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Introduction of a new bis-derivative of succinimide (Bis-Su) as a sustainable and efficient basic organo-catalyst for the synthesis of arylidene malononitrile and tetrahydrobenzo[*b*]pyran derivatives under green conditions

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Abstract

Organo-catalysts have been under great consideration for many years because of their performance and selectivity in the reactions. In this work, a new bis-succinimide (Bis-Su) compound is prepared, identified and used as a green and efficient basic organo-catalyst for the promotion of the synthesis of arylidene malononitrile and tetrahydrobenzo[*b*]pyran derivatives. Using this method, both of the reactions were performed under mild conditions, during short reaction times in high yields. The catalyst can also be recycled and reused several times without a considerable decrease in its activity.

Keywords Organo-catalyst · Bis-succinimide · Arylidene malononitriles · Tetrahydrobenzo[*b*]pyran · Water media

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Introduction

In recent years, the use of green and commercially available or easily preparable catalysts in organic reactions has been under attention because they do not contaminate the environment and are economically affordable. In this way, organo-catalysts as compounds that have the only hydrogen, carbon, nitrogen, sulfur and phosphor atoms in their structure have been extensively followed up since they are usually, inexpensive, easily available, non-toxic and non-sensitive toward moisture and oxidation. Also, these compounds can be utilized in various conversions with excellent selectivity and yields. Besides, organo-catalysts are free of metals, making them an environmentally friendly and valuable portion of green chemistry [1–4].

Succinimide is a cyclic imide which can be easily prepared from the reaction of succinic acid with ammonia in the presence of some catalysts. It can also be prepared by the thermal decomposition of ammonium succinate [5]. Some of its derivatives such as *N*-chlorosuccinimide (NCS) [6], *N*-bromosuccinimide (NBS) [7] and *N*-sulfonated succinimide are very important because that they show promoting effect in special organic transformations [8]. Despite its derivatives, there is not any report about the promoting catalytic activity of succinimide on its own.

Arylidene malononitrile compounds are important chemical intermediates that can be obtained via reactions between different types of aldehydes and malononitrile. Knoevenagel reaction that is a very important method for the formation of C=C bonds is a famous reaction to acquire such derivatives. For this reason, various types of the catalysts have been reported for this reaction such as MNPs–guanidine [9], onion extract [10], CTMAB [11], ZnO [12], [TBA][Leu] [13], Ni-NPs [14], Fe_3O_4@SiO_2-3 N [15], taurine [16], hexamethylenetetramine-bromine [17] and γ-Fe_2O_3@SiO_2@[Bis-APTES]Cl_2-NPs [18].

Multi-component reactions (MCRs) as reactions which by using them three or more reactants combine via a one-pot reaction to form a single product containing substantially all of the atoms of the starting materials have been widely used in medicinal chemistry and drug discovery programs [19], combinatorial chemistry [20], natural product synthesis [21] and agrochemistry [22].

Tetrahydrobenzo[*b*]pyrans are organic compounds that can be utilized as cosmetic pigments and photoactive materials [23, 24]. Because of their usefulness and very effective properties, different types of methods and catalysts have been used for the promotion of the synthesis of these compounds via MCRs among which taurine [16], HDMBAB [25], DBSA [26], I_2 [27], TBAC [28], L-proline [29], [Ch][OH] [30] and glutamic acid [31] are examples.

Although the reported methods and catalysts for the synthesis of arylidene malononitriles and tetrahydrobenzo[*b*]pyrans are useful, most of them suffer from disadvantages including long reaction times, low yields, difficulties in the preparation of the catalyst, non-recyclability of the catalyst, harsh reaction conditions and so on. Therefore, the introduction of new catalysts which their utilizations are not included the mentioned drawbacks is still in demand.

In this research, for the first time, we designed, synthesized and characterized a bis-compound of succinimide as a basic catalyst having two active sites and used

it in the promotion of the preparation of arylidene malononitrile and benzo[*b*] pyran derivatives which are suitable reference reactions to show the ability of the basic and acidic catalyst.

Experimental

Materials

All chemicals including succinimide (Merck ≥ 98%), 1,4-dichlorobutane (Sigma-Aldrich ≥ 98%), aldehydes, malononitrile (Sigma-Aldrich ≥ 99%), dimedone (Merck 95%) and solvents were purchased from Merck (Munich) and Sigma-Aldrich (Mumbai) chemical companies and were used without further purification. The purity determination of the substrates and reaction monitoring was accompanied by TLC using silica gel SIL G/UV 254 plates. All solvents were kept sealed in airtight bottles as well to minimize the absorption of atmosphere moisturize. Moreover, they had gotten distilled before being used. Products were characterized by their physical constants and comparison with authentic samples and those reported in the literature.

Instrumentation

Melting points were determined using a Buchi B-545 apparatus. The FT-IR spectra were recorded on a PerkinElmer 781 spectrophotometer. The NMR spectrums were recorded using Bruker Avance 400 MHz and 500 MHz instruments using TMS (0.00 ppm) as internal standard and DMSO-d_6 as the solvent.

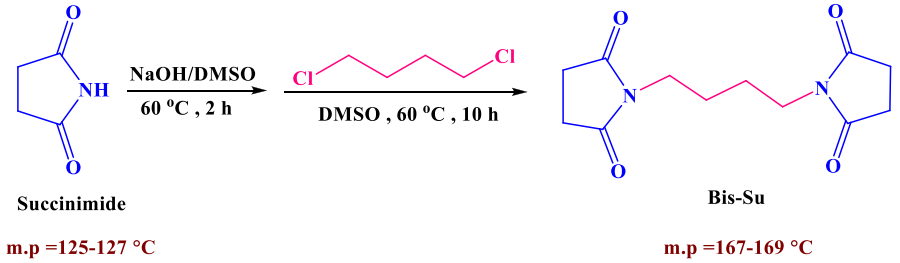
Preparation of 1,1′-(butane-1,4-diyl)bis(pyrrolidine-2,5-dione) (Bis-Su)

In a 50-mL round-bottomed flask, a mixture of succinimide (0.99 g, 10.0 mmol) and NaOH (0.4 g, 10.0 mmol) in DMSO (5 mL) was stirred for 2h at 60 °C. After that, 1,4-dichlorobutane (0.54 mL, 5.0 mmol) was added to the mixture and stirred for 10 h at 60 °C until a very viscous solution was formed. After that, it was washed by diethyl ether for three times (3 × 5 mL) and ethanol (3 × 10 mL) to remove any unreacted substrates; then, the white solid was obtained. The resulting product was dried using a vacuum pump (2.42g, yield 93.6%) (Scheme 1) (Fig. 1).

Physical and spectral data of Bis-Su: White solid; M.P. 167–169 °C; FT-IR (KBr, cm^{-1}) υ_{max}: 3445, 2998,1692, 1401, 666; ^{1}H NMR (500 MHz, DMSO-d_6) (ppm): 1.47–1.52 (m, 4H), 2.69 (s, 8H), 3.40–3.43 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) (ppm): 25.0, 28.6, 37.9, 178.4.

General procedure for the preparation of arylidene malononitrile derivatives

In a 25-mL round-bottomed flask, a mixture of aromatic aldehyde (1.0 mmol), malononitrile (1.1 mmol) and Bis-Su (0.04 mmol ~ 10 mg), in H_2O/EtOH [5 mL



Scheme 1 Synthesis of 1,1′-(butane-1,4-diyl)bis(pyrrolidine-2,5-dione) (Bis-Su)

Fig. 1 Synthesized bis-succinimide (Bis-Su)

(1:1)] was stirred at 60 °C for an appropriate time. After completion of the reaction, which was monitored by TLC [*n*-hexane:EtOAc (7:3)], the mixture was cooled to room temperature and the solid product was filtered, and washed several times with cold distilled water (2 mL) to obtain essentially pure product. The solid product can be recrystallized from ethanol if necessary (1a–1s).

Spectroscopic data of (E)-2-(3-(2-nitrophenyl) allylidene)malononitrile [1 s] (new product)

Yellow solid; M.P. 191–194 °C; IR (KBr) ν_{max} (cm^{-1}) = 3422, 3100, 2927, 2224, 1609, 2224, 1571, 1522, 1349; ^{1}HNMR (400 MHz, DMSO-d_6) (δ, ppm) = 7.32 (dd, J_1 = 15.0 Hz, J_2 = 11.2 Hz, 1H), 7.79 (dd, J_1 = 8.0 Hz, J_2 = 1.6 Hz, 1H), 7.88 (dd, J_1 = 15.0 Hz, J_2 = 11.2 Hz, 1H), 7.95 (d, J = 15.0, 1H), 8.16 (tt, J_1 = 8.0 Hz, J_2 = 1.2 Hz, 1H), 8.51 (dd, J_1 = 11.6 Hz, J_2 = 0.8 Hz, 1H); ^{13}CNMR (100 MHz,

DMSO-d_6) (δ, ppm) = 83.9, 111.8, 113.7, 124.8, 126.5, 128.9, 129.4, 131.8, 133.9, 144.6, 148.5, 161.3.

General procedure for the preparation of tetrahydrobenzo[b]pyran derivatives

In a 25-mL round-bottomed flask, a mixture of an aromatic aldehyde (1.0 mmol), malononitrile (1.1 mmol), dimedone (1.0 mmol) and Bis-Su (0.04 mmol ~ 10 mg) in H_2O/EtOH [5 mL (1:1)] was stirred at 60 °C for an appropriate time. The reaction progress was monitored by TLC [*n*-hexane: EtOAc (7:3)]. After completion of the reaction, the mixture was cooled to room temperature and the solid product was filtered, and washed with cold distilled water (2 mL) to obtain the pure products. The solid product can be recrystallized from ethanol if necessary (2a-2 s).

Spectroscopic data of (E)-2-amino-7,7-dimethyl-4-(2-nitrostyryl)-5-oxo-5,6,7,8 -tetrahydro-4H-chromene-3-carbonitrile [2 s] (new product)

Pale yellow powder; M.P. 256-258 °C; IR (KBr) ν_{max} (cm^{-1}) = 3435, 3334, 2555, 3205, 2961, 2193, 1670, 1599, 1529, 1359; ^{1}HNMR (400 MHz, DMSO-d_6) (δ, ppm) = 1.01 (s, 3H), 1.05 (s, 3H), 2.19–2.47 (m, 4H), 3.88 7.79 (d, J = 6.8 Hz, 1H), 6.27 (dd, J_1 = 15.6 Hz, J_2 = 6.8 Hz, 1H), 6.67 (d, J = 15.6 Hz, 1H), 7.16 (s, 2H), 7.49 (td, J_1 = 7.6 Hz, J_2 = 16 Hz, 2H), 7.65 (td, J_1 = 7.6 Hz, J_2 = 5.6 Hz, 1H), 7.73 (dd, J_1 = 8.0 Hz, J_2 = 1.6 Hz, 1H), 7.92 (dd, J_1 = 8.0 Hz, J_2 = 1.2 Hz, 1H); ^{13}CNMR (100 MHz, DMSO-d_6) (δ, ppm) = 26.7, 28.3, 31.8, 32.7, 49.9, 54.3, 111.4, 123.5, 124.2, 127.9, 130.8, 133.2, 136.6, 147.6, 159.4, 162.7, 195.8.

Results and discussion

In this study and in continuation of our previous works on the introduction of various catalysts for the promotion of organic reactions [32–37], we wish to introduce bis-succinimide, a cheap, non-toxic and easily preparable catalyst from available reagent, as an effective accelerator for the above-mentioned target molecules. It should be mentioned that the use of this reagent is not accompanied by considerable drawbacks.

Regarding our research, the basic catalysts often have better performance when a component of a reaction is malononitrile. Since a basic catalyst can quickly remove the acidic hydrogen of the malononitrile, negatively charged carbon forms fast and enters the reaction. To prove this, we also used succinimide as the catalyst in the optimized reaction condition for Bis-Su which has relatively acidic hydrogen. It was observed that the use of Bis-Su has distinguished advantages from the viewpoints of catalyst amount in the reaction and also the rate of the

reaction. Therefore, the design and preparation of Bis-Su from the succinimide that not only is a basic reagent in its nature, but has two active catalytic sites in its structure that results in much higher performance in the reaction compared to succinimide are a valuable promotion in this new presented methodology.

Characterization of the catalyst

After the preparation of Bis-Su, various spectroscopic techniques were used to characterize the structure of the compound. Here are some elucidations in detail:

FT-IR and NMR analysis

The comparison of FT-IR spectrum of succinimide and bis-succinimide shows the presence of similar functional groups in both of the reactions. It should be emphasized that noticeable absorption bands at 2992 cm^{-1} and 2948 cm^{-1} in the FT-IR spectrum of Bis-Su are related to the stretching vibrations of the methylene groups of the linker. Also, the number of peaks in the FT-IR of Bis-Su is considerably decreased. This result can be attributed to the limitation of the vibration modes due to the locking of the rings in the prepared reagent.

In the ^{1}H NMR spectrum of 1,1′-(butane-1,4-diyl)bis(pyrrolidine-2,5-dione) as a symmetric compound, three peaks related to three types of hydrogen are observable. Two multiplet peaks (H_a and H_b) in the areas of 1.47–1.52 ppm and 3.40–3.43 ppm are related to the hydrogens of methylene groups of the linker. Besides, a single peak at 2.69 ppm (H_c) is related to the methylene groups of succinimide rings.

^{13}C NMR also shows four peaks related to four different carbons. The peaks at 25.0 ppm 28.6 ppm (C_1 and C_3) are related to the middle methylene groups of the linker. The peak at 37.9 ppm (C_3) is attributed to methylene groups of the succinimide, while the peak at 178.4 ppm (C_4) is related to carbonyl groups of the rings.

Catalytic activity

Optimization of the reaction conditions in the synthesis of arylidene malononitrile derivatives

For the optimization of the reaction conditions of preparation of arylidene malononitrile derivatives, the reaction of 4-chlorobenzaldehyde and malononitrile was selected as a model reaction for the synthesis and it was investigated under the effect of different factors including the temperature, amounts of the catalyst, solvent (dichloromethane, acetonitrile, ethanol, DMSO, H_2O and EtOH/H_2O) and also solvent-free conditions. As given in Table 1, the best results can be obtained

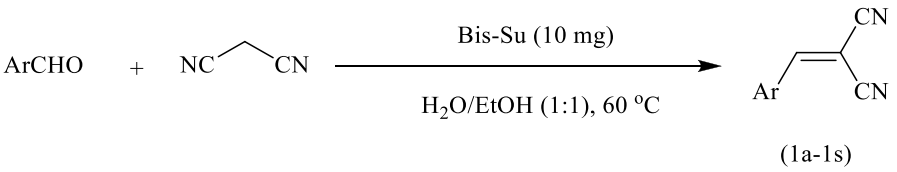
Table 1 Optimization of the reaction conditions for the preparation of arylidene malononitrile derivative of 4-chlorobenzaldehyde

Entry	Catalyst (mg)	Solvent	Temperature (°C)	Time (min.)	Yield (%)
1	No catalyst	H_2O/EtOH[b]	Reflux	100	30[a]
2	10	*n*-Hexane	Reflux	100	Trace[c]
3	10	DMSO	Reflux	100	Trace[c]
4	10	CH_2Cl_2	Reflux	100	Trace[c]
5	10	$CHCl_3$	Reflux	100	Trace[c]
6	10	CH_3CN	Reflux	100	40[c]
7	10	EtOH	Reflux	100	55[c]
8	10	H_2O	Reflux	60	81
9	10	H_2O/EtOH[b]	40	100	65[c]
10	10	H_2O/EtOH[b]	50	35	84
11	10	H_2O/EtOH[b]	60	12	97
12	10	H_2O/EtOH[b]	80	12	92
13	10	H_2O/EtOH[b]	Reflux	10	87
14	20	H_2O/EtOH[b]	60	30	85
15	5	H_2O/EtOH[b]	60	24	80

[a]Reaction conditions: 4-Chlorobenzaldehyde (1.0 mmol), malononitrile (1.1 mmol), solvent (5 mL) and required amounts of the catalyst

[b](1:1)

[c]Reaction was not completed



Scheme 2 Synthesis of arylidene malononitrile derivatives

when 10 mg of bis-succinimide is used in a mixture of H_2O and EtOH (1:1) at 60 °C (Table 1, entry 11; Scheme 2).

After the optimization of the reaction conditions, various aromatic aldehydes containing electron-releasing, electron-withdrawing and halogen substituents on their aromatic rings were examined to prepare the corresponding arylidene malononitrile derivatives. Using this method, no considerable effect of the substituent was observed and the desired products were obtained in high yields during short reaction times (Table 2).

After the successful performance of Bis-Su in the condensation of aldehydes with malononitrile, we were interested to investigate its ability in the promotion

Table 2 Synthesis of arylidenlidene malononitrile derivatives in the presence of Bis-Su as the catalyst

Entry	Aldehyde	Product	Time (min.)	Yield (%)	M.p. (°C)	
					Obs.	Lit. [References]
1	C_6H_5CHO	1a	30	86	81–84	82–84 [11]
2	2-ClC_6H_4CHO	1b	16	89	90–92	93–94 [38]
3	2-$NO_2C_6H_4CHO$	1c	20	93	136–138	139–140 [9]
4	2-$OCH_3C_6H_4CHO$	1d	18	85	81–82	80 [39]
5	3-ClC_6H_4CHO	1e	16	96	93–95	95–97[18]
6	3-$OCH_3C_6H_4CHO$	1f	20	90	106–108	107–108 [40]
7	3-$NO_2C_6H_4CHO$	1 g	20	95	102–104	103–105 [38]
8	3-BrC_6H_4CHO	1 h	30	92	156–158	155–156 [14]
9	4-ClC_6H_4CHO	1i	12	97	159–161	161–162 [38]
10	4-BrC_6H_4CHO	1j	15	94	157–159	159–160 [9]
11	4-FC_6H_4CHO	1 k	18	89	123–125	122–124 [41]
12	4-$NO_2C_6H_4CHO$	1 l	27	93	156–158	157–160 [42]
13	4-OHC_6H_4CHO	1 m	37	95	185–187	186–188 [38]
14	4-$OCH_3C_6H_4CHO$	1n	32	79	111–112	112–114 [42]
15	4-$CH_3C_6H_4CHO$	1o	40	90	137–139	137–138 [39]
16	4-$NMe_2C_6H_4CHO$	1p	28	88	188–190	190–191 [40]
17	3-Phenylpropanal	1r	32	76	194–196	195–197 [16]
18	Cinnamaldehyde	1q	26	89	123–125	124–126 [42]
19	2-Nitrocinnamaldehyde	1 s	35	91	191–194	New

[a]Yields are related to isolated products

of one-pot three-component synthesis of tetrahydrobenzo[*b*]pyran derivatives as the other biologically important organic compounds.

For the optimization of the reaction conditions, the reaction of a mixture of 4-chlorobenzaldehyde and malononitrile with dimedone (as a model for the synthesis of tetrahydrobenzo[*b*]pyrans) was studied under the influence of different factors including the temperature, amounts of the catalyst and solvent and also in the absence of solvent. The collected data in Table 3 show that the best results can be obtained in a mixture of H_2O and EtOH (1:1) at 80 °C, in the presence of 10 mg of the catalyst was used in the above-mentioned reactions, respectively (Table 3, entry 12; Scheme 3).

In continue, variety of aromatic aldehydes were used for the synthesis of benzo[b]pryan derivatives in the optimized condition. The obtained results clarified that under the selected conditions, all the expected products were obtained in high yields during acceptable reaction times (Table 4).

The reusability of Bis-Su in the studied reactions was also checked by choosing the model reactions (Table 2, 1i, and Table 4, 2i) and repeating them five times

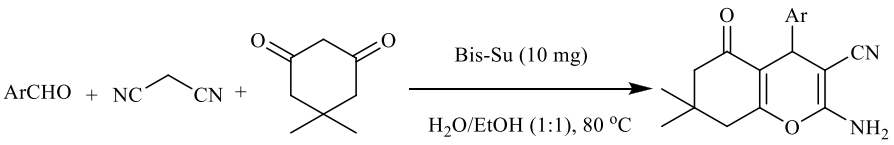
Table 3 Optimization of the reaction conditions for the preparation of tetrahydrobenzo[*b*]pyran derivative of 4-chlorobenzaldehyde

Entry	Catalyst (mg)	Solvent	Temperature (°C)	Time (min.)	Yield (%)
1	No catalyst	H_2O/EtOH[a]	Reflux	100	50[b]
2	10	*n*-hexane	Reflux	100	Trace[b]
3	10	DMSO	Reflux	100	Trace[c]
4	10	CH_2Cl_2	Reflux	100	Trace[b]
5	10	$CHCl_3$	Reflux	100	Trace[b]
6	10	CH_3CN	Reflux	100	50[b]
7	10	EtOH	Reflux	100	55[b]
8	10	H_2O	Reflux	80	88
9	10	H_2O/EtOH[a]	50	37	47[b]
10	10	H_2O/EtOH[a]	60	50	54[b]
11	10	H_2O/EtOH[a]	70	35	78
12	10	H_2O/EtOH[a]	80	20	93
13	10	H_2O/EtOH[a]	Reflux	20	87
14	5	H_2O/EtOH[a]	80	33	84
15	15	H_2O/EtOH[a]	80	28	72

Reaction conditions: 4-Chlorobenzaldehyde (1.0 mmol), dimedone (1.0 mmol), malononitrile (1.1 mmol), solvent (5 mL) and required amount of the catalyst

[a](1:1)

[b]Reaction was not completed



Scheme 3 Synthesis of tetrahydrobenzo[*b*]pyran derivatives

in the filtrated solution without the addition of any amounts of the fresh catalyst. It was observed that the stability of Bis-Su was excellent in the studied reactions (Fig. 2).

The purposed mechanism of the studied reactions in the presence of bis-succinimide as a basic catalyst is shown in Scheme 4. According to this mechanism, at the first step, malononitrile is activated by the lone pair electrons of nitrogen atoms of bis-succinimide rings. Then, it attacked the carbonyl group of the aldehydes which was activated by the conjugated acid of the catalyst. Elimination of a molecule of water led to arylidene malononitrile products (1a–1s). In

Table 4 Synthesis of tetrahydrobenzo[*b*]pyran derivatives in the presence of Bis-Su as the catalyst

Entry	Aldehyde	Product	Time (min.)	Yield (%)[a]	M.p. (°C)	
					Obs.	Lit. [References]
1	C_6H_5CHO	2a	35	84	231–233	232–234 [43]
2	2-ClC_6H_4CHO	2b	23	93	209–211	211–213 [43]
3	2-$NO_2C_6H_4CHO$	2c	32	92	227–229	228 [44]
4	2-$OCH_3C_6H_4CHO$	2d	35	83	194–196	196–199 [43]
5	3-ClC_6H_4CHO	2e	28	95	227–229	228–230 [45]
6	3-$OCH_3C_6H_4CHO$	2f	30	90	188–189	188–190 [45]
7	3-$NO_2C_6H_4CHO$	2g	30	92	213–214	211–213 [45]
8	3-BrC_6H_4CHO	2h	34	94	287–289	289–291 [45]
9	4-ClC_6H_4CHO	2i	20	93	210–212	208–210 [43]
10	4-BrC_6H_4CHO	2j	24	95	200–202	201–203 [43]
11	4-FC_6H_4CHO	2k	22	91	212–214	214–215 [43]
12	4-$NO_2C_6H_4$ CHO	2l	31	93	181–183	181–183 [43]
13	4-OHC_6H_4 CHO	2m	42	93	222–224	222–224 [46]
14	4-$OCH_3C_6H_4$ CHO	2n	30	87	197–199	196–198 [46]
15	4-$CH_3C_6H_4$ CHO	2o	50	90	217–218	217–219 [46]
16	4-$NMe_2C_6H_4$ CHO	2p	45	86	195–197	197–199 [47]
17	3-Phenylpropanal	2r	40	85	196–198	199–200 [48]
18	Cinnamaldehyde	2q	29	90	203–205	205–207 [49]
19	2-Nitrocinnamaldehyde	2s	30	89	255–257	New

[a]Yields are related to isolated products

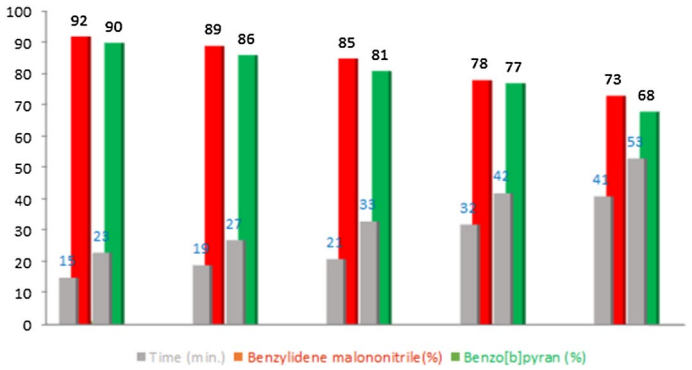
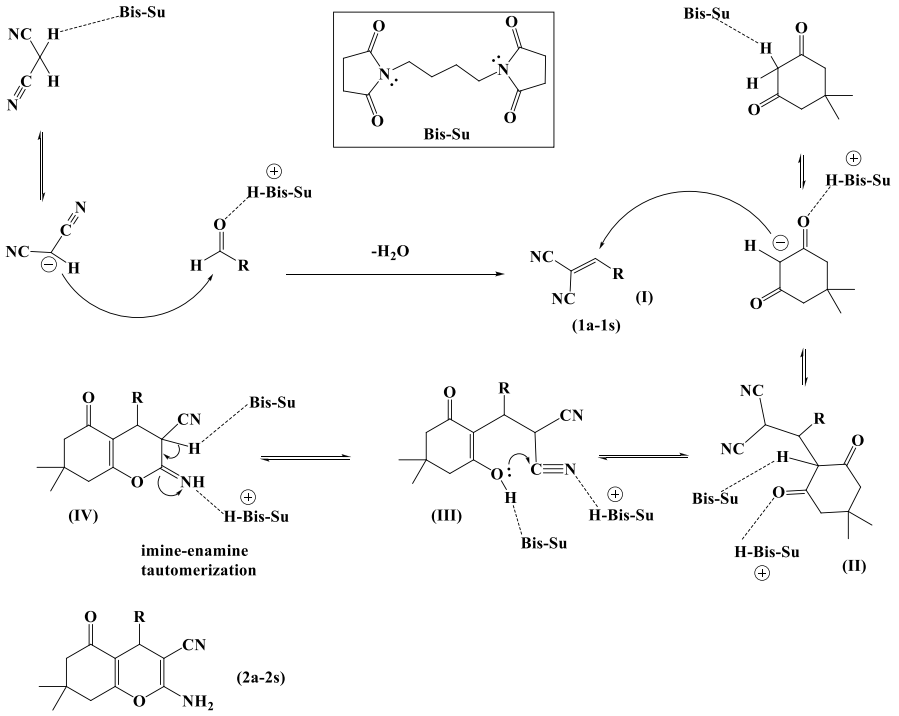


Fig. 2 Reusability of the catalyst in the synthesis of arylidene malononitrile and tetrahydrobenzo[*b*]pyran derivatives of 4-chlorobenzaldehyde

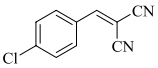
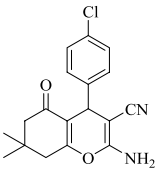


Scheme 4 Plausible pathway of the studied reactions

the presence of dimedone, intermediates (II) are produced which after cyclization forms intermediate (III) and a subsequent imine-enamine tautomerization resulted in the corresponding tetrahydrobenzo[*b*]pyran derivatives (2a–2s). The role of Bis-Su as a basic organo-catalyst is shown well in all steps of the proposed mechanism.

To show the catalytic ability of bis-succinimide in the studied reactions, a comparison between Bis-Su and other types of catalysts for all of the above-mentioned reactions is presented in Table 5. Based on this table, it can be concluded that the reaction in the presence of Bis-Su can be preferred than most of the previously reported ones regarding the catalyst amounts, reaction times and yields.

Table 5 Comparision between the ability of some catalysts and bis-succinimide in the mentioned reactions

Product	Catalyst	Amount	Conditions	Time (min.)	Yield (%)	[References]
	MNPs–guanidine	5 mg	PEG-H_2O (1:1)/r.t.	150	96	[9]
	Onion extract	2 mL	S.F/r.t.	120	90	[10]
	CTMAB	182 mg	H_2O/r.t	90	94	[11]
	ZnO	500 mg	H_2O/r.t	90	86	[12]
	[TBA][Leu]	9 mg	H_2O/50 °C	60	97	[13]
	Ni-NPs	10 mg	S.F./70 °C	45	94	[14]
	Fe_3O_4@ SiO_2-3 N	50 mg	Toluene/75 °C	20	90	[15]
	Bis-Su	10 mg	H_2O-EtOH (1:1)60 °C	12	97	[This work]
	HDMBAB	46 mg	H_2O/80–90 °C	450	90	[25]
	DBSA	65 mg	H_2O/reflux	240-420	69	[26]
	I_2	25 mg	DMSO/120 °C	210	72	[27]
	TBAC	28 mg	H_2O/reflux	120	98	[28]
	L-proline	12 mg	EtOH/reflux	120	72	[29]
	[Ch][OH]	10 mg	H_2O/80 °C	60	86	[30]
	Glutamic acid	30 mg	EtOH/reflux	40	91	[31]
	Bis-Su	10 mg	H_2O-EtOH (1:1)80 °C	20	93	This Work

Conclusions

In this article, we have synthesized and characterized bis-succinimide (Bis-Su) as a basic organo-catalyst and used it in the synthesis of biologically and pharmacologically active arylidene malononitrile and tetrahydrobenzo[*b*]pyran derivatives under mild conditions. The most important advantages of this method are included the use of non-toxic cheap organo-catalyst, mild conditions, excellent yields of the products, short reaction times, simple workup procedure and reusability of the catalyst.

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