newly synthesized products were isolated (**Scheme - 6**).



84

85 X= OH, ONa

R¹= NH₂, OH

Z= NH,

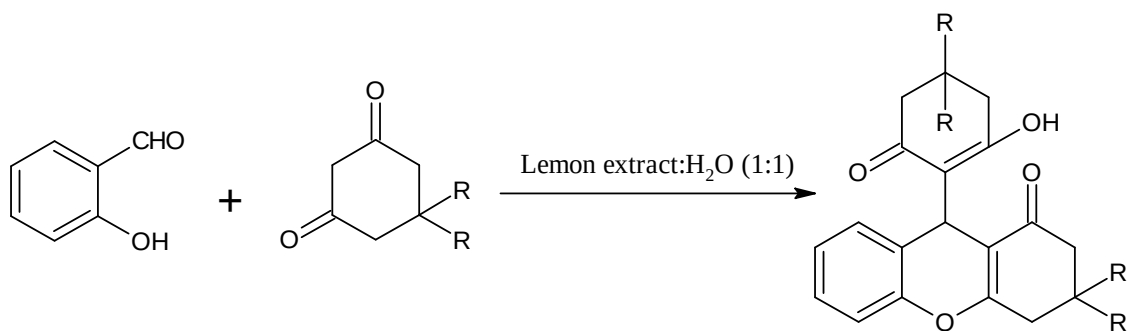
86 O

87 R= cyclopropyl, alkyl, substituted aryl

88

Scheme - 6

89 2.7 Morbale *et al* (2015) reported green and economic method for the synthesis of
 90 benzopyran synthesis. They found that lemon juice (*Citrus limonium*) act as acid catalyst for
 91 cyclocondensation of salicylaldehyde and cyclic 1,3-diketones for benzopyran formation
 92 (Scheme-7).



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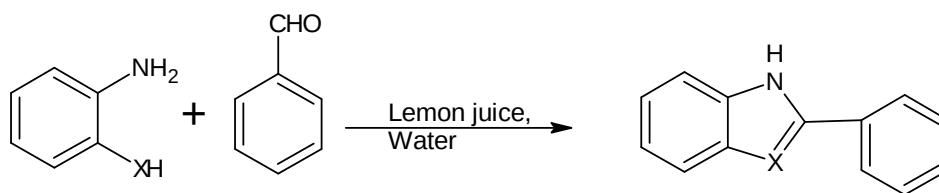
94 R= CH₃, H

95

Scheme-7

96 2.8 Patil *et al* (2017) reported the multi-component synthesis of substituted benzthiazole and
 97 benzoxazole derivatives using lemon juice in water by the reaction of 2-aminothiophenol or
 98 2-amonophenol, aryl aldehyde and lemon water mixture (1:1) was added in reaction vessel.
 99 They found that lemon juice plays a role of biocatalyst which provides non hazardous and

100 mild conditions which are basic principle of green chemistry (**Scheme-8**).

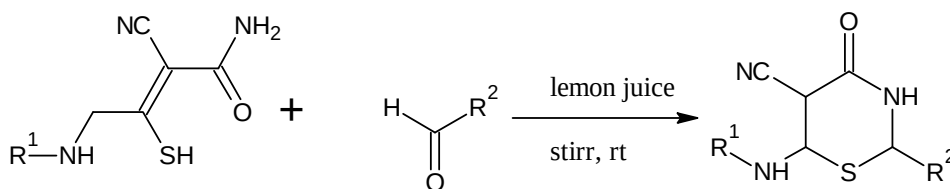


101

102 X=S, O

103 **Scheme- 8**

104 2.9 An efficient and greener approach has been developed for the synthesis of 6-arylamino-5-
 105 cyano-2,3-dihydro-1,3-thiazin-4(1*H*)-ones, using Lemon juice (*Citrus limonium*) as a natural
 106 catalyst by Ishak *et al* (2017). It was prepared *via* condensation of 3-arylamino-2-cyano-3-
 107 mercaptoacrylamides with several of aldehydes. The reaction proceeded in ecofriendly
 108 manner with excellent yields (**Scheme-9**). Most of the synthesized compounds were reprints
 109 effective promising antimicrobial activity.



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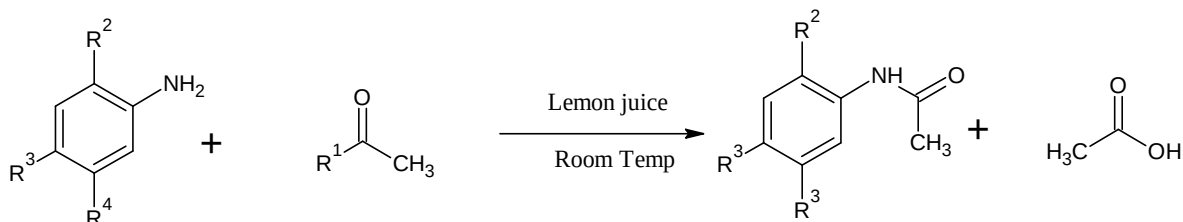
111 R¹= Ph, Ph-CH₂

112 R²= Ph, Ph-CH₃, Ph-Cl, Ph-NO₂, Ph-OCH₃

113 **Scheme-9**

114

115 2.10 Chavhan *et al* (2016) reported the acetylation of different amines and salicylic acid by
 116 using lemon juice (*Citrus limonium*) and acetic anhydride as an acetylating agent. This
 117 reaction proceeds under normal reaction condition with formation of product in high yield.
 118 This method reduce the chemical wastage and more convenient than other reported methods
 119 (**Scheme - 10**).



120

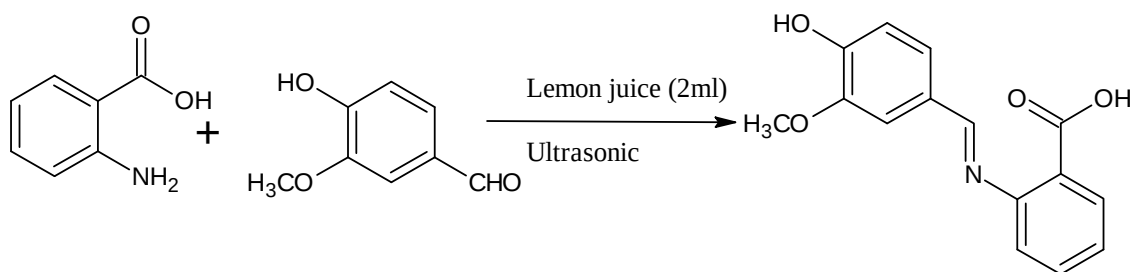
121

122 R¹= CH₃COO

$R_2, R_3, R_4 = \text{NH}_2, \text{NO}_2, \text{CH}_3, \text{OH}$

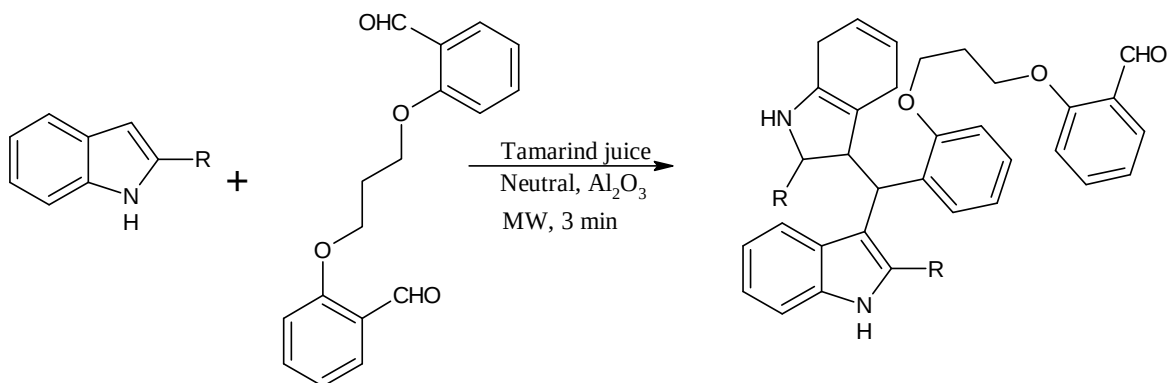
Scheme – 10

2.11 Bakhat (2015) reported a lemon juice catalysed ultrasound assisted, ecofriendly synthesis of schiff's base. A mixture of o-benzoic acid and 4-hydroxy-3-methoxy benzaldehyde was taken in sonication flask and then 2ml lemon juice was added drop wise under sonication probe (ACE probe, 20 KHz) at 40% amplitude for 2min. After complete addition of lemon juice, the reaction mixture was again exposed to acoustic cavitation for further 10 min, by keeping all the parameter constant. The completion of reaction was measured by TLC. The recrystallization was done by ethanol and a white solid pure product was obtained (Scheme - 11).



Scheme - 11

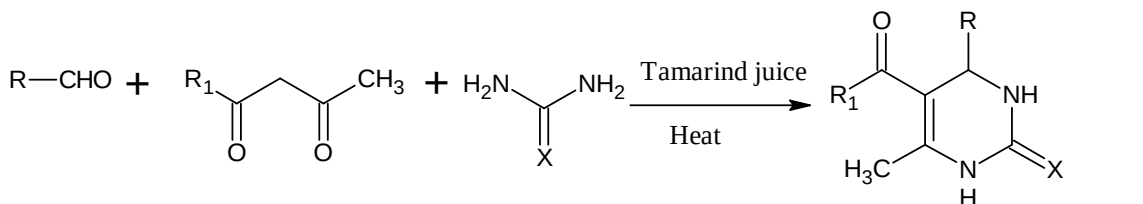
2.12 Tamarind (*Tamarindus indica*) fruit act as a biocatalyst for synthesis of bis-, and tris (indoyl) methanes and tetraindoyl compounds. This novel pathway of synthesis was first time reported by Pal (2013). Excellent yield of product have been obtained by reaction of indoles with aldehydes by using microwave irradiation under solvent free conditions (Scheme - 12).



$R = \text{H}, \text{Me}$

Scheme - 12

2.13 Nazeruddin *et al* (2014) reported the synthesized the dihydropyrimidinone derivatives by reacting between aldehyde, ethylacetoacetate, urea/thiourea and 1ml tamarind juice (*Tamarindus indica*) was irradiated in an ultrasound at 100W for few minutes. Microwave synthesis shows advantage over conventional heating by time, extent of chemicals used and by yield (**Scheme - 13**)



X= O,

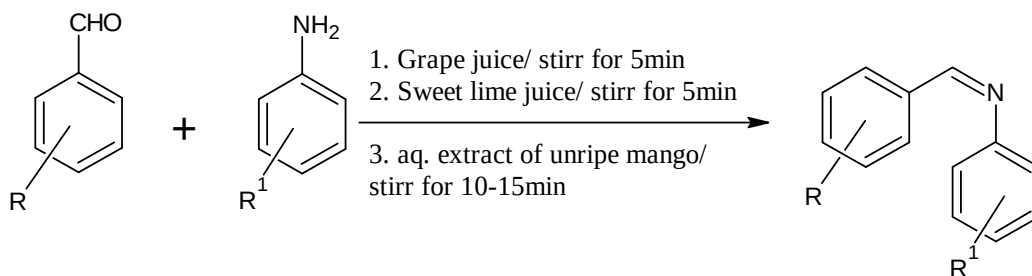
S

R= Ph, Ph-OMe, Ph-OH, Ph-NO₂, Ph-Cl,

R₁= OEt

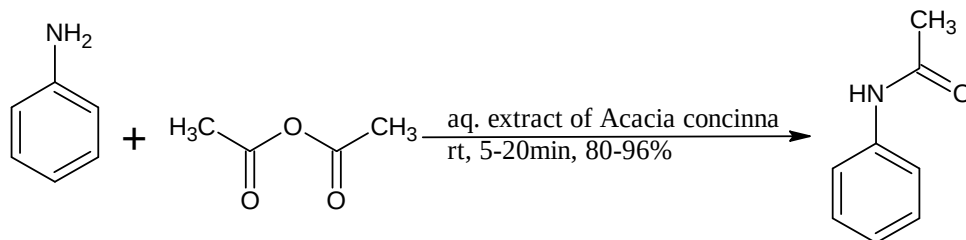
Scheme – 13

2.14 Yadav and Mani (2015) reported the green synthesis of Schiff bases with grape juice (*Vitis Linata*), sweet lemon juice (*Citrus Limetta*) and extract of unripe mango (*Mangifera indica*) under solvent free conditions. This reaction proceeds by stirring method, when the equimolar amount of benzaldehyde and aniline was taken in different beakers. In those reaction mixtures natural acid catalyst i.e. grapes juice were added in variable amounts (0.5 ml, 1 ml, 1.5 ml, 2.0 ml, 2.5 ml) and then kept for 5-10 minutes. Further each reaction mixture was stirred for 2 - 4 minutes at room temperature pale yellow solid crude product was appear after completion of reaction which was washed with distilled water and purified by recrystallization with minimum amount of ethanol. The same procedure is repeated with sweet lemon juice and aqueous extract of mango (**Scheme - 14**).



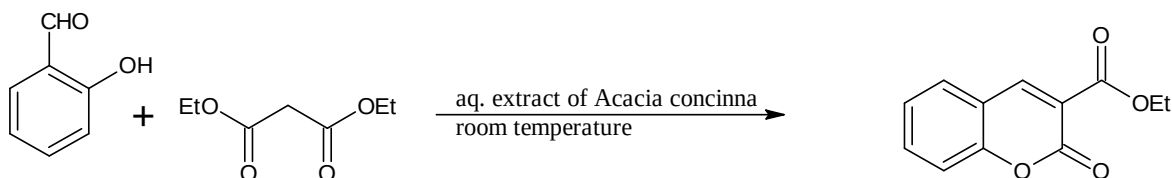
Scheme - 14

2.15 An ecofriendly and cost effective protocol was reported by Mote *et al* (2010) for the synthesis of acetanilide. The reaction proceeds by reacting aromatic primary amines and acetic anhydride, catalysed by aqueous extract of pods of *acacia concinna* fruit as a green catalyst. This catalyst shows highly acidic nature (pH 2.1) and high solubility of reactant by hydrogen bond formation in aqueous solution. This type of unique protocol shows the economic and green pathways (**Scheme - 15**).



Scheme-15

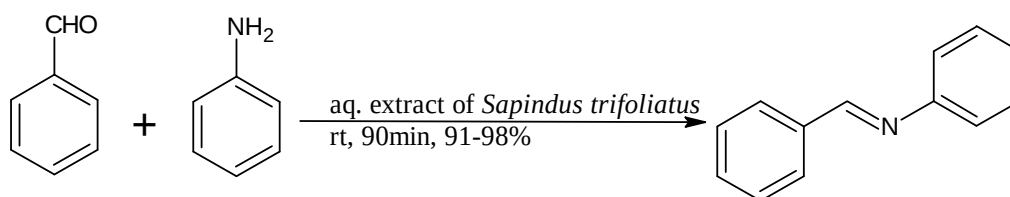
2.16 An aqueous extract of *Acacia concinna* pods has been reported as a biocatalyst by Shafqat *et al* (2017) for Knoevenagel Condensation. A equimolar mixture of salicylaldehyde and malonic ester and aqueous extract of *Acacia conccina* were taken in a round bottom flask and heated along with stirring at boiling water bath for 2h. After completion of the reaction, the reaction mixture as cooled at room temperature, the solid product was filtered off and recrystallized to get pure product (**Scheme - 16**).



Scheme - 16

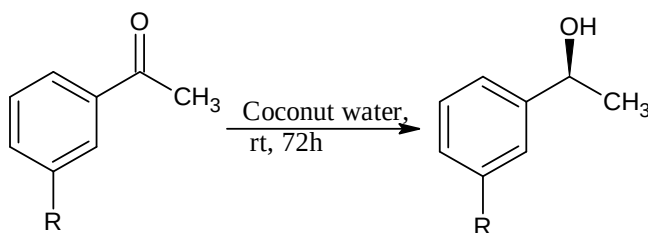
2.17 The aqueous extract of the pericarp of *Sapindus trifoliatus* acts as a biocatalyst in the synthesis of al-damines in a chemoselective manner. This reaction occurs by stirring the mixture of aromatic aldehydes and aromatic amines in presence of this fruit extract (**Scheme**

- 17). This method provides a valuable reaction pathway against conventional method reported by Pore *et al* (2010).



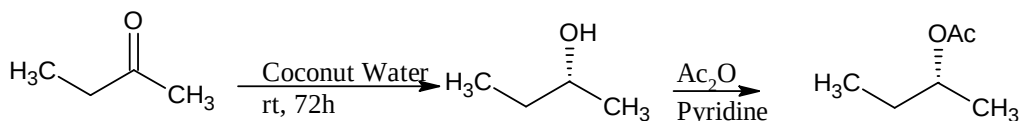
Scheme - 17

2.18 Fonseca *et al* (2009) reported the most sustainable reduction of aliphatic and aromatic aldehydes and ketones by using **fresh** coconut juice (*Cocos nucifera*) as a biocatalyst. Thus, when the various carbonyl compounds were treated with freshly prepared coconut and the mixture was shaken at room temperature for 72 h, produced alcohols (**Scheme – 18a**), (**Scheme – 18b**), (**Scheme – 18c**), (**Scheme – 18d**). Fonseca and co-workers also reported the hydrolysis of ester (**Scheme – 19a**), amide (**Scheme – 19b**) and anilides (**Scheme – 19c**) by using coconut juice. This bioconversion provides a significant role of green chemistry.

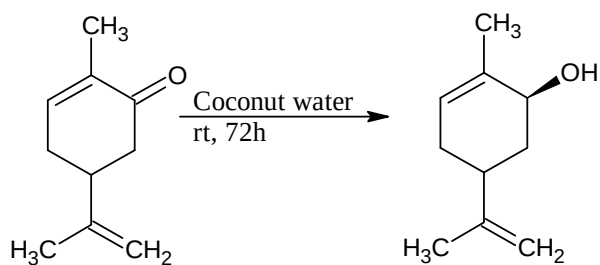


R= H, OCH₃

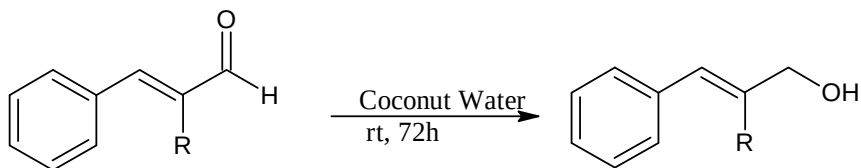
Scheme – 18a



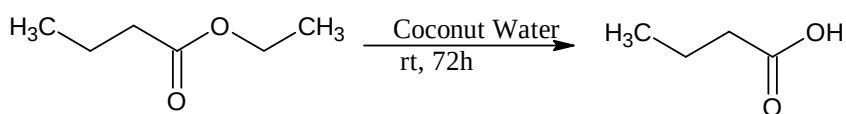
Scheme – 18b



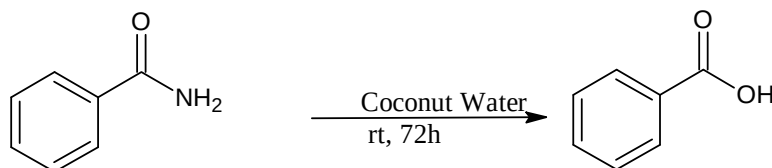
Scheme – 18c



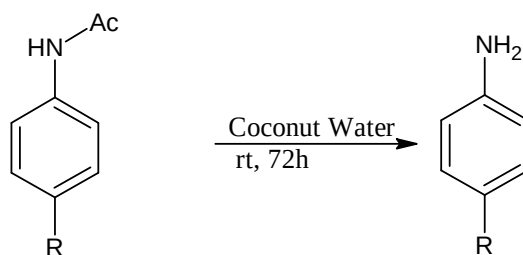
Scheme – 18d



Scheme – 19a



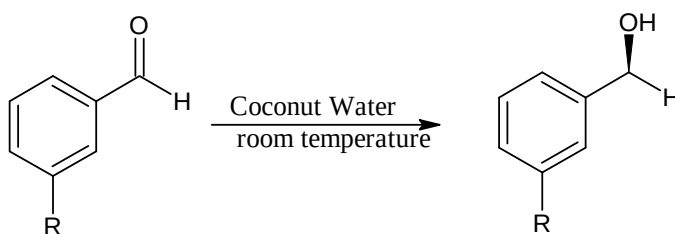
Scheme – 19b



R= OH, H

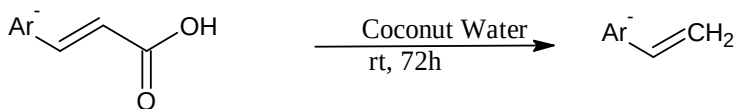
Scheme – 19c

2.19 Misra *et al* (2012) reported a new greener **alternative** for bioreduction of aromatic aldehydes (**Scheme – 20a**) and decarboxylation of substituted aromatic acids (**Scheme – 20b**) using coconut water as a biocatalyst. This is an effort towards the energy efficient, eco-friendly transformation of interesting organic molecule.



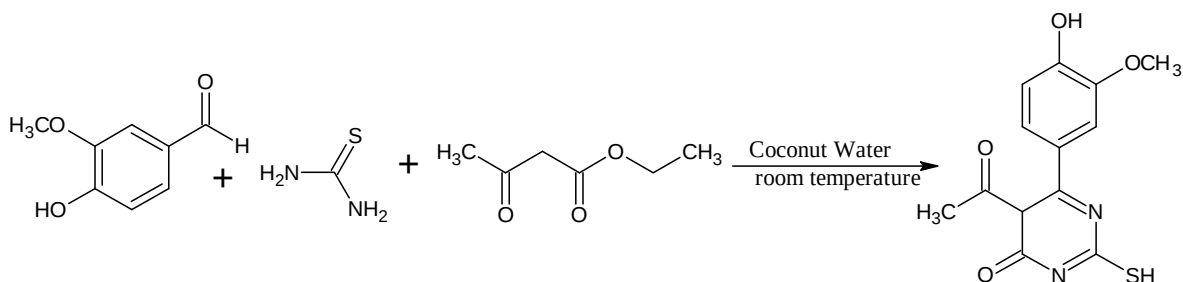
R= H, OH, OMe, Me

Scheme – 20a



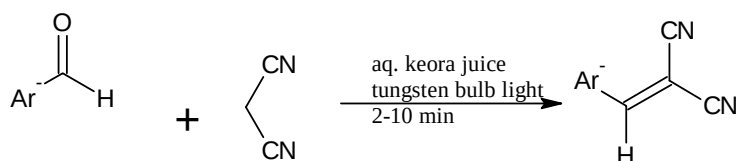
Scheme – 20b

2.20 Hajite (2017) reported the synthesis of vanillin derivative using coconut water as a green catalyst. Thus, when equimolar amount of vaniline, ethylacetoacetate, thiourea and 5ml coconut water and carried out the reaction in microwave for 120 seconds by using minimum power 180 watts. After a simple work up and recrystallization is done by proper solvent, the pure product with a good yield was obtained (**Scheme – 21**). This method is very simple, economical, mild and environmentally benign as compared to classical reactions.



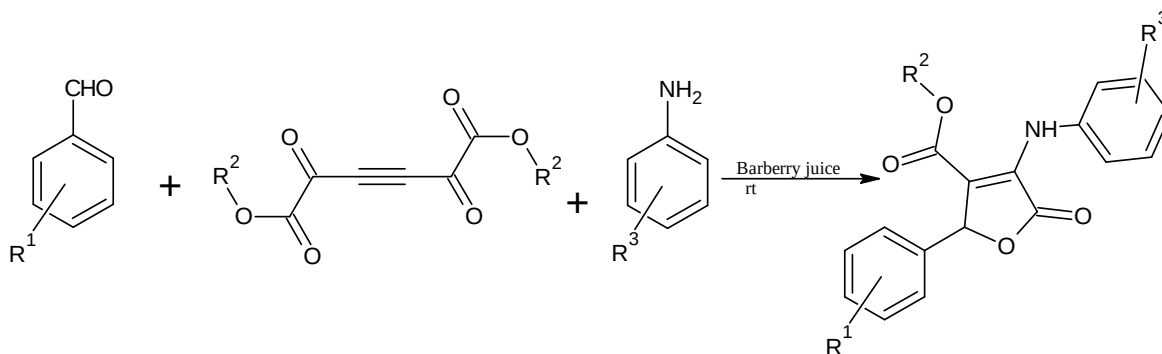
Scheme – 21

2.21 Pal (2016) reported the synthesis of arylidene malononitrile by Knoevenagel condensation reaction of aldehydes and malononitrile under visible light produced by tungsten bulb. Aqueous extract of keora fruit can catalyze the Knoevenagel condensation with a good yield of products. Aqueous extract of keora is acidic and hence it could be work as acid catalyst for reaction (**Scheme-22**).



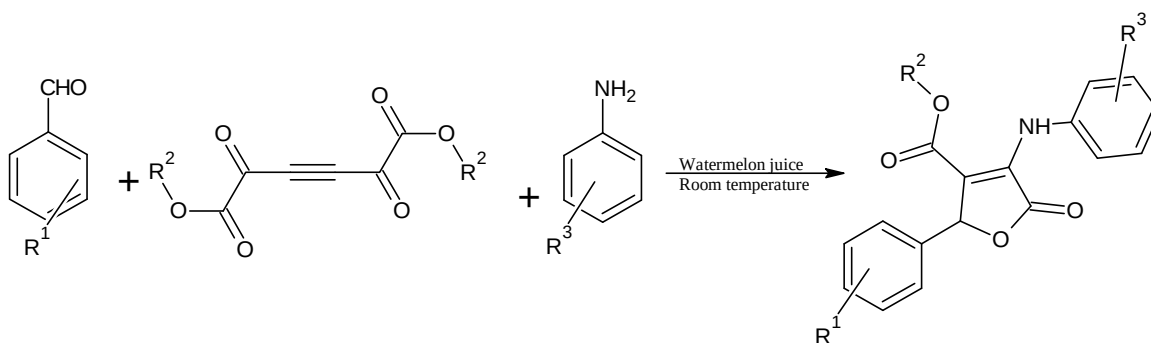
Scheme-22

2.22 A mild and environmentally benign synthesis of 3,4,5- substituted furan-2(5)-ones was reported by Hazeri *et al* (2016) employing aqueous extract of seedless barberry (*Berberis integerrima*) as a biocatalyst. The equimolar amount of substituted aldehyde, substituted amine and dialkylacetylene dicarboxylate and 5ml of barberry juice were stirred at room temperature. This method prevents the generation of waste rather than the conventional chemical reagent (**Scheme-23**).



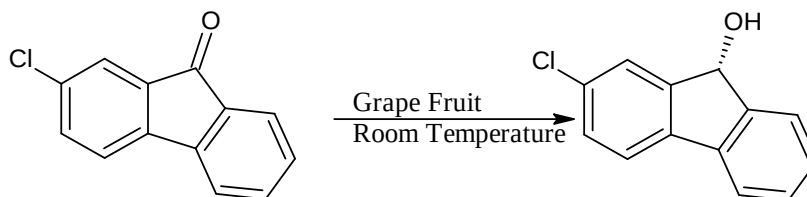
Scheme-23

2.23 Adrom *et al* (2016) reported an efficient multicomponent synthesis of 3,4,5- substituted Furan-2(5H)-ones catalysed by watermelon juice. The equimolar amount of substituted aldehyde, substituted amine and dialkylacetylene dicarboxylate and 5ml of water melon juice of watermelon were stirred at room temperature. The completion of reaction was checked by TLC, after that the reaction products were collected by filtration. Thus, the products were washed with water/ethanol (50:50) to give the pure compound. The catalyst remained in the water/ethanol filtrate (**Scheme 24**).



Scheme- 24

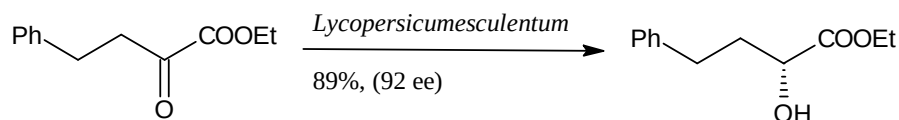
2.24 Xie *et al.*, 2012 reported a simplified green chemistry approaches in organic transformation. Grape fruit which is an efficient and mild biocatalyst for some selected organic reactions in solvent-free conditions. This catalyst is stereoselective in the biotransformation of chloro-polycyclic aromatic compounds (**Scheme – 25**).



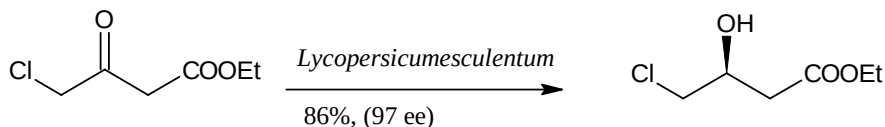
Scheme – 25

2.25 Assymetric reduction of Ketones by using Tomato juice (*Lycopersicum esculentum*) as a biocatalyst is reported by Phukan and Devi (2012). Thus, this reaction provides a biocatalytic preparative method of asymmetric alcohols (**Scheme – 26a**), (**Scheme – 26b**) and (**Scheme –**

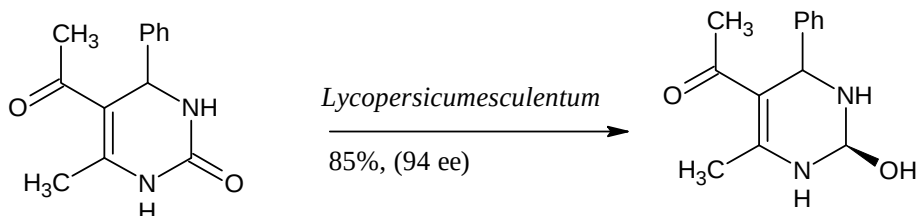
26c). This technique is more ecofriendly and provides an important approach towards green chemistry.



Scheme – 26a

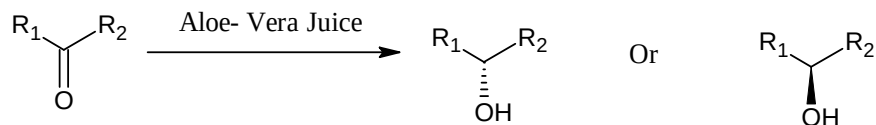


Scheme – 26b



Scheme – 26c

2.26 Fast microwave assisted bioreduction of aromatic and aliphatic aldehydes and ketones by using Aloe vera juice (**Scheme - 27**). Microwave-assisted synthesis was used in the biotransformation of these organic compounds which acts as a very useful **intermediate** in various organic reactions. This simple, efficient and ecofriendly method was reported by Leyva *et al* (2012).

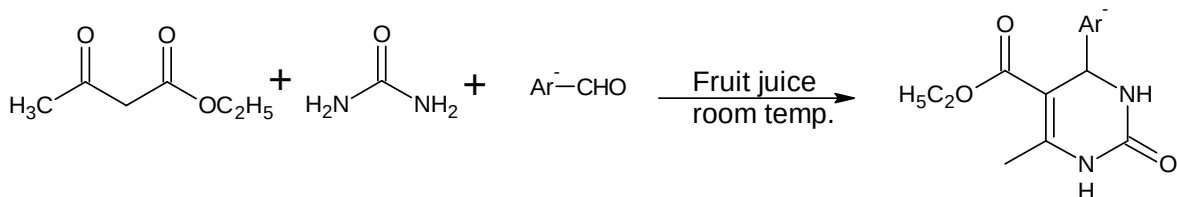


R1, R2 = H, Aliphatic, Aromatic, Saturated, Unsaturated, Cyclic etc

Scheme-27

2.27 Pranamik and Pathan (2014) reported the synthesis of Dihydropyrimidinone (DHPM) derivatives via one-pot multicomponent cyclocondensation namely Biginelli reaction employing urea, ethylacetoacetate with a series of different derivatives of benzaldehyde in

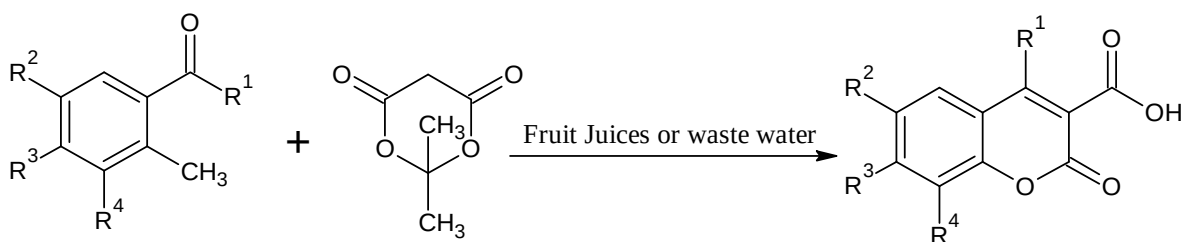
fruit juice (viz. orange juice, lime juice, amla juice) at room temperature (**Scheme-28**). They found that all the fruit juices are totally non-polluting, inexpensive and 100% biodegradable.



Ar = Ph-CH₃, Ph-Cl, Ph-OCH₃

Scheme-28

2.28 Fiorito *et al* (2016) reported a great contribution to the green chemistry by using crop-derived products, like juices obtained from edible fruits and vegetables and waste waters deriving from agriculture and industrial processing. They described the juices waste water promoted synthesis of coumarin-3-carboxylic acid (**Scheme-29**), and synthesis of cinnamic acids (**Scheme-30**) with high purity. The Knoevenagel condensation have been accomplished by using substituted aldehyde and Meldrum's acid as substrates, both are mixed in the aqueous medium represented by lemon, grapefruit, carrot, pomegranate, kiwi, vinegar, tomato and buttermilk waste water. The process was carried out for 24h under magnetic stirring at room temperature. Pure product have been isolated as solids after filtration in very good yields and the biocatalysts were recovered after filtration from all reaction media reused without any loss of activity.



Scheme-29 (Table 1)

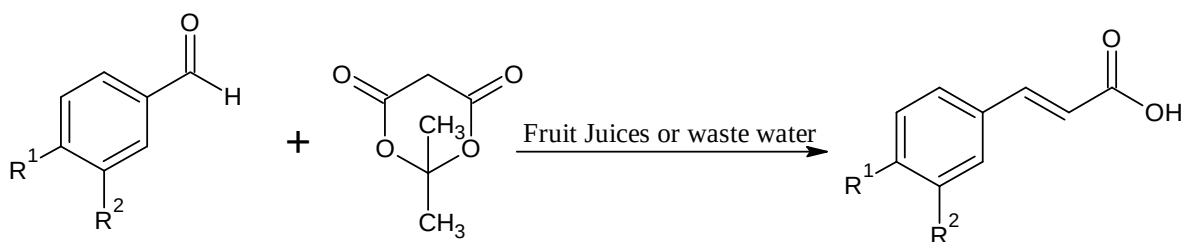
Table 1

S.N o.	Carbonyl Compounds	Product (Scheme-29)	A %	B %	C %	D %	E %	F %	G %	H %	I %	J %
1	Salicyladehyde	R ¹ =R ² =R ³ = R ⁴ =H	99	96	97	96	95	95	95	92	94	99

2	4-(Dimethylamino) salicylaldehyde	$R^1=R^2=R^4=H$, $R^3=N(Et)_2$	99	99	92	93	94	96	96	97	91	96
3	4-nitrosalicylaldehyde	$R^1=R^2=R^4=H$, $R^3=NO_2$	98	97	97	98	99	98	99	98	96	97
4	5-bromosalicylaldehyde	$R^1=R^3=R^4=H$, $R^2=Br$	99	97	99	99	98	95	97	99	99	98
5	2,4-dihydroxybenzaldehyde	$R^1=R^2=R^4=H$, $R^3=OH$	97	97	95	98	98	98	99	95	98	99
6	2-hydroxy-5-nitroacetophenone	$R^1=Me, R^2=NO_2$ $R^3=R^4=H$	96	95	98	98	98	99	98	99	96	98
7	5-chloro-2-hydroxyacetophenone	$R^1=Me, R^2=Cl$ $R^3=R^4=H$	98	97	99	99	96	98	99	96	97	98
8	2-hydroxy-4-methoxyacetophenone	$R^1=Me, R^2=OMe$ $R^3=R^4=H$	96	95	98	98	98	99	98	99	96	98
9	2,5-dihydroxyacetophenone	$R^1=Me, R^2=OH$ $R^3=R^4=H$	94	94	98	97	99	97	98	99	97	96
10	2,3,5-trihydroxyacetophenone	$R^1=Me, R^2=OH$ $R^3=H, R^4=OH$	95	96	99	96	99	99	96	99	98	95
11	2-Hydroxyacetophenone	$R^1=Me$, $R^2=R^3=R^4=H$	94	98	98	94	97	98	99	99	97	95

315 A= lemon juice, B= grapefruit juice, C= carrot juice, D= pomegranate juice, E= kiwi
316 juice, F= vinegar, G= tomato juice, H= limencello, I= olive mill waste water, J=
317 buttermilk, %Y= Percentage Yield

318



Scheme-30 (Table 2)

Table 2

S.N o.	Carbonyl Compounds	Product (Scheme-30)	A %	B %	C %	D %	E %	F %	G %	H %	I %	J %
			Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
1	Benzaldehyde	$R^1=R^2=H$	98	91	96	92	98	92	96	98	95	94
2	Vanillin	$R^1=OH,$ $R^2=OCH_3$	95	92	94	91	94	93	97	98	93	98
3	p- hydroxybenzaldehyde	$R^1=OH, R^2=H$	97	97	94	95	96	95	92	97	98	96
4	p-floro- benzaldehyde	$R^1=F, R^2=H$	92	95	94	95	97	97	93	98	97	99
5	p-chloro- benzaldehyde	$R^1=Cl, R^2=H$	93	95	98	95	96	95	98	99	99	95
6	p-bromo- benzaldehyde	$R^1=Br, R^2=H$	95	95	99	98	99	91	99	97	98	97
7	p-iodo- benzaldehyde	$R^1=I, R^2=H$	97	94	94	99	95	94	92	95	99	97
8	3,4- dihydroxybenzaldehyde	$R^1=R^2=OH$	98	99	96	99	97	98	91	98	96	97
9	p- nitrobenzaldehyde	$R^1=NO_2, R^2=H$	98	98	96	99	97	99	95	94	99	94
10	p-	$R^1=NH_2, R^2=H$	99	95	97	93	98	94	97	98	99	97

	aminobenzaldehyde											
11	4(3,3,Dimethoxyallyloxy)-3methoxybenzaldehyde	$R^1=3,3,Dimethylallyloxy$ $R^2=OCH_3$	95	99	97	92	99	96	97	97	96	99

324 **A= lemon juice, B= grapefruit juice, C= carrot juice, D= pomegranate juice, E= kiwi**
325 **juice, F= vinegar, G= tomato juice, H= limencello, I= olive mill waste water, J=**
326 **buttermilk, %Y= Percentage Yield**

327 3. CONCLUSION

328 Natural juices contain various acids act as natural acid catalyst in organic synthesis. These
329 reactions were carried out at room temperature and under solvent free conditions. This review
330 study provides an alternative for harmful catalysts for organic synthesis. These biocatalyst
331 does not harm to environment and economic in nature. This article is dedicated to green
332 chemistry and an attempt to suggest 'Go Green' for all academic and industrial researchers.

333 5. REFERENCES

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