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Tamarind juice catalyzed one pot synthesis of dihydropyrimidinone and thione under ultrasound irradiation at ambient conditions: A green approach

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ABSTRACT

Application of fruit juice as a natural and biocatalyst allows mild and highly selective transformation and synthesis in a facile and environmentally friendly manner. Moreover, fruits are inexpensive and easily available in the market, the extracted juice can be easily used as catalyst in the organic transformations. Tamarind juice catalyzed synthesis of dihydropyrimidinone and thione (DHPMs) derivatives were accomplished via Biginelli reaction between substituted aryl aldehydes, ethyl acetoacetate and thiourea/urea under ultrasound irradiation at ambient conditions. The tamarind juice can be simply prepared from fruit of Tamarindus indica in water and the reactions go to completion under ultrasound irradiation within few minutes with excellent yield. This approach is totally non polluting having several advantages such as shorter reaction time, mild reaction conditions, simple workup and moreover a green approach.

Keywords: Biocatalyst, Muticomponent Reaction, Sonicator, Dihydropyrimidinone/ thione.

INTRODUCTION

Multi-component reactions have emerged as powerful tools in the drug discovery endeavour [1-4], because of their potential for the generation of molecular diversity in a single synthetic step. The multicomponent reactions (MCR's) are one of the most important protocols in organic synthesis and medicinal chemistry. [5] This unique characteristics of the multicomponent reaction have attracted the attention of organic chemists. Moreover, higher selectivity is usually observed and the products can be easily isolated with good chemical purity by simple filtration avoiding more time consumption and tedious extractive workup. The concept of "Green Chemistry" [6] has been widely adopted to meet the fundamental scientific challenges of protecting human health and environment while simultaneously achieving commercial viability. One of the thrust areas for achieving this target is to explore aqueous reaction medium for accomplishing the desired chemical transformation and eliminating the use of organic solvents.[7] Heterocyclic molecules are of biological interest due to their potential physical and chemical properties.[8] 3,4-Dihydropyrimidin-2(1H)-ones and thiones derivative have attracted increasing interest owing to their therapeutic and pharmaceutical properties, such as antiviral, antibacterial, anti-inflammatory and antitumor activities.[9-11] Development of non-hazardous synthetic methodologies for various organic reactions is one of the latest challenges to the organic chemists. More emphasis is now given for the development of eco-friendly and economic processes. Tamarind juice as natural catalyst, due to its acidic nature (pH= 3) has been found to be a suitable replacement for various homogeneous acid catalysts. The role of naturally available fruit juice in organic synthesis has attracted the interest of chemists, particularly from the view of green chemistry. In literature, a number of organic reactions using natural catalysts such as clay [12-13], natural phosphates [14-16], animal bone [17] and various fruit juices are reported. Due to acidic nature aqueous fruit juice like lemon [18-23], pineapple [24-25], coconut [26], *Acacia concinna* [27], *Sapindus trifolistus* [28] and *Tamarindus indica* [29] fruits have been found to be a suitable replacement for various homogeneous acid catalysts. In recent years, organic research is mainly focused on the development of greener and eco-friendly processes which involve in the use of alternative reaction media to replace toxic and expensive catalysts or volatile and hazardous solvents like benzene, toluene and

methanol, commonly used in organic synthesis. Nowadays, many organic transformations have been carried out in water [30-32]. Water is unique solvent because it is readily available, inexpensive, nontoxic, safer and environmentally benign. The use of water as a reaction medium is not only inexpensive and environmentally benign but also provides completely different reactivity.[33] The applications of an aqueous extract of different fruit juice have witnessed a rapid development. This growing interest in fruit juice is mainly because of its biocatalyst behaviour, environmentally benign character and non hazardous and cost effectiveness. Fruit juice is also naturally occurring which was used as a biocatalyst in organic synthesis. Fruit juice is now being routinely used in organic synthesis as homogeneous catalysts for various selective transformations of simple and complex molecules.

Tamarind (*Tamarindus indica*) is grown extensively in Africa, South Asia, Northern Australia, South East Asia, Taiwan and China. Tamarind has long been one of the most popular of the non-citrus tropical and subtropical fruits, largely because of its attractive flavour and refreshing sugar-acid balance. The main ingredients [34] of 100 gm of pulp of tamarind fruit contain water (15-30%), protein (2-9%), fat (0.5- 3%), total carbohydrate (56-82%), edible fiber (2.2-18.3%), ash (2.1-3.3%), calcium (81-466 mg), phosphorous (86- 190 mg), iron (1.3-10.9 mg), sodium (23-28 mg), potassium (62-570 mg). It also contains 41-58% sugars of which 25- 45% is in the form of reducing sugars and 16% is in the form of non-reducing sugars and tartaric acid is (8-18%) and ascorbic acid is (3-9 mg). The composition of the tamarind fruit juice varies with geographical, cultural and seasonal harvesting and processing. An aqueous extract of tamarind fruit juice is acidic due to presence of tartaric acid and ascorbic acid and acidity percentage is 50.3% and hence it will be work as an acid catalyst for condensation of aldehydes and active methylene compound. This is continuation of our previous work wherein we have reported synthesis of the same under thermal condition [35]. The utilization of ultrasound energy in organic chemistry has been better known from the 1970s [36]. Ultrasonication, based on cavitation effects leading to mass transfer improvement, is an important technique that is widely used today in organic synthesis and has a profound impact on the way chemists approach organic and parallel synthesis. Reductions in reaction times, improved yields and suppression of side products, relative to traditional thermal heating, are benefits of this technology [37]. As increasing environmental consciousness in chemical research and industry, the challenge for a sustainable environment calls for clean procedures. Ultrasound as a green synthetic approach has gradually been used in organic synthesis over the last three decades. Compared with the traditional methods, it is more convenient, easier to be controlled, and consumes less power. With use of ultrasound irradiation, a large number of organic reactions can be carried out in milder conditions with shorter reaction time and higher product yields [38]. Ultrasound-accelerated chemical reactions are well-known and proceed via the formation and adiabatic collapse of transient cavitation bubbles. Ultrasound irradiation has been demonstrated as an alternative energy source for organic reactions ordinarily accomplished by heating. Many homogeneous and heterogeneous reactions can be conducted smoothly by sonication to provide improved yields and increased selectivities [39]. Therefore, ultrasound irradiation has been established as an important technique in organic synthesis [40].

MATERIALS AND METHODS

All melting points were measured in open capillary and are uncorrected. The products were characterized by IR spectra, 1H NMR. IR spectra were recorded on Perkin–Elmer FT-IR-1710 Instrument. 1H NMR was recorded on BrukerMSL-300 instrument using TMS as an internal standard. For ultrasound assisted organic reactions, the ultrasonicator was used having the following specifications.

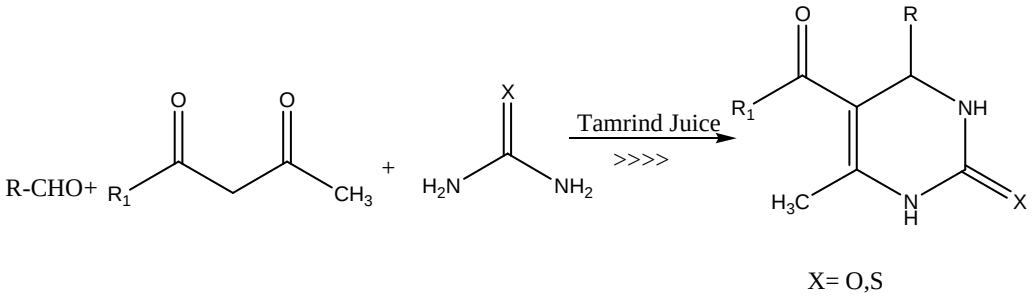
Electric supply: 230 v A.C. 50 Hz, 1phase.
Ultrasonic frequency: 36 ±3 KHz. Ultrasonic power: 100 watts.
All reagents were purchased from Merck and Loba and used without further purification.

Preparation of aqueous extract of tamarind juice:
The tamarind fruits were purchased from the local market. The upper shell of fruit and its inner grain were removed. The brown material (pulp, 10 g) was dissolved in water (50mL), and it was centrifuged using micro centrifuge (REMI RM-12C). The clear portion of the aqueous extract (pH=3) of the tamarind fruits was used as catalyst for the reactions.

General method for the preparation of dihydropyrimidinone/thiones (DHPMs) derivatives:
The mixture of 10 mmol of aldehyde, 10 mmol of ethyl acetoacetate, 10 mmol of urea / thiourea and 1 ml tamarind juice was irradiated in an ultrasound at 100 W for few mins at room temperature with monitoring by TLC. Then the reaction mixture was filtered, washed with water and the crude product was crystallised by ethanol. Its identity was confirmed by IR and NMR and its melting point.

RESULTS AND DISCUSSION

This synthesis of dihydropyrimidinone and thiones (DHPMs) derivatives which was accomplished by Biginelli reaction between substituted aryl aldehydes, ethyl acetoacetate and urea/thiourea (Scheme1) using natural and biocatalyst *Tamarindus indica*, under ultra sound irradiation. The results are presented in Table-1. The reaction was accompanied having green chemistry approach, shorter reaction time, mild reaction conditions and simple workup procedure along with excellent yield. As compared to other catalyst used under different conditions as depicted in Table-2. This is a mild and highly selective transformation and synthesis is a facile and environmentally benign.



Scheme1: Synthesis of dihydropyrimidinone/thiones

Table 1: Tamarind juice catalyzed synthesis of 3, 4-dihydropyrimidin-2 (1H)-ones and thiones under ultrasound irradiation

Sr. No	R	R_1	X	Yield (%)	M.P (° C) Observed	M.P (° C) Reported
1.	C_6H_5	OEt	O	94	202-203	(201-203)[41]
2.	4-OMe-C_6H_5	OEt	O	95	201-203	(199-201) [41]
3.	4-OH-C_6H_5	OEt	O	93	225-226	(226-228)[42]
4.	2-OH- C_6H_5	OEt	O	83	200-202	(200-202) [43]
5.	3-NO_2- C_6H_5	OEt	O	93	228-230	(230) [44]
6.	C_6H_5	OEt	S	92	208-210	(210-212)[45]
7.	4-OMe-C_6H_5	OEt	S	84	136-138	(137-139)[46]
8.	2-Cl-C_6H_5	OEt	S	88	202-204	(205-206)[47]
9.	2-OH- C_6H_5	OEt	S	80	188-190	(183-185)[48]
10	4-Cl- C_6H_5	OEt	S	92	182-184	(180-182)[49]

Table 2: Comparison for different catalyst used for the synthesis of DHPM (R= $OCH_3C_6H_4$)

Entry	Catalyst	Time	Temperature	Yield (%)
1	p-TSA[50]	1 hr	Refluxed in EtOH	90
2	$ZnCl_2$ [51]	30sec	M.W irradiation	94
3	$ZnBF_4$ [52]	4 hrs	Stirring at R.T	71
4	HSO_3Cl [53]	10 mins	Ultrasound, room temp	93
5	$CaCl_2$ [54]	2 hrs	Refluxed in EtOH	98
6	$InBr_3$ [55]	7 hrs	Refluxed in EtOH	97
7	$Mg(NO_3)_2$ [56]	45 mins	Refluxed	90
8	P_2O_5 [57]	10 mins	M.W irradiation	90
12	$Pb(NO_3)_3$ [58]	180 mins	Refluxed in Acetonitrile	89
13	5-sulfosalicylic acid [59]	3.3 mins	M.W irradiation	75
15	Tamarind juice	3.0 mins	Ultrasound irradiation	95

Spectral Data:

1)5-Ethoxycarbonyl-6-methyl-4-(phenyl)-3,4-dihydropyrimidin-2(1H)-one. m.p. 202-203ºC; IR (KBr): 3242, 3117, 2980, 1722, 1645, 1600,1462,1388,1091,781 cm^{-1}; 1H NMR (DMSO-d6): 1.12 (t, 3H, OCH_2CH_3), 2.27 (s, 3H, 6-CH_3), 4.02 (q, 2H, OCH_2CH_3), 5.11 (s, 1H, CH), 7.18-7.28 (m, 5H, aromatic), 7.31 (s, 1H, N-H), 9.37 (s, 1H, N-H).

2)5-Ethoxycarbonyl-6-methyl-4-(4-methoxyphenyl)-3,4-dihydropyrimidin-2(1H)-one. m.p.201-203ºC; IR KBr): 3234, 3110, 2933, 2833, 1703, 1649, 1511,1455,1276,1175,791 cm^{-1}; 1H NMR (DMSO-d6): 1.12 (t, 3H, OCH_2CH_3), 2.27 (s, 3H, 6-CH_3), 3.72 (s, 3H, OCH_3), 4.02 (q, 2H, OCH_2CH_3), 5.23 (s, 1H, CH), 6.75-7.25 (m, 4H, aromatic), 7.22 (s, 1H, N-H), 9.47 (s, 1H, N-H).

3)5-Ethoxycarbonyl-6-methyl-4-(4-hydroxyphenyl)-3,4-dihydropyrimidin-2(1H)-one. m.p.225-226ºC; IR KBr): 3348, 3244, 3082 2989, 2845, 1686, 1638, 1515,1462,1232,1087,759 cm^{-1}; 1H NMR (DMSO-d6): 1.12 (t, 3H, OCH_2CH_3), 2.27 (s, 3H, 6-CH_3), 4.02 (q, 2H, OCH_2CH_3), 5.11 (s, 1H, CH), 6.61-7.05 (m, 4H, aromatic), 7.61 (s, 1H, N-H), 9.45 (s, 1H, N-H), 9.80 (s, 1H, OH).

4)5-Ethoxycarbonyl-6-methyl-4-(2-hydroxyphenyl)-3,4-dihydropyrimidin-2(1H)-one. m.p.200-202ºC; IR KBr): 3348, 3244, 3082 2989, 2845, 1686, 1638, 1515,1462,1232,1087,759 cm^{-1}; 1H NMR (DMSO-d6): 1.12 (t, 3H, OCH_2CH_3), 2.27 (s, 3H, 6-CH_3), 4.02 (q, 2H, OCH_2CH_3), 5.11 (s, 1H, CH), 6.61-7.04 (m, 4H, aromatic), 7.61 (s, 1H, N-H), 9.45 (s, 1H, N-H), 9.83 (s, 1H, OH).

5)5-Ethoxycarbonyl-6-methyl-4-(3-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one. m.p. 228-230°C; IR (KBr): IR (KBr) : 3348, 3244, 3082, 2989, 2845, 1686, 1638, 1515,1462,1232,1087,759 cm^{-1}; 1H NMR (DMSO-d6):1.12 (t, 3H, OCH_2CH_3), 2.27 (s, 3H, 6-CH_3), 4.02 (q, 2H, OCH_2CH_3), 5.11 (s, 1H, CH), 7.45-8.16 (m, 4H, aromatic), 7.61 (s, 1H, N-H), 9.47 (s, 1H, N-H).

6)5-Ethoxycarbonyl-6-methyl-4-(phenyl)-3,4-dihydropyrimidin-2(1H)-thione. m.p. 208-210°C; I.R (KBr): 3327.32, 3176.87, 2983.98, 1672.34, 1572.04 cm-1. 1H NMR ($CDCl_3$): 1.14 (t, 3H, OCH_2CH_3), 2.2 (s, 3H, CH_3), 4.1 (q, 2H, OCH_2CH_3), 5.3 (s, 1H, NH), 6.8- 7.9 (m, 5H, aromatic), 8.1 (s, 1H, NH).

7)5-Ethoxycarbonyl-6-methyl-4-(4-methoxyphenyl)-3,4-dihydropyrimidin-2(1H)-thione. m.p. 136-138°C; I.R(KBr): 3315, 3173, 2985, 2937, 1720, 1664.64, 1570, 1510, 1454 cm-1. 1H NMR ($CDCl_3$): 1.16 (t, 3H, OCH_2CH_3), 2.34 (s, 3H, CH_3), 3.76 (s, 3H, CH_3), 4.08 (q, 2H, OCH_2CH_3), 5.3 (d, 1H, CH), 6.8 (d, 2H, aromatic), 7.18 (d, 2H, aromatic), 7.83 (s, 1H, NH), 8.47 (s, 1H, NH).

8)5-Ethoxycarbonyl-6-methyl-4-(2-chlorophenyl)-3, 4-dihydropyrimidin-2(1H)-thione. m.p. 202-204°C; I.R (KBr): 3240, 3109, 2976, 1703, 1653, 1460, 1226, 1093, 783. 1HNMR ($CDCl_3$): 7.30(m, 5H, aromatic), 7.07(s, 1H, NH), 6.50(d, 1H, CH), 6.24(dd,.4Hz, 1H, CH),5.35(s, 1H, NH), 5.01(s, 1H, CH), 4.20(q, 2H, OCH_2CH_3), 2.32(s, 3H, CH_3),1.21(t, 3H, OCH_2CH_3).

9)5-Ethoxycarbonyl-6-methyl-4-(2-hydroxyphenyl)-3,4-dihydropyrimidin-2(1H)-thione. m.p. 188-190°C; I.R(KBr): 3363, 3171, 3082, 2993, 1728, 1560, 1485, 1329, 1153, 1091, 765. 1H NMR ($CDCl_3$): 7.46(brs, 1H, NH), 7.12(d, 1H, aromatic), 6.94(s, 1H, NH), 6.94(t, 2H, aromatic), 6.85(d, 1H, aromatic), 4.71(d, 1H, CH), 4.24(q, 2H, OCH_2CH_3), 3.13(brs, 1H, OH), 1.91(s, 3H, CH_3),1.30(t, 3H, OCH_2CH_3).

10)5-Ethoxycarbonyl-6-methyl-4-(4-chlorophenyl)-3,4-dihydropyrimidin-2(1H)-thione. m.p. 182-184°C; I.R(KBr): 3327, 3176, 3105, 2985, 1672, 1575, 1460, 1330, 1284, 1026, 754. 1H NMR ($CDCl_3$): 8.10(s, 1H, NH), 7.61(s, 1H, NH), 7.25(d, 4H, aromatic), 5.37(s, 1H, CH), 4.09(d, 2H, OCH_2CH_3), 2.36(s, 3H, CH_3), 1.19(d, 3H, OCH_2CH_3).

CONCLUSION

We have developed an eco-friendly and a simple process for the synthesis of DHPMs by tamarind juice as a natural catalyst under ultra sound irradiation with several advantages such as shorter reaction time, mild reaction conditions, and simple work-up and reduces environmental impact with good yield of the product.

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