Stereoselective Crossed-Aldol Condensation of Hetarylmethyl Ketones with Aromatic Aldehydes inWater : Synthesis of (2*E*)-3-Aryl-1-hetarylprop-2-en-1-ones

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Aldol condensation of 2-acetylthiophene, 2-acetylpyrrole and 2-acetylpyridine with different aromatic aldehydes were carried out in water in heterogeneous phases in the presence of cetyltrimethylammonium bromide as cationic surfactant at room temperature. All the reactions occur in a short time with excellent yields of steroselective hetarylpropanones in water as environmental friendly solvent.

Key Words : Crossed-Aldol. Hetarylpropenones. Chalcones. Synthesis in water.

Introduction

Chalcones are α,β-unsaturated ketones and they have great existence in the plant kingdom. It is well known that most of natural or synthetic chalcones are highly biologically active with a great pharmaceutical and medicinal applications [1]. Recently they are used as anti-AIDS agents [2], cytotoxic with antiangiogenic activity [3,4], antimalarial [5,6], anti-inflammatory [7,8] and antitumor [9,10] .

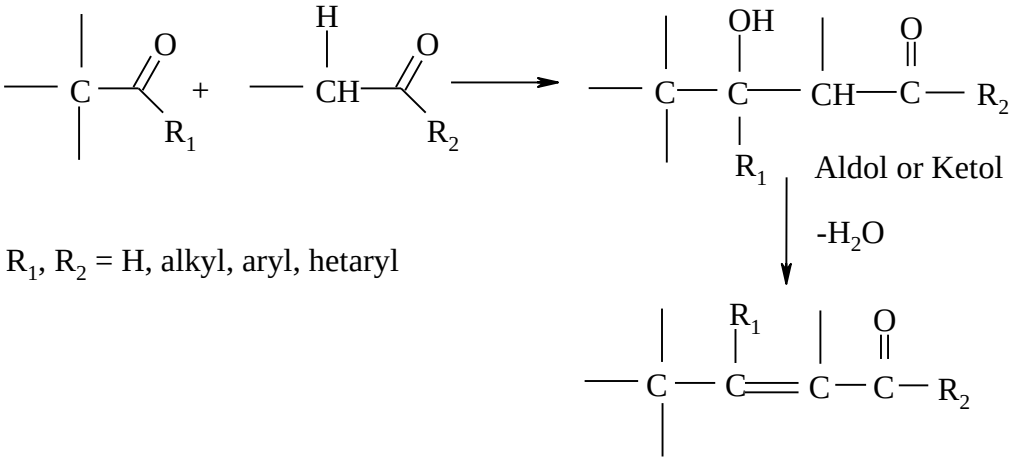
The U.S. Environmental Protection Agency (EPA) has suggested a drastic reduction of using of more than ten of hazard common organic solvents in the industrial production of chemicals. We are dealing in this paper a cleaner and safe production of high yield of

stereoselective hetarylchalcones as an important biologically active compounds in water as a cheap solvent and environmental friendly reaction medium.

Recently, water is an attractive medium for many organic reactions [11]. The important advantageous of aqueous medium with respect to organic solvents are less expensive, healthy, safe and environmentally friendly. Also, it allows the pH control and the use of surfactants as micro aggregates.

The hydrophobic effect and the large cohesive energy of water are considered to be the main factors responsible for increasing reactivity and selectivity of the reactions [12,13].

Mixed or crossed aldol condensation is a base-catalyzed addition of different aldehydes and ketones one of them must contain at least one α-hydrogen to give an aldol or ketol which are dehydrated to α,β-unsaturated aldehydes or ketones.



The classical reaction conditions of aldol condensation are NaOH solution in hydroalcoholic medium which are, often, yielded a mixture of (*E*) and (*Z*) chalcones [14,15].

Recently, aldol reaction can, also, be carried out in an aqueous medium in the presence of catalysts to increase molecular aggregations and stereoselectivity [16-19]. It is considered cleaner conditions of the production of some known and unknown chalcones.

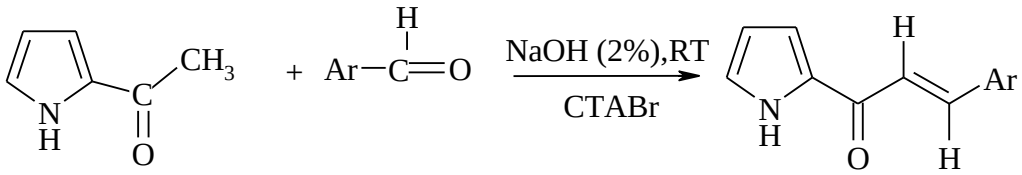
Results and Discussion

We extended the previous investigations [16-19] to carbon-carbon bond formation and we focus in this paper the crossed-aldol condensation of some hetarylmethyl ketones with a variety of different aromatic aldehydes in water at room temperature and in the presence of cetyltrimethylammoium bromide (CTABr) as the proper cationic surfactant for the synthesis of (2*E*)-3-aryl-1-hetarylprop-2-en-1-ones in an excellent yields with high stereoselectivity. Analytical gas chromatography proved that, only, *E*-isomers of isolated propenones were detected. The ^{1}H-NMR coupling constants (*J*) of C2-H and C3-H of the isolated hetarylpropenones are in the range of 15.5-16.0 Hz which are characteristic to *E*-propenones.

We are expected that the synthesized hetarylpropenones might have a biological and medicinal activities in analogous to the biologically active amino chalcones [9], quinolinyl chalcones and some ferrocenyl chalcone [5] .

Efficient stirring of an equimolar amount of 2-hetaryl methyl ketones (**1**, **3**, **5**) and aromatic aldehydes in aqueous NaOH solution and in the presence of cetyltrimethylammoium bromide (CTABr) as surfactant at room temperature, underwent stereoselective crossed-aldol condensation with precipitation of the 3-aryl-1-hetarylprop-2-en-1-ones in high yields within a short reaction time (t) as shown in **Table 1, 2, 3**. It is shown from the Tables that electron donating substituents of aromatic aldehydes decrease the reaction period and increase the yield of hetarylchalcones.

Table 1 : Crossed-Aldol condensation of 2-acetylpyrrol (**1**) with aromatic aldehydes. Synthesis of (2*E*)-3-aryl-1-(pyrrol-2'-yl)prop-2-en-1-ones (**2a-h**)**.**



(1) **(2)**

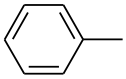
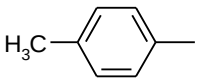
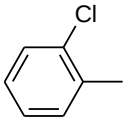
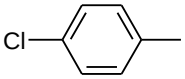
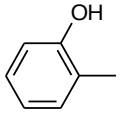
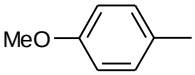
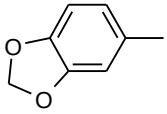
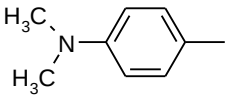
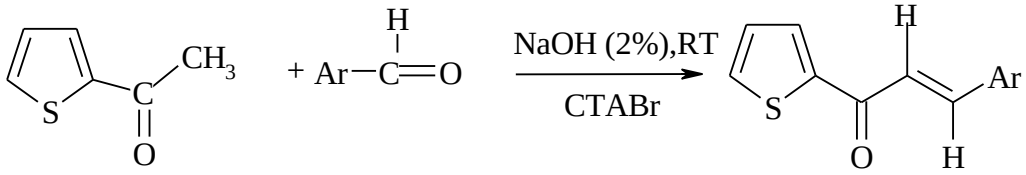
Product No.	Ar	t (min)	Yield (%)
2a		120	65
2b		140	73
2c		120	80
2d		90	82
2e		160	62
2f		30	88
2g		20	96
2h		20	92

Table 2 : Crossed-Aldol condensation of 2-acetylthiophene (**3**) with aromatic aldehydes. Synthesis of (2*E*)-3-aryl-1-(thien-2'-yl)prop-2-en-1-ones (**4a-h**).



(3) **(4)**

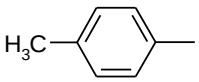
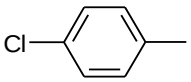
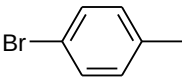
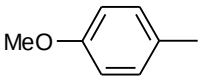
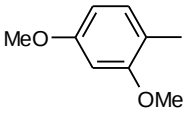
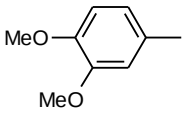
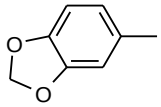
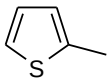
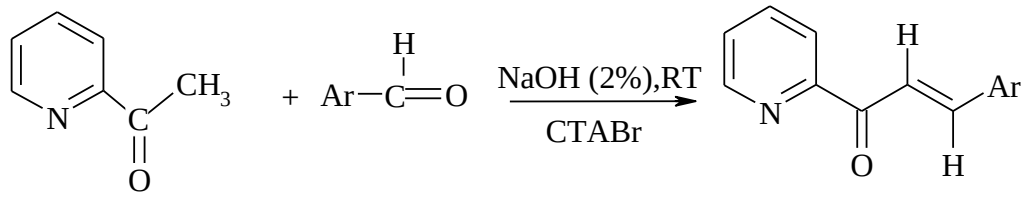
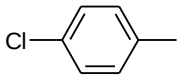
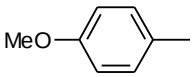
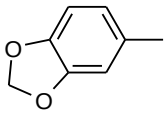
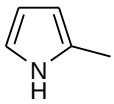
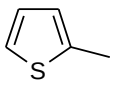
Product No.	Ar	t (min)	Yield (%)
4a		130	72
4b		100	76
4c		100	75
4d		45	83
4e		20	92
4f		30	84
4g		30	86
4h		100	74

Table 3 : Crossed-Aldol condensation of 2-acetylpyridine (**5**) with aromatic aldehydes. Synthesis of (2*E*)-3-aryl-1-(pyrid-2'-yl)prop-2-en-1-ones (**6a-e**).



(5) **(6)**

Product No.	Ar	t (min)	Yield (%)
6a		40	88
6b		45	92
6c		20	95
6d		80	77
6e		60	82

Experimental Section

All melting points reported are uncorrected. IR spectra were recorded using Perkin Elmer's Spectrum RXIFT-IR spectrophotometer (υ in cm^{-1}.) The NMR spectra were recorded on Bruker Avance DPX400 spectrometer, using $CDCl_3$ as solvent and TMS as internal standard (chemical shifts in δ values in ppm, *J* in Hz). Elemental analyses were preformed on Perkin Elmer 2400, series II microanalyzer.

General procedure:-

Hetarylmethyl ketones (**1, 3, 5**, 100 mmol), aromatic aldehydes (100 mmol) and cetyl trimethylammonium bromide (CTABr) (5.46g, 15 mmol) were added to an aqueous solution of NaOH (200 ml, 0.5 M). The mixture was vigorously stirred at 20 °C for the time reported in **Table 1, 2**, and **3**. The reaction was monitored by TLC and GC of dissolving sample of reaction mixture in CH_2Cl_2 during the reaction period. The solid products were filtered off, washed with water (3x25 ml), dried and crystallized from the proper solvent. The yields of the purified specimens are listed in **Table 1, 2**, and **3**.

The general procedure offers the following significant advantages over conventional procedures:

1- Improved reaction rates and increased yields through suppression of side reactions.
2- Clean, safe, and simple methodology.
3- Modifications of stereo-selectivity.
4- No need for expensive and hazard organic solvents.
5- Aqueous alkali metal hydroxides replace alkoxides.
6- Lower reaction temperatures and easier work-up.

(*2E*)-3-Phenyl-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2a): Pale yellow crystals from methanol; m.p. 136-138 °C; $C_{13}H_{11}NO$ (197.24); calcd.: C, 79.16; H, 5.62; N, 7.10; found: C, 79.07; H, 5.54; N, 7.13. IR: 1642 (C=O), 2851, 2918, 3026 (CH), 3268 (NH). ^{1}H-NMR: 6.38 (m, 1H), 7.07 (d, 1H), 7.18 (d, 1H), 7.32 (d, 1H, C_2-H; *J*=15.60), 7.51-7.65 (m, 5H), 7.85 (d, 1H, C_3-H; *J*=15.58), 10.32 (s, 1H).

(*2E*)-1-(1H-pyrrol-2`-yl)-3-(4`-tolyl)prop-2-en-1-one (2b): Pale yellow crystals from ethanol; m.p. 148-150 °C; $C_{14}H_{13}NO$ (211.26); calcd.: C, 79.60; H, 6.20; N, 6.63; found: C, 79.51; H, 6.13; N, 6.57. IR: 1642 (C=O), 2995 (CH), 3257 (NH). ^{1}H-NMR: 2.41(s, 3H), 6.37 (m, 1H), 7.12 (d, 1H), 7.23 (s, 1H), 7.27 (d, 2H), 7.36 (d, 1H, C_2-H, *J*=15.62), 7.56 (d, 2H), 7.85 (d, 1H, C_3-H; *J*=15.68), 10.34 (s, 1H).

(*2E*)-3-(2`-Chlorophenyl)-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2c): Pale green crystals from ethanol; m.p. 121-123 °C; $C_{13}H_{10}ClNO$ (231.68); calcd.: C, 67.40; H, 4.35; N, 6.05; found: C, 67.32; H, 4.30; N, 5.97. IR: 1645 (C=O), 2874, 2986 (CH), 3274 (NH). ^{1}H-NMR: 6.37 (m, 1H), 6.93 (d, 1H), 7.10 (s, 1H), 7.25 (s, 1H), 7.27 (d, 1H), 7.32 (d, 1H, C_2-H; *J*=15.69), 7.44 (d, 1H), 7.76 (t, 1H), 8.23 (d, 1H, C_3-H; *J*=15.69), 10.12 (s, 1H).

(*2E*)-3-(4`-Chlorophenyl)-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2d): Pale green crystals from ethanol; m.p. 154-156 °C; $C_{13}H_{10}ClNO$ (231.68); calcd.: C, 67.40; H, 4.35; N, 6.05; found: C, 67.32; H, 4.30; N, 5.97. IR: 1645 (C=O), 2874, 2980 (CH), 3274 (NH). ^{1}H-NMR: 6.36 (m, 1H), 7.10 (s, 1H), 7.16 (s, 1H), 7.36 (d, 1H, C_2-H; *J*=15.78), 7.38 (d, 2H), 7.56 (d, 2H), 7.78 (d, 1H, C_3-H; *J*=15.72), 10.34 (s, 1H). ^{13}C-NMR: 111.32 (CH), 117.11 (CH), 122.80 (CH), 124.40 (CH), 128.11 (CH), 129.63 (C), 131.38 (2xCH), 134.19 (C_2-H), 134.65 (C), 137.22 (C), 142.31 (C_3-H), 179.77 (C=O).

(*2E*)-3-(2`-Hydroxyphenyl)-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2e): Yellow crystals from methanol; m.p. 167-168 °C; $C_{13}H_{11}NO_2$ (213.23); calcd.: C, 73.23; H, 5.20; N, 6.57; found: C, 73.11; H, 5.14; N, 6.50. IR: 1632 (C=O), 2853, 2919 (CH), 3258 (NH), 3453 (OH). ^{1}H-NMR: 6.25 (m, 1H), 6.59 (m, 1H), 6.86 (d, 1H), 7.08 (m, 1H), 7.11 (d, 1H), 7.27 (d, 1H, C_2-H; *J*=15.58), 7.38 (d, 1H), 7.63 (m, 1H), 8.14 (d, 1H, C_3-H; *J*=15.66), 10.04 (s, 1H), 10.33 (s, 1H).

(*2E*)-3-(4`-Methoxyphenyl)-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2f): Pale yellow crystals from pet. ether 60-80 °C; m.p. 135-137 °C; $C_{14}H_{13}NO_2$ (227.26); calcd.: C, 73.99; H, 5.77; N, 6.16; found: C, 73.86; H, 5.71; N, 6.05. IR: 1641 (C=O), 2842, 2970 (CH), 3259 (NH). ^{1}H-NMR: 3.85 (s, 3H), 6.36 (m, 1H), 6.94 (d, 2H), 7.10 (d, 1H), 7.15 (d, 1H), 7.28 (d, 1H, C_2-H; *J*=15.62), 7.61 (d, 2H), 7.83 (d, 1H, C_3-H; *J*=15.59), 10.46 (s, 1H). ^{13}C-NMR: 57.86 (CH_3),

111.72 (CH), 115.31 (2xCH), 117.10 (CH), 118.94 (C), 121.63 (CH), 126.42 (C), 130.20 (2xCH), 131.73 (C_2-H), 143.84 (C_3-H), 163.21 (C), 180.90 (C=O).

(*2E*)-3-(3`,4`-Methylenedioxyphenyl)-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2g): Pale yellow crystals from ethanol; m.p. 140-142 °C; $C_{14}H_{11}NO_3$ (241.42); calcd.: C, 69.65; H, 4.59; N, 5.80; found: C, 69.57; H, 4.53; N, 5.71. IR: 1642 (C=O), 2830, 2962 (CH), 3223 (NH). ^{1}H-NMR: 6.06 (s, 2H), 6.37 (m, 1H), 6.85 (d, 1H), 7.09-7.15 (m, 3H), 7.17 (d, 1H), 7.23 (d, 1H, C_2-H; *J*=15.63), 7.97 (d, 1H, C_3-H; *J*=15.62), 10.49 (s, 1H). ^{13}C-NMR: 103.27 (CH_2), 105.65 (CH), 107.63 (CH), 109.85 (CH), 116.22 (CH), 119.48 (C_2-H), 124.23 (CH), 129.45 (C), 133.11 (C), 142.53 (C_3-H), 148.27 (C), 148.33 (C), 178.84 (C=O).

(*2E*)-3-(4`-N,N-dimethylaminophenyl)-1-(1H-pyrrol-2`-yl)prop-2-en-1-one (2h): Deep yellow crystals from methanol; m.p. 192-194 °C; $C_{15}H_{16}N_2O$ (240.30); calcd.: C, 74.98; H, 6.71; N, 11.58; found: C, 74.92; H, 6.65; N, 11.53. IR: 1638 (C=O), 2907 (CH), 3241 (NH). ^{1}H-NMR: 3.04 (s, 6H), 6.34 (m, 1H), 6.70 (d, 2H), 7.05 (m, 2H), 718 (d, 1H, C_2-H; *J*=15.58), 7.54 (d, 2H), 7.81 (d, 1H, C_3-H; *J*=15.53), 10.38 (s, 1H).

(*2E*)-1-(2`-Thienyl)-3-(4`-tolyl)prop-2-en-1-one (4a): Pale yellow crystals from ethanol; m.p. 112-114 °C; $C_{14}H_{12}OS$ (228.31); calcd.: C, 73.65; H, 5.30; S, 14.04; found: C, 73.58; H, 5.27. IR: 1589 (C=C), 1647 (C=O), 2918, 3083 (CH). ^{1}H-NMR: 2.40 (s, 3H), 7.18 (m, 1H), 7.21 (d, 2H), 7.39 (d, 1H, C_2-H; *J*=15.65), 7.55 (d, 2H), 7.67 (d, 1H), 7.83 (d, 1H, C_3-H; *J*=16.13), 7.87 (d, 1H). ^{13}C-NMR: 21.59 (CH_3), 120.59 (CH), 128.25 (CH), 128.56 (2xCH), 129.74 (2xCH), 131.71 (C_2-H), 131.97 (C), 133.78 (CH), 141.21 (C), 144.21 (C_3-H), 145.68 (C), 182.18 (C=O).

(*2E*)-3-(4`-Chlorophenyl)-1-(2`-thienyl)prop-2-en-1-one (4b): Yellow crystals from ethanol; m.p. 118-120 °C; $C_{13}H_9ClOS$ (248.73); calcd.: C, 62.78; H, 3.65; S, 12.89; found: C, 62.66; H, 3.62. IR: 1591 (C=C), 1645 (C=O), 3089 (CH). ^{1}H-NMR: 7.20 (m, 1H), 7.23 (d, 2H), 7.41 (d, 1H, C_2-H; *J*=15.65), 7.64 (d, 2H), 7.72 (d, 1H), 7.85 (d, 1H, C_3-H; *J*=15.79),

7.93 (d, 1H). ^{13}C-NMR: 121.96 (CH), 128.28 (CH), 129.22 (2xCH), 129.60 (2xCH), 131.90 (C_2-H), 133.13 (C), 134.11 (CH), 136.45 (C), 142.55 (C_3-H), 145.31 (C), 181.74 (C=O).

(*2E*)-3-(4`-BromoPhenyl)-1-(2`-thienyl)prop-2-en-1-one (4c): Pale yellow crystals from ethanol; m.p. 131-133 °C; $C_{13}H_9BrOS$ (293.18); calcd.: C, 53.26; H, 3.09; S, 10.94; found: C, 53.19; H, 3.05. IR: 1581 (C=C), 1649 (C=O), 2903-3085 (CH). ^{1}H-NMR: 7.21 (m, 1H), 7.48 (d, 1H, C_2-H; *J*=15.58), 7.52 (d, 2H), 7.57 (d, 2H), 7.72 (d, 1H), 7.80 (d, 1H, C_3-H; *J*=15.58), 7.89 (d, 1H). ^{13}C-NMR: 122.07 (CH), 124.84 (C), 128.29 (CH), 128.77 (2xCH), 131.93 (C_2-H), 132.18 (2xCH), 133.56 (C), 134.13 (CH), 142.62 (C_3-H), 145.30 (C), 181.72 (C=O).

(*2E*)-3-(4`-Methoxyphenyl)-1-(2`-thienyl)prop-2-en-1-one (4d): Yellow crystals from ethanol; m.p. 144 -146 °C; $C_{14}H_{12}O_2S$ (244.31); calcd.: C, 68.83; H, 4.95; S, 13.13; found: C, 68.77; H, 4.90. IR: 1590 (C=C), 1647 (C=O), 2838-3082 (CH). ^{1}H-NMR: 3.83 (s, 3H), 6.96 (d, 2H), 7.24 (m, 1H), 7.32 (d, 1H, C_2-H; *J*=15.62), 7.38 (d, 1H), 7.65 (d, 2H), 7.72 (d, 1H), 7.84 (d, 1H, C_3-H; *J*=15.62).

(*2E*)-3-(2`,4`-Dimethoxyphenyl)-1-(2`-thienyl)prop-2-en-1-one (4e): Yellow crystals from ethanol; m.p. 113-115 °C; $C_{15}H_{14}O_3S$ (274.34); calcd.: C, 65.67; H, 5.14; S, 11.69; found: C, 65.58; H, 5.07. IR: 1564 (C=C), 1635 (C=O), 2839-3090 (CH). ^{1}H-NMR: 3.85 (s, 3H), 3.90 (s, 3H), 6.52 (d, 2H), 7.15 (m, 1H), 7.45 (d, 1H, C_2-H; *J*=15.62), 7.55 (d, 1H), 7.63 (m, 1H), 7.83 (s, 1H), 8.07 (d, 1H, C_3-H; *J*=15.62).

(*2E*)-3-(3`,4`-Dimethoxyphenyl)-1-(2`-thienyl)prop-2-en-1-one (4f): Yellow crystals from ethanol; m.p. 119-121 °C; $C_{15}H_{14}O_3S$ (274.34); calcd.: C, 65.67; H, 5.14; S, 11.69; found: C, 65.61; H, 5.10. IR: 1578 (C=C), 1647 (C=O), 2847-3105 (CH). ^{1}H-NMR: 3.88 (s, 3H), 3.92 (s, 3H), 6.83 (d, 1H), 6.99 (s, 1H), 7.13 (d, 1H), 7.18 (m, 1H), 7.34 (d, 1H, C_2-H; *J*=15.64), 7.58 (d, 1H), 7.65 (d, 1H, C_3-H; *J*=15.64), 7.68 (d, 1H).

(*2E*)-3-(3`,4`-Methylenedioxyphenyl)-1-(2`-thienyl)prop-2-en-1-one (4g): Pale yellow crystals from ethanol; m.p. 117-119 °C; $C_{14}H_{10}O_3S$ (258.30); calcd.: C, 65.10; H, 3.90; S, 12.41; found: C, 64.96; H, 3.85. IR: 1587 (C=C), 1645 (C=O), 2906-3108 (CH). ^{1}H-NMR: 6.07 (s, 2H), 6.76 (d, 1H), 7.15 (d, 1H), 7.22 (m, 2H), 7.32 (d, 1H, C_2-H; *J*=15.62), 7.63 (d, 1H), 7.78 (d, 1H, C_3-H; *J*=15.69), 7.86 (d, 1H). ^{13}C-NMR: 101.63 (CH_2), 106.62 (CH), 108.67 (CH), 119.84 (CH), 125.34 (C_2-H), 128.19 (CH), 129.19 (C), 131.54 (CH), 133.66 (CH), 143.90 (C_3-H), 145.65 (C), 148.36 (C), 149.94 (C), 181.93 (C=O).

(*2E*)-1,3-Di-(2`-thienyl)prop-2-en-1-one (4h): Orange crystals from ethanol; m.p. 136-138 °C; $C_{11}H_8OS_2$ (220.32); calcd.: C, 59.97; H, 6.10; S, 29.11; found: C, 59.91; H, 6.07. IR: 1572 (C=C), 1639 (C=O), 3092 (CH). ^{1}H-NMR: 7.09 (d, 1H), 7.17 (m, 1H), 7.23 (d, 1H, C_2-H; *J*=15.72), 7.37 (s, 1H), 7.42 (m, 1H), 7.72 (m, 1H), 7.88 (s, 1H), 8.03 (d, 1H, C_3-H; *J*=15.63). ^{13}C-NMR: 120.34 (CH), 128.21 (CH), 128.35 (CH), 128.85 (C_2-H), 131.65(C_3-H), 132.20 (CH), 133.84 (CH), 136.46 (CH), 141.48 (C), 149.97 (C), 181.57 (C=O).

(*2E*)-3-(4`-Chlorophenyl)-1-(pyrid-2`-yl)prop-2-en-1-one (6a): Pale yelloe crystals from ethanol; m.p. 103-105 °C; $C_{14}H_{10}ClNO$ (243.69); calcd.: C, 69.00; H, 4.14; N, 5.75; found: C, 68.91; H, 4.09; N, 5.70. IR: 1568, 1606 (C=C, C=N), 1673 (C=O), 3020-3081 (CH). ^{1}H-NMR: 7.38 (d, 2H), 7.50 (m, 1H), 7.66 (d, 2H), 7.87 (d, 1H, C_2-H; *J*=16.15), 7.88 (d, 1H), 8.19 (d, 1H), 8.28 (d, 1H, C_3-H; *J*=16.15), 8.74 (d, 1H). ^{13}C-NMR: 121.29 (CH), 122.96 (CH), 127.10 (C_2-H), 129.14 (CH), 129.96 (CH), 133.63 (C), 136.42 (C), 137.08 (C_3-H), 143.19 (CH), 148.87 (CH), 154.00 (C), 189.27 (C=O).

(*2E*)-3-(4`-Methoxyphenyl)-1-(pyrid-2`-yl)prop-2-en-1-one (6b): Pale yellow crystals from ethanol; m.p. 120-122 °C; $C_{15}H_{13}NO_2$ (239.27); calcd.: C, 75.30; H, 5.48; N, 5.85; found: C, 75.22; H, 5.43; N, 5.76. IR: 1570, 1596 (C=C, C=N), 1666 (C=O), 2845-3052 (CH). ^{1}H-NMR: 3.86 (s, 3H), 6.94 (d, 2H), 7.48 (m, 1H), 7.69 (d, 2H), 7.86 (m, 1H), 7.92 (d, 1H, C_2-H; *J*=15.95), 8.17 (d, 1H, C_3-H; *J*=15.89), 8.19 (d, 1H), 8.74 (d, 1H). ^{13}C-NMR:

55.40 (CH_3), 114.32 (CH), 118.48 (CH), 122.87 (CH), 126.74 (CH), 126.94 (C_2-H), 127.94 (C), 130.67 (CH), 136.99 (C_3-H), 144.71 (CH), 148.79 (CH), 154.45 (C), 161.73 (C), 189.40 (C=O).

(*2E*)-3-(3`,4`-Methylenedioxyphenyl)-1-(pyrid-2`-yl))prop-2-en-1-one (6c): Yellowish green crystals from ethanol; m.p. 148-150 °C; $C_{15}H_{11}NO_3$ (253.26); calcd.: C, 71.14; H, 4.38; N, 5.53; found: C, 71.08; H, 4.34; N, 5.47. IR: 1583 (C=C, C=N), 1656 (C=O), 2905-3054 (CH). ^{1}H-NMR: 6.03 (s, 2H), 6.84 (d, 1H), 7.20 (d, 1H), 7.29 (s, 1H), 7.49 (m, 1H), 7.85 (d, 1H), 7.87 (d, 1H, C_2-H; *J*=15.88), 8.06 (d, 1H, C_3-H; *J*= 15.93), 8.17 (m, 1H), 8.74 (d, 1H). ^{13}C-NMR: 101.62 (CH_2), 107.08 (CH), 108.59 (CH), 111.85 (CH), 118.89 (CH), 122.88 (CH), 125.68 (CH), 126.80 (C_2-H), 129.72 (C), 137.00 (C_3-H), 144.68 (CH), 148.37 (C), 148.82 (CH), 149.98 (C), 154.35 (C), 189.33 (C=O).

(*2E*)-1-(Pyrid-2`-yl)-3-(1H-pyrrol-2`-yl)prop-2-en-1-one (6d): Orange crystals from ethanol; m.p. 124-126 °C; $C_{12}H_{10}N_2O$ (198.22); calcd.: C, 72.71; H, 5.08; N, 14.13; found: C, 72.64; H, 5.06 N, 14.08. IR: 1573 (C=C, C=N), 1654 (C=O), 2998-3112 (CH), 3304 (NH). ^{1}H-NMR: 6.34 (m, 1H), 6.73 (d, 1H), 7.00 (d, 1H), 7.26 (s, 1H), 7.48 (m, 1H), 7.85 (d, 1H, C_2-H; *J*=16.26), 7.88 (m, 1H), 8.19 (d, 1H, C_3-H; *J*=16.28), 8.71 (d, 1H), 8.88 (s, 1H). ^{13}C-NMR: 111.46 (CH), 113.94 (CH), 116.82 (CH), 122.79 (CH), 123.29 (CH), 126.60 (C_2-H), 134.14(CH), 136.28 (C), 137.05 (C_3-H), 148.57 (CH), 153.93 (C), 193.22 (C=O).

(*2E*) -1-(Pyrid-2`-yl)-3-(2`-thienyl)prop-2-en-1-one (6e): Pale green crystals from ethanol; m.p. 76-78 °C; $C_{12}H_9NOS$ (215.28); calcd.: C, 69.51; H, 4.21; N, 6.51; S, 14.90; found: C, 69.47; H, 4.19; N, 6.43. IR: 1582 (C=C, C=N), 1665 (C=O), 3010, 3061 (CH), 3316 (NH). ^{1}H-NMR: 7.08 (m, 1H), 7.45 (m, 2H), 7.49 (m, 1H), 7.86 (d, 1H, C_2-H; *J*=15.54), 8.07 (s, 2H), 8.17 (d, 1H, C_3-H; *J*=15.52), 8.74 (d, 1H). ^{13}C-NMR: 119.76 (CH), 122.81 (CH), 126.84 (C_2-H), 128.24 (CH), 129.18 (CH), 132.17 (CH), 136.97 (C_3-H), 137.22 (CH), 140.99 (C), 148.82 (CH), 155.32 (C), 193.21 (C=O).

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