J. Braz. Chem. Soc., Vol. 20, No. 10, 1895-1900, 2009.
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0103 - 5053 $6.00+0.00

Article

A Green, Inexpensive and Efficient Organocatalyzed Procedure for Aqueous Aldol Condensations

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É apresentado um procedimento simples e geral para a condensação aldólica cruzada dupla de cetonas cíclicas com vários aldeídos aromáticos catalisada por dietilamina em condições aquosas. Foram obtidos 3,5-bisarilmetilidenos de cetonas homocíclicas e heterocíclicas em um procedimento one-pot em excelentes rendimentos. Além disto, a metodologia é aplicada eficientemente à síntese de chalconas a partir das metil cetonas correspondentes. Na maioria dos casos estudados, os produtos precipitam das misturas de reação e o meio é reciclado em várias reações subseqüentes sem perda significativa de atividade.

A facile and general procedure is presented for diethylamine-catalyzed double crossed aldol condensation of cyclic ketones with various aromatic aldehydes under aqueous conditions. Excellent yields of 3,5-bisarylmethylidenes of homocyclic and heterocyclic ketones are achieved in a one-pot procedure. Furthermore, the methodology is efficiently applied to the synthesis of chalcones from their corresponding methyl ketones. In the majority of the cases studied, products precipitate from the reaction mixtures and the medium is recycled in subsequent several reactions without significant loss of activity.

Keywords: aldol condensation, aqueous conditions, organocatalyst, bisarylmethylidenes, chalcones

Introduction

The use of water, the most abundant chemical on earth, as a solvent has been neglected for many years by organic chemists since water has been traditionally considered to have destructive effects on many reagents and synthetic reactions, unless water is used as a reagent or in workup procedures. It was believed so until pioneering experiments by Breslow's[1] and Grieco's[2] groups conducted in aqueous media led to unexpected rate and selectivity enhancement in Diels-Alder reactions. Since then, numerous papers[3] and reviews[4] on various aqueous-conditioned organic transformations have been released and more additions are expected in the upcoming years. On another "green chemistry" front, acceleration of chemical reactions caused by addition of substoichiometric quantities of organic compounds containing no metal in their structures, known as organocatalysis, has increasingly gained popularity among synthetic organic chemists in recent years.[5]

Crossed aldol condensation of cyclic ketones with aromatic aldehydes has been extensively employed for the synthesis of bis(arylmethylidene)cycloalkanones.[6] Many efforts have been devoted in recent years to widen the synthetic scope of bis(arylmethylidene)cycloalkanones by using microwave radiation,[7] ultrasound irradiation,[8] and Lewis acid catalysis.[9] Despite these developments, even very recent reports still involve the use of either acidic or alkaline conditions or require organic solvents during the reaction or at workup stage.[10] In the framework of our studies on aldol condensation reactions[11] and in

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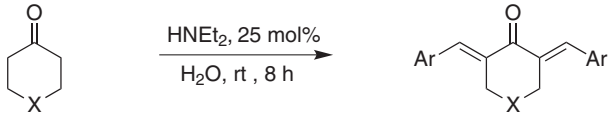
continuation of our previous research on the development of environmentally friendly processes,[12] we would like to report herein an efficient protocol for aldol condensation of ketones with various aldehydes using deficient quantities of diethylamine (Et_2NH) under aqueous conditions (Scheme 1). As far as we know, this is one of the most inexpensive and environmentally friendly procedures offered so far for aldol condensation reactions.



1: X = S; **2**: X = O
3: X = CH_2; **4**: X = none

Scheme 1.

Results and Discussion

Initially, thiopyran **1** was subjected to condensation with benzaldehyde in the presence of an amine and water (Table 1). Optimum results were obtained by the use of deficient amounts of Et_2NH leading to formation of **1a** in 92% yield after 8 h treatment at room temperature (entry 1). The generality of the method was demonstrated by the synthesis of similar products **1b-f** obtained from the reactions of other aromatic aldehydes with **1** under the same conditions (entry 2 - 6). Similarly, pyran **2** was subjected to react with the same aldehydes under the cited conditions and complete formation of **2a-f** was observed (entry 7 - 12). All reactions proceeded rapidly at room temperature and complete conversions were observed in less than 8 h to obtain 90 - 96% of the respective products. In all cases, products separated out spontaneously allowing an easy solvent-free workup and efficient reuse of the H_2O/Et_2NH filtrate in the next reactions.

Table 1. Et_2NH/H_2O promoted condensation of heterocyclic ketones with aldehydes

Entry	Aldehyde	Ketone	Product[a]	Yield (%)[b]
1	benzaldehyde	**1**	**1a**	92
2	*p*-methylbenzaldehyde	**1**	**1b**	90
3	*p*-anisaldehyde	**1**	**1c**	95
4	*p*-chlorobenzaldehyde	**1**	**1d**	91
5	Thiophene-2-carbaldehyde	**1**	**1e**	93
6	nicotinaldehyde	**1**	**1f**	93
7	benzaldehyde	**2**	**2a**	90
8	*p*-methylbenzaldehyde	**2**	**2b**	91
9	*p*-anisaldehyde	**2**	**2c**	96
10	*p*-chlorobenzaldehyde	**2**	**2d**	92
11	Thiophene-2-carbaldehyde	**2**	**2e**	94
12	nicotinaldehyde	**2**	**2f**	93

[a]A mixture of **1** or **2** (1 mmol), aldehyde (2 mmol), and Et_2NH (0.25 mmol) was treated in 0.5 mL H_2O for 8 h. All products precipitate spontaneously in the reaction mixtures; [b]Isolated yields.

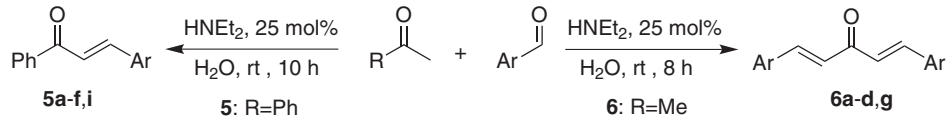
In comparison with many previously related reports,[7,8,10] the present procedure is very mild, takes place at room temperature, and involves a very straightforward and easy workup. Therefore, the procedure was next employed to evaluate the same chemistry in homocyclic ketones. In similar series of reactions, **3** and **4** were subjected to condensation with aldehydes to synthesize their respective bisarylmethylidene products, as summarized in Table 2. All aldehydes reacted in a similar manner producing high yields of the expected products. The products were easily solidified and separated by a simple filtration after acidification of the reaction mixtures by dilute HCl solution.

Table 2. Et_2NH/H_2O promoted condensation of homocyclic ketones with aldehydes

Entry	Aldehyde	Ketone	Product[a]	Yield (%)[b]
1	benzaldehyde	**3**	**3a**	90
2	*p*-methylbenzaldehyde	**3**	**3b**	91
3	*p*-anisaldehyde	**3**	**3c**	90
4	*p*-chlorobenzaldehyde	**3**	**3d**	94
5	Thiophene-2-carbaldehyde	**3**	**3e**	96
6	nicotinaldehyde	**3**	**3f**	95
7	*p*-nitrobenzaldehyde	**3**	**3g**	84
8	benzaldehyde	**4**	**4a**	96
9	*p*-anisaldehyde	**4**	**4c**	94
10	*p*-chlorobenzaldehyde	**4**	**4d**	96
11	furanaldehyde	**4**	**4h**	95

[a]A mixture of **3** or **4** (1 mmol), aldehyde (2 mmol), and Et_2NH (0.25 mmol) was treated in 0.5 mL H_2O for 8 h; [b]Isolated yields.

Moreover, methyl ketones were subjected to condensation with aldehydes in the presence of an amine and water (Scheme 2) under the same conditions. Condensation of acetophenone **5** with various aromatic aldehydes bearing electron-releasing or electron-withdrawing substituents led to rapid and high yield formation of chalcones, which are the central core of a variety of important biologically active compounds (Table 3, entries 1 - 7).[13] Under the above conditions, acetone **6** also efficiently yielded its respective bisarylmethylidene products (entries 8 - 12). For reactions of **6**, products spontaneously precipitated in the medium and allowed a convenient solvent-free workup and efficient reuse of the H_2O/Et_2NH filtrate in subsequent reactions,



Scheme 2.

Table 3. Et_2NH/H_2O promoted aldol condensation of methyl ketones

Entry	Substrate	Aldehyde	Product[a,b]	Yield (%)[c]
1	**5**	Benzaldehyde	**5a**	92
2	**5**	*p*-methylbenzaldehyde	**5b**	93
3	**5**	*p*-anisaldehyde	**5c**	94
4	**5**	*p*-chlorobenzaldehyde	**5d**	90
5	**5**	thiophene-2-carbaldehyde	**5e**	89
6	**5**	Nicotinaldehyde	**5f**	82
7	**5**	Formaldehyde	**5i**	61
8	**6**	Benzaldehyde	**6a**	97
9	**6**	*p*-methylbenzaldehyde	**6b**	92
10	**6**	*p*-anisaldehyde	**6c**	90
11	**6**	*p*-chlorobenzaldehyde	**6d**	85
12	**6**	*p*-nitrobenzaldehyde	**6g**	95

[a]A mixture of **5** (1 mmol), aldehyde (1 mmol), and Et_2NH (0.25 mmol) was treated in 0.5 mL H_2O for 10 h. [b]The same conditions were employed for **6**, except 2 mmol of aldehyde were used and the reactions completed in 8h. Products **6a**-**d**,**g** precipitate spontaneously in the reaction mixtures; [c]Isolated yields.

Table 4. Effect of different additives on aldol condensation of **1** (or **5**) with benzaldehyde

Entry	Additive (0.5 mL)	Yield of **1a** (%)[a,b]	Yield of **5a** (%)[a,c]
1	H_2O	61	58
2	-	0	15
3	NaCl (1.5 M)	41	30
4	NaCl (3.0 M)	22	10
5	LiCl (1.5 M)	27	15
6	LiCl (3.0 M)	0	5
7	$LiClO_4$ (1.5 M)	80	70
8	$LiClO_4$ (3.0 M)	82	74
9	GnCl (1.5 M)	65	65
10	GnCl (3.0 M)	74	76

[a]GC yields; [b]A mixture of **1** (1 mmol), PhCHO (2 mmol), and Et_2NH (0.25 mmol) was treated for 3 h; [c]A mixture of **5** (1 mmol), PhCHO (1 mmol), and Et_2NH (0.25 mmol) was treated for 5 h.

while in the case of **5** separation of the products takes place after acidification of the mixtures by dilute HCl.

Usually, in aqueous-conditioned organic transformations, either the hydrophobic interaction of the reactants in water[14] or the activation of organic functional groups by water molecules through hydrogen bonding[15] is known to be responsible for the progress of the reactions. To shed light on the mechanism of these water-mediated aldol condensations, we designed several parallel experiments for the reactions of benzaldehyde with ketones **1** or **5** as summarized in Table 4. For a meaningful comparison, all reactions were stopped before reactants had been completely consumed.

Entry 1 shows the results for the reactions conducted under the optimized H_2O/Et_2NH conditions. In the absence of water, a dramatic decrease in the rates of both reactions was observed illustrating the crucial role of the aqueous medium (entry 2). When NaCl (entries 3 - 4) and LiCl (entries 5 - 6) solutions were used, a descending pattern in the yields of both reactions was observed. This rate reduction which is more obvious at higher concentrations of NaCl and LiCl, disfavours the "salt-out"[16] effect and excludes the hydrophobic interactions from having a major role in the catalysis of the reaction. Conversely, a noticeable rate acceleration was observed for the reactions conducted in the presence of $LiClO_4$ (entries 7 - 8) and guanidinium chloride (entries 9 - 10). This could be rationalized by proposing a favourable hydrogen-bonded association of the reactants with water molecules to facilitate the amine-catalyzed enolization process, as proposed by others for similar observations.[17] Such hydrogen bonding activation (Figure 1) can lower the energy profile of the process similar to a Lewis acid mediated reaction.

Conclusions

In conclusion, we have reported a general and efficient protocol for the preparation of 3,5-bisarylmethylidenes of a variety of ketones at room temperature by using an environmentally friendly medium consisting of water and Et_2NH. Formation of products in high yields within relatively short time periods, use of no toxic organic solvent, ease of operation, and no special handling requirements makes this protocol an attractive addition to the present literature archive. Recoverability of the reaction medium in many of the cases is an additional advantage. The efficient reuse of the medium in seven consecutive reactions in a row is illustrated in Figure 2 for the synthesis of **1a**.

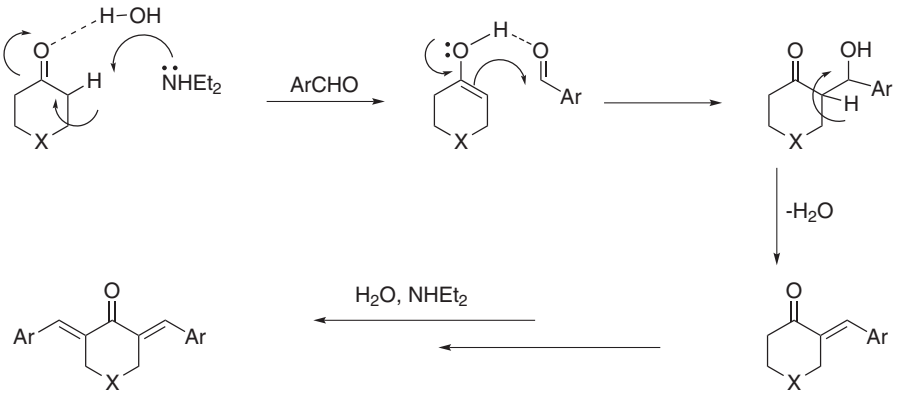


Figure 1. Suggested mechanistic overview of the reaction.

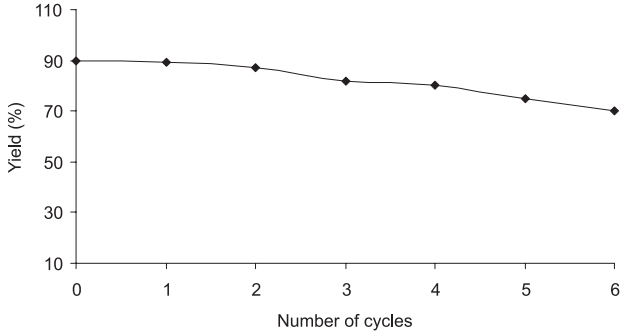


Figure 2. Efficient recovery of the catalyst for the synthesis of **1b**.

Experimental

Reactions were monitored by TLC using silica gel coated plates and ethyl acetate/hexane solutions as the mobile phase. Melting points are uncorrected. FT-IR spectra were recorded using KBr disks on a Bruker Vector-22 infrared spectrometer and absorptions are reported as wave numbers (cm^{-1}). ^1H NMR and ^{13}C NMR spectra were obtained on a Bruker AC 80 MHz or Bruker Ultra Shield™ (500 MHz) instrument as $CDCl_3$ solutions and the chemical shifts are expressed as δ units with Me_4Si as the internal standard. Mass spectra were obtained on a Finnigan Mat 8430 apparatus at ionization potential of 70 eV. Compound **1** was prepared using available methods.[18] All other reagents were purchased from commercial sources and were freshly used after being purified by standard procedures.

*General synthetic procedure for the reactions of ketones **1**, **2**, and **6***

A ketone (1.0 mmol) was added to a mixture of an aldehyde (2.0 mmol), H_2O (0.5 mL), and Et_2NH (25 mol%) and the mixture was stirred at room temperature for 8 h. The course of the reaction was monitored by TLC. After completion of the reaction, the precipitated product was filtered and purified by recrystallization from ethyl acetate.

*General synthetic procedure for the reactions of ketones **3**-**4***

A ketone (1.0 mmol) was added to a mixture of an aldehyde (2.0 mmol), H_2O (0.5 mL), and Et_2NH (25 mol%) and the mixture was stirred at room temperature for 8 h. The course of the reaction was monitored by TLC. After completion of the reaction, the mixture was treated with 3 mL of HCl (5%) to solidify the product. The precipitated product was filtered and purified by recrystallization from ethyl acetate.

*General synthetic procedure for the reactions of ketones **5***

Ketone **5** (1.0 mmol) was added to a mixture of an aldehyde (1.0 mmol), H_2O (0.5 mL), and Et_2NH (25 mol%) and the mixture was stirred at room temperature for 10 h. The course of the reaction was monitored by TLC. After completion of the reaction, the mixture was treated with 3 mL of HCl (5%) to solidify the product. The precipitated product was filtered and purified by recrystallization from ethyl acetate.

Selected spectral data

*(3Z,5Z)-3,5-Dibenzylidene-tetrahydrothiopyran-4-one (**1a**)*

Obtained as yellow solid, yield 92%, mp 142-144 °C. IR (KBr) ν_{max}/cm^{-1}: 1599, 1444, 1269. ^1H NMR ($CDCl_3$) δ 3.84 (s, 4H, SCH_2), 7.30 (s, 10H, Ar-H), 7.72 (s, 2H, =CH). ^{13}C NMR ($CDCl_3$) δ 30.0 (SCH_2), 128.4, 128.7, 129.8, 133.7, 134.9, 136.6, 188.6 (C=O). MS *m/z* 292 (M^+, 27%), 147 (40), 115 (100).

*(3Z,5Z)-3,5-Bis(4-methylbenzylidene)-tetrahydrothiopyran-4-one (**1b**)*

Obtained as yellow solid, yield 90%, mp 186-188 °C.

IR (KBr) ν_{max}/cm^{-1}: 1657, 1595, 1275. ^{1}H NMR ($CDCl_3$) δ 2.31 (s, 6H, CH_3), 3.84 (s, 4H, SCH_2), 7.20-7.45 (m, 8H, Ar-H), 7.68 (s, 2H, =CH). ^{13}C NMR ($CDCl_3$) δ 21.4 (CH_3), 30.1 (SCH_2), 129.1, 129.9, 132.4, 133.3, 136.8, 139.2, 185.5 (C=O). MS *m/z* 320 (M^+, 21%), 305 (19), 147 (30), 130 (62), 115 (100).

*(3Z,5Z)-3,5-Bis(4-methoxybenzylidene)-tetrahydro-thiopyran-4-one (**1c**)*

Obtained as yellow solid, yield 95%, mp 174-176 °C. IR (KBr) ν_{max}/cm^{-1}: 1654, 1592,1505, 1252. ^{1}H NMR ($CDCl_3$) δ 3.76 (s, 6H, OCH_3), 3.80 (s, 4H, SCH_2), 6.85 (d, 4H, *J* 9.8 Hz, Ar-H), 7.30 (d, 4H, *J* 9.8 Hz, Ar-H), 7.66 (s 2H, =CH); ^{13}C NMR ($CDCl_3$) δ 30.2 (SCH_2), 55.3 (OCH_3), 114.1, 127.5, 131.9, 136.1, 160.2, 185.4 (C=O). MS *m/z* 352 (M^+, 49%), 146 (98), 103 (100).

*(3E,5E)-3,5-Bis(4-chlorobenzylidene)-tetrahydropyran-4-one (**2d**)*

Obtained as yellow solid, yield 92%, mp 168-170 °C. IR (KBr) ν_{max}/cm^{-1}: 1671, 1612, 1559, 1263, 1090. ^{1}H NMR ($CDCl_3$) δ 4.80 (s, 4H, OCH_2), 7.25 (d, 4H, *J* 6.4 Hz, Ar-H), 7.40 (d, 4H, *J* 6.4 Hz, Ar-H), 7.70 (s, 2H, =CH). ^{13}C NMR ($CDCl_3$) δ 68.2 (OCH_2), 128.0, 128.8, 131.4, 133.1, 133.3, 135.0, 188.4 (C=O). MS *m/z* 344 (M^+, 22%), 253 (13), 141 (82), 115 (100).

*(3E,5E)-Tetrahydro-3,5-bis((thiophen-2-yl)methylene) pyran-4-one (**2e**)*

Obtained as yellow solid, yield 94%, mp 195-197 °C. IR (KBr) ν_{max}/cm^{-1}: 1662, 1592, 1186. ^{1}H NMR ($CDCl_3$) δ 4.90 (s, 4H, OCH_2), 7.00-7.50 (m, 6H, H's of thienyl), 7.87 (br s, 2H, =CH). ^{13}C NMR ($CDCl_3$) δ 68.3 (OCH_2), 127.9, 128.2, 130.9, 133.3, 138.3, 184.2 (C=O). MS *m/z* 288 (M^+, 20%), 260 (6), 122 (100).

*(3E,5E)-Tetrahydro-3,5-bis((pyridine-3-yl)methylene) pyran-4-one (**2f**)*

Obtained as yellow solid, yield 93%, mp 192-194 °C. IR (KBr) ν_{max}/cm^{-1}: 1672, 1616, 1272. ^{1}H NMR ($CDCl_3$) δ 4.86 (s, 4H, OCH_2), 7.18-7.70 (m, 8H, H's of pyridyl), 8.50 (br s, 2H, =CH). ^{13}C NMR ($CDCl_3$) δ 68.2 (OCH_2), 123.4, 130.4, 132.8, 134.6, 136.9, 149.9, 150.9, 187.0 (C=O). MS *m/z* 278 (M^+, 18%), 117 (100), 90 (75).

*2,6-Dibenzylidenecyclohexanone (**3a**)*

Obtained as yellow solid, yield 90%, mp 113-114 °C. IR (KBr) ν_{max}/cm^{-1}: 1661, 1607. ^{1}H NMR ($CDCl_3$) δ 1.76-1.83 (m, 2H, -CH_2-), 2.96 (t, *J* 6.5 Hz, 4H, CH_2), 7.30-7.49 (m, 10H, Ar-H), 7.81 (s, 2H, =CH). ^{13}C NMR δ 23.5 (CH_2), 28.9 (CH_2), 128.8, 129.0, 130.8, 136.4, 136.6, 137.4, 190.8 (C=O). MS *m/z* 274 (M^+, 85%), 273 (100), 129 (36), 115 (90).

*2,6-Bis-thiophen-2-yl-methylenecyclohexanone (**3e**)*

Obtained as yellowish brown solid, yield 96%, mp 142-143 °C. IR (KBr) ν_{max}/cm^{-1}: 1663, 1608. ^{1}H NMR ($CDCl_3$) δ 1.95-2.00 (m, 2H, -CH_2-), 2.94 (t, *J* 5 Hz, 4H, CH_2), 7.16 (t, *J* 4 Hz, 2H, H of thienyl), 7.40 (d, *J* 2.5 Hz, 2H, H of thienyl), 7.54 (d, *J* 5 Hz, 2H, H of thienyl), 7.98 (s, 2H, =CH). ^{13}C NMR ($CDCl_3$) δ 22.1 (CH_2), 28.6, 128.0, 130.1, 130.3, 133.3, 133.4, 140.0, 189.4 (C=O). MS *m/z* 286 (M^+, 100%), 229 (77), 115 (28).

*2,5-Dibenzylidenecyclopentanone (**4a**)*

Obtained as yellow solid, yield 96%, mp 186-187 °C. IR (KBr) ν_{max}/cm^{-1}: 1655, 1625. ^{1}H NMR ($CDCl_3$) δ 3.12 (s, 4H, CH_2), 7.35-7.62 (m, 12H, Ar-H and =CH). ^{13}C NMR ($CDCl_3$) δ 27.0 (CH_2), 129.2, 129.8, 131.2, 134.3, 136.3, 137.7, 196.8 (C=O). MS *m/z* 260 (M^+, 84%), 129 (42), 115 (95).

*2,5-Bis-(4-chlorobenzylidene)-cyclopentanone (**4d**)*

Obtained as yellow solid, yield 96%, mp 226-227 °C. IR (KBr) ν_{max}/cm^{-1}: 1694, 1607. ^{1}H NMR ($CDCl_3$) δ 3.01 (s, 4H, CH_2), 7.30-7.45 (m, 10H, Ar-H and =CH). ^{13}C NMR δ 26.8 (CH_2), 129.5, 132.3, 133.1, 134.6, 135.9, 137.9, 196.3 (C=O). MS *m/z* 328 (M^+, 40%), 207 (42), 115 (100).

Acknowledgements

The Ministry of Science, Research, and Technology of Iran has partially financial supported this work.

Supplementary Information

^{1}H NMR, ^{13}C NMR, mass and IR spectra of the data mentioned above are available free of charge at http://jbcs.sbq.org.br, as a PDF file.

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Received: June 8, 2009
Web Release Date: November 12, 2009